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Preface

Radioactive Cs (^{137}Cs) is of special concern due to its high radioactivity and long half-life (30.2 years). Cesium has 40 isotopes, more than any other chemical element, with Cs-133 as the only naturally occurring and stable isotope. Testing of nuclear weapons and accidents at both military and civil nuclear facilities led to a large-scale release of radioactive Cs isotopes (Cs-134 and Cs-137) into the environment. ^{137}Cs is a ^{235}U fission product of high yield; ^{134}Cs is an activation product. Cs is a very volatile element and distributes fast after release. Hence, ^{134}Cs and ^{137}Cs contribute a major part to the radiation dose to population and workers. The ^{137}Cs release from the Chernobyl accident (8.5×10^{16} Bq) amounts to ca 10 % of the ^{137}Cs released by atmospheric nuclear weapons testing (1×10^{18} Bq) in the 1960s; the accident at Fukushima Daiichi Nuclear Power plant added another ca 1×10^{16} Bq. The Cs^+ ion is rather easily taken up by plants, animals, and humans due to its chemical similarity with K^+ causing internal exposure. On the other hand, the 661 keV gamma line of its short-lived ^{137m}Ba daughter causes external radiation exposure, both from gamma submersion and radiation of ground deposition.

The Fukushima Daiichi Nuclear Power Plant (FDNPP) accident led to the discharge of a large quantity of radioactivity into the environment. A total of nearly 9×10^{17} Bq of radionuclides was released in the environment and distributed into an area of 30 km around the power plant. From the FDNPP, radioactive Cs released into the air was estimated at 10^{16} Bq for ^{134}Cs and ^{137}Cs each; it was also reported that ca. 5×10^{15} Bq each of ^{134}Cs and ^{137}Cs were released into the ocean.

There is no doubt that ionizing radiation from radioactive elements can cause several adverse health effects on exposed populations. Several reports showed that the fate of ^{134}Cs and ^{137}Cs derived from the nuclear accident fallout and associated radiological hazards are mainly dependent on their movement in surface soils. Hence, the pathways from contaminated soils via plant uptake and into the human food chain must be understood. Radiocesium is quite soluble, but is strongly adsorbed on soil clays and resists leaching through soils. Cs also behaves similarly to potassium and sodium in terrestrial ecosystems. Potassium generally enters cells through passages and high-affinity transporters, and not all of them show the same permeability for Cs^+ and Na^+ or the same pH or Ca^{2+} regulation. Inward and outward

rectifying K^+ channels show a restricted permeability to Cs^+ and also are highly selective for K^+ over Na^+.

Plant uptake is the major pathway for the migration of radiocesium from the soil to the human diet. Two major regulatory mechanisms are involved in maintaining K^+ homeostasis, i.e., K^+ flow across the plasma membrane and utilization of vacuolar K^+ reserve. The role of K^+ in enzyme stimulation and protein biosynthesis is based on its high and steady concentration in the cytoplasm.

Even though there are some resemblances in the chemistry of K and Cs, there are numerous differences between the two alkali metals. For example, Cs has a much higher atomic weight and ionic radius than potassium. It is known that the K regime and pH affect the activity of K^+ transporters, and the interaction between K^+ and Na^+ uptake is also well known. Apart from that, it is well known that Ca^{2+} stimulates K^+ uptake in terrestrial plants. In fact, it has been proven that Ca^{2+} stimulates Cs uptake in terrestrial plants and that the K^+ regime affects Cs uptake rates in some grasses. However, voltage-insensitive monovalent-cation channels have been described recently in some plant cells which are permeable to a range of monovalent cations, including K^+, Cs^+, and Na^+ and which are inhibited by extracellular Ca^{2+}. High-affinity K^+ transporters are considered the main way by which Cs enters into plants. Molecular studies have exposed that some transporters demonstrated poor perception between K^+, Cs^+, and Na^+ which has been shown to reduce K^+ uptake through them. It seems also that these transporters could carry K^+ coupled with the entrance of protons, which has been generally considered the mechanism for the high-affinity K^+ transports in plants.

The soil to plant transfer factor (TF) is an important descriptive parameter that quantifies the uptake of radionuclides into plants and is a valuable tool to estimate dose to the population through ingestion. There are many studies regarding the TF values for ^{137}Cs performed for a specific site. They prove that a single TF for a certain RN – plant couple does not appropriately describe the complicated chemistry involved. In practice, several reasons are proposed to alter the degree of Cs accumulation in plants such as plant species/cultivars, Ca^{2+} levels, plant-associated microorganisms, and Cs location in the soil as well as atmospheric CO_2 levels. In an experiment with sagebrush, wheatgrass, or rabbitbrush it was clear that Cs-137 had a minimum activity in the leaves of the three vegetation species, but increased in the stem and roots; this may be an indication of rapid translocation of ^{137}Cs through the plant physiologic arrangement. There are some experimental confirmations which prove that soluble Cs is transferred directly from foliage surfaces into vegetal materials. It is observed that radiocesium is relocated within the trees by its radioactivity appearing oil leaves, which had developed in few weeks or months after contamination (in the case of the Chernobyl accident).

Plants generally accumulate radionuclides by two principal mechanisms: (1) direct deposition from the atmosphere and (2) root uptake. Reducing the amount of Cs-polluted soils and water is an urgent task following an accident. It is necessary now to develop a technique for solubilizing Cs from soil matrix. River waters, ponds, and sea as well riverbanks are also polluted by Cs. Radioactive Cs has been reported to tightly bind to fine soil particles in water and sediment at narrow depth.

Radioactive Cs is exceptionally tough to remove from the sediment particles and needs to be treated with extremely strong acid for re-solubilizing. Many native bioremediation methods have employed on terrestrial plants to reduce radionuclides from the environment after the Chernobyl accident in 1986. The accumulation of radioactive Cs into terrestrial plants such as tea, rice, sunflower, and tomato has been reported. In a study, it was reported that *Helianthus Annuus* absorbed up to 150 μg of Cs in 100 h, whereas a *Vetiveria zizanoides* absorbed 61 % of ^{137}Cs in 168 h from Chernobyl accident radio-polluted water. It is also reported that brown alga (*Laminaria digitata*) adsorbed higher than 80 % ^{134}Cs when the membrane was phosphorylated artificially under high pH conditions.

In existing exposure situations on Cs-contaminated sites, there is a need to remove radioactive Cs from the human habitat. One promising possibility is phytoremediation. However, the accumulation capacity as well as the absorption and the accumulation mechanism of Cs in plants must be studied to choose appropriate plants. In terrestrial plants, Cs is mainly taken up by roots and then translocated to the leaf and stem through the xylem vessel. In rice and sunflower, Cs is mainly distributed to young leaves. In *Arabidopsis thaliana*, Cs mostly accumulates in rosette leaves. Several physiological experiments have demonstrated that the mechanism of Cs absorption via roots is similar to that of K. Through aerial deposition, Cs accumulation in the leaves may disturb the basic physiological functions of plants. The first observed reaction in the Cs-treated plants is decreased stomatal opening. The control function of stomata in respect to photosynthetic CO_2 assimilation and transpiration was modified by the presence of Cs. In an experiment, it was noticed that decreased stomatal opening limits transpiration and uptake of water by roots, but photosynthetic CO_2 incorporation did not change during short-term contact to CsCl. Stomatal closure in the presence of Cs may be a result of a reduction in osmotic potential (due to Cs accumulation in vacuoles), the role of Ca^{2+} and K^+ in the regulation of stomatal opening, or disturbance of the signal transduction pathway that is already demonstrated by several researchers. In most cases, possibly Cs inhibits the channels responsible for K^+ transport into guard cells, which influence reduced stomatal opening.

The most noteworthy features of this book are related to how cesium is released in the environment and its translocation from soil to plants and lastly to animals and human. Chapters 1–5 deal with the bioavailability of cesium in the environment and its translocation via soil to plants. Chapters 6–14 focus on the effective half-life of cesium in plants and how different cultivars are responding in the accumulation of cesium; some chapters focus on cesium impact on single cell to higher plants and also on remediation measures and transfer factors. However, the information compiled in the volume will bring in-depth knowledge of cesium uptake and translocation and its toxicity in plants after the Chernobyl and Fukushima accidents.

Dr. Dharmendra K. Gupta and Prof. Clemens Walther personally thank the authors for contributing their valuable time, knowledge, and keenness to bring this book into its present shape.

Hannover, Germany

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Contents

Distribution of Caesium in Soil and its Uptake by Plants

Sergiy Dubchak

Abstract The speciation of radiocaesium forms in the soil horizons that determine its further behaviour in the environment was considered. The present-day equilibrium in radiocaesium migration processes demonstrates that 70–90 % of the radionuclide was retained in the fixed form in upper 5–20 cm soil layer regardless of the type of soil and nature of contamination. In the forest ecosystems the litter layer continues to be a major accumulator of radiocaesium and contains 40–80 % of ^{137}Cs being the main biochemical barrier to its vertical and horizontal migration. It was shown that radiocaesium can be easily absorbed by plant roots from soil solution and translocated to the above-ground plant biomass. The main environmental factors responsible for the variability in radiocaesium uptake by higher plants from soil are described. The uptake of radiocaesuim by plant species of herbal-shrubby storey in forest ecosystems was considered. It was shown that various plant species reveal the differences in ^{137}Cs uptake via significantly different rates of the radionuclide accumulation during their vegetation period.

Keywords Radiocaesium speciation • Soil type • Migration processes • Plant species • Forest ecosystems • Transfer coefficients

1 Introduction

The chemical element caesium was discovered in a Bavarian mineral spring by Bunsen and Kirchhoff in 1860 (Avery 1996). It is a Group I alkali metal present in solution as the free hydrated monovalent cation Cs^+ with little or no tendency to form soluble complexes in the soil with chemical properties similar to potassium and rubidium. ^{133}Cs is the only stable caesium isotope, however it exists in various isotopic forms with atomic masses in the range from ^{112}Cs to ^{151}Cs (Audi et al. 2003). Altogether, caesium has 40 isotopes, more than any other chemical element.

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The further fast development of nuclear industry resulted in large scale release of Cs radioactive isotopes into the environment. In the process of various uranium, plutonium and thorium isotopes fission or as the result of ^{133}Cs or ^{136}Ba neutron bombardment, seven radiologically significant radioisotopes of Cs are produced (Nichols and Hunt 1998). Among them ^{137}Cs and ^{134}Cs have a particular environmental importance. The ^{137}Cs (half-life $T_{½}=30.17$ years, β and γ emitter; main emission γ-line is $E_{\gamma}=0.662$ MeV with quantum yield $k_{\gamma}=0.892$; maximum β-energy is $E_{\beta max}=1.173$ MeV; daughter product ^{137}Ba) was discovered by in the late 1930s by Seaborg and Melhase. It is an ordinary fission-product produced in nuclear reactors and aboveground nuclear weapons explosions with ca. 3–7 % fission yield (Unterweger 2002; Audi et al. 2003).

The ^{134}Cs ($T_{½}=2.06$ years, β and γ emitter; main emission γ-lines are $E_{\gamma 1}=0.605$ MeV with $k_{\gamma 1}=0.975$ and $E_{\gamma 2}=0.796$ MeV with $k_{\gamma 2}=0.851$; β-energy is $E_{\beta max}=2.059$ MeV; daughter product ^{134}Ba) is produced within the nuclear reactors only, not in the process of uranium or plutonium fission, but as a result of neutron activation (Nichols and Hunt 1998; Unterweger 2002). Both these Cs radioisotopes are released into the environment during intentional and unintentional discharges from nuclear installations. They corresponds a significant environmental risk due to fast incorporation into biological systems and further efficient transfer through the food chains, emission of harmful β and γ radiation during the decay and (in case of ^{137}Cs) relatively long half-life (Bell et al. 1988). The study of caesium isotopes behavior in the soil and their radioecological significance received further attention due to the large-scale release of radioactivity occurred as a result of Chernobyl catastrophe in former USSR in 1986 and the most recent accident at Fukushima NPP in 2011. Nowadays ^{137}Cs had become the radionuclide that mainly contributes to the overall level of the external and internal exposure at contaminated areas, and thus determines the long term radiological impact of Chornobyl and Fukushima accidents.

1.1 Caesium in the Environment and its Bioavailability in Soils

The naturally occurring stable ^{133}Cs, whose main natural source is pollucite (an aluminosilicate mineral), is the rarest of alkali metals with petty economic value. Up to date, no substantial biological role of caesium has been found (Avery et al. 1992), although its trace quantities occur in most living organisms (Komarov and Bennett 1983). Concentrations of natural caesium in different environments variate essentially. According to Coughtrey and Thorne (1983), the average concentration of ^{133}Cs in different soil types varies between 0.3 and 25.7 mg kg^{-1} dry soil weight. This corresponds to micromolar Cs^{+} concentrations in soil solutions.

The caesium can be easily absorbed by plant root system from solution and translocated to the above-ground plant parts (Zhu and Smolders 2000). The role of

stable ^{133}Cs in plant nutrition is not studied completely, but excessive concentration of this element (above 200 μM) can be toxic to plants (Marschner 1995). The toxicity appearances caused by unnaturally high external Cs^+ concentrations could include shoot and root tissues apobiosis (Kordan 1987). Due to the absence of any substantial biological role of stable caesium and to the fact that it is not toxic in natural concentrations for plants and soil organisms, this element received minor attention until the middle of the twentieth century, when the nuclear power was invented. Following the surface deposition, Cs^+ (the prevalent species of caesium in aqueous solution) interacts intensively with many types of soil since it has little tendency to hydration and higher polarizability than ions of smaller size like K^+ (Shainberg and Kemper 1997). A considerable part of Cs^+ is fixed efficiently with clay minerals and becomes unavailable for biota (Kirk and Staunton 1989). The radiocaesium binding in soils significantly depends on the concentration of inorganic minerals relative to organic matter. It is also sensitive to density of plant roots, degree of cation saturation (e.g. NH_4^+ and K^+), as well as to stable Cs content in soil (Shenber and Eriksson 1993).

As a monovalent cation, radiocaesium is generally assumed to exhibit cation-exchange characteristics and its behaviour is supposed to be partly pH-dependent. The radiocaesium mobility and solubility in soils and its bioavailability increase with soil pH reduction, since Cs^+ ions coherent to clay particles may be replaced by hydrogen ions H^+ (Bakken and Olsen 1990). Thus, Heinrich (1992) demonstrated that the acidic nature of soils with a high concentration of humus resulted to increased uptake of Cs^+ by mushrooms. From the previous researches on the environmental fate of radiocaesium, a general conclusion emerged that biogeochemical behaviour of Cs is rather similar to that of alkali metal potassium. This fact implies that in terrestrial ecosystems Cs^+ demonstrates low mobility in the soil profile (Rosen et al. 1999), but its movability in biological systems is relatively high (Carter 1988). Thus, in semi-natural and tillable soils with high content of clay materials, radiocaesium demonstrates low bioavailability as well as a minor vertical migration rate (Rosen et al. 1999). In forest soils poor with nutrients, the rate of ^{137}Cs vertical migration is also low, but the bioavailability is often high, especially for fungal species (Yoshida and Muramatsu 1994; Huang et al. 2016).

Due to low rate of vertical migration (usually several millimeters per year), the radiocaesium isotopes deposited from fallouts have been mainly retained in the upper 10–15 cm horizon of soil (Mahara 1993). The distribution pattern of radiocaesium isotopes differs from that of natural stable ^{133}Cs which is distributed much more uniformly through different soil profiles (Bakken and Olsen 1990). Such localization of radiocaesium is problematic, since the upper soil layers are extensively explored by plant roots, fungal mycelium and soil microorganisms (Thiry and Myttenaere 1993). Due to the ubiquity in the environment and the interaction with plant roots and soil, the fungi and other microorganisms may have a significant influence on the biogeochemical cycling of radiocaesium. Thus, the microbe-associated ^{137}Cs represents between 1 and 56 % of its total content in upper organic layer of forest soils (Bruckmann and Wolters 1994).

Table 1 Biological and environmental half-lives of ^{137}Cs (Adopted from Conkic and Ivo (1990); Daroczy et al. (1994))

	Organism/Environment	Half-life
Biological	Moss	4–5 years
	Lichen	5–8 years
	Grass	14 days
	Plant leaf surface	14 days
	Hen	1–5 days
	Cow	3 days
	Fish	70–300 days
	Child	57 days
	Woman	84 days
	Man	105 days
Environmental	Lake	2-7 years
	River	1–4 years
	Airborne dust	270 days

Being involved in the environmental food-chains, radiocaesium may be accumulated in higher organisms including humans that results in occurrence of sublethal and lethal ionization effects at the molecular level (Ghosh et al. 1993). The timescale of Cs retention in higher organisms and environmental objects (biological and environmental half-lives) demonstrates that accumulation of this element does not last to indefinite extend (Table 1). In case of single intake of certain radiocaesium activity to living organisms or ecosystems, the radionuclide is eliminated from these objects for the definite period of time. However in case of permanent intake of certain radiocaesium activity, a steady internal concentration of the radionuclide in living organisms and ecosystems is finally reached.

1.2 Radiocaesium Migration in Soil After Chornobyl Catastrophe

The significant release of radiocaesium isotopes to the environment occurred during the accident at the Unit 4 (high-power pressure-tube reactor RBMK-1000, electrical power 1000 MW, thermal power 3200 MW) of Chornobyl nuclear power plant that took place on April 26, 1986. During the accident, about 3.8×10^{16} Bq of ^{137}Cs, 1.9×10^{16} Bq of ^{134}Cs and 0.9×10^{16} Bq of short-lived ^{136}Cs ($T_{½} = 13.1$ days) were released into the environment. As a result, nearly 340,000 km^2 were contaminated with released radionuclides to the levels exceeding 37,000 Bq m^{-2} (the threshold for taking countermeasures in former USSR). Within these areas, ^{137}Cs surface contamination from Chornobyl fallouts exceeded the mentioned 37 kBq m^{-2} threshold nearly at 160,000 km^2 (Morrey et al. 1988).

Nearly half of all released radionuclides had been deposited within 30 km Chornobyl zone, namely 1.0×10^{16} Bq of ^{137}Cs, 0.5×10^{16} Bq of ^{134}Cs, 0.6×10^{16} Bq of ^{90}Sr and 3.3×10^{14} Bq of transuranium elements, i.e. U, Am, Pu, Np and Cm iso-

topes. Nowadays, within Chornobyl zone the inventories of ^{137}Cs in the soil reach more than 37 MBq m^{-2} (Kholosha et al. 1999).

After several months since the accident and complete decay of released short-lived radionuclides, the deposited ^{137}Cs and ^{134}Cs had become the main sources of the population exposure. Initially their contribution constituted only about 10 % (Israel and Vakulovskij 1990). With the time, the importance of ^{137}Cs relative to ^{134}Cs increases due to its longer half-life and practically complete decay of ^{134}Cs. Thirty years after the Chornobyl accident, about 50 % of ^{137}Cs from its initial release is still remained in the environment.

There were two main types of Chornobyl fallouts both containing radiocaesium isotopes. The fuel component of fallouts is typical for the near 10–30 km zone and represents tiny particles of dispersed nuclear fuel and its conglomerates including graphite and construction materials. The condensation component dominates at longer distances from the reactor. It represents radionuclides condensed on surface of various metals, non-metals, carbon compounds and dust particles (Rybalka et al. 2001). The principal part of ^{134,137}Cs (about 60–75 %) was deposited within the 30 km Chornobyl zone was initially in the structure of fuel particles. Outside of Chornobyl zone, the mentioned radiocaesium isotopes were deposited mostly in the condensation form of fallouts. The large areas within Chornobyl zone are characterized with superposition of condensation and fuel components of fallouts. The radiocaesium mobility in such conditions depends mainly on the ratio of mentioned forms of fallout. The dynamics of radiocaesium mobile form concentration in this area depends on physical-chemical properties of fuel particles and characteristics of soil (Kashparov 2001).

During the first years after Chornobyl accident, the content of radiocaesium exchangeable forms in soils on the territories contaminated with the condensation component of fallout was higher than that in the same soils contaminated with the fuel component (Ivanov et al. 1997). Later the chemical destruction of fuel particles and leaching of ^{137}Cs from particles matrix due to interaction with the soil solution resulted in tens of times increase of the radiocaesium mobile (especially water-soluble) fraction in areas located within Chornobyl zone. The behavior of ^{137}Cs represented by the condensation component of fallout in the soil is very similar to behavior of this radionuclide deposited earlier due to the global fallout. The concentration of radiocaesium mobile forms in the soil is predetermined mainly by sorption-desorption mechanisms by solid phase of the soil and decreases with time depending on specific landscape-geochemical conditions, acidity and soil solution, granulometric and mineralogical composition of soil, water regime of soil, etc. (Isaksson et al. 2001).

Radiocaesium has an intermediate place in the mobility chain of radioactive elements in the soil formed according to this property: ^{90}Sr > ^{106}Ru > ^{137}Cs > ^{144}Se > ^{129}I > ^{239}Pu. Similarly to other radionuclides, ^{137}Cs absorbed in the soil can be found in different physical and chemical forms that differ in their mobility and thus in availability of uptake by the root system of plants. The following basic forms of radiocaesium in the soil are distinguished: water soluble, exchangeable, movable (acid-soluble) and fixed (Ivanov 2004).

The forms of radiocaesium in the soil determine its further behavior and speciation in the vertical horizons, namely the migration in the soil profile. In particular, the water soluble and exchangeable forms of radiocaesium determine its uptake by plants, fungi and microorganisms. The transfer of radiocaesium changes its distribution in the plant root layer of soil that affects the radionuclide availability by root systems of different plant species. Due to desorption process, the water soluble and exchangeable forms of radiocaesium are transferred to the soil moisture and surface water, thus being dissolved and involved in the processes of migration (Lee and Lee 2000). These forms also take part in the processes of the radionuclide sorption and desorption by soil-absorbing complex. The significant part of ^{137}Cs in the soil migrates mainly in the structure of water-soluble organic compounds– the metabolic products of the soil flora and fauna. The non-exchangeable forms of radiocaesium can migrate in the soil only in the structure of solid particles to which they are included (Ivanov 2004). The numerous studies demonstrated that the key factors of radiocaesium migration in the soil profile are the diffusion processes, biogenic migration and lessivage (Pronevych 2014).

Under the influence of many environmental factors, the radiocaesium is being leached out from the fuel particles, transformed in mobile forms and enters the soil solution. The migration rate of radiocaesium soluble forms to the lower soil horizons is strongly dependent on internal water runoff in the soil, especially in Ukrainian Polissya region, that is characterized with 600 mm annual rainfall and light granulometric composition of soils (Pronevych 2014).

The granulometric composition of soil significantly affects the strength of radiocaesium fixation. Thus, the heavy soils absorb ^{137}Cs more efficiently than light-textured ones. The strength of ^{137}Cs fixation is increased with the decrease of soil fractions size (Absalom et al. 1995). The fine fractions of natural clay and silt (usually with the size about 0.001 mm) have the highest absorptive capacity and thus reduce the radiocaesium mobility in soil and fix it most firmly as compared to the coarser soil fractions.

The absorbed ^{137}Cs in contrast to the ^{90}Sr is more firmly sorbed by various soil minerals. The strongest fixation of radiocaesium was found for the minerals of montmorillonite group: askanite, gumbrine, and glimmer and hydro-glimmer, phlogopite, hydro-phlogopite and vermiculite. Unbound ^{137}Cs can be absorbed by the soil via its sorption on the surface of the multi-layer minerals (Anenkov 1991). During this process, the crystal lattice of the minerals is changed and slightly expanded, so the radionuclide can be absorbed and included in the crystalline structure. Being included in the crystal lattice, ^{137}Cs can substitute to some extent potassium, ammonium and stable caesium. With the time, the physical-chemical forms of ^{137}Cs are changed due to so-called aging process of the radionuclide in the soil, i.e. resulting from complex of chemical reactions associated with the inclusion of radiocaesium into the crystal lattice of clay minerals, ion exchange and chemical co-precipitation (Pronevych 2014). The kinetics of radiocaesium sorption in the soil includes two basic processes: (1) ion exchange that is limited by external diffusion and (2) irreversible absorption (fixation) limited by a direct interaction of radiocaesium with the soil matter. Diffusion is considered to be the most important process

that determines radionuclide migration after Chornobyl accident. Thus, 60–99 % of ^{137}Cs is estimated to be distributed in soil due to diffusion processes (Loshchilov et al. 1990).

In general, the soddy-podzolic, podzolic, sandy, peat, and peat-bog soils that mainly represent contaminated regions of Ukraine, Belarus and Russia provide favorable conditions for radiocaesium migration. These soils typically have a low humus content, base saturation, pH and exchange capacity. Their mineralogical composition was characterized with low amount of clay minerals and weather able materials (Shcheglov et al. 2001). These features result in low soil capacity for both exchangeable and irreversible sorption of ^{137}Cs except for the forest litter with a thin (1–2 cm) mineral level beneath it.

The migration rate of radiocaesium increases from the soddy-podzolic dry sandy soils to wet meadow peat soils, where it has reached a depth of 27–30 cm due to the high soil humidity and lack of sorption minerals in the soil structure (Pronevych 2014). All upper mineral layers of soil are characterized with the increase of ^{137}Cs content over a certain period of time only. This period was longer for the soil layers located deeper. Thus, the peak of ^{137}Cs concentration for 0–2 cm layer in studied soils accounts for 1997, for 2–4 cm layer this peak was observed in 1999, and for 4–6 cm layer in 2001 (Karachov 2006). After reaching the maximum concentration, the slow reduction of ^{137}Cs activity in corresponding soil layers was occurred, predominantly due to two processes—migration in deeper soil layers and physical decay of the radionuclide. Nowadays, the main part of radiocaesium was located within the 5–10 cm horizon in the most of soil types within contaminated regions (Konopleva et al. 2009).

The radionuclide contamination of the soil after the Chornobyl accident has a distinct spatial heterogeneity, so the specific activity of radionuclides in the soil can vary at a short distance by two to three orders of magnitude (Kashparov 2001). As radiocaesium was included in the biological turnover, the heterogeneity of soils and forest litter horizontal contamination was being reduced. The analysis of radiocaesium distribution in the soil demonstrated that its further migration depends on the environmental landscape factors such as surface relief, vegetation cover, and presence of river valleys, watersheds, sandy hillocks and forests (Shcheglov et al. 2001).

Nowadays the stabilization of radioecological situation and the dynamic equilibrium in migration processes could indicate that the current state of affairs will not be changed for the next few decades. This means that 70–90 % of radiocaesium will be retained in the upper 5–20 cm soil layer. Regardless of the type of soil and nature of contamination (fuel or condensation component of fallout), nearly 85–98 % of ^{137}Cs in the upper soil layer was found in the fixed form. Presently, the different soil types have less than 1 % of ^{137}Cs in water-soluble form (Anenkov et al. 2004). According to Pronevych (2013), in the recent decades the ratios of radiocaesium different forms in the soils have not been changed significantly. Depending on the soil type, the amount of ^{137}Cs water-soluble and exchangeable forms in the upper (0–20 cm) soil layer varies between 0.3 and 0.5 % and between 2.11 and 11.70 % correspondingly (Table 2, according to Pronevych (2013)).

Table 2 Fraction of ^{137}Cs organic-mineral solutions in the upper soil layer (% from radionuclide total activity, as of 1992 and 2006)[a]

Type of soil	Physical-chemical forms of ^{137}Cs in soil							
	Water-soluble		Exchangeable		Movable (acid-soluble)		Fixed	
	1992	2006	1992	2006	1992	2006	1992	2006
Soddy-podzolic natural	0.40	0.62	11.7	5.41	3.10	4.08	84.8	89.89
Soddy-podzolic gleyed	0.42	0.47	6.22	10.86	2.65	7.74	90.71	80.93
Peat lowland	0.29	0.38	2.11	1.12	5.21	2.13	92.39	96.37
Soddy-podzolic, cultivated grasslands	0.48	0.56	8.71	5.17	3.64	3.30	87.17	90.97
Peat lowland, cultivated grasslands	0.35	0.42	3.76	3.68	2.23	2.46	93.66	93.44

[a]Depth of soil layer for natural grasslands is 0–10 cm, for cultivated grasslands 0–20 cm

1.3 Migration of ^{137}Cs in Forest Ecosystems

The forests ecosystems of Ukrainian Polissya region firmly fix a significant amount of radiocaesium, thereby protecting surface and ground waters and adjacent landscapes against radioactive contamination. These forests are mainly represented by pure or mixed pine plantations that form rather vigorous layers of litter reaching 10–15 cm in thickness (Shcheglov et al. 2001; Delvaux et al. 2001). 30 years after the Chornobyl accident namely the forest litter continues to be a major accumulator of radiocaesium. In the mentioned pine forest stands the forest litter contains 40–80 % of ^{137}Cs being the main biochemical barrier to vertical and horizontal migration of the radionuclide in forest landscapes. The ground biomass of the forest stands (without the forest litter) retains only 2.8 % of ^{137}Cs. According to Shcheglov et al. (2001), each year nearly 1–2 % of radiocaesium activity is washed with vertical internal soil drainage from the forest litter to mineral soil horizons. In contrast to grassland soils, the overall pattern of ^{137}Cs vertical distribution in forest soils represents the exponential decrease of its activity with the depth (Karachov 2006). The mobility of radiocaesium in the forest litter and its vertical migration rate in the soil are strongly affected by the vital activity of soil fauna. As a result of such activity, mixing the soil horizons, decomposition of tree wastes and forest litter, and biochemical transformation of the litter usually take place. All these processes intensify leaching of radiocaesium from the upper organic soil layer to the mineral fraction of soil and increase the content of radionuclide mobile fractions (Baryakhtar 1996).

It is likely that the significant part of ^{137}Cs in soils is associated with soil organic matter, as evidenced by the high correlation coefficient (0.95) between the specific activity of the radionuclide in organic matter and amount of organics in different soil layers. Thus it was found that the concentration and composition of finely dispersed fraction and organic matter are the main factors that determine the mobility of radiocaesium in the upper layers of soil (Koarashi et al. 2012).

In damp and wet forest conditions, the accumulation of ^{137}Cs is several times higher than in dry and fresh ones. The highest increase of ^{137}Cs migration intensity and plant uptake with the gain of humidity was observed for the peat-bog soil. This may indicate a weak character of radiocaesium binding in organogenic soils. Thus, the organogenic hydromorphic soils of Polissya region are considered as the most critical in terms of migration mobility and bioavailability of ^{137}Cs (Garger et al. 2002). These soils are characterized with very small content of clay minerals which mainly sorb radiocaesium, so the significant fraction of this radionuclide is remained in water-soluble and exchangeable forms. This fact determines the considerable intensity of ^{137}Cs vertical redistribution in hydromorphic organogenic soils.

1.4 Radiocaesium in Soil After Fukushima Accident

The most recent heavy accident with radiocaesium isotopes release occurred on March 11, 2011 at the Fukushima Daiichi nuclear power plant in Japan. It caused a release nearly 3×10^{16} Bq of ^{137}Cs and 5×10^{16} Bq of ^{134}Cs with a subsequent contamination of soils in terrestrial ecosystems. The Fukushima accident represents a major potential long-term source for mobile forms of ^{137}Cs and ^{134}Cs that had deposited on the ground (Hirose 2012).

Five years after the accident, when more than 80 % of ^{134}Cs has decayed, ^{137}Cs became to contribute principally to the soil and vegetation contamination as well as to the population exposure in radioactively contaminated areas in Japan. However the mobility of radiocaesium in Fukushima forest soils was differed from that in Chornobyl grassland or forest soils derived from loess and glacial deposits. Thus, even 30 years after the Chornobyl accident, the larger proportions of ^{137}Cs (as compared to those in Fukushima forests) are retained within thick organic layers of the forest litter. According to Nakanishi et al. (2014), the major part of ^{137}Cs and ^{134}Cs in studied forest ecosystem around Fukushima NPP were leached from the organic layer of the litter and moved into the mineral soil within several years after the accident. Such a rapid loss of radiocaesium isotopes from the organic layer was considered to be caused by rapid wash-out of deposited ^{137}Cs and ^{134}Cs from a thin organic layer of the local forests and rapid decomposition of forest litter, which can mobilize radionuclides associated with organic matter. The smaller radiocaesium retention capacities of forest soils in Fukushima could be explained by rich organic matter (Koarashi et al. 2012) formed due to large amounts of precipitation and volcanic ash deposition (Shoji et al. 1985). Currently the most of radiocaesium isotopes (59–73 %) in contaminated Japanese forest ecosystems are accumulated in the upper soil layer (0–5 cm). The topsoil prevented migration of ^{137}Cs and ^{134}Cs, and only 2 % of these isotopes in the leachate from litter and humus layer penetrated below a 10 cm depth (Nakao et al. 2014).

1.5 Radiocaesium Uptake by Plants

The study of the uptake and accumulation of radiocaesium by plants only gained a real importance after the Chornobyl catastrophe. Shortly after this accident, the levels of plant contamination by radionuclides had become to be predetermined almost by the root uptake processes depending on the soil type and thus the biological characteristics of plants (Anenkov 1991). This resulted in a growing number of studies on ^{137}Cs transfer from soil to plants in the last decades (Dupré de Boulois et al. 2008). The principal route of radiocaesium entry into biological food chain in terrestrial ecosystems is the soil-to-plant pathway. Nowadays the ^{137}Cs absorption by plants and its accumulation, therefore, represents the main source of human exposure to this radionuclide.

The tendency of radiocaesium to accumulate in food chains via plant uptake from soil had been further highlighted by several reports (Bergeijk et al. 1992; White and Broadley 2000; Zhu and Smolders 2000). Moreover, the studies of radiocaesium transfer to various fungal species had also revealed that its accumulation was higher in relation to potassium, the biologically-significant alkali metal, which is chemically similar to caesium (Horyna and Randa 1988; Bakken and Olsen 1990). The further laboratory researches have demonstrated that Cs^+ is accumulated via active K^+ transport systems in yeasts (Perkins and Gadd 1993). Also, the common mechanisms for Cs^+ and K^+ transport in *Arabidobsis thaliana* plants were revealed (Sheahan et al. 1993; Hampton et al. 2004; Kanter et al. 2010). However, the mechanisms by which radiocaesium is taken up by plant roots are not completely understood. Evidently, at low potassium concentrations Cs^+ was absorbed by the K^+ uptake system of the root since the caesium uptake was strongly suppressed by increased potassium content (Zhu and Smolders 2000). The further physiological studies confirmed that Cs^+ and K^+ competed for influx to plant roots. Consequently, it was suggested that the influx of above mentioned cations to root cells is stipulated by the same molecular mechanisms (White and Broadley 2000). Such similarity of root uptake mechanisms for Cs^+ and K^+ implies an important hazard in terrestrial ecosystems for radiocaesium contamination of the aboveground vegetation. However, there are some other processes influencing the radiocaesium transfer to plants. Thus, Avery (1996) distinguished three main environmental factors responsible for the variability in Cs^+ uptake by higher plants:

1. Soil type: The uptake of radiocaesium by plants is higher in soils characterized with a high content of organic matter. Thus, tenfold augmentation of Cs^+ transfer to plants was observed with the increase of organic matter content in soil from 5 to 50 % (Bergeijk et al. 1992).
2. Seasonal changes: plant speciation and plant ageing. Seasonal fluctuations in different climatic conditions may contribute to 3–50-fold variation in radiocaesium uptake by the same plant species. Also, a distinct reduction in Cs^+ transport was observed with ageing of pants (Noordijck et al. 1992).
3. Ion content of soil: The presence of increased NH_4^+ and particularly K^+ content in soil can result in appreciable reduction of radiocaesium uptake by ground

vegetation. Thus, the addition of potassium fertilizers to soils decreases soil-plant Cs^+ transfer factors by nearly 60 % (Pietrzak-Flis et al. 1994), accordingly it was suggested that the Cs^+ uptake and translocation by plants is more strongly related to Cs/K ratio of soils rather than to total Cs^+ content in soil.

The radiocaesium accumulation by different plant species is strongly dependent on peculiarities of plant mineral nutrition, duration of the vegetation period, the distribution and localization of root system in the soil and other biological characteristics of plants. Thus, the interspecies differences in accumulation of radiocaesium via root uptake can reach 10–60 times (Prister 2007).

The essential peculiarity of radiocaesium migration in soil-plant system is the extremely high mobility of this radionuclide in areas with light (according to granulometric type) sandy and sabulous soils of podzolic and bog type. The excessive soil acidity of Polissya region, wide spreading of peat swamps, low content of clay minerals, hydromorphism and the low absorbing capacity of the soil solid phase result in very high ^{137}Cs transfer factors from soil to plants. Depending on soil and meteorological factors and biological characteristics of plants, the radiocaesium transfer factors can be changed from 20 to 30 times for the same plant species (Gudkov and Vinnychuk 2003). The transfer of radiocaesium to plants also depends on the agrochemical properties of soils. Thus, the previous studies (Grytsyuk 2001; Anenkov et al. 2004) demonstrated that regardless of the plant species and the year after the accident, the transfer factors of ^{137}Cs from different soil types to plants are reduced in the following order: peatbog soil > soddy-podzolic soil > grey forest soil > black soil.

The forests and natural grasslands are the most critical landscapes within the territory contaminated after the Chornobyl disaster. They are characterized with substantially higher transfer factor of ^{137}Cs and other radionuclides from soil to vegetation as compared to arable lands. Such criticality of Polissya landscapes in terms of ^{137}Cs migration intensity in food chains is primarily predetermined by the soil type. The typical soils of this region are mainly peat, peat-gley and peat-bog soils. They are characterized with a high content of organic matter (from 20 to 60 %), very low content of clay minerals and silt fractions, acidic reaction of soil solution (pH (KCl) = 4.2–5.4) and high moisture. The transfer factors of ^{137}Cs in the "soil-plant" system for these soils may exceed the corresponding values for soddy-podzolic soils in 4–30 times (Prister 2007). The accumulation of radiocaesium in particular plant species also depends on number of additional factors. They could be combined into two groups in terms of plant organism: *internal* (inherent to a particular species) and *external* (related to the ecosystem). The group of internal factors consists of the following biological characteristics of plant species (Prister 2007):

- systematic position;
- life form;
- formation of symbiosis with mycorrhizal fungi;
- need for K+ and other cations;
- depth of the root system location in the soil;
- ecological amplitude.

The numerous studies related to ^{137}Cs accumulation in plant species of herbal-shrubby storey of contaminated forest ecosystems demonstrate that plants are characterized with the species-specific accumulation of radiocaesium. This phenomenon is revealed in all types of phytocenoses and conditions of plant growth. In particular, the interspecies differences of ^{137}Cs accumulation by vascular plants in fresh sudubravas of Ukrainian Polissya reach 50 times for herbal and lily species in the oak forest cenosis (Baryakhtar 1996). In the fresh pine forests cenosis these differences can be even 250-fold for the herbal and blueberry species. Hence, the studied plant species could be grouped according to their intensity of ^{137}Cs accumulation. Usually the aggregated transfer factor (T_{agg}) of radiocaesum from the soil to plant biomass is used to describe this process. This is the ratio of activity concentration in plant biomass (Bq kg^{-1}) to the radionuclide inventory in the soil (Bq m^{-2}). In terms of T_{agg} value (m^2 kg^{-1}), the intensity of ^{137}Cs transfer to the aboveground plant biomass can be conditionally divided into very strong ($T_{agg} > 100$), strong ($100 > T_{agg} > 50$), moderate ($50 > T_{agg} > 10$), weak ($10 > T_{agg} > 1$) and very weak ($T_{agg} < 1$). The average values of T_{agg} for ^{137}Cs obtained for some plant species of herbal-shrubby storey in fresh pine forests and fresh sudubravas of Ukrainian Polissya region are listed in the Table 3. The systematic affiliation of plants significantly affects the accumulation of radiocaesium. Also the increased uptake of this radionuclide is characteristic for ferns and mosses. In pine forests the plant species from the heather and cranberry families are the main accumulators of ^{137}Cs, while in sudubravas radiocaesium is accumulated predominantly by plants representing the Rosaceae and lily families (Orlov and Krasnov 1996).

The plant species of different taxonomic groups reveal the differences in ^{137}Cs uptake via significantly different rates of the radionuclide accumulation during the subsequent stages of the vegetation period as well as in different redistribution of radiocaesium between aboveground and underground parts of plants. Thus, the activity of ^{137}Cs in aboveground biomass of species from sedge family is decreased from the beginning of vegetation period till its end, while corns are characterized with the gain of radiocaesium concentration at the end of vegetation. Also it was demonstrated (Shcheglov et al. 2001) that the intensity of ^{137}Cs uptake by plants is significantly influenced the horizontal length of their root system, degree of soil horizons filling by roots, and chemistry of root secretions. The uptake of radiocaesuim by plant species of herbal-shrubby storey is the result of complex interaction of internal factors briefly reviewed above and external factors, in particular the following soil parameters:

- type of soil;
- mineralogical and granulometric composition of soil;
- soil richness (concentration of mineral nitrogen);
- soil humidity;
- soil acidity (pH);
- soil organic matter;
- content of exchangeable K^+;
- degree of saturation with bases of soil absorbing complex.

Table 3 Intensity of ^{137}Cs accumulation by aboveground phytomass of plant species representing herbal-shrubby storey (Adopted from Orlov and Krasnov 1996)

Intensity of ^{137}Cs accumulation by aboveground phytomass	Plant species	Transfer coefficient (TC), $m^2\ kg^{-1}$
Fresh pine forest		
Very strong (TC > 100)	*Dryopteris carthusiana*	590
	Pteridium aquilinum	500
	Maianthemum bifolium	170
	Melampyrum pratense	160
Strong (100 > TC > 50)	*Trientalis europaea*	72
	Rubus saxatilis	56
	Calluna vulgaris	45
	Convallaria majalis	40
	Vaccinium myrtillus	29
	Vaccinium vitis-idaea	28
	Potentilla alba	23
	Cárex	18
	Fragaria vesca	17
	Hypericum perforatum	11
Weak (10 > TC > 1)	*Chimáphila umbelláta*	9.6
	Festuca ovina	9
	Orthília secúnda	4.4
	Geranium sanguineum	2.3
Fresh sudubravas		
Moderate (50 > TC > 10)	*Dryopteris carthusiana*	47
	Dryopteris filix-mas	15
	Athyrium filix-femina	13
	Potentilla alba	13
	Carex brizoides	13
	Maianthemum bifolium	12
	Melampyrum nemorosum	10
Weak (10 > TC > 1)	*Rubus saxatilis*	9.9
	Convallaria majalis	8.6
	Fragaria vesca	8.3
	Carex montana	6.3
	Polygonatum odoratum	4.6
	Origanum vulgare	2.6
	Vaccinium myrtillus	2.4
	Vaccinium vitis-idaea	2.3
	Hypericum perforatum	1.6
Very weak (TC < 1)	*Geranium sanguineum*	0.9

Considering these factors, it was shown (Shcheglov et al. 2001) that ^{137}Cs accumulation by plants from soil is higher in case of lower soil fertility and higher soil moisture. The intensity of radiocaesium accumulation by plant phytomass is higher for soils characterized with lower content of fine fractions (silt and clay), lower pH values and K^+ exchangeable forms, and lesser saturation degree with bases of the soil absorbing complex. During the recent two decades, the uptake of ^{137}Cs by plants in terrestrial ecosystems of Polissya region demonstrates a general decrease trend with time. It is caused mainly by reduction of the radionuclide mobility in the soil due to gradual increase of its absorption and fixation strength. Thus, the most widespread species of *Vacciniaceae* family in Polissya forests are characterized with significant decrease of ^{137}Cs specific activity for the period from 1991 to 2011 (Furdychko 2012). In particular, the concentration of radiocaesium in fresh berries of cranberry was decreased in 3–4 times, while in those of blueberry the decrease of ^{137}Cs reached five times. The estimated half-cleaning period from ^{137}Cs for plant species depends on conditions of specific biocenosis. It is 7.5 years both for cranberry and blueberry growing in humid pine forests, while for cranberry from wet pine and broad-leaved forests the half-cleaning period accounts 7.7 and 5.5 years correspondingly. The similar differences in ^{137}Cs accumulation were revealed also for agricultural plants. According to intensity of radiocaesium uptake, the certain agricultural plant spices can be placed in the following three ascending orders (Prister 2007):

1. *Cereals and legumes*: buckwheat < soy < bean < pinto < peas < oats < rye < barley < millet < triticale < corn < wheat.
2. *Forage crops (fresh biomass)*: yellow lupine < kale < vetch < sunflower < clover < timothy < awnchaff < corn.
3. *Industrial crops*: oil radish < rape < sugar beets < sunflower < flax.

In some cereal crops (winter wheat, barley, rye and oats) the most intensive accumulation of radiocaesium is found in their leaves and stems, while in reproductive organs of plants this radionuclide is accumulated to a much lesser extent (Kotkova 2004). Depending on the soil properties, the levels of soil contamination, cultivated plant species and possible ways of harvest use, the various countermeasures can be applied that reduce radiocaesium activity in crop production. Usually five major complex reduction systems of ^{137}Cs and other radionuclides intake to plants are distinguished: (1) techniques of soil cultivation; (2) use of chemical fertilizers and ameliorants; (3) change of plant species in crop rotation; (4) change of irrigation mode and (5) use of special remedies (Prister 2007). Additionally, the physical countermeasures such as removal of upper soil layer and application of soil amendments have been used (Zhu and Smolders 2000), but these methods are rather expensive and often unjustified. Therefore other methods, such as extraction of radiocaesium by plants, were widely considered in the recent time. However application of this technique is still questionable since the phytoextraction of radiocaesium may be unsuitable for agricultural lands used for food production (White et al. 2003).

2 Conclusion

During the last decades the ratios of radiocaesium fraction in the soils have not been changed considerably. The uptake of ^{137}Cs by plants in terrestrial ecosystems of Ukrainian Polissya region reveals a general decrease trend. It is caused mainly by reduction of radiocaesium mobility in the soil due to increase of its absorption and fixation strength. The distribution of radiocaesium in the soil demonstrated that the radionuclide migration depends on the many environmental landscape factors, primarily from as surface relief, vegetation cover and type of ecosystem.

The research concerned to ^{137}Cs accumulation in plant species of herbal-shrubby storey of forest ecosystems show that plants are characterized with the species-specific accumulation of the radionuclide. This is typical for all types of studied phytocenoses and conditions of plant growth. The accumulation of radiocaesium in a plant also depends on number of internal (inherent to a particular species) and external (related to the ecosystem) factors.

The forests and natural grasslands are suggested to be the most critical landscapes in relation to radiocaesium speciation in soil and further intake to food chains. These ecosystems are usually characterized with the highest transfer factors of ^{137}Cs and other radionuclides. Such criticality of Polissya landscapes in terms of ^{137}Cs migration is primarily caused by the soil type. The relatively smaller (in comparison with Polissya region) radiocaesium retention in forest soils in Fukushima could be explained by richer organic matter formed due to large amounts of precipitation and volcanic ash deposition.

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Factors Influencing the Soil to Plant Transfer of Radiocaesium

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Abstract Radiocaesium isotopes are among the long-lived radionuclides which were released in largest quantities into the environment as a consequence of the atmospheric nuclear weapon tests and accidents involving nuclear material (Chernobyl and Fukushima, etc.). Its transfer to plants, especially those for human or animal food, can be a major pathway for human intake and, therefore, have a significant radiological impact. There are many factors that can affect the radiocaesium transfer to plants, which are reviewed in this chapter, such as the considered plant species, its habitat, climatic conditions, type of soil (clay content, physico-chemical characteristics, organic matter, use of amendments, etc.).

Keywords Plant • Transfer factor • Speciation • Organic matter • Clay • Soil

1 Introduction

The occurrence of radiocaesium is ubiquitous in the environment, as a result of the global fallout caused by the atmospheric nuclear weapon tests carried out in the 1950–1960s (UNSCEAR 2000). Accidents involving nuclear material, such as Chernobyl and Fukushima Dai-ichi also released large quantities of radiocaesium and, as a result, vast areas were contaminated (UNSCEAR 2000; IAEA 2006, 2014). Radiocaesium is one of the long-lived radionuclides that are highly significant from the radiological protection point of view. Due to the chemical similarities with potassium, it enters the food chain pathway and may cause a health hazard to humans. Plant consumption, either as direct foodstuff or indirectly as animal foodstuff, is a major pathway for radiocaesium incorporation. In fact, its content in foodstuff has been controlled since the Chernobyl accident happened, with temporary permissive limits for different foodstuff after an emergency (IAEA 2006; EU 2009; CA 2011; Hamada and Ogino 2012). One of the main interests of the study of the

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soil to plant transfer processes of radiocaesium is its quantification and later use to predict its content in plants for human and/or animal consumption. This knowledge is necessary in order to carry out remedial actions to decrease the radiocaesium incorporated by men, reducing the potential health hazard. In this chapter, the most usual ways to quantify the radiocaesium soil-to-plant transfer are presented, along with its problems and factors that influence its great range of variation.

2 Quantification of the Transfer Process

The concept of transfer factor (usually TF, CR or F_v) is the most usual and straightforward approach to quantify this process. It is usually defined as the ratio between the radionuclide content in the plant or plant compartment and its content in the soil (see Eq. 1) (IAEA 2010).

$$\mathrm{TF, CR, F_v} = \frac{\mathrm{Bq / kg\ d.w.\ plant}}{\mathrm{Bq / kg\ d.w.\ soil}} \tag{1}$$

The radionuclide content in plant is often expressed as dry weight basis, in an attempt to decrease the data variability. In case of fruits or when data are used to estimate internal dose rates, F_v values are usually given as fresh weight. These data can be converted to dry weight basis, using dry matter content reference values, such as those presented in IAEA TRS 472 (IAEA 2010). Calculation of the transfer factor values, F_v depends on the depth of soil considered, which is especially important in cases in which the distribution of radionuclides is inhomogeneous in depth. In order to reduce the influence of this variable, the International Union of Radioecology (IUR) recommended a standardized root location in soil. This approach assumes that all roots and all radionuclides present in the rooting zone are in that soil layer. The soil depth is 10 cm for grass, and 20 cm for all other crops, including trees (IUR 1992). The use of standard soil depth for agricultural systems is reasonable since ploughing is usually part of soil preparation for growing crops. Thus, the radionuclide content of this soil layer becomes homogeneous. The concept of transfer factor also implies, by definition, that the transfer process proportional to the radionuclide content in the soil, and that the soil-plant system is in equilibrium or quasi-equilibrium. This condition is achieved in most cases, assuming that the radionuclide flow from soil to plants is negligible compared to the total amount of radionuclides present in soil (IAEA 2010).

Figure 1 shows the range of variation of ^{137}Cs transfer factors worldwide for different plants/crops. The soil-plant transfer factors for radiocaesium present a large range of variation, about 4–5 orders of magnitude, reflecting the fact that transfer processes are complex and affected by many variables. Wild products, such as mushrooms or berries, usually have high F_v values, followed by grass, non-leafy vegetables and stems of cereals. This high range of variation for F_v implies that they

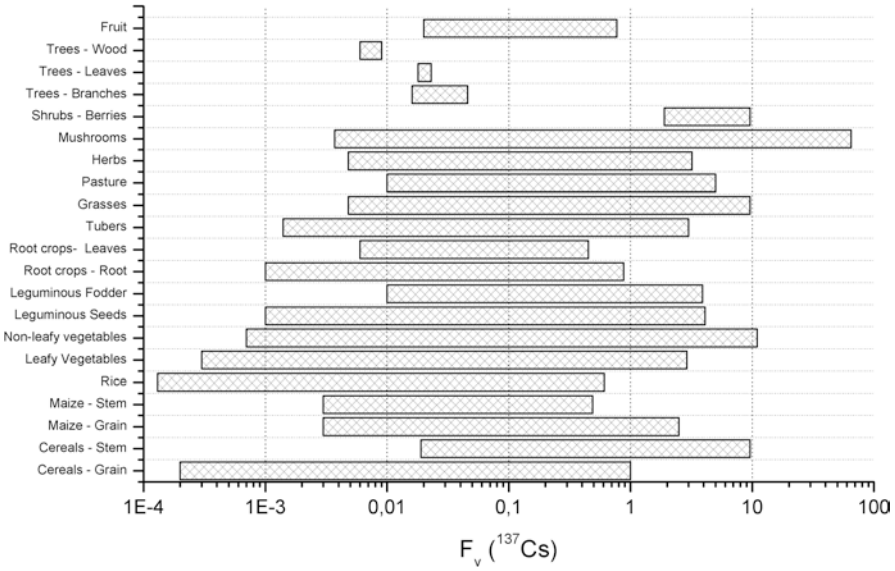

Fig. 1 Worldwide range of variation of soil-to-plant transfer factor F_v for ^{137}Cs. Data adopted from Ban-nai and Muramatsu (2002), Baeza et al. (2005), Kuwahara et al. (2005), Al-Oudat et al. (2006), Gaso et al. (2007), Handl et al. (2008), Moran-Hunter and O'Dea (2008), IAEA (2010), Zhiyanski et al. (2010), Karadeniz and Yaprak (2011), Velasco et al. (2012), Kobayashi et al. (2014), Yamashita et al. (2014), Godyń et al. (2016)

should be considered valid at local level, due to a great number of factors. As cereals are a wide extended crop, the FAO/IAEA/IUR workgroup proposed to consider it as reference crop in an attempt to reduce variability, taking into account the ratios between different crops and cereals (Frissel et al. 2002)

In the case of a radioactive fallout event, the aggregated transfer factor, T_{agg}, is also frequently used to estimate the transfer, because it relates the activity in plant compartments with that deposited on soil (see Eq. 2). Thus, they are used to predict the transfer of radionuclides to plants, essential for stakeholders to manage emergencies.

$$T_{agg}\left(m^2 kg^{-1}\right) = \frac{Bq / kg\ d.w.\ plant}{Bq / m^2 deposited\ on\ soil} \tag{2}$$

The T_{agg} values are also used in natural and semi-natural ecosystems, such as forests, in which the distribution of anthropogenic radionuclides is inhomogeneous in depth. This is especially important for radiocaesium, because after radioactive fallout, it is deposited on the surface layer of soil and then it migrates downwards to deeper layers. Figure 2 shows the range of variation of T_{agg} for ^{137}Cs for different plants in forest ecosystems worldwide. As it occurred for F_v values, the range of

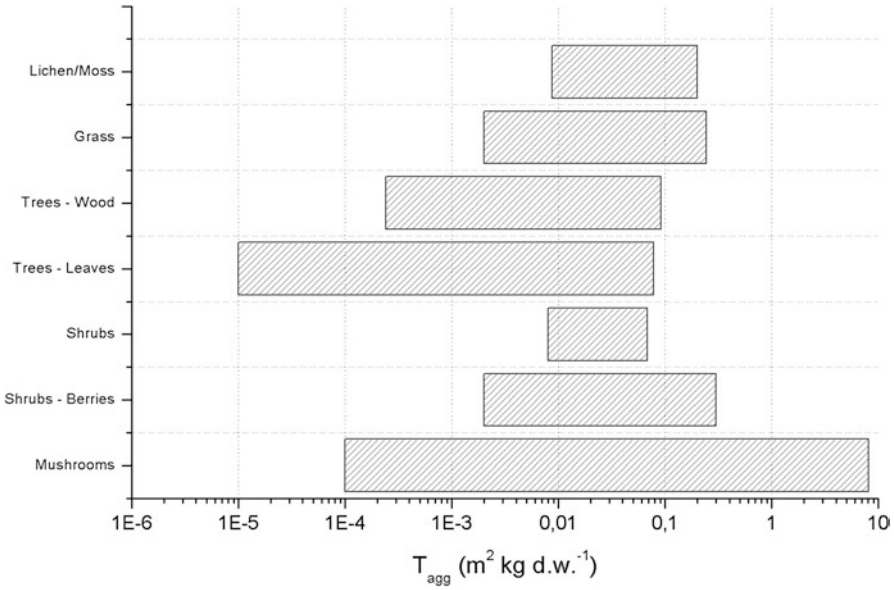

Fig. 2 Worldwide range of variation of soil-to-plant aggregated transfer factor, T_{agg}, for ^{137}Cs. Data adopted from Strandberg (1994), Fesenko et al. (2001b), Kaduka et al. (2006), IAEA (2010), Karadeniz and Yaprak (2011), Nakai et al. (2015)

variation of T_{agg} values is about 5–6 orders of magnitude. Mushrooms presented again the highest T_{agg} values.

Another important integrating transfer factor for the ecosystems is the geochemical transfer factor, defined as the ratio between the radionuclide total activity in vegetation collected from the certain area, expressed as Bq/m^2, and its deposition in the same area, expressed as Bq/m^2.

$$T_{geo} = \frac{\text{Bq / m}^2\text{in plant biomass}}{\text{Bq / m}^2\text{in soil}} \tag{3}$$

The F_v values can also be used in forest ecosystems, especially when the source term of the deposition of radionuclides occurred long time ago, as it is the case of areas in which the main deposition event was the global fallout from atmospheric nuclear tests in the 1950–1960s (UNSCEAR 2000). However, in this case it is very important to define precisely the depth of the soil layer used in its calculation. For wild grass, as it is a Reference Animal and Plant (RAP) used to analyze the dose rate to wildlife (ICRP 2008), the soil depth used is standardized to 10 cm. This depth of soil has also been considered in software to assess the radiological impact of radionuclides in non-human biota, such as ERICA (Brown et al. 2008). In the case of mushrooms, some authors took into account the radiocaesium content of the soil

layer in which mycelium was located. This location was performed by using different techniques: mechanical isolation of fungal mycelium directly from the soil (Nikolova et al. 2000); the determination in the soil of chemical compounds present in the cell walls of the mycelium (Baeza et al. 2005); and the comparison of the ratio $^{137}Cs/^{134}Cs$ in different layers of soil and in the fruit bodies (Rühm et al. 1997). Regarding trees, a high percentage, 54–70 %, of fine roots of pine (*Pinus sylvestris*) and birch (*Betula pendula*) were detected at 40 cm depth in soil (Fesenko et al. 2001a). Therefore, the depth of the soil layer should be given when calculating F_v values in forest ecosystems.

Other radioisotopes of caesium also occur in the environment, such as ^{134}Cs or stable ^{133}Cs. The source term for both ^{137}Cs and ^{134}Cs is usually the same, and their ratio, $^{134}Cs/^{137}Cs$, depends on the characteristics of the fallout event. Differences in the transfer process due to isotopic effect are almost negligible, being the F_v values for ^{134}Cs and ^{137}Cs the same within measuring uncertainties (Kobayashi et al. 2014). In fact, ^{134}Cs is used in transfer experiments under controlled conditions (Carini and Lombi 1997). The stable isotope of caesium, ^{133}Cs, has been considered as a key element to predict long-term behaviour of radiocaesium in the environment, i.e. the transfer of ^{137}Cs and stable caesium is expected to be the same long time after the fallout event. The correlation of the ^{137}Cs and stable caesium is considered as an indication of equilibrium in forest ecosystems (Yoshida et al. 2004; Kuwahara et al. 2005)

There are also other ways to quantify the transfer processes, such as compartmental models (Kichner 1998; Absalom et al. 1999, 2001; Baeza et al. 2001). These models are capable of determining the effect of several variables affecting the radiocaesium transfer processes: influence of clay, organic matter, bioavailability of radiocaesium, exchangeable potassium, etc. However, they require a high degree of knowledge of the compartments involved in the transfer, and are difficult to implement for screening purposes.

3 Factors Affecting the Caesium Soil-to-Plant Transfer

The fact that the soil-to-plant transfer factors for radiocaesium vary widely over several orders of magnitude implies that there are factors influencing the transfer processes other than its content in soil and plant compartment considered. Climatic conditions can affect the transfer processes, since they control major variables affecting the plant growth, as temperature, water regime, humidity, etc. Temperate environments a generally located between the Tropics and the Polar Regions, in which the temperatures are relatively moderate, and changes between winter and summer are also moderate. Tropical environments are typically found in the Tropics, with mean monthly temperatures about 18 °C, and seasonal variations dominated by precipitation. In these environments, there is a rapid decomposition of almost all organic materials deposited on surface soil, and high mineral weathering rates in soil. Subtropical climates are roughly located in areas between the Tropics and the

38th parallel in each hemisphere. Mediterranean climate is a particular variety of subtropical climate characterized by warm to hot, dry summers and mild to cool, wet winters.

Table 1 shows the range of variation of radiocaesium F_v values for several plant groups in different climates (IAEA 2010). In the case of cereal grain and root crop plant groups, these ranges do not vary greatly, and are about the same order of magnitude temperate, subtropical and tropical climates. In the case of root crops in tropical climate, the lower range can be due to the limited number of samples considered. Regarding grass plant group, tropical environments seem to present a wider range of variation. Thus, the direct influence of climatic conditions on the radiocaesium transfer seems to be minimal, but its indirect effect, through changes in soil and crop properties can be significant (IAEA 2010). However, in Mediterranean environments, in which there is an alternation between hot dry seasons (mainly summer) and cold wet seasons (mainly winter), the radiocaesium transfer to grass showed a seasonal dependence reflecting the variation of nutrient availability in each season (Baeza et al. 2001).

Soil characteristics play a crucial role on radiocaesium transfer. As a way of example, Table 2 shows the range of variation of radiocaesium F_v for several plant groups in different soil groups, based on their texture, cation exchange capacity and organic matter content. Clay soils show lower F_v values than other soil groups. This is because clay content in soil acts as a sink for radiocaesium (Ohnuki 1994). It is found usually that the higher the clay content the higher the radiocesium content in it (Apostolakis et al. 1991), because its sorption on clay minerals (Ohnuki 1994; Atun et al. 1996; Kruyts and Delvaux 2002; Stauton et al. 2002). This sorption can

Table 1 Range of variation of radiocaesium transfer factors, F_v for several plant groups in different climatic conditions

	Environment		
Plant group	Temperate	Subtropical	Tropical
Cereal-grain	2×10^{-4}–0.9	1×10^{-3}–2.6×10^{-2}	6×10^{-2}–1.0
Root crops-root	1×10^{-3}–0.88	1.4×10^{-3}–0.23	0.13–0.81
Grass	4.8×10^{-3}–0.99	6×10^{-3}–3.7	1.5×10^{-4}–13

Data adopted from IAEA (2010)

Table 2 Range of variation of radiocaesium transfer factors, F_v for several plant groups in different soil groups

	Soil group			
Plant group	Sand	Loam	Clay	Organic
Cereals/grain	2×10^{-3}–0.66	8×10^{-4}–0.20	2×10^{-4}–9×10^{-2}	1×10^{-2}–0.73
Root crop/root	8×10^{-3}–0.40	1×10^{-4}–0.16	5×10^{-3}–6×10^{-2}	1.6×10^{-2}–0.88
Grass	1×10^{-2}–4.8	1×10^{-2}–2.6	1×10^{-2}–1.2	0.3–5.0

Data adopted from IAEA (2010)

be carried out in two different ways: sorption sites on regular exchange sites (RES) and on frayed edge sites (FES). These sorption sites have different properties. RES are reversible and non-specific; while sorption on FES is irreversible and specific, due to the small hydration energy for caesium ions. The energy required for caesium desorption from FES was found to be so large that desorption was energetically unfavorable (Stauton et al. 2002). The FES selectivity for monovalent cations decrease in the order: $Cs^+ > NH_4^+ > Rb^+ > K^+ > Na^+ > Li^+$ (Rigol et al. 2002; Stauton et al. 2002). The K^+ content in soil can cause the collapse of the expanded interlayers (Rigol et al. 2002). In this case the caesium binded inside the interlayers is blocked and unavailable for transfer processes. Climate can also affect the type and exchange capacity of clays. The occurrences of low exchange capacity clays, such as kaolinite, are more common in the Tropics than in temperate climates, due to the high mineral weathering rate (IAEA 2010).

Soil organic matter content can also influence the radiocaesium transfer, since it can influence its association with soil components. The organic matter compounds comprise humin and humic substances present in soil. The humic acids are macromolecules with carboxylic and phenolic functional groups. In the pH range 3–6, the humic substances chemistry is dominated by carboxylic acid groups (Lofts et al. 2002). The addition of humic substances to mineral clays reduced its radiocaesium adsorption (Dumat et al. 1997; Dumat and Stauton 1999). This reduction was not due to the direct sorption of caesium on humic substances. In fact, its association with humic and fulvic acids is very low, about 5 % (Dumat and Stauton 1999; Amano et al. 1999; Vinichuk et al. 2005; Guillén et al. 2015). The presence of organic matter can produce a dilution effect on FES, increasing the availability of radiocaesium (Kruyts and Delvaux 2002). The association of organic compounds can also impede the collapse of interlayers in clay minerals, maintaining them open for reversible exchange sites (Rigol et al. 2002). However, even a small content of clay, about 1–6 %, can bind caesium effectively (Forsberg et al. 2001; Lofts et al. 2002; Rigol et al. 2002). Only in soils with more than 95 % content of organic matter and no clay, the adsorption occur in non-specific sites (Rigol et al. 2002).

Microflora can also retain effectively the radiocaesium present in the soil. This association can be assessed by killing all living organisms present in the soil by autoclaving, γ-ray sterilization, fungicides, or chloroform fumigation (Brückman and Wolters 1994; Guillitte et al. 1994; Stemmer et al. 2005). The caesium retention by microflora was about the 1–56 % (Brückman and Wolters 1994; Guillitte et al. 1994; Stemmer et al. 2005). After these treatments, it was observed an increase of the labile caesium (Stemmer et al. 2005). The soil organic matter and its mineralization potential controlled the radiocaesium transfer in soddy-podzolic and peat bog soils, since the mineralization is accompanied by the release of ^{137}Cs and mineral nitrogen (Tulina et al. 2010).

Fungal material present in soil also participated greatly in the radiocaesium cycling in forests. The fungal mycelium in soil can retain about 0.1–32 % of the total inventory in soil (Olsen et al. 1990; Vinichuk and Johansson 2003, Vinichuk et al. 2005). The increase of labile radiocaesium after the elimination of microflora

may be attributed to its release from fungal mycelium. About 42–83 % of the radiocaesium associated with fungal mycelium can be extracted with distilled water (Vinichuk et al. 2005). Larger amounts of caesium were found in white spots on fungal mycelium, mainly associated with polyphosphates (Landeweert et al. 2001). The importance of fungal mycelium can be due to their ability to exude organic acids, such as citric and oxalic acid, which can complex metals in the surrounding (Gadd 1999). The concentration of these acids in bulk soil is usually low, but high in the microenvironments near the hyphaes (Landeweert et al. 2001). The occurrence of mycorrhizal fungi living in symbiotic relationship with plant hosts can also influence the radiocaesium transfer. They usually present higher radiocaesium content than other fungi with saprophyte or parasitic nutritional mechanisms (Guillitte et al. 1994; Kammerer et al. 1994; Yoshida and Muramatsu 1994). This suggested that mycorrhizic fungi act as a "filter" for the host plant, accumulating non essential elements such as caesium (Guillitte et al. 1994; Kammerer et al. 1994). Laboratory experiences growing Norway spruce (*Picea abis*) seedlings inoculated with the ectomycorrhizal fungus *Hebeloma crustiliniforme* and without it showed a reduction in the uptake of ^{134}Cs, and a higher accumulation in the hypae (Brunner et al. 1996; Riesen and Bunne 1996). The arbuscular mycorrhizal colonization of several species of grass by *Glomus mossea* also resulted in reduction of caesium content in shoots and root (Berreck and Haselwandter 2001). However, this does not reflect the whole tendencies. According to the previous research, the uptake of caesium by plants in the presence of arbuscular mycorrhiza was either lower (Berreck and Haselwandter 2001), similar (Rosén et al. 2005) or higher (Entry et al. 1996) as in nonmycorrhizal plant species.

In the discussion of the previous factors affecting the radiocaesium transfer, the concept of bioavailability has been mentioned, as the fraction of the radionuclide pool in soil that it is able to be transferred to a plant compartment. Its empirical determination is based on sequential extraction procedures in which soil is attacked by a series of reagents with increasing replacement/extraction power. One of the main handicaps of this approach to the transfer is the fact that there is no unified procedure/reagent to carry out (Guillén et al. 2014). There is in the literature a great number of modifications on these procedures varying reagents, their concentrations, contact times, and order of application. This fact implies that sometimes the comparison of the results from different sequential extraction methods can be complex, because although the extractants are usually designed to attack a single geochemical phase, they are not completely specific (Schultz et al. 1998). The bioavailable fraction can be subdivided into two different sub-fractions: readily available (water soluble and exchangeable) and potentially available (associated with oxides and carbonates). However, the number of reagents usually involved in the bioavailable fraction is quite standard, at least for the readily available fraction. Water soluble fraction present the weakest attachment with soil particles, as the reagent used is distilled water. Radiocaesium associated with this fraction is usually minimal and sometimes it is omitted from some sequential procedures (Riise et al.

1990). Exchangeable fraction comprises the radionuclides associated with ion exchange sites. The NH_4AcO 1 M at pH 7 is the reagent usually selected for this fraction, and is considered to be a robust extractant for acidic and neutral soils, but not for alkaline soils, although it can be buffered to extend its range of application (Kennedy et al. 1997). Its validity for radiocaesium is based on the fact that NH_4^+, along with K^+, are competitors with Cs^+ in soils. Other reagents based on divalent ions, such as $MgCl_2$ or $CaCl_2$, are also able to desorb caesium from exchange sites, but are considered to be less effective in clay interlayers (Rigol et al. 2002). The exchangeable fraction (extracted with NH_4OAc) of radiocaesium in soils is in the range 1.8–29 % of the total content of soil (Riise et al. 1990; Bunzl et al. 1997; Lee and Lee 2000; Forsberg et al. 2001; Vinichuk et al. 2005). The exchangeable fraction presented a dependence of the layer of soil considered, increasing with the depth of that layer (Bunzl et al. 1997). This fraction was not constant with time, but it decreased while increasing the lapsus of time since the deposition of radiocaesium occurred (Cheshire and Shand 1991; Krouglov et al. 1998; Baeza et al. 1999; Forsberg et al. 2001), which is usually known as ageing effect, due to the irreversible sorption of caesium. This ageing effect caused the reduction of the transfer factors to rice after Fukushima accident (Fujimara et al. 2015). The definition of the potentially available fraction is more diffuse, and only considered in one of the sequential extraction procedure used in agriculture testing (Pavlotskaya 1974). The use of dilute inorganic acids, HCl 1 M, after the exchangeable fraction can remove cations from exchange complexes in soil, and also dissolve oxides, hydroxides, carbonates and some alkaline earth compounds. It has been considered potentially available to plants (Fesenko et al. 2001a). Other reagents can be considered partially equivalent to this potentially bioavailable fraction. The radionuclides associated with the carbonated fraction can be extracted with sodium acetate in acetic acid (Tessier et al. 1979) or with NH_4OAc after the extraction of the exchangeable fraction with $MgCl_2$ (Schultz et al. 1998). The reducible fraction, also named in some procedures bound to Fe and Mn oxides, are obtained by the application of a reducing extractant, generally $NH_2OH \cdot HCl$ (Tessier et al. 1979; Riise et al. 1990; Schultz et al. 1998).

Finally, the definition of transfer factor does not take into account the stage of maturity of the plants (see Eqs. 1 and 2). They are usually derived from activities in soil and plant compartments at the end of the growing period, which may lead to high values (IAEA 2010). In this sense, the transfer factors can be considered as integrators of the whole growth period, providing a mean value of the period. However, there are variations due to the different nutritional requirements in each stage of development. In the case of fruits, the transfer factor was found to be maximum at the initial stages and decrease as fruit developed, being lowest at the maturation period (Velasco et al. 2012). Experiences of ^{134}Cs contamination of *Pleurotus eryngii* fungus species at different stages of development showed an increase of the transfer of ^{134}Cs as the contamination occurred closer to maturity stage (Baeza et al. 2002).

4 Modification of the Radiocaesium Transfer

The modifications of the radiocaesium transfer processes are a key factor to controlling its accumulation in plant and thus remediate contaminated sites. The modification can be carried out in two opposite ways: enhancement and inhibition. Phytoremediation uses the first one. Its objective is to locate hyperaccumulators and thus maximize the soil-to-plant transfer and therefore remove radiocaesium from soil (Cook et al. 2009; Jagetiya et al. 2014). In this case, the management of the plants after harvest has also to be considered. Other remediation techniques are focused on reducing the transfer. Ploughing was considered to reduce the radiocaesium content in soil after a deposition by mixing the surface soil with that at different depth with lower content, thus diluting the deposited radiocaesium. In areas affected by the Chernobyl accident, the combination of ploughing and reseeding was effective to reduce the radiocaesium transfer (Camps et al. 2004). However, ploughing in areas affected by Fukushima accident did not result in reducing the transfer factors (Kobayashi et al. 2014). This might be as consequence of the quantity of radiocaesium deposited, and not being able to dilute it effectively.

The radiocaesium transfer can also be modified by the application of inorganic fertilizers and soil amendments. The final effect depends on the fertilizer composition. Potassium based fertilizer are used to reduce the radiocaesium transfer, by saturating the soil with an additional supply of nutrients chemically analogous to the radionuclides (Nisbet et al. 1993). There is a critical threshold in the concentration of potassium in soil solution. At lower concentrations, the root uptake mechanism is unable to distinguish between alkaline elements Cs^+, Rb^+, and K^+ (Nisbet et al. 1993; Shaw 1993; Zhu et al. 2000). Above ca. 20 μM, wheat root uptakes K^+ preferentially to Cs^+ (Shaw 1993). These fertilizers can reduce the radiocaesium transfer about 40–60 %, being more successful the lower K^+ available in soil (Jacob et al. 2009; Rosén et al. 2011). The reduction effects are lasting and observable during long periods of time, 10–34 years after fertilization (Kaunisto et al. 2002; Robison et al. 2009; Rosén et al. 2011). The addition of fertilizers that supply NH_4^+ can also modify the content of ^{137}Cs in the soil solution, but in the opposite way. It increased the transfer by a factor of 3–4, due to its competition with Cs^+ for exchange sites (Nisbet et al. 1993). The application of NH_4^+ and manure can also reduce the uptake of ^{137}Cs, probably due to the release of potassium and other ions from the manure when NH_4^+ is applied (Fuhrmann et al. 2003).

5 Conclusion

The knowledge of the factors affecting the radiocaesium soil-to-plant transfer is one of the key factors to manage the consequences of nuclear accidents and the health hazard involved in plant consumption, either directly or indirectly via animal foodstuff. Transfer factors defined as the ratio between what is in the plant and what is

in the soil are the most usual way to quantify the transfer processes. However, their uses have some difficulties. The range of variability for transfer factors for the same plant groups is great, about 4–5 orders of magnitude, which limits their applicability. The definition of soil compartment is also not perfectly defined. In the case of agricultural crops, some international standards were proposed to limit this variability, mainly the depth of soil considered. But in the case of natural or semi-natural environments, such as forest, it is not so well established. There are also some factors responsible for the variation of the radiocaesium transfer factors. Climate contributes to it, but indirectly, as it conditions the type of plants able to grow and the type and characteristics of soil. The latter is the main factor affecting the radiocaesium transfer. Soil clay content is a major factor, as caesium can be adsorbed irreversibly on them and, therefore unable to be transferred to plant. Organic matter present in soil and its mineralization potential can modify this adsorption, making radiocaesium more available to transfer. The bioavailability concept is linked to the transfer process as the fraction of the total radiocaesium that can be effectively be transferred to plant. The methods for its evaluation are based on sequential extraction techniques, and it can be classified into readily available (water soluble and exchangeable fractions) and potentially available (extractable with diluted inorganic acids). The bioavailable fraction can be modified by the addition of fertilizers and soil amendments. The transfer factor is reduced when potassium based fertilizer or amendments are added to the soil, so that there is more potassium bioavailable. When it surpasses a concentration threshold, potassium is taken preferentially to caesium up. The addition of NH_4^+ has the opposite effect, increasing the transfer.

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Role of Mycorrhizal Fungi in Caesium Uptake by Plants

Sergiy Dubchak

Abstract It was demonstrated that rhizospheric processes involving mycorrhizal fungi can influence root uptake of radiocaesium. The ability of both ectomycorrhizal and endomycorrhizal fungi to limit radiocaesium availability to their host plants was considered. The ectomycorrhizal fungi grown in forest ecosystems were suggested to immobilize between 10 and 100 % of the total ^{137}Cs activity. Radiocaesium was found to be accumulated in mycelium and fruit bodies of ectomycorrhizal fungi by simple diffusion and facilitated transport. Ectomycorrhizal fungi was considered to be efficient indicators of cumulative biogeochemical fluxes of radiocaesium in terrestrial ecosystems and thus to be appropriate candidates for phytoremediation technique.

The significance of arbuscular mycorrhizal fungi as participants in radiocaesium cycle in the upper soil layers was discussed. Up to date, their role in processes of radiocaesium uptake by plants remains incompletely understood and controversial. It was demonstrated that arbuscular mycorrhizal fungi could accumulate ^{137}Cs in their extraradical or intraradical structures, transport the radionuclide to their hosts and influence its distribution among plant roots/shoots. Depending on arbuscular mycorrhizal fungi and plant species used, the significant reduction or increase of radiocaesium transfer to aboveground plants biomass was found. The perspectives of arbuscular mycorrhizal fungi application in phytoremediation techniques were discussed.

Keywords Radiocaesium • Ectomycorrhizal fungi • Arbuscular mycorrhizal fungi • Plant species • Fungal mycelium • Caesium translocation

1 Introduction

During the last decades the alternative strategies, oriented towards the use of plants and micro-organisms, separately or in combination, have been proposed for removing or immobilizing radiocaesium in the soil (Entry et al. 1999; Zhu and Shaw 2000;

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Singh et al. 2008; Fulekar et al. 2010; Wei et al. 2014). Among these microorganisms, mycorrhizal fungi received a particular attention. An estimated 90 % of terrestrial plants exist in a symbiotic association with soil fungi forming mycorrhizal associations (Smith and Read 1997). It was demonstrated that rhizospheric processes involving mycorrhizal fungi, also influenced root uptake of various plant pollutants including radiocaesium (Gadd 1996; Joner and Leyval 1997).

Thus, the mycorrhizal fungi could play a key role in soil remediation strategies of pollutants, including radionuclides (Entry et al. 1996). The symbiotic fungi develop an active continuum between an extraradical network in the soil and an intraradical network within the roots of their host plants. This continuum allows bidirectional exchange of carbohydrates from plant to fungus and minerals (mainly phosphorous and nitrogen) from fungus to plant. Such exchange suggests an intense involvement of mycorrhizal fungi in carbon and various mineral elements cycling and their influence on ecosystem processes (Rillig 2004). Therefore, the potential impact of mycorrhizal fungi on biogeochemical cycles and plant transfer of radiocaesium and other radionuclides was suspected. Nevertheless, the impact of these fungi on radiocaesium accumulation by plants was not investigated (Declerck et al. 2003). Recent studies reported the ability of these plant-associated organisms to restrict radiocaesium transfer to plants (Dupré de Boulois et al. 2008).

2 Radiocaesium Accumulation by Ectomycorrhizal Fungi

The first studies of radiocaesium uptake by mycorrhizal fungi started when high concentrations of this radioisotope were recorded in mushrooms (Grueter 1971). After the Chernobyl accident, Kalac (2001) and Mietelski et al. (2010) observed that ectomycorrhizal (ECM) fungi have an extremely high capacity to concentrate ^{137}Cs and accumulate this radionuclide 10–150 times more from soil habitats as compared to plants growing in their nearest vicinity. It has been calculated that the ECM fungal component of soils is able to immobilize between 10 and 100 % of the total radiocaesium activity (Dighton et al. 1991). Besides, the average ^{137}Cs concentration in symbiotic ECM fungi was also considerably higher as against saprophytic and parasitic fungi. Radiocaesium originated from various technogenic sources is generally distributed in differing depths within soil profile. This fact often influences the radionuclide concentration in aboriginal fungi. Thus, organisms with deeper penetrating mycelia have a tendency to preferentially accumulate increased inventories of ^{137}Cs originated from nuclear weapon testing, while those with shallow mycelia are predisposed to uptake the radiocaesium deposited from Chornobyl fallout (Mietelski et al. 2010).

It was observed that ^{137}Cs levels tend to be lower in the underground ECM fungal mycelia than in the aboveground fruiting bodies (Vinichuk and Johanson 2003). Heinrich (1992) noted that the stems and caps or fruiting mushrooms bodies are richer in ^{137}Cs as compared to their mycelium. Furthermore, Cs^+ is accumulated in mycelium and fruit bodies of ECM fungi by simple diffusion and facilitated transport.

Consequently most Cs^+ bounds within the ECM fungal tissues rather than the fungal surface. Based on these reports, it was estimated that significant part of radiocaesium isotopes located in the soil is accumulated and retained by fungi in an "immobile" condition for a long time. Due to such continuous upward remobilization, the mycelia of ECM fungi could slow down the vertical migration of radiocaesium in soils and consequently maintain this radionuclide in upper soil layers replete with plant roots and mycorrhizal mycelium. In turn, the radiocaesium immobilized in fungal structures could be transferred by the mycelia to their plant hosts. Besides, Thiry et al. (2009) showed that following aboveground deposition, radiocaesium present in plant tissues could be transferred backward to the ECM fungi and accumulate in their fruit bodies. All these facts enable to use ECM fungi as indicators of cumulative biogeochemical fluxes of radiocaesium in terrestrial ecosystems and suggest that ECM fungi could participate in plant decontamination. However, only a few studies were concerned with the analysis of mycorrhizal fungi role in the transport of radiocaesium from soil to plants (Clint and Dighton 1992; Riesen and Brunner 1996). In these studies plants associated with ECM fungi had lower root and shoot Cs content as compared to nonmycorrhizal ones. Consequently, it was suggested that ECM fungi could immobilize radiocaesium and reduce its availability to their host plants. Thus, they appear to be appropriate candidates for phytoremediation techniques aimed to stabilize Cs isotopes in the soil.

The estimation of integrated radiocaesium accumulation by ECM fungi within the large areas remains quite problematic due to apparent difficulties in measurement of the underground mycelium biomass and variable yield of fruit bodies as well as several "generations" of fruit bodies during the vegetative period. Thus, in boreal forest ecosystems, the fruit body biomass may vary from 5 to 100 kg ha^{-1} (Shcheglov et al. 2001) and the total reserves of fungal mycelium in the soil can reach 200 g^{-2}. The fruit body/mycelium weight ratio varies from 1/63 to 1/154 for saprotrophic and symbiotrophyc fungi, respectively. The pine forests growing on podzolic and soddy-podzolic soils are characterized with the maximum reserve of total ECM fungal biomass (including mycelium). It was found (Shcheglov et al. 2001) that in some cases the weight of ECM fungal mycelium may reach 20 % of the total weight of the forest litter. Fungal mycelium is the most significant component of soil microbiota since it contributes 88–99 % of the total microbial biomass in the soil. The average yield of ECM fungus fruit bodies in forests ecosystems of Ukrainian Polissya is 15 kg ha^{-1}. The highest mycelium content is attributed to the forest litter and decreases down the soil profile. However, the maximum reserves of mycelium are located within the mineral soil layers because of its higher bulk density (Polaynskaya 1996). Considering the mycelium content, the total fungus biomass is estimated from 213 to 290 kg ha^{-1} in the mixed (deciduous-pine) stand and pure pine stand correspondingly. The experimental data suggest that ^{137}Cs transfer factors (TF, defined as the ratio between the radionuclide activity concentration in the plant (Bq kg^{-1}, d.w.) and its activity concentration in the soil (Bq kg^{-1}, d.w.)) in the fungal mycelium of higher (basidial) fungi is higher than that in fruit bodies by a factor 1.5–2 depending on environmental conditions and availability of radiocaesium. It was demonstrated that 10–62 % of ^{137}Cs activity in the forest soils was accumulated in the fungal mycelium (Bruckmann and

Wolters 1994). With the time, the significant fraction of ^{137}Cs and other radionuclides and nutrients is translocated to their fruit bodies. The fungal biomass demonstrates large seasonal and multilayer fluctuations which indicate a high intensity and relativity of the production processes. Thus, the mycelium biomass may increase in 2–3 times during the growing season (Polaynskaya 1996). Therefore, it is difficult to estimate the corresponding annual production and die back of the mycelium. Taking into account that the average life cycle of fungal mycelium in the soil is about two years, it was assumed that annual mycelium production was about 50 % of the total fungal biomass (Shcheglov et al. 2001).

The maximum intensity of ^{137}Cs accumulation in forests ecosystems of Ukrainian Polissya is usually characteristic for all studied fungal species; however they have a significant variation of TF average values (Kurbet 2007). Thus, in dry forest edatope (a specific combination of soil moisture and nutrient regimes within a certain area) even at levels of ^{137}Cs inventory in the soil about 37 kBq m^{-2}; the concentration of radiocaesium in fresh fruit bodies of edible mushrooms significantly exceeds the permissible levels (Kurbet 2007). For such forest ecosystem, the studied fungal species form the following chain according to a decrease of TF in their fresh fruit bodies: *Paxillus involutus* > *Xerocomus badius* > *Boletus luteus* > *Lactarius rufus* > *Russulales* > *Boletus edulis* > *Lepista nuda* > *Leccinum scabrum* > *Armillaria mellea* > *Clitocybe odora*. The level of ^{137}Cs accumulation in fungal fruiting bodies, regardless of their type, is caused by conditions of the local forest ecosystem. The intensity of ^{137}Cs uptake by fungi is increased with the gain of soil moisture and reduction of soil fertility.

The concentration of ^{137}Cs in the most species of edible fungi with the mycelium located in forest litter (*Xerocomus badius, Cantharellus cibarius* and *Clitocybe robusta*) had been reduced by 30–40 % for the recent (2006–2012) years. However, in fungal species whose mycelium is located in the upper soil layers (*Boletus edulis* and *Russula vesca*), the significant increase of ^{137}Cs activity in fruit bodies is noticed due to downward migration of radiocaesium (Furdychko 2012).

2.1 Arbuscular Mycorrhizal Fungi and Their Role in Radiocaesium Accumulation by Plants

Along with ECM fungi, other soil fungi are involved in radiocaesium immobilization (Rafferty et al. 1997). These fungi are important participants in the Cs cycle in the upper layers of forest soils. They have a strong impact on the mobility of radiocaesium in the soil and result to unavailability of this radionuclide to the other components in ecosystems (Dupré de Boulois et al. 2008). Among the mentioned soil fungi, the obligate arbuscular mycorrhizal (AM) fungal symbionts are supposed to have a principal role (Entry et al. 1996, 1999). Arbuscular mycorrhiza is the eldest and most widespread among all types of mycorrhizas (Smith and Read 1997; Brundrett 2002). Arbuscular mycorrhizas appeared on earth at least 350–400 million years ago, in the early Devonian (Remy et al. 1994). Since then, arbuscular

mycorrhizal fungi have spread throughout the majority of land ecosystems and developed symbiotic interactions with about 80 % of land plant species including agricultural ones (Schüßler et al. 2001). These fungi commonly form associations with herbs, shrubs and tropical trees. AM fungi were recently placed in a new monophyletic phylum, the *Glomeromycota*. Nowadays nearly 150 AM fungal species have been identified. Rather frequently, these fungi constitute the largest part of the biomass among other soil microorganisms and thus occupy an essential position at the soil/root interface (Peterson et al. 2004). AM fungi are obligatory biotrophs. It means that they can develop without a contact with plant roots at only short period of time. The associations of AM fungi with plants were formed during a long co-evolutionary process. The morphological changes in plants resulted in their accommodation to fungus and development of nutrients exchange between these two partners (Karandashov and Bucher 2005). The comprehensive success of arbuscular mycorrhizas in evolution is stipulated by the key role of AM fungi in the capture of nutrients from the soil and in their transfer to the host plant. As a direct consequence, they determine ecosystem variability, plant biodiversity and productivity of plant communities (Turnau and Haselwandter 2002). The AM fungal colonization of plant roots includes stages of signals aesthesis, enhancement and transformation in the root tissues of host plants. After inter- and intracellular colonization of the root cortex, AM fungi develop branched network of extraradical mycelium, which spreads beyond the depletion zone around plant roots. Accordingly, the plant root system obtains access to considerably larger volume of soil. It results to enhancement of the plant/soil nutrients interface (i.e. enlargement of the nutrient absorptive surface zone around roots) and accretion of AM fungal biomass due to the growth of extraradical hyphal network (Smith et al. 2004). The mutual interaction of extraradical hyphal network and intraradical structures of AM fungi stipulates an intensive symbiotic bilateral transport of carbon compounds from plant to AM fungi and nutrients from AM fungi to plant.

In both epidermal and cortical cells of plant roots, the AM fungus is surrounded by a membrane of host origin. The formation of this apoplastic interface causes an essential structural re-organization of the plant cells with all their organelles. The bidirectional transfer of nutrients from the plant to the fungi and vice versa occurs through the arbuscules or intraradical hyphae (Brundrett 2002). The plants provide AM fungi with organic carbon in form of carbohydrates (mainly as hexoses) contained in plant photosynthesis products and required to complete the fungal life cycle. In this way, up to 20 % of the host plant's photosynthate carbon may be transferred to the fungal structures (Pfeffer et al. 1999). In turn, AM fungi alleviate the plant with the uptake of nutrients from the soil, such as nitrogen compounds and phosphates.

The AM fungi also act as a physical barrier preventing root infections induced by fungal pathogens (Sikes 2010). Besides, AM fungi protect plants from osmotic stress, participate in soil structuration by producing glycoproteins, modify gene expression of their host and affect plant community structure and ecosystem processes (Dupré de Boulois 2007). Due to such beneficial properties and significant role of AM fungi in plant growth, they attract considerable attention during the

recent decades. The numerous studies of the acquisition of various nutrients by AM fungi had revealed that fungal species could also provide their host plants with elements poorly mobile or/and those with naturally low content in soils. Leyval et al. (1997) reviewed the capacity of AM fungi to transport some nutrients and mineral pollutants to host plants as well as fungal tolerance/sensitivity to those pollutants impact. It was found that such elements as P, N, Zn, Na, S, Cd, Se, Rb, Sr, Y and U were efficiently taken up and transported by AM fungi. At the same time, it is likely that AM fungi can transport Be, Sc, Cr, Mn, Fe, Co, Zr and Tc, but in negligible amounts.

It was demonstrated as well that AM fungi could contribute to the accumulation of various radionuclides in plants by transporting them to their hosts (Joner and Leyval 1997). At the same time, Chen et al. (2005) showed that AM fungi could transform and immobilize radiocaesium and correspondingly limit their toxicity and bioavailability to plants and spread into the soils. Accordingly, plants growing in contaminated soil could obtain benefit from their AM fungal symbiotic partners. Such significant role of AM fungi in stable and radioactive pollutants accumulation by plants could be explained by their influence on plant physiology and gene regulation. Indeed, plants also have mechanisms allowing them to tolerate soil contamination. The modification of plant physiology following mycorrhizal association could influence plant tolerance/sensitivity to environmental pollutants, but also the partition of the pollutants between roots and shoots. Such observations gave an opportunity to develop potential strategies of radioactively contaminated areas phytoremediation using mycorrhizal fungi (Leyval et al. 1997; Gaur and Adholeya 2004).

Nevertheless, the role of AM fungi on the acquisition of radiocaesium by plants remains incompletely understood and controversial. The lack of clear results on the capacity of AM fungi to accumulate or transport Cs could be principally attributed to different and inadequate experimental systems used in previous studies. Also, the concentration of potassium in the soil could have interfered with the capacity of AM fungi to accumulate or transport Cs. Furthermore, the various AM fungi and plants studied could also explain the controversial conclusions obtained since AM fungi and plants symbioses have probably different capacity to accumulate and transport radiocaesium. It is successively suspected that AM fungi could accumulate radiocaesium in their extra radical or intra radical structures transport Cs to their hosts and influence on its distribution among plant roots/shoots.

The number of pot studies of radiocaesium transfer to plants was performed by Campbell and Davies (1997); Gerzabek et al. (1998) and Dushenkov et al. (1999), however, in these experiments authors used soils contaminated only artificially with radionuclides in easily soluble forms (in water solutions). The studies on artificially contaminated substrata showed the various results of AM fungi impact on radiocaesium transport and its acquisition and accumulation by plants (Dighton and Terry 1996; Entry et al. 1999; Berreck and Haselwandter 2001; Rosen et al. 2005). Cs uptake by plants in the presence of mycorrhiza was either lower (Dighton and Terry 1996; Berreck and Haselwandter 2001; Gyuricza et al. 2010a), similar (Rosen et al. 2005; Vinichuk et al. 2013) or higher (Entry et al. 1996; Joner and Leyval 1997; Kripka et al. 2003) as in nonmycorrhizal ones.

The pot experiments concerned the impact of mycorrhiza on ^{137}Cs and ^{90}Sr uptake by plants grown on radioactively contaminated soil from Chornobyl area were carried out by Kripka (2005), who demonstrated the significant increase of radionuclide concentration both in roots and shoots of *Zea mays*, *Sorghum vulgare* and *Medicago sativa* plants colonized with *Glomus intraradices*. These results are contradictory to research with *Plantago lanceolata* plants cultivation (Dubchak 2013) although, the fungal species was the same, but selection of plant species was different what might explain the different results. In the abovementioned study, the pot experiments were carried out on soddy-podzolic soil sampled in highly contaminated areas of Chornobyl zone. The activity concentration of ^{137}Cs in roots *P. lanceolata* plants mycorrhizal with AM fungus *G. intraradices* and that in non-mycorrhizal plants did not differ significantly. At the same time, the 40–70 % decrease of ^{137}Cs activity concentration, and correspondingly the TF values, was observed in aboveground part of mycorrhizal *P. lanceolata* when compared to non-mycorrhizal ones, and it was confirmed by recalculation of radionuclide concentration per single plant. The most of radiocaesium activity in studied plants (60–73 %), both inoculated and non-inoculated with *G. intraradices*, was localized within their shoots.

These results are in good agreement with those obtained by Berreck and Haselwandter (2001) who observed the significant (ca. 30 %) reduction of stable Cs concentration in shoots of pot cultures of *Agrostis tenuis* colonized with AM fungus *Glomus mosseae*. Also, Gyuricza et al. (2010a) found that inoculation of *Medicago truncatula* plants grown into in vitro culture system with *G. intraradices* led to about double reduction of ^{134}Cs activity concentration in plant shoots. The similar pot experiments were carried out with four plant species (*P. lanceolata*, *M. truncatula*, *Lolium perenne* and *Helianthus annuus*) cultivated on soil spiked with ^{134}Cs and inoculated with AM fungus *G. intraradices* (Dubchak 2015). It was demonstrated that arbuscular mycorrhiza changed significantly the uptake of ^{134}Cs and influenced the radionuclide translocation within the plants. In particular, the mycorrhizal colonization resulted in considerable reduction of ^{134}Cs activity concentration, and thus the TF values, in shoots of *P. lanceolata* (20–24 %), *M. truncatula* (31–35 %) and *L. perenne* (52–60 %) as compared to nonmycorrhizal plants (Table 1). The concentration of ^{134}Cs in the root system of *P. lanceolata* and *M. truncatula* species didn't differ for mycorrhizal and nonmycorrhizal plants. The quite contrary results were obtained in the case of *H. annuus* (Table 1), where the AM plants had nearly tenfold higher radiocaesium activity concentration both in their roots and shoots (Dubchak 2015). The un-inoculated plants of *H. annuus* had lowest TF values in their roots and shoots when compared to other studied plant species. Also, *H. annuus* was the only plant species which shoot biomass was significantly higher in mycorrhizal plants (Table 1).

The sunflower was previously shown to be an efficient hyperaccumulator of As and Cd under hydroponic conditions (January et al. 2008) and bioaccumulator of ^{137}Cs and ^{60}Co (Hornik et al. 2005), although, the impact of radioactivity on mycorrhiza colonization in the case of this species has not been studied. According to results obtained by Dubchak et al. (2010), *H. annuus* revealed its ability of ^{134}Cs hyper-accumulation only in the presence of the mycorrhiza. The most significant

Table 1 ^{134}Cs activity concentration (kBq kg^{-1}), ^{134}Cs total activity in roots and shoots of single plant (Bq per plant tissue), ^{134}Cs transfer factors (TFs), root-to-shoot ratios of ^{134}Cs activity concentration and dry weights (mg) of *Plantago lanceolata, Medicago truncatula, Lolium perenne* and *Helianthus annuus* mycorrhizal (M) or not (NM) with *Glomus intraradices* and cultivated on soil spiked with ^{134}Cs (77 kBq kg^{-1})

	Activity concentration in kBq kg^{-1}		Total activity in Bq per plant tissue		Transfer factor (TF)		Root/Shoot ratio	Dry weight of single plant tissues, mg	
	Roots	Shoots	Roots	Shoots	Roots	Shoots		Roots	Shoots
Plantago lanceolata									
NM	60.62 ± 13.61^{a}	87.50 ± 12.33^{b}	0.46 ± 0.16^{a}	2.80 ± 0.58^{a}	0.79 ± 0.18^{a}	1.14 ± 0.16^{b}	0.69 ± 0.16^{a}	7.6 ± 2.3^{a}	32.0 ± 5.0^{a}
M	51.32 ± 9.13^{a}	66.8. ± 11.03^{a}	0.44 ± 0.11^{a}	2.73 ± 0.60^{a}	0.67 ± 0.12^{a}	0.87 ± 0.14^{a}	0.77 ± 0.19^{a}	8.5 ± 1.8^{a}	40.0 ± 7.0^{a}
Medicago truncatula									
NM	106.40 ± 19.98^{a}	132.10 ± 15.51^{b}	2.3 ± 0.2^{a}	12.3 ± 0.6^{b}	1.38 ± 0.21^{a}	1.72 ± 0.20^{b}	0.81 ± 0.28^{a}	24.0 ± 5.0^{a}	93.0 ± 21.0^{a}
M	126.44 ± 11.46^{b}	86.89 ± 20.02^{a}	3.1 ± 0.1^{b}	9.3 ± 0.6^{a}	1.64 ± 0.15^{b}	1.13 ± 0.26^{a}	1.45 ± 0.41^{b}	25.0 ± 5.0^{a}	107.0 ± 25.0^{a}
Lolium perenne									
NM	153.57 ± 9.64^{b}	58.34 ± 12.32^{b}	0.9 ± 0.1^{b}	1.2 ± 0.3^{b}	1.99 ± 0.13^{b}	0.76 ± 0.16^{b}	2.72 ± 0.76^{a}	5.9 ± 1.9^{a}	21.0 ± 5.0^{a}
M	74.43 ± 8.97^{a}	24.19 ± 3.85^{a}	0.6 ± 0.1^{a}	0.7 ± 0.2^{a}	0.97 ± 0.12^{a}	0.31 ± 0.05^{a}	2.82 ± 0.75^{a}	8.1 ± 2.4^{a}	28.0 ± 6.0^{a}
Helianthus annuus									
NM	38.41 ± 3.93^{a}	26.20 ± 1.15^{a}	0.4 ± 0.1^{a}	6.4 ± 1.8^{a}	0.50 ± 0.05^{a}	0.34 ± 0.04^{a}	1.47 ± 0.21^{a}	11.0 ± 3.0^{a}	248.0 ± 36.0^{a}
M	378.93 ± 10.23^{b}	245.35 ± 4.50^{b}	6.5 ± 1.2^{b}	104.1 ± 4.6^{b}	4.92 ± 0.08^{b}	3.19 ± 0.06^{b}	1.54 ± 0.10^{a}	17.0 ± 6.0^{a}	424.0 ± 11.0^{b}

Adopted from Dubchak (2015)

The results are presented as mean ± SD (values within a column followed by the different letters differ significantly at $p < 0.05$)

(about twofold) reduction of ^{134}Cs activity concentration found in shoots and roots of mycorrhizal *L. perenne* plants (Table 1) contradicts the data obtained by Rosen et al. (2005) who revealed that inoculation with arbuscular mycorrhiza significantly enhanced uptake of ^{137}Cs by *L. perenne*. The authors explained their results by the fact that root density of *L. perenne* is extremely high, and in the presence of branched extraradical AM hyphae, it increased ^{137}Cs accumulation in plant shoots. Such contradictory findings demonstrated the importance of different parameters under laboratory conditions influencing the final effect of AM fungi application.

The application of *H. annuus* plants which revealed the hyperaccumulation properties regarding radiocaesium conditioned by mycorrhiza is also questionable for the phytoremediation. Thus, it was demonstrated (Dubchak 2013) that total activity of ^{134}Cs accumulated in biomass of *H. annuus* during three months was 220 Bq. This is only 2.2 % from total ^{134}Cs activity in the pot (100,000 Bq). Extrapolating these data for a longer term and assuming the plant active growth period is about 6 months per year, we can roughly estimate that nearly two decades are needed to remove radiocaesium completely from the soil. This assumption does not take into consideration the environmental factors, such as the Cs inhomogeneous distribution in the soil, the presence of different physicochemical and insoluble forms of the radionuclide in soil and its possible leaching below the 30–40 cm (i.e. outside of plant roots zone) as well as potential impact of another AM fungi and various soil microorganisms on the radionuclide uptake by plants.

2.2 Distribution of Caesium in Plant and AM Fungal Tissues

Only a few studies are related to analysis of caesium uptake and distribution in plant root tissues and AM fungal structures at cellular level (Kripka 2005; Dubchak 2013). Thus, the evidence of AM fungi participation in accumulation and transport of stable Cs isotope to plants was shown in from results of X-ray microanalysis of *P. lanceolata* mycorrhizal root material (Dubchak 2013). Being added in moderate concentration (400 mg kg^{-1}) to the soil substrate as the complete analogue of radiocaesium, the stable ^{133}Cs was found both in extraradical and intraradical structures of AM fungi *G. intraradices* and *G. mosseae* used in this research (Fig. 1a, b). The concentration of ^{133}Cs in the cortex and vascular tissue of plants colonized with *G. mosseae* was approximately the same, whereas plants mycorrhizal with *G. intraradices* had about threefold higher caesium concentration in the root cortex than in the vascular tissue. This fact shows the significant reduction of Cs translocation to aboveground part of plants in case of their colonization with *G. intraradices*.

The numerous arbuscules of *G. mossseae* and *G. intraradices* fungi located within cortical layer cells were revealed and analysed with SEM and EDX techniques in root fragments of *P. lanceolata* plants (Fig. 2a–d). The significant differences in colonization degree of *P. lanceolata* with *G. intraradices* or *G. mosseae* fungal species (Dubchak 2013) confirm the results previously obtained by Berta et al. (1995) and Calvet et al. (2004), who found a greatly *higher* extent of G. intraradices colonization

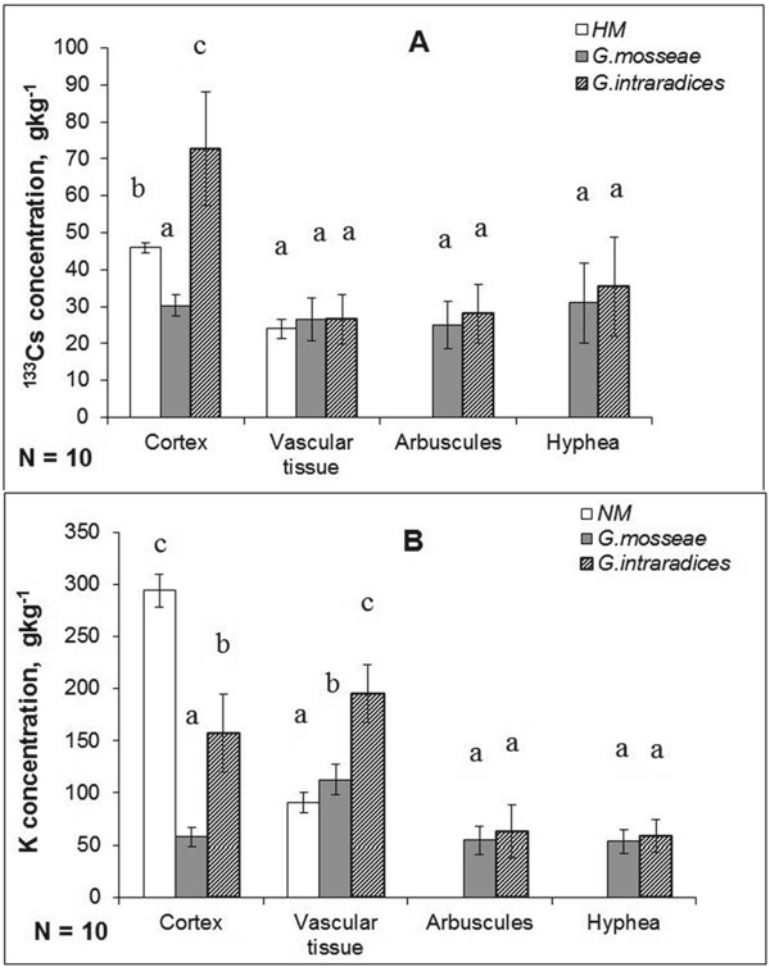

Fig. 1 Concentrations of ^{133}Cs (**a**) and K (**b**) in cortex and vascular tissue of *Plantago lanceolata* mycorrhizal or not (NM) with *Glomus moseae* or *Glomus intraradices*; arbuscules and extraradical hyphae of *Glomus mosseae* and *Glomus intraradices* (g kg^{-1}). Plants were cultivated on substrata spiked with ^{133}Cs (0.4 g kg^{-1}). The results are presented as mean ± SD; the different letters above bars mean statistically significant differences at $p < 0.05$ (Reproduced from Dubchak 2013)

as compared with *G. mosseae*. The distribution of stable Cs and K in arbuscules and extraradical hyphea of both studied AM fungi correlated well (Fig. 1a, b), since ratios of K/Cs concentrations in these structures did not differ significantly. Such similarity in K and Cs localization suggests the existence of common uptake and translocation mechanisms for these elements by AM fungi. The similar conclusions were made by Dupré de Boulois et al. (2005), who supposed that the uptake of Cs is mediated by K transporters as observed for plants.

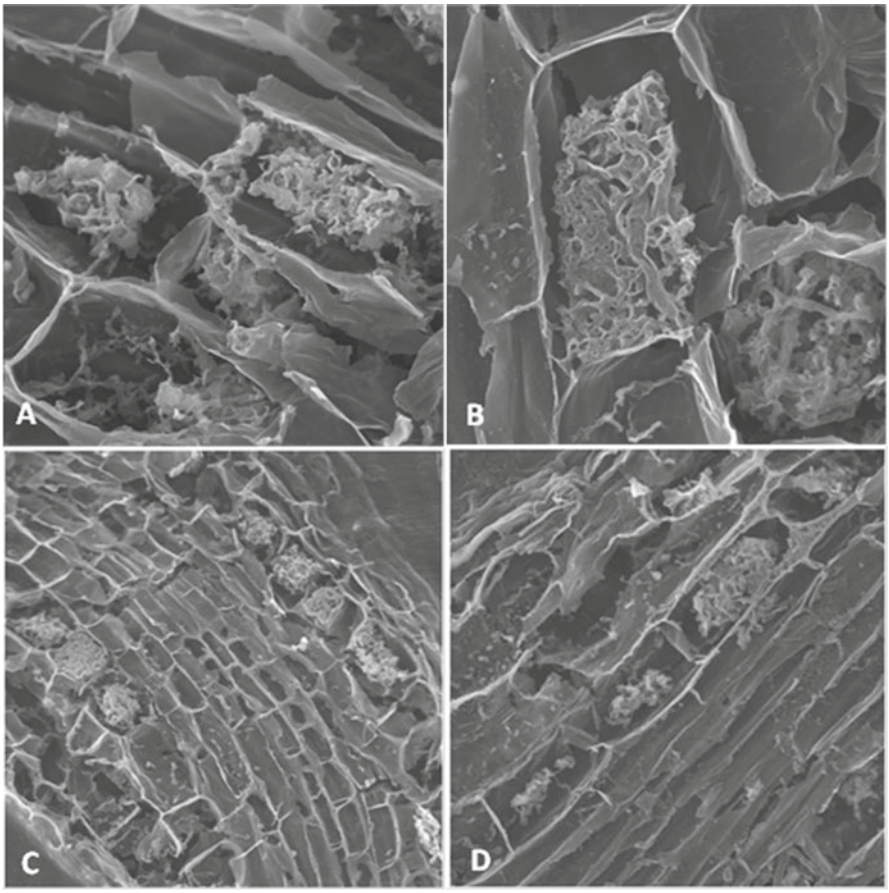

Fig. 2 Arbuscules of *Glomus mosseae* (**a**) ×1500; (**c**) ×500) and *Glomus intraradices* (**b**) ×2000; (**d**) ×750, localized in cortical root cells of *Plantago lanceolata* grown on substrata spiked with ^{133}Cs (400 mg kg^{-1}) (Reproduced from Dubchak 2013)

The translocation of Cs and K within roots (i.e. the ratio of element concentrations in vascular and cortical root tissues) in *P. lanceolata* colonized with *G. intraradices* and *G. mosseae* was differed considerably (Dubchak 2013). Thus, the concentration of K in mycorrhizal plants rose in their vascular tissues in comparison with the root cortex, however such tendency was not noticed for Cs. Accordingly the translocation of K from cortical to vascular tissue of mycorrhizal *P. lanceolata* plants was more intensive than Cs translocation within root tissues of these plants. It demonstrates that significant amount of Cs is retained in the root cortex of mycorrhizal plants since it is not transferred to vascular tissue while K is more efficiently transported from cortical to vascular tissue (Kripka 2005; Dubchak 2013). This result suggests that mycorrhiza limits the Cs transport and at the same time enhances the K transport to aboveground part of plants.

The availability of potassium in the soil strongly affects Cs accumulation in plants due to some chemical similarities of these elements potassium (Zhu and Smolders 2000). The ratios of bulk content of stable K and Cs in the soil are on the order of 100,000 (Joner et al. 2004). This fact as well as the limited bioavailability of radiocaesium in the soil, makes the effective $K/^{134}Cs$ and $K/^{137}Cs$ ratios orders of magnitude higher. But the previously obtained results related to AM fungi influence on K/Cs distribution in plants are rather contradictory. Thus, Gyuricza et al. (2010b) found that additional K content resulted in a lower accumulation of ^{137}Cs in *M. truncatula* plants, but the root to shoot translocation of Cs was not affected. On the other hand, studies carried out by White and Broadley (2000) and Zhu and Smolders (2000) have shown that K can significantly influence ^{137}Cs root to shoot translocation. In the experiments reported by Dubchak (2013), the presence of AM fungi significantly enhanced K accumulation both in underground and aboveground parts of plants. Thus, it was found that the K/Cs ratios in roots and shoots of studied mycorrhizal plant species were about twofold higher than those of nonmycorrhizal ones, except for *P. lanceolata* grown on soil spiked with ^{133}Cs, where the AM colonization resulted in twofold reduction of K concentration in roots. Also, the K/Cs ratios in roots and shoots of *H. annuus* plants mycorrhizal with *G. intraradices* were about ten times lower than those of nonmycorrhizal plants due to the effect of Cs hyperaccumulation by this species (Dubchak 2013). The number of authors proposed using of AM fungi in phytoremediation strategies for radiocaesium-contaminated areas to enhance radionuclide removal by plant biomass (Entry et al. 1996; Zhu and Shaw 2000; Dubchak et al. 2010; Dutov and Dubchak 2014). On the other hand, the effects of AM fungi on Cs accumulation could be applied in strategies to develop crops with smaller soil-to-plant transfer factors which accumulate less Cs (White et al. 2003; Willey 2005; Gyuricza et al. 2010a). Such plant species may be potentially grown within areas with moderate radiocaesium contamination levels and further used for agricultural purposes, if the radionuclide concentration in their biomass does not exceed the prescribed permissible levels.

Summarizing aforesaid, the additional data are needed to test the sensitivity of broader group of plant species to joint impact of radiocaesium and other AM fugal species. The pilot pot and field experiments (Gyuricza et al. 2010b; Vinichuk et al. 2013; Wiesel et al. 2015) demonstrated that different AM fungi could have different effects on radiocaesium uptake by plant species. Obviously, under the field conditions plants could be colonized with the much broader range of AM fungal species. Thus, the presence of five AM fungi morphotypes was revealed in roots of *P. lanceolata* collected from two sites within the 10 km zone around Chornobyl nuclear power plant. The most common species was *Glomus irregulare,* and slightly less common were *Glomus clarum*, *Glomus tenue*, *G. intraradices* and *Scutellospora* (Dubchak 2013). As opposed to laboratory experiments considered above, such variability of AM fungal species in the soil may drastically change the uptake and translocation of radiocaesium by plants.

3 Conclusions

It was demonstrated that the mycelia of ECM fungi could slow down the vertical migration of radiocaesium in soils and retain significant amount of this radionuclide in upper soil layers via accumulation in mycelium and fruit bodies. This fact allows applying ECM fungi as bioindicators of radiocaesium fluxes in forest ecosystems and also as biological organisms that could participate in plant decontamination. However, the common estimation of integrated radiocaesium accumulation by ECM fungi within the large areas is problematic due to difficulties in measurement of the underground mycelium biomass. The studied fungal species in forests ecosystems of Ukrainian Polissya are characterized with the maximum intensity of ^{137}Cs accumulation, but they have a significant variation of TF values

The concentration of ^{137}Cs in the most ECM fungal species with the mycelium located in forest litter had been reduced at 30–40 % for the recent decades. On the contrary, ECM fungi with a mycelium location in the upper soil layers become to be characterized with the considerable increase of the radionuclide activity due to radiocaesium downward migration. The AM fungi are involved in radiocaesium immobilization being important participants in the Cs cycle in the upper soil layers soils. They have strong impact on mobility of radiocaesium bioavailability in the soil to the plant uptake. Nevertheless, the role of AM fungi on the acquisition of radiocaesium by plants remains incompletely understood and controversial. It was demonstrated that uptake of caesium by different plants in the presence of different AM fungi can be either lower, similar, or even higher in comparison to nonmycorrhizal plants.

It was revealed that the activity concentration of radiocaesium in the root system of mycorrhizal plants is not changed significantly. However, the mycorrhizal colonization resulted in considerable reduction of radiocaesium in aboveground biomass of studied plant species as compared to non-mycorrhizal ones. The application of plant species which demonstrated the hyperaccumulation properties regarding radiocaesium under the mycorrhiza impact is considered as rather problematic for the phytoremediation since under the field conditions plants could be influenced by the broad range of AM fungal species.

It is shown that considerable amount of caesium is retained in AM fungal structures as well as in the root cortex of mycorrhizal plants since it is not transferred to vascular tissue, while it's chemical analogue potassium is more efficiently transported from cortical to vascular tissues. It suggests that mycorrhiza limits the Cs transport to plants and at the same time enhances the K transport to aboveground plant biomass.

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Influence of Biologically Active Preparations on Caesium-137 Transition to Plants from Soil on the Territories Contaminated after Chernobyl Accident

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Abstract Wood ecosystems with various density of radioactive pollution of Cherikov district of Mogilyov area was an object of the article. The research of influence of biologically active preparations of "Gidrogumat", "Ekosil" and «Baikal—EM1» on an immobilization of ^{137}Cs in soil and its transition to plants of natural ecosystems of Cherikov district of Mogilyov area was the objective of the work. Experimental platforms that are located in Cherikov district, representative communities with various density of radioactive pollution at the level of 74–185 and 185–555 kBq/m^2, were chosen.

Spectrometer analyses of the ^{137}Cs content in soil and vegetative tests at the selected sites were executed. Factors of accumulation and transition factors were calculated. The least ^{137}Cs accumulation at various densities of radioactive pollution was observed at *Potentilla argentea* L., maximum—at a *Calluna vulgaris* L., among grass- at *Convallaria majalis* L. The definition of the content of forms accessible and inaccessible to plants ^{137}Cs in average samples of soil of the skilled site, selected before the processing by biological products was spent, the quantity of accessible forms varied from 1.66 to 4.31 %. The greatest influence on transition minimization radionuclides in grassy vegetation against 74–185 kBq/m^2 was rendered by a regulator of growth of plants of "Ekosil". There are weak dependences between activity

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of vegetative samples and the content of accessible forms ^{137}Cs in soil, and also the fact of the considerable reduction of the level of the interfaced variation between specific activity of soil and vegetative samples after processing by biologically active preparations was elicited.

Keywords Wood ecosystems • Radio ecological monitoring • ^{137}Cs • Biologically active preparations

1 Introduction

One of the rehabilitation problems of radioactive contaminated territories is the involvement of the polluted territories with high levels of radioactive pollution of soils in economic activities. Therefore it is necessary to develop action complexes directed on manufacture of standard-net production. The research methods of use of biologically active preparations in natural ecosystems is necessary to reveal their influence on an immobilization of ^{137}Cs in soil of natural ecosystems of the Mogilyov area. This will allow estimating possibility of working out of the actions, directed on reducing radioactive polluted production of natural ecosystems. The work objective was the influence of biologically active preparations on a caesium-137 immobilization in soil and its transition to plants of natural ecosystems at Cherikov district of the Mogilyov area.

To achieve this aim the following problems will be solved in the accounting year:

1. To research ^{137}Cs vegetation accumulation of wood ecosystems of Cherikov district of the Mogilyov area at different levels of radioactive pollution of soil.
2. To research ^{137}Cs content by interfaced tests of soil and vegetation at processing carrying out by biologically active preparations in radioactive polluted ecosystems of Cherikov district of the Mogilyov area.
3. To research the influence of biologically active preparations on ^{137}Cs transition from soil to plants.
4. To reveal the ^{137}Cs content in soil in forms accessible to plants.
5. To estimate the influence of biologically active preparations on structure communities of soil organisms.

On the basis of the spent researches, the validity of efficiency of application of biological products for transition decrease of radionuclides in wood vegetation, it will be possible to estimate the possibility of their use at industrial cultivation wood grassy and Low shrubs plants for the purpose of reception of its standard-net production. The researches on use of biologically active preparations in agriculture show their efficiency on the increase of productivity of agricultural cultures, and, thanks to the effect of «biological concentration fall» accumulation decrease radionuclides in them. The mentioned above scientific workings out were spent by employees of Mogilyov branch Research Institute of Radiology together with the Belarus State Agricultural Academy.

In Belarus the Institute of Wood, Research Institute of Radiobiology, Research Institute of Radiology are engaged in problems of pollution of wood ecosystems, but almost all their works are connected with the pollution of woods of the Gomel area and Polesye Radiation Ecological Reserve. The measurement of the content of radionuclides in the production of wood and the forest products directed for processing and other economic needs, is spent by radiological laboratories and posts industrial Wood-economy associations.

2 Objects and Methods of Research

The object of the research is the vegetation of the bottom circle of natural wood biogeocenosis of Cherikov district of the Mogilyov area, located on polluted by radionuclides territories. Cherikov district of the Mogilyov area is one of the most radioactive polluted areas of Belarus with a problem of radioactive contaminations of the wood production. For researches the following methods were used: field experiments, laboratory analyses, and also a rather-analytical method. The selection of the representative communities located on territories with various density of radioactive contaminationat with the level of pollution of 74–185 kBq/m^2 (by 64th wood quarter of the Veprin forest area) and 185–555 kBq/m^2 (65th wood quarter of the Veprin forest area) were made in Cherikov district of the Mogilyov area, where experimental platforms have been placed. On average on a quarter of pollution of №64 density makes 85.1 kBq/m^2, on a quarter N^o 65–392.2 kBq/m^2.

The scheme of carrying out the experiments includes a control background—without processing by biological products and double spraying of plants during the vegetative period on experimental platforms biological products—«Baikal EM-1», "Gidrogumat" and "Ekosil" at two levels of radioactive contaminations. During the experiment there was a triple frequency. The area of an allotment 25 m^2 (5 × 5 m), the area of a variant 75 m^2, frequency triple. Before and after the second processing by biological products on the specified sites selection of the interfaced tests of vegetation (grass and low shrubs and soils) from each site for carrying out the analyses of specific activity ^{137}Cs was led.

«Baikal EM-1» is (EM—effective microorganisms) the microbiological preparation of a new generation combining symbiotic, anabiotic microorganisms, products of their ability to live and a complex of biologically active substances stimulating the growth and development of plants. As the world practice of its use has shown, the given preparation is rather effective for the increase of productivity of agricultural crops, decrease the level of their diseases, and, somewhat, for the increase of qualitative characteristics of received production. The regulator of the growth of plants «Gidrogumat» is a preparation from the peat, consisting from humines and humin similar acids (70–80 %), biologically active low-molecular carbol acids (15–20 %), amino acids (4–5 %). The regulator of the growth of plants with fungicides properties, inductor of immunity of plants "Ekosil" is a preparation of three terpenes acids of wood greens of a Fir Siberian (Evdokimova et al. 2002).

Two processings a year by biological products of “Gidrogumat”, «BaikalEM-1» and “Ekosil” at the chosen experimental sites of wood ecosystems by the spraying method were made. The expense of a working liquid is 20 sm^3/m^2 (200 L h^{-1}). Spray doses of preparations were defined according to the recommendations of the manufactors and the results of the research of application of used preparations for cultural Landings of berries: for «Baikal EM-1»0.5 mL L^{-1}, “Gidrogumat” 0.6 mL L^{-1}, “Ekosil” it is 0.15 mL $L^{-1}$of water. Spectrometers MKC-AT1315 (manufactures of “Atomtex”, Belarus) were used as a measuring apparatus of specific ^{137}Cs activity.

The measurement of a specific activity was made according to the normative document MH 1181-2007 «The Technique of doing measurements performance of volume and specific activity of ^{90}Sr, ^{137}Cs and ^{40}K on spectrometer MKC-AT1315, of volume and specific activity of gamma radiating of ^{137}Cs and ^{40}K on a gamma spectrometer EL 1309 (MKG-1309) in food, drinking water, soil, agricultural raw materials and forages, production of a forestry, other objects of the environment». All applied devices have passed metrological certification and have operating certificates on the state checking.

For the estimation of the volume of transition of caesium-137 in forest production accumulation factors (Кн) were used. Calculation of the factors was made under the formula:

$$\mathrm{KH} = \text{specific activity of the vegetative sample}\left(\mathrm{Bq\ kg^{-1}}\right) : \text{specific activity of soils}\left(\mathrm{Bq\ kg^{-1}}\right)$$

(Gilyarov and Striganova 1987).

Microbiological researches have been spent in fresh samples of soils and in the vegetative rests. In natural mineral soil samples were taken on genetic horizons. The research of number, structure and specific structure were done by using mikrobiotes spent crops method on dense nutrient mediums and direct microscopic methods. A wide set of the nutrient mediums allowing to consider and if necessary to allocate microorganisms with various requirements for nutritious and power substances was used. The number and a biomass of bacteria, the length of the mushroom mycelium and its biomass (without the weight of mushroom dispute) were defined by using the direct methods of fluorescent microscopy with the use of phaeochrous filters with diameter of a 0.2 μm for bacteria and 0.8 μm for mushrooms. The filters for the account of bacteria were painted acrid orange, and filters for the account of mushrooms-dye FITC (fluorestsin-5-izotiotsianat). The estimation was spent in 30 fields of the vision of a standard grid for this method, inserted into an eyepiece. The recalculations the length of mycelium mushrooms and the number of bacterial cages were spent by the standard techniques. All analyses were executed in fresh soil samples, calculations were made on absolutely dry soil. A mushroom biomass was defined, accepting the weight of one millimeter mycelium equal 1.1×10^6. For calculation of a biomass of bacteria the weight of one bacterial cage is considered equal to 4×10^{-14}. The analysis of a natural variety of soil microorganisms was spent on the basis of methods of their detection, allocation, cultivation and identification (Gilyarov and

Striganova 1987). An estimation of fauna of the test wood laying and humus horizon were assorted manually under a binocular magnifier at 16-fold increase. For allocation of hidden meso- and micro faunae they were warmed up within a day by a method of thermogradient extraction. The definition of live weight of Invertebrata was made after movement terminations by their steams of aether by weighing on analytical scales. Invertebrata was identified using Bondarenko and Glushchenko's (1985) determinants, Ilyinsky (1962) and «To the determinant of harmful and useful insects and pincers of annual and long-term grasses and leguminous cultures in the USSR» (Kopaneva 1983). Our researches have shown the possibility of application of biologically active preparations for decrease of radionuclides in wood production. The absence of essential distinctions between communities causes presentation of Invertebrate in an average kind at various density of the radioactive pollution. The factor of a variation of the number and biomass soil Invertebrata paid off, reliability of the received data was estimated by Student's *t*-test (Dospekhov 1985) with the use of the standard software. The comparison of structure of communities in the soil control and processed by biological products was made on ecological indexes (Sorokina 2012). In the course of researches standard materials, results before the spent scientific researches were used.

2.1 ^{137}Cs Vegetation Accumulation in Wood Ecosystems of Cherikov District of Mogilyov Area at Different Levels of Radioactive Pollution of Soil

Radiological investigations of several types of natural wood ecosystems in Cherikov district have been carried out for selection of the most representative communities with the greatest differentiation on radionuclides accumulation in the higher vegetation of the bottom circle. The type of edaphotopes was defined on method of P.S. Pogrebnjak, the wood formations were structured on method of I.D. Yurkevich. The results of the measurements are presented in Tables 1 and 2 in the interfaced tests of soil and grassy vegetation, and also soil and bushes to vegetation taking into account types of conditions of a habitat (edaphotopes). The received results testify to a considerable variation of the level of radioactive pollution of soil and vegetation in the communities.

The tendency of stronger accumulation of^{137}Cs in the vegetative samples growing on more damp soils (B_3—damp soils, B_4—crude soils) is observed. The herbaceous vegetation on fresh (B_2) at a density of soils contamination74–185 kBq/m^2 was observed more in comparison with low shrub vegetation, accumulation of ^{137}Cs. The results of calculations of factors of ^{137}Cs accumulation, made on the basis of our measurements of specific activity in the interfaced tests of soil and grassy vegetation, and also soil and low shrubs vegetation accordingly, are presented to Table 3.

Analyzing ranks of formations of wood on ^{137}Cs transition from soil in vegetation of the bottom circle, based on average on a formation to values of factors of accumulation

Table 1 The content of ^{137}Cs in the interfaced tests of soil and grassy vegetation

	Edafotopyes					
	B_2	B_3	B_4	B_2	B_3	B_4
Wood formations	Activity of soil, Bq kg^{-1}			Activity of vegetative tests, Bq kg^{-1}		
Density of polluted territories 74–185 kBq/m^2						
Pinewood	344.0	775.4	832.8	926.7	1318.0	1416.5
Birchwood	484.4	648.7	953.5	935.9	1266.6	1722.7
Firforest	588.57	646.8	878.5	946.7	1037.9	1506.3
Aspenwood	653.5	750.8	972.5	1018.4	1373.3	1755.1
Density of polluted territories 370–555 kBq/m^2						
Pinewood	1051.6	1927.5	1951.0	1655.8	2405.5	2409.9
Birchwood	1026.6	1573.1	2265.7	1609.5	2214.3	2546.3
Firforest	1135.6	1518.8	2533.3	1707.5	2211.3	2917.8
Aspenwood	1260.3	1296.5	2233.9	2014.9	2054.7	2538.3

Table 2 The content of ^{137}Cs in the interfaced tests of soil and low shrubs vegetation

	Edafotopyes					
	B_2	B_3	B_4	B_2	B_3	B_4
Wood formations	Activity of soil, Bq kg^{-1}			Activity of vegetative tests, Bq kg^{-1}		
Density of polluted territories 74–185 kBq/m^2						
Pinewood	891.6	632.1	667.6	627.7	576.6	487.0
Birchwood	987.3	1026.6	467.8	949.7	948.3	829.5
Firforest	1037.6	1162.2	1007.8	794.8	1082.5	532.9
Aspenwood	610.9	519.9	912.6	868.5	485.7	925.0
Density of polluted territories 370–555 kBq/m^2						
Pinewood	1279.6	1261.0	1780.6	1869.7	2065.6	4211.4
Birchwood	1517.9	1736.6	1392.4	2532.0	2123.1	2536.1
Firforest	1262.0	1424.9	1470.6	2001.4	2104.5	3211.0
Aspenwood	1416.6	1158.2	1513.6	2203.6	1829.7	3266.8

in vegetation, it is necessary to notice that at fir groves the minimum averages on a formation of value of factors of accumulation were observed, and we should take into account their strong variation depending on Edafotopes.

At other analyzed formations the change of ranks with various density of radioactive pollution was observed. In particular, birch forests have replaced a rank from the first on level of values of factors of accumulation ^{137}Cs against 74–185 kBq/m^2—with the second against 370–555 kBq/m^2, pine forests—from the third on the first, aspen forests—from the second on the third accordingly.

The influence type of Edafotopes on values of factors of ^{137}Cs accumulation ambiguously also depends on the vegetation form. The increase in degree of humidifying from fresh (B_2) to crude (B_4) soils slightly raises ^{137}Cs transition in grassy vegetation without dependence from density of radioactive pollution and strongly increases transition in low shrubs. Thus, at higher level of radioactive pollution

Table 3 ^{137}Cs accumulation factors in wood vegetation

Ranks of formations	Wood formations	Edafotopyes					
		B_2	B_3	B_4	B_2	B_3	B_4
		Accumulation factors in grassy vegetation			Accumulation factors in low shrubs vegetation		
Density of polluted territories 74–185 kBq/m^2							
1	Birchwood	1.9321	1.9525	1.8067	0.9619	0.9237	1.7732
2	Aspenwood	1.5584	1.8291	1.8047	1.4217	0.9342	1.0136
3	Pinewood	2.6939	1.6998	1.7009	0.7040	0.9122	0.7295
4	Firforest	1.6081	1.6047	1.7146	0.7660	0.9314	0.5288
	Average	1.9481	1.7715	1.7567	0.9634	0.9254	1.0113
Density of polluted territories 370–555 kBq/m^2							
1	Pinewood	1.5987	1.5848	1.1363	1.5556	1.5798	2.1583
2	Birchwood	1.5746	1.2480	1.2352	1.4612	1.6381	2.3652
3	Aspenwood	1.5036	1.4560	1.1518	1.5859	1.4769	2.1835
4	Firforest	1.5678	1.4076	1.1238	1.6681	1.2226	1.8214
	Average	1.5612	1.4241	1.1618	1.5677	1.4794	2.1321

^{137}Cs transition in low shrubs the vegetation raises depending on a hydrotop in 1.6–2.1 times. Besides, it is necessary to notice that the maximum values of factors of ^{137}Cs accumulation are observed at low shrubs vegetation on crude soils against 370–555 kBq/m^2 in birch forests, aspen forests and pine forests, slightly lower in fir groves. In grassy vegetation, on fresh (B_2) as a hydrotop soils at density of radioactive pollution of 74–185 kBq/m^2 highest^{137}Cs transition in plants was observed, in comparison with low shrubs vegetation.

Summarizing, it is necessary to notice that the obtained data shows a considerable variation of ^{137}Cs pollution grassy and low shrubs to wood vegetation. The least value of factors of ^{137}Cs accumulation in vegetation of the bottom circle on the average on phones of radioactive contamination is noted in fir forests.

At density of radioactive pollution of 370–555 kBq/m^2 ^{137}Cs transition in low shrubs the vegetation raises depending on a hydrotop in 1.6–2.1 times in comparison with density of pollution of 74–185 kBq/m^2. At the same time analyzing the transition of radio caesium in grassy vegetation it is possible to notice that the increase of humidifying of soil does not lead to transition substantial increase radionuclides in plants. Thus, it is necessary to note ambiguous influence edaphic conditions on ^{137}Cs transition in wood vegetation of the bottom circle. We consider that it is necessary to continue the research of the presented problem.

2.2 Research of Specific Specificity of ^{137}Cs Accumulation by Plants of Wood Ecosystems

The results of research of species-specific accumulation radionuclides in wood vegetation have shown that the greatest ^{137}Cs accumulation was typical for long-term low shrubs forms. Thus, maximum accumulation was in bilberries (*Vaccinium myrtillus* L.), the ^{137}Cs content in which was in a range from 873.1 Bq kg^{-1} (at the density of the territory pollution of 74–185 kBq/m^2) to 5879.3 Bq kg^{-1} (at the density of 370–555 k Bq/m^2), and in berries from 843.7 to 1857.4 Bq kg^{-1} respectively. Cowberry (*Viccinium vitis-idaea* L.) stood second in accumulation of radionuclides. In the given kind the ^{137}Cs content fluctuated in a range from 456.7 to 3578.3 Bq kg^{-1}. Among the research of grassy vegetation the highest ^{137}Cs content was observed at a lily of the *Convallaria majalis* L.; it was in a range from 475.3 to 1956.1 Bq kg^{-1}. Other kinds were less accumulating. In the *Sedum telephium* L. the ^{137}Cs content fluctuated from 356.4 to 1712.7 Bq kg^{-1}, in the *Potentilla argentea* L. —from 283.1 to 1473.6 Bq kg^{-1}, *Potentilla erecta* (L.) Hampe—from 271.4 to 1289.9 Bq kg^{-1}, in the *Asarum europaeum* L.—from 267.1 to 1258.3 Bq kg^{-1}.

2.3 Definition, Forms of ^{137}Cs Content in Soil

The content of ^{137}Cs in accessible and inaccessible forms in plants allocated in soil were researched. It is necessary to notice that without dependence from density of radioactive pollution of the territory under grassy vegetation accessible ^{137}Cs forms in soil were low: from 1.66 to 3.28 %. However, observable considerable ^{137}Cs accumulation in vegetation can be connected to plants act as accumulators of given radionuclide because of the morpho-physiological features.

Analyzing content of ^{137}Cs forms in soil under Low shrubs vegetation, it is possible to note a similar picture: accessible forms make from 1.79 to 4.31 %. Thus, it is possible to make the assumption: despite low level of accessible ^{137}Cs forms in soil there is its accumulation both in low shrubs, and in grassy vegetation that, probably, is connected with features of a metabolism of plants. We consider that this point requires the further, deeper research.

2.4 Factors of ^{137}Cs Accumulation and Transition in Wood Vegetation

^{137}Cs accumulation factors, calculated on the basis of spectrometer measurements of the specific activity of the interfaced soil and vegetative tests, are resulted in Table 4. The analysis of the presented data shows that in comparison with control the greatest influence on accumulation decrease of radionuclides in grassy vegetation at the

Table 4 ^{137}Cs accumulation factors in vegetation of wood ecosystems

No.	The variant of application of a preparation	Vegetation forms	
		Grassy	Low shrubs
Density of polluted territories 74–185 kBq/m^2			
1	Control (withoutprocessing)	6.529	13.137
2	Baikal EM-1	4.304	3.408
3	Gidrogumat	6.621	11.904
4	Ekosil	2.490	9.189
Density of polluted territories 370–555 kBq/m^2			
5	Control (withoutprocessing)	2.162	2.802
6	Baikal EM-1	5.886	1.537
7	Gidrogumat	7.462	4.289
8	Ekosil	5.615	3.766
The least essential difference		0.846	1.148

density of pollution of soils of 74–185 kBq/m^2 was rendered by a regulator of growth of plants «Ekosil». Quite good results were received at preparation using of «Baikal EM-1». However at processing of the grassy vegetation growing on soils with density of pollution of 370–555 kBq/m^2, the return tendency is revealed. Accumulation factors at the use of all specified biological products have appeared higher, than on a control site. Estimating the influence of biological products on low shrubs vegetation it is necessary to notice that the maximum effect on receiving radionuclides decrease in low shrubs vegetative forms has rendered by a microbiological preparation «Baikal EM-1» which action was noticed on the territories with various density of radioactive pollution. At the same time at processing by other biological products insignificant decrease in factors of accumulation was observed in low shrubs the vegetation growing only on soils with density of pollution of 74–185 kBq/m^2. The data of transition factors presented in Table 5 confirms earlier specified supervision about efficiency of the use of bioactive preparations “Ekosil” and «Baikal EM-1» for radionuclides transition decrease in wood plants. Estimating transition factors, it is necessary to note their considerable variation: from 0.0051 to 0.0438, at the same time their smaller values are characteristic for low shrubs vegetation against 370–555 kBq/m^2; thus they are authentic lower, than against 74–185 kBq/m^2. Thus, it is possible to make the assumption that biologically active preparations make various impacts on transition radionuclides from soil to a plant, but deeper research of this problem is required.

2.5 Induced Succession of Rhizosphere Communities

The most serious characteristics successions processes in cenoses are changes of number (pool), a biomass and specific structure of communities. In Table 6 the results of the research of an aggregate number and a biomass of bacterial cages in gumus-accumulative (A_1) and alluvial (A_2B) horizons of soil of an experimental site are presented.

Table 5 ^{137}Cs transition factors in vegetation of wood ecosystems

No.	The variant of application of a preparation	Vegetation forms	
		Grassy	Low shrubs
Density of polluted territories 74–185 kBq/m^2			
1	Control (withoutprocessing)	0.0218	0.0438
2	Baikal EM-1	0.0143	0.0114
3	Gidrogumat	0.0221	0.0397
4	Ekosil	0.0083	0.0306
Density of polluted territories 370–555 kBq/m^2			
5	Control (withoutprocessing)	0.0072	0.0093
6	Baikal EM-1	0.0196	0.0051
7	Gidrogumat	0.0249	0.0143
8	Ekosil	0.0187	0.0126
The least essential difference		0.0011	0.0009

Table 6 The Aggregate number (×10^{9}cell g^{-1} soils) and a biomass (mg g^{-1}) of bacterial cages in soil of a skilled site

No variant	An experience variant	Horizon	Number, ×10^{9}cell g^{-1} soils	Biomass, mg g^{-1}
1	Control	A_1	19.11	0.78
		A_2B	4.43	0.21
2	Processing by a preparation of "Gidrogumat"*	A_1	19.41	0.81
		A_2B	4.61	0.23
3	Processing by a preparation of "Ekosil"*	A_1	24.07	0.97
		A_2B	6.34	0.31
4	Processing by a preparation «Baikal EM-1»*	A_1	41.71	1.55
		A_2B	9.57	0.49

The note—*—is authentic at p = 0.05

The maximum pool of microorganisms in gumus-accumulative horizon is noted in a variant, with processing plants by microbiological preparation «Baikal EM-1». Processing by it has the most considerable contribution to the growth of quantity of bacterial cages (on 118.3 % in comparison with control) and their biomass (on 98.7 % in comparison with control) in soil of the site. At comparison of effects of influence of investigated preparations on number and on a biomass rhizosphere's microbiota's, it is necessary to notice that application of "Gidrogumat" does not promote its authentic changes. At the same time the application of "Ekosil" has led to the growth of number of bacterial cages in comparison with the control on 26.0 %, and a biomass on 24.4 % at the expense of processes of stimulation of the growth and development of organisms. Some impact on the number of bacteria and their biomass in alluvial horizon (A_2B) soils «Baikal EM-1» is made. Plants processing by this preparation led to increase of the general pool of microorganisms in 2.16 times in comparison with control, and a biomass in 2.33 times in comparison with control.

Thus, it is possible to make a conclusion that processings by a microbiological preparation «Baikal EM-1» lead to considerable increase of the general pool (in two and more times) and a biomass of bacterial cages both in humus-accumulative and in podzolic horizons of a skilled field. In Table 7 the results of the research of the taxonomic variety and frequency of occurrence of bacteria in soil of the site are presented. It is necessary to notice that in control the highest occurrence is noted at *Bacillus pumilus, Meyeret gottheil, Bac. subtilis Cohn* and *Cyanobacterium* sp. The application of preparations "Gidrogumat" and "Ekosil" has led to the increase in frequency of occurrence of the above-stated microorganisms. The use of microbiological fertilizer «Baikal EM-1» has led to change of a gradient of occurrence (more than in four times in comparison with control frequency of occurrence *Clostridium nitrificiens* has increased), substantial growth of frequency of occurrence and occurrence of new kinds (*Azotobacter* sp., *Lactobacterium* sp.) in soil community, i.e. to start of succession microcenosis. In Table 8 the calculated indexes of Menhinika, Margalefa, Berger–Parker, Simpson, Shannon–Weaver are presented (Sorokina 2012). The data presented in Table 8 shows change of a parity of dominating kinds in community after the application of the preparation «Baikal EM-1». Polydomination proves to be true low degree of domination on Simpson, high values of indexes of a variety of Shennon–Weaver ($H' = 3.282 - 3.451$ bitg^{-1}), values of Menhinika's index,

Table 7 A taxonomic variety and frequency of occurrence of bacteria in soil of a skilled site, %

	Experience variants			
Sort, kind	Control	Gidrogumat*	Ekosil*	Baikal EM-1*
Azotobacter sp.	0	0	0	8
Bacillus cereus Frankland et Frank.	8	7	11	23
B. Laterosporus Laubach	12	11	13	14
B. licheniformis Chester	9	12	12	23
B. megaterium de Bary	14	14	16	21
B. Mesentericus trevisan	15	16	16	17
B. pumilus Meyer et Gottheil	18	19	21	24
B. subtilis Cohn	16	16	17	31
B. firmus Bredemann et Wemer	13	14	13	21
Clostridium nitrificiens	11	12	14	46
Cyanobacterium sp.	16	17	19	33
Lactobacterium sp.	0	0	0	45

The note—*—is authentic at $p = 0.05$

Table 8 Ecological indexes of a biodiversity microcenosys

Index	Control	Gidrogumat	Ekosil	Baikal EM-1
Menhinika's index	1.044	1.022	0.973	0.686
Margalefa's index	1.562	1.547	1.518	1.332
Berger–Parker's index	0.136	0.138	0.138	0.150
Simpson's index	0.099	0.099	0.098	0.096
Shennona–Weaver's index	3.282	3.278	3.294	3.451

Margalefa's and Berger–Parker's indices confirm absence of mono domination of the kinds. Thus, application of a microbiological preparation «Baikal EM—1» leads to occurrence in soil of the bacteria which before were not seen in control and increase quantity of microorganisms, participating in processes of biodegradation and transformation of organic substances in soil of a skilled site (Shchur and Shumigay 2012a; Shchur et al. 2012a).

In Table 9 the results of the research of a taxonomic variety and frequency of occurrence of mushrooms in soil of a skilled site are presented. From the revealed biodiversity the most often met mushrooms community (mycocenosis) in soil, representatives of sort *Penicillium* are widely presented. And, the use of studied preparations led to the increase in their number that indicates strengthening of the processes of biodegradation of organic substance in soil as the presented mushrooms are saprotrophic according to the type of nutrition. Besides, it is necessary to notice that mentioned mushrooms are capable to develop an antibiotic the penicillin possessing antibiotical and allelopathic action that promotes decrease in number of pathogenic organisms in soil. Besides the described sort, there was an increase of number of mushrooms in soil: *Acremonium butyri* W. Gams, *Mortiereiia longicollis* Dixon-Stewart, *Mortiereiia* sp., *Aureobasidium*

Table 9 A taxonomic variety and frequency of occurrence of mushrooms in soil of a skilled site, %

	Experience variants			
Sort, kind	Control	Gidrogumat*	Ekosil*	Baikal EM-1*
Penicillium canescens Sopp	15	19	22	19
P. cyaneum (Bainier et Sartory) Biourge	12	13	16	11
P. cydopium Westling	10	10	13	12
P. implicatum Biourge	22	21	25	23
P. frequentans Westling	24	25	28	26
P. oxalicum Currie	21	22	25	23
P. puberulum Bainier	19	20	22	20
P. spinulosum Thorn	14	14	16	17
P. steckii Zaleski	17	20	18	22
P. thomii Maire	23	23	25	26
P. varlabile Sopp	17	17	22	24
Penicillium sp.	35	37	39	37
Acremonium butyri W. Gams	21	22	31	32
Fusarium sp.	62	50	32	28
Mortierella longicollis Dixon-Stewart	12	32	31	27
Mortierella sp.	16	34	32	34
Mucor sp.	50	32	31	29
Aureobasidium sp.	21	22	25	22
Trichoderma sp.	13	14	14	20
Mycelia sterilia	0	0.2	0.4	0.5
Ulodadium sp.	7	8	8	10

The note—*—is authentic at p = 0.05

sp., *Trichoderma* sp., *Mycelia sterilia*, *Ulodadium* sp. Probably, it is connected with the action on soil biotas studied preparations. Simultaneously, in soil the frequency of occurrence of some pathogenic, in particular, various representatives of sort *Fusarium* sp. have gone down (Shchur et al. 2012b; Shchur and Shumigay 2012b).

In Table 10 the calculated indices of Menhinik, Margalef, Berger–Parker, Simpson, Shennon–Weaver are presented. The data presented in Table 10 shows similar to microcenosis picture change of a parity of dominating kinds in mycocenosis influenced by the preparation «Baikal EM-1», but thus degree of influence of the specified preparations is not as high as in the case of with microcenosis. Hence it is possible to make a conclusion that the preparations positively influence the number of soil saprotrophic mushrooms, at the same time leads to reduction of number of some pathogenic organisms.

3 Invertebrates Natural Forest Ecosystems

3.1 The Taxonomic Diversity of Soil Fauna

We have researched two size-functional groups of invertebrates that inhabit the natural forest podzols region: micro- and meso fauna. Micro fauna is presented with microarthropoda—Collembola, mites and Acari: Oribatei, Gamasoidea. As part mesofauna is represented with 20 invertebrate taxa: Oligocheta: Lumbricidae, Enchytraeidae; Gastropoda: Stylommatophora; Arachnida: Araneae, Opiliones; Myriapoda: Lithobiidae; Insecta: Homoptera: Cicadellidae, Psyllidae, Aphididae; Hemiptera; Coleoptera: Cantharidae larvae, Carabidae, Curculionidae, Elateridae larvae, Staphylinidae; Formicigae, Hymenoptera, Diptera, Lepidoptera (Lepidoptera larvae), Thysanoptera. The representatives of Diplopoda were not found and neither Isopoda was, usually typical for forest soils of Belarus. This may be due to the dynamic processes in populations and migration. The taxonomic composition of invertebrate fauna is not constant. In different seasons and years in the soil no representatives of various taxa. In particular, a variety of meso fauna increased from 7–8 groups in May to 18–20 in August and decreased to 10–11 groups by the end of October. The increasing diversity in the warmer months of the growing season is connected with the emergence of phytotrophs: bedbugs, psyllites, aphids, thrips, leafhoppers, Hymenoptera, Lepidoptera. In colder months they are partially or completely absent.

Table 10 Ecological indexes of a biodiversity mycocenosys

Index	Control	Gidrogumat	Ekosil	Baikal EM-1
Menhinika's index	1.012	0.984	0.963	0.976
Margalefa's index	2.285	2.265	2.249	2.259
Berger–Parker's index	0.144	0.110	0.082	0.080
Simpson's index	0.066	0.057	0.053	0.053
Shannon–Weaver's index	4.106	4.197	4.248	4.258

3.2 The Number of Soil Fauna

In the researched communities the basis of soil fauna has been formed by microarthropods, which accounted for 98 % of the total density of invertebrate's meso fauna share did not exceed 2 %. Collembola Ticks predominated over, in some embodiments exceeding 91 % of the total microarthropoda (Table 11). To estimate population density, involved in community of invertebrates; the figures of "dynamic density", which characterizes the median number of individuals (instances) per unit area for a certain period were used. The total dynamic density of micro- and macro fauna was about 23×10^3 ind/m^2, dynamic density mite populations 21×10^3, springtails 2×10^3 ind/m^2; meso fauna 522 specimens/m^2.

The variation of the total number of invertebrates was not more than 100 %, indicating the stability of the community in natural forest soils of the region. Acidic sod-podzolic forest soils are characterized by the predominance of small forms and low density of large invertebrates. It happens due to the relatively low productive capacity

Table 11 Soil in vertebrates population density in the control and application of biologically active agents, individual/m^2

No	Groups invertebrates	Applications				Cv ± m, %
		Control	Baikal EM-1	Gidrogumat	Ekosil	
1	Gastropoda	9.7	10.1	10.3	10.2	50.1 ± 14.7
2	Lumbricidae	50.3	53.1	57.7	51.4	14.8 ± 4.1
3	Enchytraeidae	61.3	64.1	61.2	62.9	35.1 ± 8.1
4	Lithobiidae	53.1	49.9	58.3	57.5	27.1 ± 7.3
5	Araneae	88.3	92.1	91.5	93.3	44.1 ± 13.8
6	Opiliones	2.3	2.1	2.4	2.3	13.1 ± 2.3
7	Cicadellidae	1.8	1.9	1.7	2.1	11.3 ± 3.1
8	Psyllidae	2.1	1.9	2.3	2.1	10.9 ± 3.3
9	Aphididae	0.91	1.1	0.87	0.94	8.1 ± 2.7
10	Formicigae	3.7	4.1	3.9	3.6	14.7 ± 3.9
11	Hymenoptera	2.3	2.5	2.4	2.5	6.5 ± 1.8
12	Thysanoptera	102.4	89.7	114.1	109.1	67.3 ± 16.3
13	Lepidopteralarvae	1.3	1.2	1.8	1.5	35.4 ± 9.4
14	Diptera	55.6	60.4	58.0	57.1	39.1 ± 13.3
15	Cantharidae larvae	26.3	22.1	24.6	23.4	28.6 ± 8.1
16	Carabidae	12.1	14.3	13.5	14.2	31.1 ± 12.5
17	Curculionidae	6.3	6.1	6.4	6.2	8.9 ± 1.7
18	Elateridae larvae	5.8	5.3	6.1	5.9	12.3 ± 4.1
19	Staphylinidae	23.8	21.9	26.6	22.7	27.3 ± 7.6
20	Collembola	1.9×10^3	2.1×10^3	2.0×10^3	1.9×10^3	6.7 ± 1.1
21	Acari	20.1×10^3	22.2×10^3	21.1×10^3	22.1×10^3	21.3 ± 7.8
Student'scriterion, t_f		1.85				–

Note: Student criterion, t_{theor} (0.05) = 2.01

of the soil. It was found that the density of radioactive pollution in the studied limits, did not affect the dynamic processes and the number of invertebrate populations. Our results in species composition and abundance of invertebrates and their relations of size-functional groups are comparable with those of other researchers on soil fauna forest (Panfilov 1966; Gilyarov and Chernov 1975; Kozlovsky 1981; Maximova 1998; Stepanov et al. 1991; Laskova 1994). Among the representatives mesofauna in forest soils are most numerous soils and litter moisture-loving forms, thrips, spiders, Diptera, lumbricid, enchytraeids, centipede-litobiitsidy, soldier beetle, rove. They formed the dominant complex of meso fauna, which accounted for 95 % of the total number of its members and the level of total meso fauna throughout the growing season. The remaining taxa are small; their total share does not exceed 10 % of the density meso fauna. Presented in Table 11 data show no significant effects of bioactive agents on the number of invertebrates, as evidenced by the value of *t*-test.

3.3 Biomass Soil Fauna

The quantity of biomass mesofauna averaged 18.32 g/m^2. These data are consistent with the results obtained by other researchers (Panfilov 1966; Gilyarovand Chernov 1975; Kozlovsky 1981; Maximova 1998; Stepanov et al. 1991; Laskova 1994). The dominant groups by weight were molluscs and oligochaetes, as well as the related trophic litobiidy on these groups accounted for 90 % of the total macro fauna biomass (Table 12). The total biomass of the representatives of the remaining taxa does not exceed 10 %, of which only 3 % accounted for insects. The high level of abundance and biomass of earthworms in natural forest soils at low values of these parameters for the whole meso fauna testified a favorable for worms hydrothermal and trophic regime of these soils. Estimating the biomass of invertebrates on options for the use of biologically active compounds, it should be noted the lack of significant differences, deviations from the average for the options do not exceed 5 %. Statistical analysis confirmed the absence of significant differences. Contamination density had no significant effect on the described index.

3.4 Structure of Soil Fauna Community

Describing the structure of domination as the dominant the taxa with abundance and biomass fraction of individuals of 10 % or more of the total value of these indicators was taken, as subdominants taxa—with a share of 5–10 %.

The results show that the representatives of about half of the identified taxa meso fauna sub dominated and dominate in numbers; the percentage of each taxon is not more than 20 % (Table 13). It allows us to consider the dominance largest polydominant structure as consisting of nine dominant groups, small groups (1–5 %) and five taxa represented by single specimens (less than 1 %). Polydominant confirmed a low degree of

Table 12 Biomass in controlling so invertebrate and application of biologically active compounds, mg/m^2

No	Groups invertebrates	Applications				Cv ± m, %
		Control	Baikal EM-1	Gidrogumat	Ekosil	
1	Gastropoda	98.57	102.64	103.67	103.65	34.1 ± 13.2
2	Lumbricidae	6.77	7.15	7.77	6.92	41.3 ± 17.1
3	Enchytraeidae	95.78	100.15	95.62	98.28	13.5 ± 6.3
4	Lithobiidae	151.03	141.93	165.82	163.55	28.1 ± 12.3
5	Araneae	77.21	80.54	80.01	81.59	16.3 ± 7.5
6	Opiliones	It wnd	It wnd	It wnd	It wnd	–
7	Cicadellidae	1.27	1.34	1.20	1.48	85.7 ± 32.1
8	Psyllidae	0.51	0.46	0.56	0.51	88.3 ± 33.7
9	Aphididae	0.18	0.22	0.18	0.19	85.4 ± 39.3
10	Formicigae	1.53	1.69	1.61	1.49	13.1 ± 6.4
11	Hymenoptera	0.49	0.54	0.52	0.54	92.7 ± 43.7
12	Thysanoptera	2.58	2.26	2.87	2.75	29.1 ± 14.7
13	Lepidopteralarvae	10.04	9.27	13.90	11.58	89.3 ± 43.1
14	Diptera	22.55	24.50	23.53	23.16	63.2 ± 32.7
15	Cantharidae larvae	19.02	15.98	17.79	16.92	57.8 ± 24.5
16	Carabidae	17.92	21.18	20.00	21.03	20.1 ± 11.3
17	Curculionidae	7.44	7.21	7.56	7.32	15.7 ± 8.7
18	Elateridae larvae	83.01	75.86	87.31	84.44	89.7 ± 45.1
19	Staphylinidae	20.56	18.92	22.98	19.61	4.8 ± 0.7
20	Collembola	It wnd	It wnd	It wnd	It wnd	It wnd
21	Acari	It wnd	It wnd	It wnd	It wnd	It wnd
Student'scriterion, t_f		1.85				–

Note: Student criterion, t_{theor} (0.05) = 2.01; «It wnd» – It was not defined; « – » – Value did not pay off

Table 13 Performance dominance of soil invertebrates in numbers

No.	Index	Applications			
		Control	Baikal EM-1	Gidrogumat	Ekosil
1	Numbermesofaunagroups	21	21	21	21
2	The number of dominant groups	9	9	9	9
3	The number of dominant groups, %	87.8	88.1	86.7	87.9
4	The number of small groups	7	7	7	7
5	The share of small groups, %	10.1	10.3	10.9	9.8
6	Number of rare groups	5	5	5	5
7	The proportion of the rare groups, %	2.1	1.6	2.4	2.3
8	Shannon–Weaver's index	3.17	3.15	3.11	3.16
9	Simpson's index	0.19	0.18	0.17	0.18
10	Pielou's index	0.77	0.81	0.69	0.78
11	Menhinika's index	0.842	0.846	0.815	0.826
12	Margalefa's index	2.002	2.004	1.98	1.989
13	Berger–Parker's index	0.201	0.183	0.21	0.206

domination by Simpson, and high values of diversity indices of Shannon–Weaver (H′ = 3.11–3.17 bits/ind) and aligned Pielou (e = 0.69–0.81), the values of the indices of Menhinik, Margalef and Berger–Parker monodominant confirm the absence of the species. The stability of the dominant complex dynamics during the year on the number of groups and their share of taxonomic composition and size of dominance indices confirms our conclusion about the meso fauna community's stability in natural forest soils and the absence of significant effects of bioactive agents on the number of animals and the dominant structure in cenosis. Dominance structure meso fauna biomass is also characterized by the presence of polydominant (Table 14). It provides with the less number of taxa—5. The basis of the dominant group are oligochaetes, mollusces and centipedes. At the same time it increases up to 7 groups of rare invertebrates, due to their low biomass. The presence of polydominant Community confirms the importance of Simpson dominance index (C = 0.11–0.13), and high values of diversity indices of Shannon–Weaver (H′ = 3.21–3.29 $bitg^{-1}$) and aligned Pielou (e = 0.67–0.71). Assessing the impact of biologically active preparations on the estimated parameters, should be noted the lack of significant differences in the calculated indices, which shows the lack of the influence on the structure of domination in biomass of the studied communities (Shchur et al. 2010, 2013).

4 Conclusions

The research results showed a significant radioactive contamination of grass and shrub forest vegetation, even at relatively low densities of radioactive contamination, indicating the unsuitability of the specified plants for the use as raw materials

Table 14 Performance dominance of soil in vertebrate biomass

No.	Index	Applications			
		Control	Baikal EM-1	Gidrogumat	Ekosil
1	Number mesofauna groups	18	18	18	18
2	The number of dominant groups	5	5	5	5
3	The number of dominant groups, %	85.4	86.1	84.7	85.8
4	The number of small groups	6	6	6	6
5	The share of small groups, %	12.9	11.3	13.2	12.7
6	Number of rare groups	7	7	7	7
7	The proportion of the rare groups, %	1.7	2.6	2.1	1.5
8	Shannon–Weaver's index	3.29	3.21	3.23	3.27
9	Simpson's index	0.12	0.13	0.11	0.12
10	Pielou's index	0.71	0.67	0.68	0.69
11	Menhinika's index	0.765	0.768	0.744	0.748
12	Margalefa's index	1.94	1.94	1.92	1.93
13	Berger–Parker's index	0.245	0.232	0.254	0.254

in the national economy, the need to conduct continuous monitoring of forest products, and to continue to research the accumulation of radionuclides in forest vegetation. The results indicate the presence of the species specificity of ^{137}Cs accumulation in vegetation and significant effects on the figure of life forms of plants. The lowest accumulation in different densities of contamination was observed in *Potentilla argentea* L., maximum- among shrub vegetation—from *Calluna vulgaris* L., among herbaceous in *Convallaria majalis* L. There was a significant variation of ^{137}Cs in vegetation natural communities. The range of variation of this indicator herbaceous vegetation ranged from 196.2 to 4807.4 Bq kg^{-1}, shrub- from 393.7 to 8358.3. Soil contamination was at the level of 202.8–19329.2 Bq kg^{-1}. By analyzing the shape of ^{137}Cs in the soil under the vegetation, we can conclude that the available forms ranging from 1.66 to 4.31 %. Despite the low number of available forms of ^{137}Cs in the soil under the vegetation, there is its accumulation in perennial vegetation, and in the first year, which is likely, due to the peculiarities of plant metabolism. We believe that this issue requires further, deeper research.

The highest impact, compared to the control, minimizing the transfer of radionuclides in the herbaceous vegetation on the background of 74–185 kBq/m^2 had a regulator "Ecosil" plant growth. Considering the impact of biological products on the shrub vegetation, it should be noted that in the study of the levels of radioactive contamination, the maximum effect to reduce the radionuclide in plant shrub form, has a microbiological preparation "Baikal EM-1".

The coefficients of accumulation of pollution density of 74–185 kBq/m^2 ranged from 0.9272 to 2.5321 in herbaceous vegetation, and from 1.8688 to 5.8739 at the shrub. When the density of contamination of 370–555 kBq/m^2 accumulation coefficients ranged from 1.1003 to 3.7406 in herbaceous vegetation, and from 1.3181 to 5.4899 at the shrub. The coefficients of transition at the density of radioactive contamination of 74–185 kBq/m^2 range from 0.0031 to 0.0084 in herbaceous vegetation, and from 0.0062 to 0.0196 at the shrub. When contamination density of 370–555 kBq/m^2 these parameters herbaceous vegetation ranged from 0.0037 to 0.0125, shrub vegetation—from 0.0044 to 0.0183.

There is a weak relationship between the activities of plant samples and content of available forms of ^{137}Cs in the soil, and also revealed the significant decrease in the level of variation of the dual between the specific activity of the soil and plant samples after treatment with biologically active agents. Practical application of "Ecosil" is recommended for the drug twice a year processing natural berry, located on the territory with the density of radioactive contamination up to 74 kBq/m^2 by a fine spray. Working fluid flow is 20 cm^3/m^2 (200 L/ha). Doze of the product is 0.15 mL L^{-1} of water. This will effectively reduce the content of radionuclides in the treated plants and their products. The use of analysed preparations increases the frequency of occurrence, common pool and biomass of beneficial microorganisms involved in the process of biodynamics and transformation of organic matter in the soil plot. The test drugs to varying degrees are capable of inducing succession microbiocenosys root layer of soil, affecting to varying degrees on the abundance, biomass and occurrence microbiota. The most effective application of the "Baikal EM-1", promoting the emergence of new forms of community of microorganisms

and increase the autochthonous microflora actively involved in nitrogen fixation processes mobilisation of phosphatesand biodegradation of organic matter (Panfilov 1966; Laskova 1981; Stepanov et al. 1991; Maximova 1998; Shchur et al. 2010, 2013, 2014, 2015a, b).

Studies of biological active agents "Baikal EM-1", "Gidrogumat" and "Ecosil" on fauna of soil invertebrates have shown no how any significant effects of these drugs on the abundance, biomass and community structure of invertebrates in intact forest ecosystems in conditions of radioactive contamination of soils. It is also worth noting that the use of biologically active compounds "Baikal EM-1" and "Ecosil" allows you to reduce the content of radionuclides in forest ecosystems in the lower tier of vegetation (Shchur et al. 2010, 2013, 2014, 2015a, b). Therefore, we can conclude that it is safe to use such preparations in the conditions of forest ecosystems to reduce the transfer of radionuclides in products.

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The Distribution of ^{137}Cs in Selected Compartments of Coniferous Forests in the Czech Republic

Ivan Suchara

Abstract Sources of the airborne radionuclide ^{137}Cs and published data about the input and redistribution of ^{137}Cs activity concentrations in temperate coniferous forest are briefly evaluated. Information about the crucial global- and Chernobyl-derived ^{137}Cs fallout loads and a most recent minute ^{137}Cs deposition rate derived from the Fukushima radioactive leak across the Czech Republic is reported, along with selected results from two large-scale investigations of the distribution of ^{137}Cs activity concentrations in forest floor humus (in O_h horizons) and in outer spruce bark throughout the Czech Republic (1995–2010). The ^{137}Cs activity concentration distributions and speed of vertical migration of ^{137}Cs were evaluated in forest soil profiles at six localities affected by the Chernobyl fallout. The ^{137}Cs activity concentrations measured in annual and biennial spruce and pine needles, two grass species and one moss at three localities are also presented. Finally, a summary of other sources of data on ^{137}Cs in forest compartments in the country are given.

Keywords ^{137}Cs concentrations • Coniferous forests • Forest humus • Spruce bark • Forest soils • Czech Republic

1 Introduction

Along with other new anthropogenic radionuclides, the technogenic radioactive isotope ^{137}Cs has accompanied human nuclear activities since the mid of 1940s. The first amounts of ^{137}Cs were injected into the atmosphere during tests of nuclear weapons and from accidents in several nuclear power plants. Since then, this radionuclide has become an important component of atmospheric deposition loads entering the environment. Gamma radiation emitted by ^{137}Cs can be dangerous to people and other biota over long-term exposure. Caesium is highly soluble in water and

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hence a very reactive and mobile element in ecosystems. Due to the physical-chemical similarities with hydrated ions of the macronutrients potassium and ammonium, ^{137}Cs is easily absorbed by living organisms and enters food chains. Since the radioactive decay of ^{137}Cs is relative slow (with a physical half-life of 30.17 years), it can endure in ecosystem compartments and in biota for long periods. However, a certain proportion of ^{137}Cs is usually transported out of soil horizons or is excluded by organisms. Hence the effective, ecological and biological half-lives are shorter than the physical half-life. Nevertheless, due to the health risks the activity concentrations of ^{137}Cs in the environment, food and biota have been frequently monitored worldwide.

Early studies soon found that temperate forests are significant interceptors and accumulators of airborne ^{137}Cs. Forests are assemblages of both abiotic and biotic compartments with complex interactions maintained by the cycling of matter and energy. For this reason, it is desirable to investigate the inputs, distribution and fate of ^{137}Cs in forest ecosystems. In line with this, there have been numerous studies on the input and redistribution of deposited ^{137}Cs in cultivated forests along with relationships to other environmental interactions. Knowledge about the distribution of ^{137}Cs in temperate forests is also important because these forests occupy considerable areas of the landscape, where they play environmental protective functions. Moreover, people utilise forest products (timber, mushrooms, edible fruits, game, etc.) and use forests for recreational activities. This chapter provides a short bibliographic summary of the data concerning the behaviour of ^{137}Cs in abiotic and biotic compartments in temperate forests. Then we provide information about the deposition loads of ^{137}Cs across the Czech Republic caused by global fallout and accidents involving nuclear material. Finally, some data are presented about the distribution of ^{137}Cs in selected compartments of coniferous forests as obtained through large-scale and local monitoring campaigns in the Czech Republic between 1995 and 2013.

2 Coniferous Forests as an Important Receptor and Reservoir of Airborne ^{137}Cs

The leak of ^{137}Cs caused by the Chernobyl accident on April 26, 1986 triggered numerous investigations of ^{137}Cs in the environment, including possible health effects, methods of decontamination, etc. Forests in the surroundings of the Chernobyl power plant (30–100 km) were heavily affected by quickly sedimenting ^{137}Cs bound to coarse particles (“hot particles”), which behave differently than radiocaesium included in fine aerosol carriers that can be deposited more than a thousand kilometres away. Since the area of the Czech Republic was mostly affected by this latter long-range transport of Chernobyl-derived ^{137}Cs, our studies have focused mostly on how ^{137}Cs from this background fallout endures in temperate coniferous forest compartments.

Because of the large surface area of the coniferous forest canopy, dry and wet atmospheric deposition in effectively intercepted by adult coniferous forests throughout the entire year. For example, Bunzl and Kracke (1988) found a deposition velocity of airborne ^{137}Cs nine times faster in coniferous forest stands than on grassland or arable soil surfaces. The median accumulated global deposition activity of ^{137}Cs in coniferous forests of Germany was found to be about 800 Bq m^{-2} higher than in grassland before 1984, and it has frequently been found that a majority of aerosol particles containing ^{137}Cs are effectively deposited via wet deposition (Bunzl and Kracke 1988; Hirose et al. 1993; Lauritzen and Mikkelsen 1999). Thus, in the first stage of coniferous forest contamination by ^{137}Cs fallout, most (>80 %) of the deposited ^{137}Cs was intercepted by canopies (Tikhomirov et al. 1993). Due to the effects of wind and precipitation, the majority of this intercepted ^{137}Cs fell directly or via through fall, stem flow and current litter fall to the forest floor surface (Shcheglov et al. 2001). The effective half-life of ^{137}Cs in coniferous canopies is several tens of days during the vegetation period and about 3–4 times longer in winter seasons. Only a small proportion of the ^{137}Cs is absorbed by needles and retained in trees, but this can be translocated into new tissue, similarly as for potassium (IAEA 2002). In the initial phase of forest contamination, the ^{137}Cs activity concentrations in trees decrease in the order needles > outer bark > branches > inner bark > secondary wood.

In the second phase, the deposited ^{137}Cs accumulates abundantly in the forest floor. According to Bunzl et al. (1995), in adult spruce and pine forests this phase takes several months to 2 years. Depending on the intensity of precipitation ^{137}Cs is relocated to the forest floor surface or just slightly deeper. Generally, the vertical movement of ^{137}Cs in forest soils is very slow and a crucial fraction of the soil ^{137}Cs is retained in the uppermost organic soil horizons. The soil matrix itself has a certain absorption capacity, and a significant proportion of the forest soil ^{137}Cs can be accumulated in the living biomass of fungi and microedaphon (Witkamp and Barzansky 1968; Brückmann and Wolters 1994; Rafferty et al. 1997; Huang et al. 2016). Nearly all investigations of forest soils have found maximum ^{137}Cs activity concentrations in the upper humic material, whether from global fallout or more recent nuclear power plant accidents.

In the third phase, when soil ^{137}Cs reaches the fine root zone, understory plants and trees uptake ^{137}Cs and redistribute it into their roots and aboveground organs. Since hydrated ions of ^{137}Cs and potassium are similar, ^{137}Cs accompanies potassium along the transport systems and metabolic pathways. ^{137}Cs thus enters element cycling and is dynamically redistributed among individual forest compartments. Several repeated pulses of increased ^{137}Cs uptake and return to the soil through litter fall may be observed. However, only about 2 % of the total ^{137}Cs amount retained in forest ecosystems is maintained in vegetation biomass. In coniferous trunk wood in particular, ^{137}Cs accumulation is very low and may peak 15–20 years after fallout. The main differences in the redistribution of ^{137}Cs in forest soils and forest vegetation are described below.

2.1 The Movement of ^{137}Cs in Forest Soils

Deposited ^{137}Cs is easily dissolved in precipitation or in the soil solution, but the migration of ^{137}Cs in forest soils depends on physical-chemical properties of the soil matrix. Typical forest soils in temperate coniferous forests are comprised of an uppermost litter layer (horizon O_l), followed by a deeper zone of litter fermentation (O_f) and a horizon of biologically stable humus (O_h). These organic horizons contain about 5–30 % incombustible matter (1 % litter ash and the remaining mineral admixtures). Between the upper organic layers and mineral subsoil is a thin or thicker Ah transition horizon with organic content less than 30 %. Percolating precipitations can deprive upper mineral soil layers of humic, clay or iron compounds (eluvial E horizons), and deeper layers can accumulate these soil compounds (illuvial B horizons). During fallout, the majority of ^{137}Cs is accumulated on the forest floor surface and only minute amounts penetrate along channels and cracks deeper into the soil, even during rainy periods (Matsunaga et al. 2013). Humic matter has been considered to have a just a minor ability to adsorb cations of ^{137}Cs. The increasing efficiency to retain ^{137}Cs in the humus forms fibrimor < dysmor < eumor has been explained by the increasing content of clay mineral admixtures with high ability to fix ^{137}Cs (Kruyts et al. 2004). For example, Lofts et al. (2002) found that a content of the clay mineral illite in an amount less than 1 % in humus absorbed more ^{137}Cs than the remaining humus matter. However, in spite of the common opinion that ^{137}Cs can be only ineffectively and weakly adsorbed on functional groups of humic substances and that the presence of organic matter in soils can decrease adsorption of ^{137}Cs on clay minerals (Valcke and Cremers 1994; Lofts et al. 2002; Stauthton et al. 2002), a crucial proportion of ^{137}Cs from fallouts occurring some decades ago have been still retained in the upper organic horizons (O_h and O_f) of European forest soils (Mahara 1993; Rühm et al. 1996; Ramzaev et al. 2007; Zhiyanski et al. 2008). Some studies have documented that microorganisms sharing organic horizons of forest soils can retain 20–60 % of the total ^{137}Cs inventory in the biomass of organic soil horizons (Gullitte et al. 1994; Parekh et al. 2007). Shcheglov et al. (2014) found effective half-lives of ^{137}Cs in O_l, O_f and O_h horizons in contaminated coniferous forests of 2–3, 8–19 and 20–60 years, respectively.

In subsoils, the clay minerals muscovite < biotite < vermicullite < montmorillonite are the main sorbents of soil ^{137}Cs (Cornell 1993). Cations of ^{137}Cs are incorporated into the interlayer space of frayed edges of clay mineral structures. Illite in particular controls the mobility of ^{137}Cs in forest soils (Fuller et al. 2015). A variable but minor vertical migration of ^{137}Cs has been measured in soil profiles, probably due to movement of colloidal particles, fulvic acids and Fe-Al oxides carrying ^{137}Cs. The downward migration velocities of ^{137}Cs associated with fulvic acids are very small and considerably decrease with soil depth. Only about 5 % of ^{137}Cs in soils are associated with more mobile fulvic acids (Guillén et al. 2015). Bunzl et al. (1995) and Spezzano (2005) found that ^{137}Cs originating from global fallout and from the Chernobyl fallout moved in humic topsoil with comparable speed range, but in mineral subsoil Chernobyl-derived ^{137}Cs migrated twice as fast. Published annual

speeds of ^{137}Cs migration have been derived in various ways using vertical activity concentrations in soil compartments or convection-dispersion equations of spreading radionuclides in an "undisturbed homogeneous" soil matrix. However, most published values for ^{137}Cs mean annual migration speeds (cm year^{-1}) have been obtained by dividing the soil depth (cm) of maximum ^{137}Cs activity concentration or the soil depth above which 50 % of the ^{137}Cs inventory is present by the time (in years) from the relevant fallout event. The apparent burial rate is also calculated by dividing the ^{137}Cs decay constant (0.023 year^{-1}) by the regression line slope when plotting log ^{137}Cs activity concentrations and relevant soil depths (Kirchner et al. 2009; IAEA 2010; Fujiyoshi et al. 2011). Faster migration of ^{137}Cs has been frequently reported for wet soils (Ipatyev et al. 1999). Generally, three basic types of ^{137}Cs activity concentration distributions in soil profiles can be distinguished (Fujiyoshi and Sawamura 2004): (a) an isolated activity concentration in the topmost soil layer, (b) exponential decrease of the ^{137}Cs activity concentration with soil depth, and (c) two separated peaks of ^{137}Cs activity concentrations, one near the soil surface and the second much deeper. However, results of numerous soil studies have shown many variations in ^{137}Cs distributions along soil profiles, caused by various soil and stand conditions operating in the investigated forests.

2.2 *The Redistribution of ^{137}Cs in Forest Vegetation, Mosses and Fungi*

^{137}Cs in solution is easily absorbed by plant roots. The uptake of potassium and ^{137}Cs, at least by dicotyledons, corresponds more or less to the same proportions as they occur in the soil (Bunzl and Kracke 1989). Due to the retention of ^{137}Cs in organic forest soil horizons, forest plants contain higher ^{137}Cs activity concentrations in roots and aboveground biomass compared to the plants outside forests. However, the efficiency of ^{137}Cs uptake is influenced by the plant species (Broadley and Willey 1997) as well as by soil and stand factors (Zhu and Smolders 2000; Shcheglov et al. 2001). A high uptake of ^{137}Cs has been reported (Strandberg 1994; Fawaris and Johansson 1994) for small shrubs, especially heather (*Calluna vulgaris*), and an about one-third-lower uptake for loganberry (*Vaccinium vitis-idaea*) and bilberry (*V. myrtillus*). Ferns can highly accumulate not only heavy metals but with high probability radionuclides as well (Lindner et al. 1994). However, not many plant species have been recommended for the remediation of ^{137}Cs contaminated soils (Lasat et al. 1997). The efficiency of plants in the ^{137}Cs uptake from soils has been frequently evaluated (Calmon et al. 2009) by the aggregated transfer factor T_{ag} (m^2 kg^{-1}), which represents the ratio of the ^{137}Cs activity concentration in the plant body (Bq kg^{-1}) to the ^{137}Cs activity in the surrounding soil (Bq m^{-2}). Increased T_{ag} has been found for species growing in wet soils and plants with a superficial root system (Fesenko et al. 2001). Industrial soil pollution, at least by copper and zinc, changes T_{ag} and ^{137}Cs activity concentrations in plants, for example the grass

Avenella flexuosa in the surroundings of a Cu–Ni smelter had T_{ag} from 0.013 to 0.031 $m^2\ kg^{-1}$ and a ^{137}Cs content of 40–270 Bq kg^{-1} (Bunzl et al. 1999). The transfer factor (activity in plant to activity in soil) for ^{137}Cs uptake by *Avenella flexuosa* in coniferous forest was found to be 0.26–0.97 with a mean of 0.60 ± 0.40 (Wirth et al. 1994). The grass *Molinia coerulea* growing in peat soil with a ^{137}Cs activity of 3.5–7.4 Bq m^{-2} showed ^{137}Cs activity concentrations of 195–342 Bq kg^{-1} (McGee et al. 1992), with the content of ^{137}Cs in this grass peaking in spring and gradually decreasing during the vegetation period (Bunzl and Kracke 1989).

The ratio of ^{137}Cs to stable ^{133}Cs in various parts of growing pine was found to be similar to the same ratio found in the upper forest soil layers (Fogh and Andersson 2001). However, cations of elements with a similar hydrated radius as caesium (mainly K^+ and NH_4^+ but Rb^+ and Ca^{2+} as well) can compete with $^{137}Cs^+$ uptake or adsorption (Shaw and Bell 1991; Marčiulionienė et al. 2015). The potassium transport systems transferring ^{137}Cs discriminate potassium from ^{137}Cs (Zhu and Smolders 2000). However, potassium starvation of plants may increase ^{137}Cs influx rates several times compared to plants fed sufficient potassium (Zhu et al. 2000). In line with this, fertilizing forest stands with potassium decreases the uptake of ^{137}Cs by plants (Zibold et al. 2009). The ^{137}Cs adsorbed by plants is easily translocated from roots or old tissues to intensively growing parts. In compartments of coniferous trees, the ^{137}Cs activity concentrations decrease in the following order: young shoots and needles > outer bark > older twigs and needles > trunk wood. The radial ^{137}Cs distribution of pine trunk wood shows an exponential decrease in a few external rings and then rather stable activity concentrations toward the trunk centre. Hence the 1986 Chernobyl contamination signal is not reflected in increased ^{137}Cs activity concentrations in the rings of relevant age (Soukhova et al. 2003). Surprisingly high ^{137}Cs activity concentrations, higher than in moss or lichen bioindicators, are present in tree bark at sites more affected by fallouts. Coarse bark is a better accumulator of ^{137}Cs than smooth bark (Belivermiş et al. 2010; Cosma et al. 2016).

If pleurocarpous moss plants do not contain large amount of soil and are not affected by throughfall, the ^{137}Cs activity concentration in moss tightly correlates to that in atmospheric bulk deposition (Steinnes and Njåstad 1993). In living parts of the moss *Pleurozium schreberi* in Sweden, mean ^{137}Cs activity concentrations were found to be about 28 Bq kg^{-1} and exponentially decreased between 1961 and 1973, with the effective half-life of ^{137}Cs in mosses calculated to be approximately 4 years (Mattsson and Lidén 1975). Long-term monitoring in a birch-pine forest near Ekaterinburg (Sverdlovsk), Russia, between 1978 and 1995 showed "background" ^{137}Cs activity concentrations in a lichen (*Hypogymnia physodes*) and in ground moss-lichen vegetation that were higher (400–550 Bq kg^{-1}) than in the organic upper forest floor (100–200 Bq kg^{-1}) or tree bark (60–150 Bq kg^{-1}). Shortly after the Chernobyl fallout, the ^{137}Cs activity concentrations in the lichen and moss vegetation increased 20 times to 11,400–12,600 Bq kg^{-1} (Nifontova 1998). The respective typical ^{137}Cs activity concentrations in mosses and forest soil (0–10 cm) in western Serbia in 2005 were 1.5–3.0 and 90–120 Bq kg^{-1} (Dragović et al. 2010). The ^{137}Cs activity concentrations in the lichen *Cladonia stellaris* in southern Finland

exponentially decreased between 1987 (94,000 Bq kg^{-1}) and 2004, from 94,000 to 1600 Bq kg^{-1}, with an effective half-life of ^{137}Cs in this lichen of about 3 years (Puhakainen et al. 2007).

Fungi, in particular the mycorrhizal type, can uptake and bioaccumulate ^{137}Cs retained in humus layers. The accumulation of ^{137}Cs from the background Chernobyl fallout in fruiting bodies is highly variable (3–3000 Bq kg $^{-1}$) depending on the species and current biomass of fruiting bodies (Smith et al. 1993; Duff and Ramsey 2008; Taira et al. 2011). The application of potassium to forest stands (200 kg K Cl ha^{-1}) affected by the Chernobyl fallout of about 35,000 Bq m^{-2} in central Sweden in 1992 significantly decreased the ^{137}Cs activities in small shrubs and in the fruiting bodies of mushrooms (Nilsson 2009). Increased contamination of wild boar meat has recently been reported in central Europe. The aggregated transfer factor (T_{ag}) between forest soil and wild boar meat was found to be 0.004–0.060 m^2 kg^{-1} (Semizhon et al. 2009). The cause of this increased meat contamination proved to be earthworms and fruiting bodies of the autumn mushroom *Elaphomyces granulatus,* both accumulators of ^{137}Cs and favourite foods of wild boars. In the following section, a short survey is presented about the history of ^{137}Cs atmospheric deposition loads throughout the Czech Republic, where large-scale investigations of ^{137}Cs activities have been performed in multiple forest compartments.

3 History of the Atmospheric Deposition Loads of ^{137}Cs in the Czech Republic

The Czech Republic (78,867 km^2, 10.5 M inhabitants), the western half of the former Czechoslovakia, is situated in Central Europe (50.09°N; 14.40°E). Since the 1950s, the territory of the Czech Republic (hereafter abbreviated CZ) has been affected by fallouts of ^{137}Cs derived from three sources.

3.1 Global Fallout

The global fallout of ^{137}Cs was initiated by the first tests and use of nuclear weapons in 1945. In spite of occasional attempts to ban nuclear tests globally (the Partial Test Ban Treaty in 1963 and the Comprehensive Nuclear Test Ban Treaty in 1996) nuclear weapons have continued to be tested. An investigation of the ^{137}Cs deposition records found in dated Arctic ice core layers in Canada revealed that the global fallout of ^{137}Cs appeared in 1945, with a main peak occurring in 1962 (Kudo et al. 1998). The temporal ^{137}Cs activities in the ice core layers well correspond with the frequency, timing, and magnitude of the bombs tested. The median integrated long-term (1954–1984) deposition rates of ^{137}Cs global fallout corrected for the ^{137}Cs

physical decay to 1984 in forest stands in Bavaria (which neighbours the CZ) was 3580 Bq m^{-2}. However, the long-term dry deposition velocity of ^{137}Cs in coniferous forests in Bavaria was 9 times higher than on pastures or arable soils (Bunzl and Kracke 1988). An estimate of the average global ^{137}Cs fallout in central Europe as of January 1, 1970 was about 5050 Bq m^{-2} (Aoyama et al. 2006). Respecting the most recent knowledge of the global fallout in central Europe, we can estimate the total background ^{137}Cs fallout (1945–2015) to be about 1960 Bq m^2. However, the input of global ^{137}Cs fallout to coniferous forest soils may be significantly higher and locally modified by the distribution of forest age, canopy gaps, clear cuttings, eroded slopes, etc.

3.2 Post-Chernobyl Fallout

On April 26, 1986, after the explosion of a reactor in the Chernobyl power plant radioactive material of about 5200 PBq was released into the atmosphere. Long-lasting ^{137}Cs was an important component of this radioactive leakage. For the first few days after the accident, between April 29 and May 8, 1986, plumes crossed the area of CZ. The highest radioactivity in the atmosphere above Prague was measured on April 30, May 3–4 and May 7, 1986 (Bučina et al. 1996). Exponential decreases in ^{137}Cs activity concentrations in the air (Bq m^{-3}) and in the surface ^{137}Cs activity (Bq m^{-2}) in Prague both in 1986 and between 1986 and 2006 were published by Rosina et al. (2008). Similarly, maximum ^{137}Cs activity concentrations in the atmosphere (4 Bq m^{-3}) were measured in Bratislava (in neighbouring Slovakia) in spring 1986 (Povinec et al. 2012). Dry and wet deposition rates of Chernobyl-derived ^{137}Cs across the Europe was modelled by Evangeliou et al. (2013).

Due to the relatively long distance of the CZ territory from Chernobyl (980–1300 km), the most significant proportion of the dry atmospheric deposition in April/May 1986 in CZ was mainly composed of fine aerosol particles carrying ^{137}Cs. Large-scale monitoring of the atmospheric deposition of selected radionuclides on bare soils across Czechoslovakia was implemented during and shortly after the passing of the Chernobyl plume (IHE 1987). Calculated from more than 1100 surface soil samples from across CZ, the mean deposition load was 6500–7600 Bq m^{-2} and the maximum measured values were 80,800–95,000 Bq m^{-2} corrected to the reference date June 16, 1986 (Bučina et al. 1996; Rosina et al. 2008). The ^{137}Cs deposition rate distribution showed high variability, which conformed to the patterns of the local precipitation distribution (0–15 mm) in the period when the non-homogeneous plumes were passing CZ along intricate trajectories (Fig. 1). Generally, a wide belt extending across the mid CZ territory in a NE-SW direction was most affected by the Cs fallout (Bučina et al. 1996; Rulík and Helebrant 2011). The ^{137}Cs activity concentrations in the air and atmospheric deposition decreased exponentially in the following decade, with trace ^{137}Cs activity concentrations of about 1 μBq m^{-3} in the CZ atmosphere measured after 2000. Recent bulk deposition still contains ^{137}Cs,

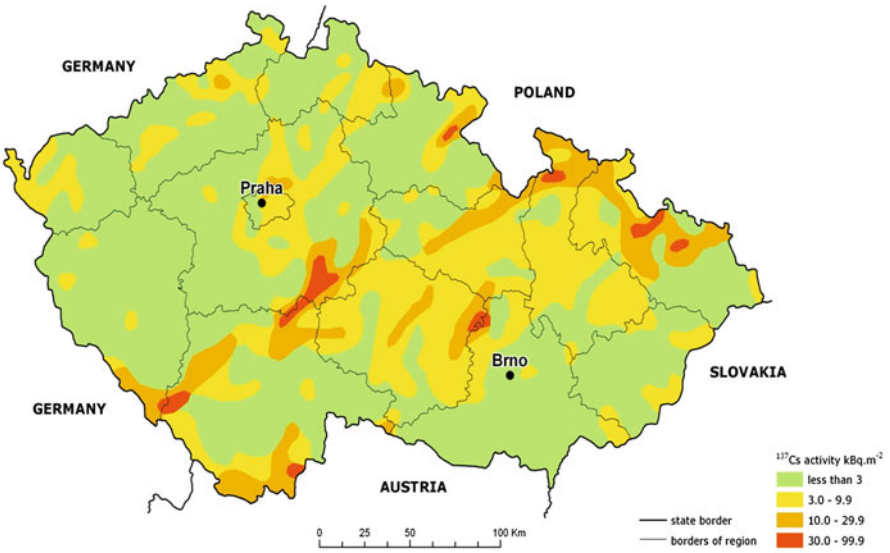

Fig. 1 The interpolated distribution of surface ^{137}Cs activities in CZ ($n = 798$) as measured in June 1986 (Adopted and modified from Rulík and Helebrant 2011). Borders of the actual administrative regions are depicted in the figure

mainly coming from re-suspended (by wind erosion) soil surface particles retaining ^{137}Cs from the former atmospheric deposition loads (Mason et al. 2013).

3.3 Post-Fukushima Fallout

The great earthquake in eastern Japan on March 11, 2011 triggered a devastating tsunami that damaged the Fukushima Dai-ichi nuclear complex and caused an accidental release of radionuclides into the atmosphere, mainly from March 13–16, 2011. The ^{137}Cs release was estimated to be 13–15 PBq (Povinec et al. 2013a, b). The first radioactive plumes from the Fukushima leak reached CZ on March 30–31, 2011. Measurements in central Europe in March and April, 2011 found ^{137}Cs activity concentrations below 0.07 mBq m^{-3}, associated with particles of low aerodynamic diameter 0.2–0.7 μm (Malá et al. 2013). The ^{137}Cs activity concentrations in the atmosphere were about four orders of magnitude lower than the ^{137}Cs activity concentrations in the Chernobyl-derived plumes crossing CZ in April and May 1986. However, the radiocaesium fallout in March 2011 quickly decreased and approached the pre-Fukushima values in May (Mason et al. 2013). The daily trends of the volume activity concentrations of ^{134}Cs and ^{137}Cs (Bq m^{-3}) in the atmosphere in CZ between March and May 2011 is available on the internet (SONS 2015), but no Fukushima-derived ^{134}Cs and ^{137}Cs activity concentrations in forest compartments in CZ have been published.

4 Material and Methods

4.1 Determining the ^{137}Cs Activity Concentrations in Forest Humus

Samples of forest humus (O_h horizon) were collected in the second half of the years 1995 (152 sampling plots) and 2005 (259 sampling plots). The litter (O_l) and organic fermentation horizon (O_f) were scraped off, and from the uncovered humus horizon (O_h) a disk of humus was cut out using a plastic pipe. Depending on the thickness of the O_h horizon 7–12 humus disks were cut out at each sampling plot and combined into one composite sample. From about 10 sampling plots three composite samples were collected in parallel in order to examine plot variability of the ^{137}Cs activity concentrations. Using an O-ring, a defined volume of 100 cm^3 of naturally compressed humus matter (O_h) was collected. These samples were dried (60 °C) and weighed, and then the reduced bulk density of humus was counted ($g_{d.w} \times 100\ cm^{-3}$). The composite humus samples were air dried between sheets of filter paper, and milled and sieved through a 2.0 mm mesh sieve. Humus pH—H_2O and pH—0.01 M $CaCl_2$ (10 g:50 mL) were measured using a glass-electrode pH meter, and the content of the mineral portion (wt.%) after combustion (450 °C/24 h) in a muffle furnace. Details about sampling and analyses of the humus samples can be found in Suchara and Sucharová (2000, 2002). In 2005 the content of total caesium and total potassium were also measured in the humus samples using an ICP-MS method (Sucharová et al. 2011). The ^{137}Cs activity concentrations in the humus samples (Bq $kg^{-1}_{d.w}$) were measured by the National Radiation Protection Institute in Prague by an accredited gamma spectrometry method with coaxial HPGe detectors (ORTEC and Canberra), using calibration sources prepared by the Czech Metrological Institute. The dried samples in containers 200 ml in volume were placed around (2–7 containers) or in front of the detector (1 container). The measuring time was adjusted so that the combined standard uncertainty of the detected ^{137}Cs activity concentration did not exceed 30 %; the average uncertainty was 9 %. Energy and efficiency calibrations were also done. Analyses of the relevant NIST, JSAC and national reference soil, sediment and plant materials were done to check quality of the ^{137}Cs measurements. The detected ^{137}Cs activity concentrations were corrected to the sampling date, and for humidity and density. The minimum detectable activity was between 0.035 and 15 Bq kg^{-1}. The ^{137}Cs activity concentrations in all humus samples were above the detection limit of the method.

4.2 Determining the ^{137}Cs Activity Concentrations in Spruce Bark

Samples of spruce bark were collected at 194 monitoring plots in 1995/1996 and at 255 plots in 2010. Outer bark was collected from adult trees of Norway spruce (*Picea abies* (L.) Karst) of typical age 60–90 years. The 1995/1996 samples were

collected from six trees at each plot, and from ten trees at each tenth plot. In 2010 the bark samples were collected from ten trees at all plots. Only the outermost easily separable pieces of bark about 2–3 mm in thickness were gently scraped off around the tree trunks at a height of 1.3 ± 0.1 m, using a chisel, and stored in individual bags separately for each tree. The collected bark samples were air-dried between sheets of filter paper and milled into powder. The ^{137}Cs activity concentrations were measured in the National Radiation Protection Institute in Prague as described in 4.1. All bark subsamples from each locality (6–10) were measured together, as if they were a composite sample. The readings of the remaining ^{137}Cs specific activities in bark were corrected for the radioactive decay of ^{137}Cs relevant to the date of bark collection. The detection limit (minimum detectable activity at the 95 % significance level) was about 0.6 Bq kg $^{-1}$. The ^{137}Cs activity concentrations in all bark samples were above the detection limit of the method. In parallel, the pH (pH—H_2O) of a bark suspension (4 g:16 mL) was measured with a pH meter and glass combined electrode. More details about collection and measuring the bark samples are available in Suchara et al. (2011a).

4.3 Determining the ^{137}Cs Activity Concentrations in Forest Soils

An investigation of the distribution of ^{137}Cs in forest soils in 2011 was aimed at recognizing the effect of different soil units and differences in the ^{137}Cs distribution in soils below tree canopies and below tree canopy gaps. A subsequent study in 2013 investigated mainly the migration speed of ^{137}Cs in forest soils loaded from the previous fallouts and effect of the Fukushima fallout. The geographical positions of the sampling points, soil and stand characteristics of these two studies are shown in Table 1.

In September 2011, soil samples were collected at three localities (I–III), with three sites below the pine canopy (*Pinus sylvestris* L.) and three below canopy gaps, where no trees had been present since 1986, and in parallel at three sites below the spruce canopy (*Picea abies* (L.) Karst.) and three sites below spruce canopy gaps. A piece of stainless steel shaped into a hollow cube frame with a 20 × 20 cm base and 20 cm in height was gradually pressed into the soil, and soil material from each of the soil horizons was successively scraped off with a steel trowel and quantitatively transferred to plastic bags. Below the forest floor humus (about 5–10 cm), two blocks of mineral soil with dimensions of 20 × 20 × 10 cm were also collected. The upper block of mineral soil contained a thin Ah transition zone between the humus and the mineral soil. Since the boundary of this horizon was usually difficult to recognize, this zone of small thickness was included in the upper block of the mineral soil. In order to investigate the ^{137}Cs activity concentrations in deeper soils affected by the percolation water or by the accumulation of dissolved humic and iron compounds (illuvial soil horizons), the pits were deepened and soil material from a depth of 30–40 cm was also collected in localities I–III. At two sites in locality

Table 1 Localities and basic stand characteristics for investigations of ^{137}Cs distribution in forest soils

Locality (Name)	I (Pivnisko)	II (Dobšice)	III (Čachrov)	IV (Přemyslov)	V (Opatovice)	VI (Průhonice)
Geografical position	49°50′36″N; 15°08′03″E	50°30′32″N; 15°07′08″E	49°15′23″N; 13°18′13″E	50°06′40″N; 17°03′54″E	49°57′45″N; 15°11′46″E	49°59′10″N; 14°32′32″E
Elevation m a.s.l.	530	290	730	800	390	330
Soil unit	Pseudogley (Stagnosol)	Arenic Podzol	Haplic Cambisol	Mesobasic Cambisol	Modal Cambisol	Modal Cambisol
Soil horizons	L-F-H-Ah/ En-(En)-Bm	L-F-H-Ah/ Ep-Ep-(Bhs)-Bs	L-F-H-Ah/Bv-Bv-Bv/IIC	L-F-H-Ah-Bv	L-F-H-Ah-Bv	L-F-H-Ah-Bv
^{137}Cs Chernobyl fallout (Bq m^{-2})	8440	2920	5600	9460	1160	5560

II, a distinct layer with an accumulation of dissolved humus matter, Al, Fe and Mn-oxides, aluminium silicates and clay 2–3 cm in thickness (B(h)s horizon) was found at the beginning of the illuvial soil zone (Bs horizon), and soil material from this horizon was also collected. In parallel, naturally compacted soil matter from the individual soil horizons was cut out using a sampling ring collector with a volume of 100 cm^3 to determine the bulk density. The soil material collected from each of the horizons was air dried, sieved through a 2 mm aperture mesh, and weighed. In the fine soil fraction, the content of organic matter, content of total C and N, quotient $Q_{4/6}$ (ratio of fulvic and humic acids), content of clay, silt and sand and reaction of suspension (pH—H_2O and pH—$CaCl_2$) were measured.

The ^{137}Cs activity concentrations in the fine soil samples (Bq $kg^{-1}_{d.w}$) were measured in the National Radiation Protection Institute in Prague as described in the previous sections. The detected ^{137}Cs activity concentrations were corrected to September 2011, and for humidity and density. The minimum detectable activity was between 0.035 and 15 Bq kg^{-1}.

In September 2013, soil samples were collected at an additional three localities (IV–VI in Table 1), with three sites at each locality sampled exclusively below the coniferous canopy with abundant spruce (*Picea abies*). Soil samples were collected the same way as in localities I–III; however, the size of the sampling cube was 10 × 10 × 10 cm. Material from soil horizons with area 10 × 10 cm was stored in plastic bags. Below the O_h horizon, a transition layer (A_h) between the organic and mineral layers was collected, always with a thickness of 1 cm, below this layer; a deeper 5-cm layer of mineral soil (Ah/Bv or Bv horizon) was quantitatively sampled. Finally, the next deeper 5 cm thick layer of mineral soil (Bv horizon) was collected at each site. The soil material was collected from the soil blocks of the upper area 10 × 10 cm and total depth of 13.1–17.6 cm. All soil samples were air-dried and sieved with a 2 mm mesh.

The fine soil fraction was analysed at the Faculty of Engineering, Hokkaido University, Japan. The pH (pH—H_2O) was measured in a soil suspension and the content of soil organic matter (SOM) by combustion in a muffle furnace. In all samples from the O_h horizons, powder X-ray diffraction (XRD) analyses (RINT-2000 X-ray difractometer, Rigaku, Japan) were carried out to identify the components of mineral admixtures in these organic horizons. Activity concentrations of ^{137}Cs were determined by a gamma spectrometry method using a HPGe detector (GMX10P, EG&G Ortec, USA). Acquisition time was set at 144,000 s for all measurements including a background check. Standard reference materials were purchased from the International Atomic Energy Agency (IAEA-372, Grass) and the Japan Society for Analytical Chemistry (Soil materials JSAC 0471, JSAC 0472 and JSAC 0473) to evaluate the activity concentration of individual samples from counting data obtained with the same geometry under identical operating conditions. The lowest detection limit of the method used for measuring ^{137}Cs activities was 0.34 Bq kg^{-1}. Activities of short-lived ^{134}Cs were also checked in the samples at a 605 keV photopeak on the gamma spectrogram under identical experimental conditions as for the determining of ^{137}Cs activities. The detection limit of the method used for measuring of ^{134}Cs activities was 0.15 Bq kg^{-1}.

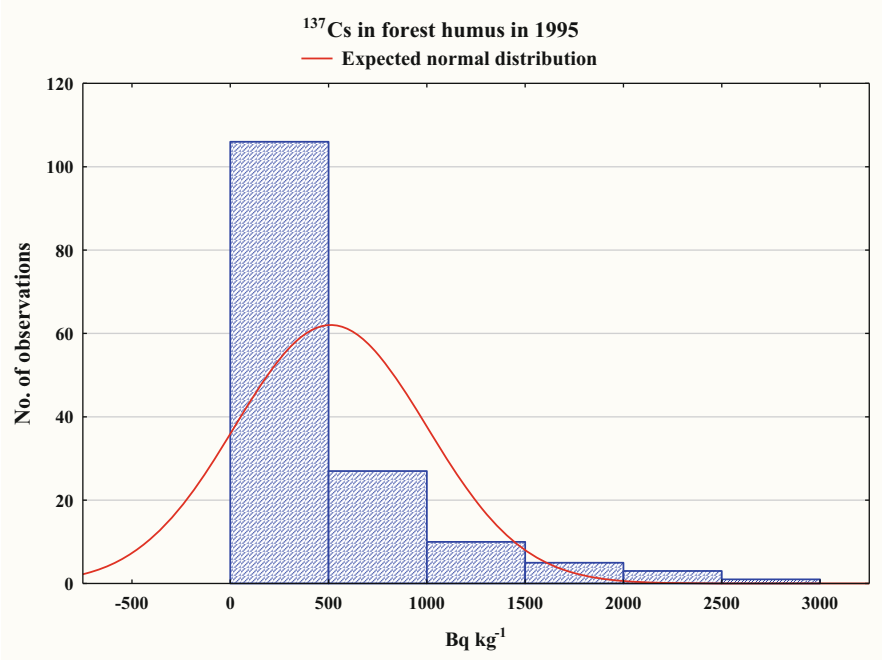

Fig. 2 The ^{137}Cs activity concentration distribution in the humus samples (O_h) collected across the Czech Republic in 1995

4.4 Determining the ^{137}Cs Activity Concentrations in Plant Material

Samples of the above-ground biomass of dominant plant species below canopy gaps and below the canopy were also collected in localities I–III. The plant samples were carefully cut off to avoid contamination by humus or soil. The samples were then dried and powdered in a mill. ^{137}Cs activity concentrations were measured by the gamma spectrometry method in the National Radiation Protection Institute in Prague by the same method used for measuring the humus, bark and soil samples.

5 Results

5.1 The Large-Scale Distribution of ^{137}Cs in Forest Floor Humus (O_h)

The ^{137}Cs activity concentrations in all measured humus samples collected in 1995 showed a log-normal distribution with a clearly positive skew (Fig. 2). Concentrations of ^{137}Cs in the composite samples from 152 monitoring plots ranged between 81 and 2590 Bq kg^{-1}. The ^{137}Cs activity concentration distribution showed the same pattern

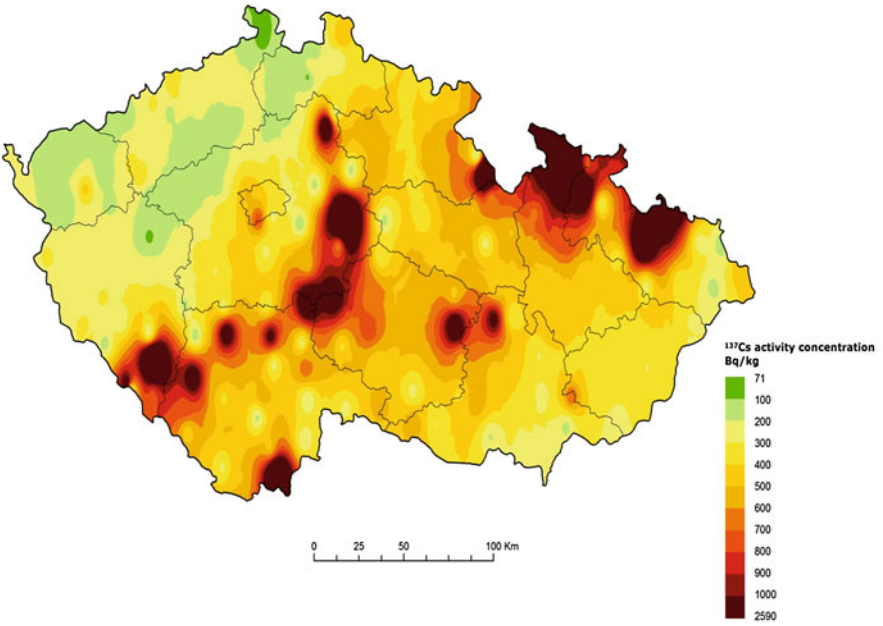

Fig. 3 Distribution of the ^{137}Cs activities in forest floor humus (O_h) in CZ in 1995

as the distribution of local precipitation sums occurring at the time that the Chernobyl-derived plumes were passing CZ (e.g., see maps in Pilátová et al. 2011a). The distribution of the wet deposition of ^{137}Cs in CZ had a crucial effect on the distribution of the total surface activities of ^{137}Cs in the spring of 1986 (Fig. 1). Increased ^{137}Cs activity concentrations in humus in 1995 were found in a wide belt across the central part of CZ in a NE-SW direction (Fig. 3). The ^{137}Cs concentration distribution in humus in 1995 well corresponded with the ^{137}Cs surface activities measured in 1986.

The average ^{137}Cs activity concentration in humus (O_h) at 152 monitoring plots in 1995 was about 350 Bq kg $^{-1}$ (geometric mean 380 ± 110 Bq kg $^{-1}$ and median 330 ± 150 Bq kg $^{-1}$). The typical thickness of the O_h horizons was 2.5–3.5 cm, and the mean reduced bulk density of humus in spruce, pine and mixed forests was 16.8, 15.7 and 15.9 g ×100 cm^{-3}, respectively (Suchara and Sucharová 2000, 2002). No significant correlation between the current humus pH–H_2O (3.02–4.19) and pH-$CaCl_2$ (2.09–3.19) and the ^{137}Cs activity concentration were found. This is in contrast to the common opinion that a majority of ^{137}Cs in forest humus is weakly adsorbed on functional groups of humic macromolecules. In addition, no significant relationship was found between the mass of mineral admixtures in humus (7.6–47.3 wt%) and the ^{137}Cs activity concentrations. This may indicate that a content of mineral admixtures in humus material above 7.6 % does not significantly affect the sorption of ^{137}Cs in forest floors. However, details on the composition of the mineral portion in humus were not analysed.

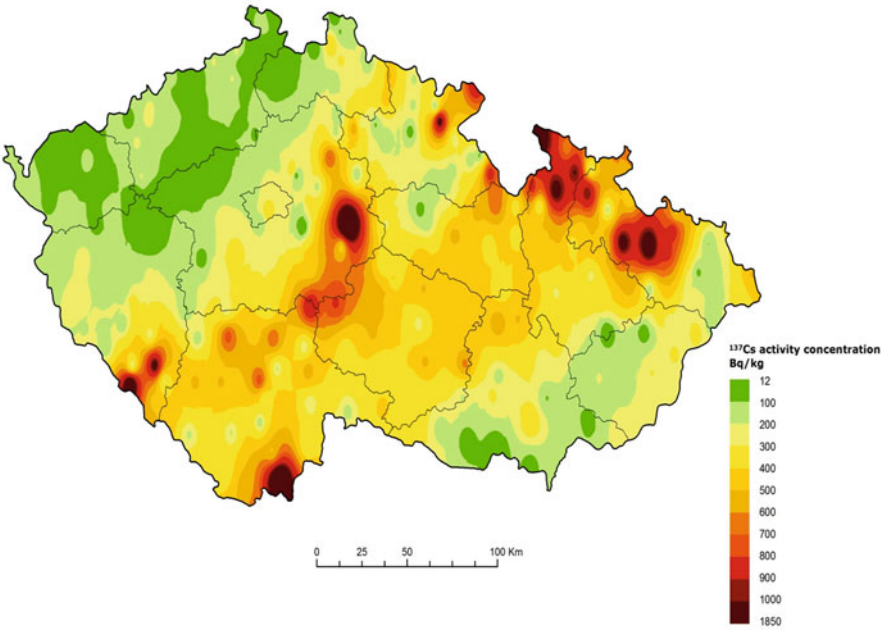

Fig. 4 Distribution of the ^{137}Cs activities in forest floor humus (O_h) in CZ in 2005

In 2005, humus samples collected at 259 monitoring plots were analysed. The ^{137}Cs activity concentrations were in the range 12.2–1850 Bq kg $^{-1}$, with a geometric mean, median and median absolute deviation of 175, 153 and 63 Bq kg $^{-1}$, respectively. These values represent an average decrease in ^{137}Cs activity concentrations by 53.5 % over ten years. Maps of the ^{137}Cs activity concentration in humus in 2005 (Pilátová et al. 2011b and Fig. 4) show a very similar distribution pattern as the ^{137}Cs activity concentration in humus in 1995 (Fig. 3), but with generally decreased ^{137}Cs concentrations. Reductions in the ^{137}Cs activity concentration in humus were evaluated in more detail by comparing radiocaesium data from 152 identical monitoring plots where humus samples were collected and analysed in 1995 and repeated in 2005. The respective median ^{137}Cs activity concentrations in humus decreased from 330 ± 150 Bq kg $^{-1}$ in 1995 to 170 ± 60 Bq kg^{-1} in 2005. When considering physical decay as the only factor reducing the ^{137}Cs activity concentrations in humus between 1995 and 2005, the corresponding median of the radiocaesium activity concentrations in humus in 2005 would have been 260 Bq kg $^{-1}$. However, the actual reduction was about one third lower, indicating the influence of other factors on the decreasing content of ^{137}Cs in O_h horizons (vertical migration, bioturbation, uptake by roots of vegetation).

No significant correlation was found between the humus characteristics (humus pH, mineral portion) and the ^{137}Cs activity concentrations. However, in the humus samples collected in 2005 ($n = 259$) total caesium concentrations (range 0.68–11.3; median 2.17 ± 0.70 mg kg^{-1}) and total potassium concentrations (range 1101–19,390; median 3240 ± 1040 mg kg^{-1}) were also measured because these elements may compete with ^{137}Cs in adsorption on humus. The ^{137}Cs activity concentrations

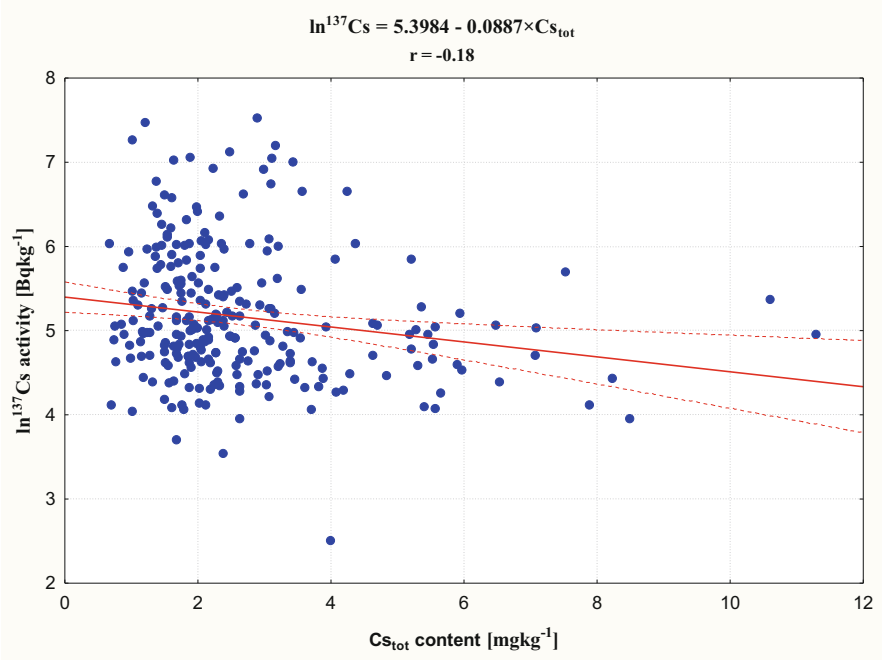

Fig. 5 Relationship between the logarithm (ln) of ^{137}Cs activity concentrations and total caesium concentrations in the forest floor humus samples (O_h) in 2005. A 95 % confidence interval for slope is delimited by dashed lines

in forest floor humus had a decreasing tendency with the rising content of total caesium in humus ($p=0.04$; $r=-0.13$ for raw data). The relationship for logarithms of the ^{137}Cs activities and total Cs content in humus ($p=0.003$; $r=-0.18$) is given in Fig. 5. A substantially increased total caesium concentration in forest floor but in soil, mosses, understory vegetation and annual needles of conifer trees, as well was found (Suchara et al. 2011b; Sucharova et al. 2011) in areas of CZ where granitic pegmatites occur with caesium-rich micas (Fig. 6). More detailed investigations of the potential competition between ^{137}Cs and stable caesium for adsorption sites and in uptake and redistribution in forest compartments are intended in these areas. Nevertheless, no significant correlation was found between ^{137}Cs and total potassium concentration in the humus samples ($r=-0.01$; $p=0.85$), even though potassium can be a stronger competitor than stable caesium.

5.2 *The Large-Scale Distribution of ^{137}Cs in Spruce Bark*

The ^{137}Cs activity concentrations detected in the spruce outer bark samples in 1995 (range 2–318, geometric mean 26 ± 2, median 24 ± 9 Bq kg $^{-1}$) showed a log-normal distribution (Pilátová et al. 2011c). Figure 7 shows the measured and interpolated

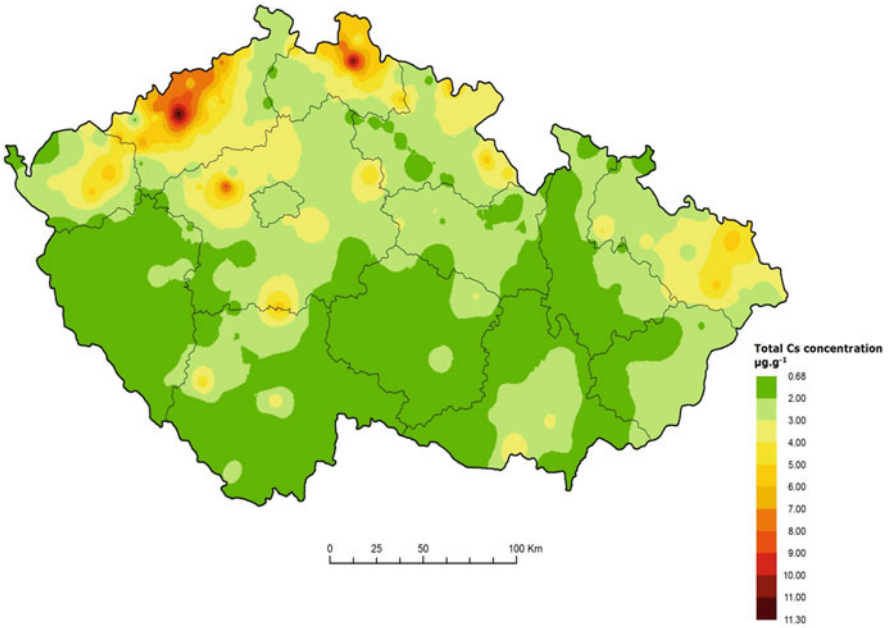

Fig. 6 Total caesium concentration distribution in forest floor humus (O_h) in CZ in 2005

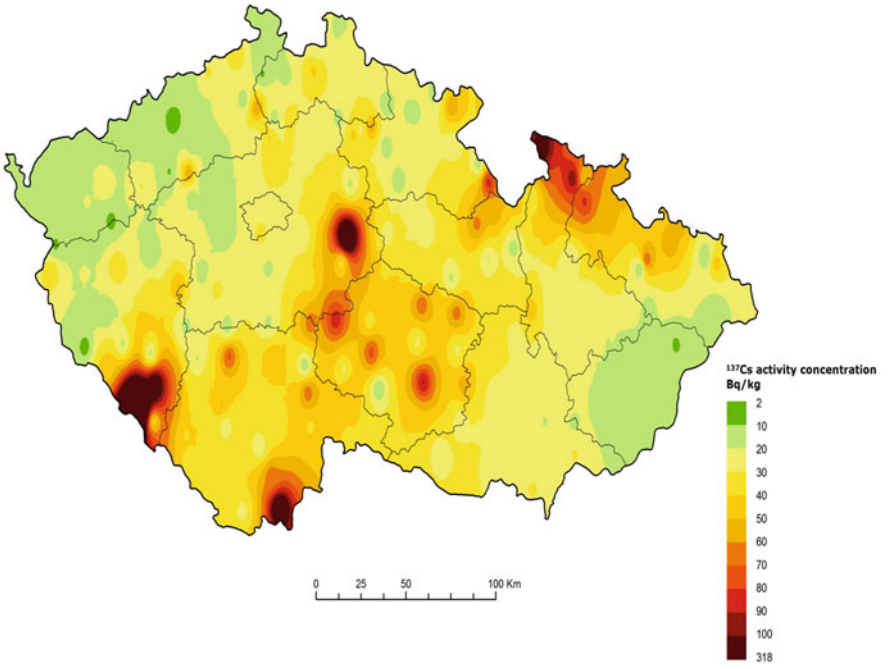

Fig. 7 Distribution of the ^{137}Cs activities in outer spruce bark in CZ in 1995

^{137}Cs activity concentrations in spruce bark in 1995. The activity concentrations of ^{137}Cs in bark significantly correlated with the ^{137}Cs soil surface activities in 1986 (Fig. 1) measured after the Chernobyl fallout (Suchara et al. 2011a), as well as with the distribution of precipitation sums when the Chernobyl plumes were passing CZ in spring 1986 (Suchara et al. 2011a). It is evident that the ^{137}Cs activity concentrations maintained in spruce bark was derived mainly from the local atmospheric deposition rates in 1986. However, the mechanism of the long-term retention of ^{137}Cs in bark proportional to the initial fallout rates on the local soil surface is not completely known. Outer bark (dead tissue that remains on spruce trunks for about 15 years) may be permanently and directly contaminated by surface particles from the local forest floor by wind erosion, bioturbation or through the correspondingly contaminated stemflow and throughfall. The geometric mean value of the aggregated transfer factor (T_{agg}) for transfer of deposited ^{137}Cs from the ground surface to bark in CZ in 1995 was 10.5×10^{-3} m^{-2} kg^{-1}. No significant differences were found in the T_{agg} values for bark in areas affected by the Chernobyl dry deposition and areas of variable wet deposition of Chernobyl-derived ^{137}Cs in 1986 (Suchara et al. 2011a). The relationship between the ^{137}Cs activity concentrations in bark in 1995 (y) and on the soil surface in 1986 (x) was well expressed by a linear equation with a slope of 3.3×10^{-3} m^{-2} kg^{-1} and an intercept at 20 Bq kg^{-1} (Suchara et al. 2011a). The intercept was interpreted as the background ^{137}Cs activity concentration in outer spruce bark across CZ caused by global fallout before the Chernobyl disaster (Suchara et al. 2011a). Considering the global fallout of about 5000 Bq m^{-2} in CZ in the 1960s, the T_{agg} for the pre-Chernobyl period was then estimated to be 4×10^{-3} m^{-2} kg^{-1}. The concentration activities of ^{137}Cs in bark in 1995 did not significantly correlate with bark pH—H_2O ($r = -0.14$, $p = 0.053$). Generally, the elevation of the sampling plots in CZ did not significantly affect the ^{137}Cs activity concentrations in outer bark ($r = -0.01$, $p = 0.85$); however, in the area affected by the wet deposition of ^{137}Cs in spring 1986 elevation played a significant and positive role in maintaining increased ^{137}Cs concentrations in bark ($r = 0.32$, $p = 0.004$).

The distribution of ^{137}Cs activity concentrations in spruce bark collected at 255 sampling sites in 2010 (Pilátová et al. 2011d) is shown in Fig. 8. The ^{137}Cs activity decay corrected to July 2010 ranged between 1.5 and 270 Bq kg^{-1}, with a geometric mean of 9.3 ± 2.2 Bq kg^{-1} (Rulík et al. 2014). These data represent about a three-fold reduction in the ^{137}Cs activity concentrations in bark in comparison to 1995. The ^{137}Cs activity distribution pattern in 2010 was very similar to the pattern in bark in 1995 and in forest floor humus in 1995 and 2005. Material from bark samples collected in 2010 was about 15-year-old and could not be exposed to the Chernobyl fallout in 1986 either in the form of inner bark. This means that other factors operate in forest ecosystems, probably re-suspensation from the forest floor surface and diffusion of ^{137}Cs into outer bark tissue, which maintain the ^{137}Cs activity concentration storage in humus and in spruce bark proportionally to the original ^{137}Cs fallout rates for long periods. In any case, the concentrations of airborne pollutants in trunk bark, including ^{137}Cs, increase from the inner bark towards the outermost bark layers (Cosma et al. 2016). Similar correlations between ^{137}Cs activity concentrations in bark in 2010 and site and bark characteristics were found to be similar to those from

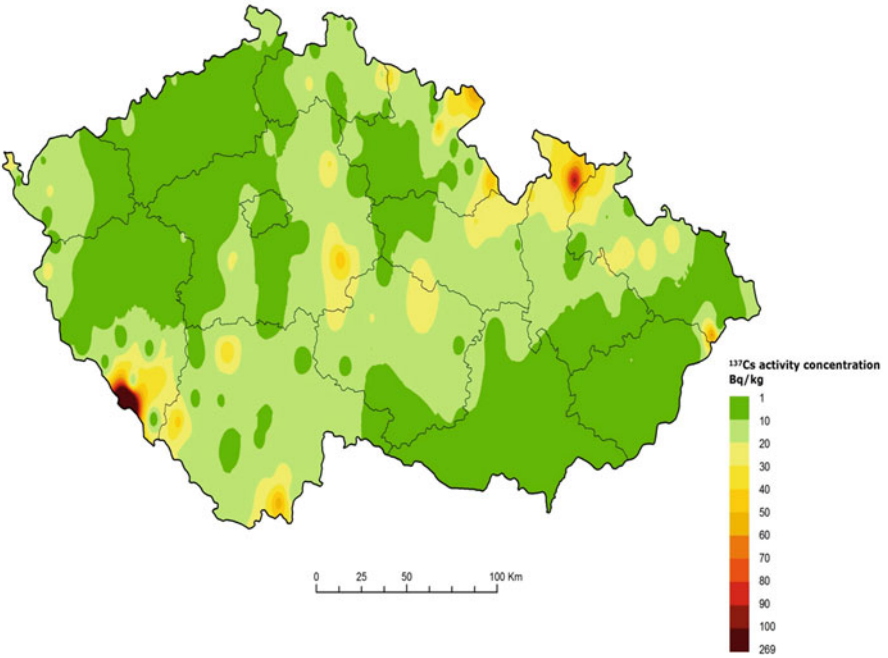

Fig. 8 Distribution of the ^{137}Cs activities in outer spruce bark in CZ in 2010

1995. If a majority of ^{137}Cs in bark came from the Chernobyl fallout, the reduction of the ^{137}Cs activity concentrations in bark in 2010 was again higher than would be expected from just physical decay. Data for ^{137}Cs activity concentrations in spruce bark obtained from 114 identical monitoring plots in CZ where bark was collected in 1995 and again in 2010 showed an effective half-life $T_{eff} = 9.6$ y and environmental half-life $T_{env} = 14.0$ years. However, T_{eff} and T_{env} significantly and positively correlated with the long-term (1961–2000) average annual precipitation sums in CZ. For seven annual precipitation classes (500–1200 mm y^{-1}), $T_{eff} = 7.1$–16 y and $T_{env} = 9.3$–34 years were found (Rulík et al. 2014). Mechanisms of how higher precipitation sums increase the retention of ^{137}Cs in bark have not yet been elucidated. Increasing stand precipitation may support the uptake of ^{137}Cs by spruce roots from the soil, leaching of ^{137}Cs from needles and wet bark, and diffusion of ^{137}Cs from stem flow into the outer bark tissue. However, higher humidity reduces the resuspension of forest floor surface material.

From the bark data in 2010 a geometric mean aggregated transfer factor $T_{agg} = 3.9 \times 10^{-3}$ m^{-2} kg^{-1} was estimated, which corresponds well to T_{agg} calculated from the 1995 bark data. A more detailed regression analysis of the current ^{137}Cs activity concentrations in spruce bark and the site soil surface ^{137}Cs activities in 1986 indicate a post-Chernobyl transfer of ^{137}Cs into spruce bark $T_{agg} = 5.3 \times 10^{-4}$ m^{-2} $kg^{-1}$and a pre-Chernobyl ^{137}Cs activity concentration in spruce bark $T_{agg} = 4.2$ Bq kg^{-1}

Table 2 Median ± median absolute deviation of ^{137}Cs activity concentrations in forest soil horizons (Bq kg $^{-1}$) in localities I–III in 2011 ($n = 12$) and localities IV–VI in 2013 ($n = 3$) (for each locality specified in Table 1)

Locality horizon	I	II	III	IV	V	VI
L	57 ± 27	23 ± 11	22 ± 7	229 ± 3	154 ± 17	11 ± 0
F	272 ± 60	62 ± 18	123 ± 50	576 ± 65	775 ± 317	33 ± 17
H	218 ± 38	65 ± 14	259 ± 81	1286 ± 78	1495 ± 94	186 ± 17
Ah	N.A.	N.A.	N.A.	601 ± 214	468 ± 85	85 ± 8
Ah/M1	35 ± 17	8 ± 3	39 ± 11	116 ± 24	286 ± 5	33 ± 2
M2	7 ± 4	4 ± 2	3 ± 1	22 ± 5	6 ± <0.05	10 ± 4
M3	0.6 ± 0.3	0.8 ± 0.2	6 ± 3	N.A.	N.A.	N.A.

N.A. not analysed, *M* mineral horizons

in CZ (Rulík et al. 2014). There are no published comparable data about the ^{137}Cs activity concentrations in outer spruce bark from CZ. However, monitoring (2000–2009) the ^{137}Cs activity concentrations in outer pine (*Pinus sylvestris*) bark in a 20 km radius around the nuclear power plant Temelín (49.18°N; 14.38°E) found typical ^{137}Cs activity concentrations of 30 Bq kg^{-1} (range 11–120 Bq kg^{-1}) with no trend over time (Thinová 2009).

5.3 The Distribution and Migration of ^{137}Cs in Forest Soils

The recent average total ^{137}Cs activity concentrations in the soil profiles from localities I–VI are proportional to the interpolated ^{137}Cs surface activity after the Chernobyl fallout in spring 1986. Table 2 summarizes the median ^{137}Cs activity concentrations measured in individual forest soil horizons. In all soil profiles, only one maximum ^{137}Cs activity concentration was found located in the upper O_h or O_f organic horizons. This indicates that the majority of the ^{137}Cs retained in forest soils in localities I–VI came from the Chernobyl fallout in 1986. The ^{137}Cs activity concentration and distribution in the forest soils below spruce and pine canopies and below spruce and pine canopy gaps, which have endured since 1986 in localities I–III, were not significantly different. These forest gaps were probably too small to show different soil ^{137}Cs inputs than soils below canopies, where increased ^{137}Cs inputs through dry deposition trapped in tree crowns was expected (Suchara et al. 2016).

The ^{137}Cs activity concentrations in the individual soil horizons of localities I-VI did not significantly correlate with soil pH or soil organic matter content, and ^{137}Cs did not significantly accumulate in elluvial soil horizons. Even in the Bhs zone with the retention of fulvic acids and Al-Fe oxides in a Sandy Podzol (locality II), there was no increased ^{137}Cs activity concentrations compared to the neighbouring soil horizons. This finding confirms really very small association of ^{137}Cs to humic and fulvic acids.

The inventory of ^{137}Cs in soil horizons from localities I–III found that these soils retained about twice the amounts of ^{137}Cs than would have been expected from ^{137}Cs decay rates after the Chernobyl fallout. However, when the estimated ^{137}Cs pre-Chernobyl fallout of about 1960 Bq m^{-2} was included in the balance, the retained ^{137}Cs in the 0–40 cm soil layer well corresponded to the sum of the global and Chernobyl fallouts.

The vertical movement of ^{137}Cs in the investigated soils was very slow at all localities. The median apparent burial rate was highest in Sandy Podzol (0.129 ± 0.017 cm y^{-1}) and lowest in Haplic Cambisol (0.085 ± 0.010 cm y^{-1}). However, at all sites below canopies ($n = 18$) the apparent burial rate was significantly faster (median 0.110 ± 0.018 cm y^{-1}) than in soils below canopy gaps (0.089 ± 0.014 cm y^{-1}), probably because of the clearly thicker organic horizons formed below the canopies compared to the canopy gaps. The median apparent burial rates for ^{137}Cs in Cambisols (locality IV–VI) were between 0.04 ± 0.01 and 0.08 ± 0.01 cm y^{-1} and the highest value was found for soil in most warm and dry area in CZ. However, dividing the depth of the middle of the soil horizon with the maximum ^{137}Cs content by the number of years since 1986, the annual migration speeds of ^{137}Cs in the forest soils in localities I–III were calculated to be 0.072–0.392 (median 0.147 ± 0.05) cm y^{-1} below the canopies, and 0.052–0.364 (median 0.163 ± 0.05) cm y^{-1}, below canopy gaps; these differences were not significant. Similarly, the vertical migration of ^{137}Cs in soils at localities IV–VI ranged from 0.019 to 0.152 (median 0.119 ± 0.03) cm y^{-1}. The migration of ^{137}Cs in the upper organic parts of forest soils may be most affected by the content of clay minerals, intensity of bioturbation and long-term biomass of soil edaphone. However, powder X-ray diffraction (XRD) analyses of humus samples from localities IV–VI did not find a significant presence of clay minerals in the O_h humus. An important proportion of the mineral admixture in all samples from O_h horizons in localities IV–VI was comprised of quartz grains, a material of low adsorption capacity. Using the ^{137}Cs activity concentrations in humus (O_h) in 1995 and 2005, the effective half-life (T_{eff}) for ^{137}Cs in the O_h horizons of localities I–VI was calculated. For various soil units (locality I–III) T_{eff} was found to be between 6 and 10 years, whereas for Cambisols (locality IV–VI) between 20 and 25 years. Bioturbation of the forest floor may be the main factor affecting T_{eff} in CZ. The effective half-life has also been calculated (Thinová and Trojek 2008) for coniferous humus at 29 sites in a 20-km radius around the nuclear power plant Temelín, with T_{eff} values found to be between 3 and 12 years for the period 2000–2007.

5.4 The Distribution of ^{137}Cs in Selected Biota

The ^{137}Cs activity concentrations were measured in some plant materials in localities I–III. Unfortunately, only small sets of samples ($n = 1$–5) were investigated. The recent litter below spruce canopies contained clearly more ^{137}Cs than the litter below

pine canopies. The highest ^{137}Cs activity concentrations in spruce and pine litter were found in locality I (31 and 20 Bq kg^{-1}) while the lowest content of ^{137}Cs was in spruce and pine litter on Sandy Podzol in locality II (13 and 11 Bg kg^{-1}).

In the late summer, the annual needles of spruce and pine trees contained more ^{137}Cs than their biennial needles. The ^{137}Cs activity concentrations in annual spruce needles were 9–42 Bq kg^{-1} (typical values about 30 Bg kg^{-1}) compared to 8–32 Bq kg^{-1} (typical values about 20 Bq kg^{-1}) in biennial needles. In pine needles, the respective annual, biennial and typical ^{137}Cs concentrations were 14–78 and 50 Bq kg^{-1} and 2–32 and 25 Bq kg^{-1}. The aggregated transfer factor for ^{137}Cs in annual and biennial spruce and pine needles related to the surface ^{137}Cs activity of $O_f + O_h$ at the relevant sites in localities I–III was 0.7–150 × 10^{-3} m^2 kg^{-1}. The typical T_{agg} for spruce needles was 5–10 × 10^{-3} m^2 kg^{-1} and for pine needles 10–50 × 10^{-3} m^2 kg^{-1}. No clear relationships between the T_{agg} for needles and soil units were found.

The ^{137}Cs activity concentrations in the grass *Avenella flexuosa* at localities I–III were between 7.5 and 127 Bq kg^{-1}; the typical values ranged between 30 and 50 Bq kg^{-1}. *Avenella flexuosa* growing below canopies usually contained more ^{137}Cs than this grass below canopy gaps. Typical values of T_{agg} were 20–30 × 10^{-3} m^2 kg^{-1}with little tendency to increase in the grass growing below canopies. The grass *Molinia coerula* occurred in locality I at sites with seasonally wet soil. Leaves of this grass contained 2–71 Bq kg^{-1} in the late summer 2011. The corresponding T_{agg} for ^{137}Cs in *Molinia* was in the range 2–43 × 10^{-3} m^2 kg^{-1}. In the moss *Pleurozium schreberi* growing in forests and below canopy gaps in locality III, nearly the same ^{137}Cs activity concentrations of about 33–34 Bq kg^{-1} were found. No direct uptake of ^{137}Cs from a forest floor by the moss is expected. Nevertheless, the aggregated transfer factor for ^{137}Cs in *Pleurozium* was T_{ag} = 24–35 × 10^{-3} m^2 kg^{-1}. Published ^{137}Cs activity concentrations in the moss *Pleurozium schreberi* from 6 sites around the nuclear power plant Temelín in 2009 were from 6.4 ± 1.4 to 263.1 ± 9 Bq kg $^{-1}$ with a median about 24 Bq kg $^{-1}$ (Drábová et al. 2013). Fruiting bodies of fungi, which frequently accumulate ^{137}Cs, were not analysed in these studies. However, some data from the CZ territory, showing high site and species variably, have been published (Vobecký and Těthal 1993; Kalač 2001; Dvořák et al. 2006; Škrkal et al. 2012, 2013).

6 Conclusions

The measured activity concentrations of ^{137}Cs in forest floor humus, outer spruce bark and soil samples collected in 1995, 2005, 2010, 2011 and 2013 correlated significantly with the surface ground activities in CZ investigated after the Chernobyl-derived fallout in 1986. However, the mechanism of the long-term retention of ^{137}Cs in forest floor humus and outer spruce bark proportional to the initial fallout rates on the local soil surface is not completely known. Mainly the forest floor resuspensation effects may be speculated. The average reduction in the ^{137}Cs activity

concentrations in humus between 1995 and 2005 was about one third higher than would have corresponded to the effect of only physical decay. No significant correlation was found between the humus characteristics (humus pH and content of mineral portion) and the ^{137}Cs activity concentrations. However, the ^{137}Cs activity concentrations had a decreasing tendency in forest floor humus (O_h) with the increasing total caesium concentration in humus.

The geometric mean value of the aggregated transfer factor for transfer of deposited Chernobyl-derived ^{137}Cs from the ground surface to spruce bark in CZ in 1995 and 2010 was 10.5×10^{-3} and 3.9×10^{-3} m^{-2} kg^{-1}, respectively. The regression line of the temporal ^{137}Cs activity concentrations in bark delimited an intercept 20 Bq kg $^{-1}$, which can be interpreted as the background ^{137}Cs activity concentration in outer spruce bark in CZ caused by global fallout before the Chernobyl disaster. The concentration activities of ^{137}Cs in bark did not significantly correlate with bark pH–H_2O and the elevation of the sampling plots. However, in the area affected by the wet deposition of ^{137}Cs in spring 1986 elevation played a significant and positive role in maintaining increased ^{137}Cs concentrations in outer spruce bark. The measured ^{137}Cs in the bark samples collected in 1995 and again in 2010 showed an effective half-life $T_{eff} = 9.6$ y and environmental half-life $T_{env} = 14.0$ y. However, T_{eff} and T_{env} significantly and positively correlated with the long-term average annual precipitation sums in CZ.

The recent average total ^{137}Cs activity concentrations in forest soil profiles in CZ is proportional to the interpolated ^{137}Cs surface activity after the Chernobyl fallout in spring 1986. The vertical movement of ^{137}Cs in the investigated soils was very slow at all localities. The majority of ^{137}Cs in forest soils is retained in O_h horizons and exclusively only one peak of ^{137}Cs activity concentration distribution was found in forest soil profiles in CZ. The median apparent burial rate of ^{137}Cs in humus (O_h) was between 0.04 ± 0.01 and 0.129 ± 0.017 cm y^{-1}. The apparent burial rate was slightly and significantly faster below tree canopies than in soils below canopy gaps. The investigated characteristics of humus and mineral soil matrices (pH, mineral and clay contents and ratio of fulvic and humic acids) had no significant effect on the ^{137}Cs retention in soil horizons. The inventory of ^{137}Cs in soil horizons well corresponded to the sum of the global and Chernobyl fallouts in CZ. The ^{137}Cs activity concentration range and the aggregate transfer factors for ^{137}Cs and annual and biennial pine and spruce needles, two common forest grasses and one forest moss in three forest stands are presented as well.

The chapter provides more complex insight into the ^{137}Cs distribution and behaviour in coniferous forests in the Czech Republic.

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Cesium Uptake in Plants: Mechanism, Regulation and Application for Phytoremediation

Ryoung Shin and Eri Adams

Abstract Radiocesium is mainly generated through anthropogenic activities and poses a great threat to health and the environment. As an alternative to costly physical and chemical methods to remediate the contaminated soil and water, phytoremediation, a technique making use of plants to remove or stabilise contamination, is receiving increasing attention. Selection of plant species that accumulate high levels of radiocesium has, therefore, been intensively investigated. In recent years, molecular techniques have enabled researchers to elucidate cesium transporters and regulatory mechanisms through which cesium uptake occurs in plants. Some proteins have been predicted to mediate cesium and many players, including cations and phytohormones, have been suggested as regulators. Although the molecular understanding of cesium uptake in plants is only just being revealed, the knowledge can readily be applied to radiocesium phytoremediation. In parallel, efforts to improve phytoremediation efficiency have sought the aid of biotic and abiotic factors such as microorganisms and chemicals. In this chapter, findings on the molecular mechanisms and regulation of cesium uptake in plants, what is known and what needs to be researched, are discussed. Then follows a discussion of the plant species suitable for radiocesium phytoremediation and the factors which improve phytoremediation efficiency.

Keywords Cesium • Potassium • Phytoremediation

1 Introduction

The accident at the Fukushima Daiichi Nuclear Power Plant in Japan on 11 March 2011 following one of the world's largest recorded earthquakes resulted in the release of significant amounts of radioactive material such as radiocesium which

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spread over a swath of eastern Japan. While efforts to decontaminate affected areas have had some success, in some areas, farming has not yet been restarted due to radiocesium contamination in the soil. The development of phytoremediation techniques to remove radiocesium from the soil thus allowing a resumption of farming has become the focus of attention.

Various plant species have been tested for removing radiocesium from the Chernobyl (Broadley and Willey 1997; Willey et al. 2005) and Fukushima contaminated areas (Saito et al. 2014). However, no specific plant species has been demonstrated to show high enough efficiency for practical use in radiocesium remediation. For example, sunflower (*Helianthus* spp.) and amaranth (*Amaranthus* spp.) shown to accumulate reasonable amounts of radiocesium in Chernobyl (Moogouei et al. 2011; Tang et al. 2011) did not show much remediation effect on the contaminated soil in the Fukushima area (Saito et al. 2014). This difference may be due to the different physicochemical properties and compositions of soil, soil microorganisms and plant growth conditions. To improve remediation efficiency, ways to modify cesium uptake in plants, such as genetic modification of the potassium uptake system and chemical treatment, have been considered (Adams et al. 2015).

Although cesium has no known nutritional value or beneficial function in plants (White and Broadley 2000), this monovalent cation is known to enter into plant root cells and to be transported across the plant via the potassium uptake system (Middleton et al. 1960; Broadley and Willey 1997). Potassium low-affinity and high-affinity transporters have been suggested as possible cesium transporters (Hampton et al. 2004; Nieves-Cordones et al. 2010; Qi et al. 2008; Rubio et al. 2000) and shaker-type potassium channels have also been considered as cesium ports of entry (Hoth et al. 1997; Ichida et al. 1999; Ichida and Schroeder 1996; Schachtman et al. 1992). Other data have suggested that voltage-insensitive channels such as CYCLIC NUCLEOTIDE-GATED CHANNELs (CNGCs) and GLUTAMATE RECEPTORs (GLRs) work as predominant cesium channels in plants (Chiu et al. 2002; Demidchik et al. 2002a, b; Hampton et al. 2004; Tapken and Hollmann 2008). Although multiple channels and transporters are thought to mediate cesium, the detailed mechanisms and regulation of cesium uptake are still unclear. It is known that plants grown with cesium encounter potassium deficiency to a certain extent (Adams et al. 2013; Hampton et al. 2004; White and Broadley 2000). Thus it has been thought that the signal cascades induced by cesium may be indistinguishable from potassium deficiency signaling. However, the accumulated information has been pointing towards the existence of cesium dependent regulatory components which are independent of potassium deficiency but the current knowledge on cesium induced signaling is too fragmented to draw a conclusion. In this chapter, the reported information on cesium transport mechanism, its regulation and application for phytoremediation are summarized and discussed.

2 Cesium Uptake Mechanism

Plants take up mineral nutrients from the soil and these are transported via the xylem. As cesium belongs to the same alkali metal group as potassium and shares similar chemical properties (Clarkson et al. 1988; White et al. 1987), it is likely that it would be absorbed and transported in the same way as potassium. Many researchers have suggested that potassium transporters and channels are possible ports of entry for cesium. Collander has first reported that plants accumulate cesium via potassium uptake mechanisms (Collander 1941) and there are several reports on the potassium transporters and channels which are involved in cesium transport from soil to roots (Hampton et al. 2004; White and Broadley 2000). Although the channels or transporters which mediate cesium into cytoplasm and vacuoles are still obscure, a ^{133}Cs Nuclear Magnetic Resonance (NMR) study has shown that cesium is localized in cytoplasm and vacuoles and has suggested the involvement of channels or transporters in translocation of cesium in plant cells (Le Lay et al. 2006). It is tempting to speculate that the channels and transporters which mediate cesium are those that translocate potassium in plants.

Although potassium and cesium are considered to share uptake mechanisms, several reports have suggested that there is a discrimination mechanism against cesium when potassium is available (Epstein and Hagen 1952; Middleton et al. 1960), and this has been directly confirmed in research on barley (*Hordeum vulgare*) roots by measuring relative uptake of ^{137}Cs and ^{42}K (Bange and Overstreet 1960). This indicates that there are complicated mechanisms at work for cesium uptake in plants but the actual port of entry of cesium has not been fully revealed. Elucidation of cesium uptake mechanisms by plants is necessary if efforts to control cesium uptake and develop plants for radiocesium phytoremediation are to be made (Kobayashi et al. 2010). In this section, research on possible membrane proteins which mediate cesium uptake and transport in plants is summarized and discussed.

2.1 KT/KUP/HAKs

The plant KT/KUP/HAK proteins have been isolated as homologs of bacteria K^+ UPTAKE PERMEASE (KUP) proteins and yeast HIGH-AFFINITY K^+ TRANSPORTER (HAK) proteins and function as high-affinity or low-affinity H^+/K^+ symporters (Ahn et al. 2004; Banuelos and Rodriguez-Navarro 2001; Epstein and Kim 1971; Santa-Maria et al. 1997). KT/KUP/HAK proteins poorly discriminate between potassium, rubidium and cesium (Very and Sentenac 2003). Thus the KT/KUP/HAK family is considered to be one of the important cesium transporters in plants.

Arabidopsis thaliana has 13 family members of KT/KUP/HAK in the genome (Ahn et al. 2004). *Arabidopsis* mutants lacking *KUP2*, *KUP3* or *HAK5* have been

demonstrated to accumulate less cesium in an environment of sufficient potassium (20 mM) with 33 μM or 1 mM cesium than the wild type (Qi et al. 2008; White et al. 2004). *Arabidopsis* KUP9 has been shown to mediate cesium uptake in *Escherichia coli*, whereas *KUP9* knockout plants do not show any difference in cesium uptake compared to the wild type plants (Kobayashi et al. 2010). In addition, the involvement of high-affinity potassium transporters, including HAK5, for cesium uptake has been reported by several researchers (Aleman et al. 2014; Hampton et al. 2004; Nieves-Cordones et al. 2010; Qi et al. 2008; Rubio et al. 2000). It has been indicated that a single mutation on *HAK5* at F130S confers dramatic tolerance to cesium and sodium and reduces cesium accumulation in yeast compared to yeast expressing non-mutated *HAK5* (Aleman et al. 2014). The authors have speculated that high-affinity potassium transporters including HAK5, have an important function in cesium accumulation in low potassium conditions. Two different pathways have been proposed for cesium transport in plants: the calcium-sensitive cesium uptake pathway which mainly functions in potassium sufficient plants and the calcium-insensitive cesium uptake pathway that has been thought to involve high affinity transporters which are inhibited by ammonium as well as gadolinium (Caballero et al. 2012; Hampton et al. 2004; Heredia et al. 2002). However, recent reports have shown that calcium regulates the high-affinity potassium uptake system in plants (Ragel et al. 2015; Scherzer et al. 2015), which suggests that as-yet-unknown machinery plays a role in the calcium-insensitive pathway.

Besides *Arabidopsis*, KT/KUP/HAK transporters from other plant species have also been suggested as possible cesium transporters. A *Physcomitrella patens* KT/KUP/HAK transporter, HAK1 has been shown to regulate potassium homeostasis and plant morphology under potassium sufficient conditions and contributes to high-affinity rubidium and cesium uptake during potassium starvation (Garciadeblas et al. 2007). A barley potassium transporter, HAK1 has been reported to possess cesium uptake abilities in yeast (Rubio et al. 2000). Although multiple KT/KUP/HAKs have been suggested as cesium transporters, only a few cases have been proven to be functional in plants. Further investigation is also required on the role of low-affinity KT/KUP/HAKs in cesium uptake.

2.2 HKT

A wheat (*Triticum aestivum*) HIGH-AFFINITY K^+ TRANSPORTER1 (HKT1), has been identified as a sodium and potassium co-transporter (Schachtman and Schroeder 1994). However, later works have shown that monocotyledonous HKTs mediate sodium and potassium (Gassman et al. 1996; Rubio et al. 1995; Schachtman 2000) but dicotyledonous HKTs do not transport potassium but only transport sodium (Liu et al. 2001; Uozumi et al. 2000). Wheat HKT1 has independent binding sites for potassium and sodium. The sodium binding site is very specific for sodium but the potassium binding site is relatively non-discriminative for other ions including cesium (Gassman et al. 1996; Schachtman 2000). Although the possibility that

HKTs transport cesium has been suggested based on the yeast and *Xenopus laevis* oocytes data, only monocotyledonous HKTs which possess the potassium binding site may mediate cesium, and not dicotyledonous HKTs which only possess the sodium binding site in plants.

2.3 Voltage-Gated Potassium Channels

In *Arabidopsis*, an inward-rectifying potassium channel ARABIDOPSIS K^+ TRANSPORTER1 (AKT1) is known to be a major player in low-affinity potassium uptake (Broadley et al. 2001; Caballero et al. 2012; Gaymard et al. 1998; Harada and Leigh 2006; Nieves-Cordones et al. 2012) and other inward-rectifying channels KAT1, KAT2 and AKT2/3 are involved in long-distance potassium transport and intracellular potassium movements (Adams and Shin 2014; Ichida et al. 1999; Schachtman et al. 1992). An *Arabidopsis* outward-rectifying potassium channel, STELAR K^+ OUTWARD RECTIFIER (SKOR), takes part in xylem loading and GUARD CELL OUTWARD-RECTIFYING K^+ (GORK) is required for controlling the stomata closure via increasing potassium efflux (Ache et al. 2000; Schroeder et al. 2001). There has been many reports that cesium blocks potassium channel activities (Bertl et al. 1997), however, whether these channels directly mediate cesium or not is barely known. KAT1 has been shown to be permeable to cesium (Schachtman et al. 1992) and less inhibited by cesium than AKT1 (Ichida and Schroeder 1996). Although Broadley and colleagues have claimed that the effect of AKT1 on cesium influx and accumulation can be negligible, mutation studies of AKT1 have suggested that some mutations alter the sensitivity and selectivity to cesium (Broadley et al. 2001; Hoth et al. 1997; Ichida et al. 1999).

2.4 Voltage Insensitive Channels

There are three channel families that contribute to transient calcium release in order to control cytosolic free calcium levels (Sanders et al. 1999; Ward et al. 2009). Among them, CNGCs and GLRs are also known as possible potassium channels and these possibly work as cesium channels as well. In *Arabidopsis*, there are 20 CNGC members and potassium inward channel activities have been confirmed in CNGC1, CNGC2, CNGC4 and CNGC10 (Demidchik et al. 2002a, b). Also, plasma membrane localized *Arabidopsis* GLRs, GLR1.1 and GLR1.4 have been shown to mediate potassium in roots (Chiu et al. 2002; Jammes et al. 2011; Tapken and Hollmann 2008). Theoretical modelling has suggested that cesium uptake from soil to root cells is predominantly through CNGCs and GLRs in *Arabidopsis* (Hampton et al. 2004; White and Broadley 2000). Gene expression analyses have also indicated that *CNGC1*, *GLR1.2* and *GLR1.3* are induced in cesium stressed roots (Hampton et al. 2004; Kanter et al. 2010). Moreover, it has been shown that a

calcium-sensitive system mediates potassium uptake as well as cesium uptake in a *HAK5* and *AKT1* double mutant (Caballero et al. 2012). It has been speculated that the CNGC and GLR families are the alternative system for potassium and cesium uptake in this double mutant.

Quantitative Trait Locus (QTL) analysis of different ecotypes in various cesium concentrations has suggested that at least 16 QTLs are involved in cesium and/or strontium accumulation in *Arabidopsis*. Among them, CNGC1, CNGC19, CNGC20 and CATION/H^+ EXCHANGER16 (CHX16) have been thought to be candidate proteins involved in cesium accumulation (Kanter et al. 2010). CNGC1 seems to be the most promising protein among these candidates since its expression is induced in cesium treated roots and its genetic location is also very close to one of the reported QTLs (Kanter et al. 2010; White and Broadley 2000). Polymorphism analyses have suggested that the amino acids 297-302 which are located in the transmembrane domains S5 and S6 and are close to the potassium pore may be a crucial region of CNGC1 for cesium accumulation (Kanter et al. 2010). Altogether, CNGC1 may be the best candidate for mediating cesium influx to roots in *Arabidopsis* at this point in time, further experimental confirmation is required. Also, the possibility cannot be ruled out that other CNGCs and GLRs, yet to be identified, more predominantly mediate cesium transport.

2.5 Other Membrane Proteins

Arabidopsis ZINC-INDUCED FACILITATOR-LIKE2 (ZIFL2) has been confirmed to mediate potassium and cesium influx when expressed in yeast. A *ZIFL2* loss of function mutant in *Arabidopsis* has shown decreased sensitivity to cesium while *ZIFL2* overexpression plants have displayed increased sensitivity to cesium and altered cesium partitioning across the plant. The authors have speculated that ZIFL2 restricts cesium xylem loading by releasing cesium into the apoplast thus controlling excessive transport of cesium from the roots to shoots and thereby regulates cesium/potassium homeostasis (Remy et al. 2015). Meanwhile, wheat LOW-AFFINITY CATION TRANSPORTER1 (LCT1) has been confirmed to mediate potassium, rubidium, sodium, calcium, cadmium (Clemens et al. 1998; Schachtman et al. 1997; White and Broadley 2000) and cesium (Schroeder et al. 1997). By contrast, it has been shown that a yeast mutant which lacks a Soluble *N*-ethylmaleimide-sensitive factor Activating REceptor (SNARE) gene, *Sec22p*, accumulates less than 50 % cesium compared with the wild type but there are no differences in accumulation of other cations such as potassium, sodium, calcium and magnesium (Draxl et al. 2013). Mathematical modeling has indicated that decreased accumulation of cesium in the *sec22p* mutant is due to repressed vacuolar sequestration of cesium. In plants, a homolog of Sec22p in *Arabidopsis*, SEC22, is predicted to function very similarly, that is, the *sec22* mutants accumulate less cesium but there are no changes in accumulation of other cations including potassium and rubidium (Draxl et al. 2013).

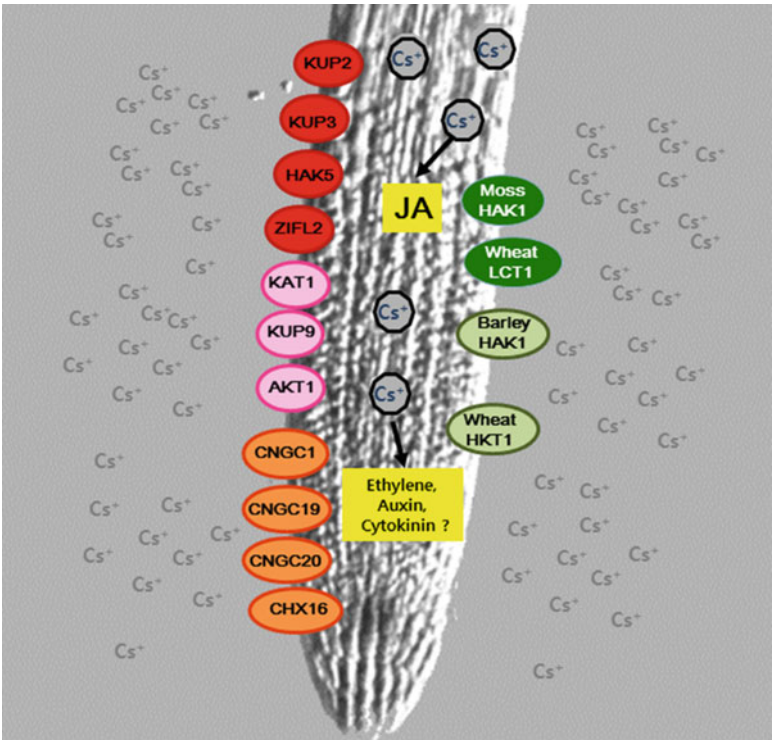

Fig. 1 Overview of possible cesium transporters in plants. Red circles indicate *Arabidopsis* membrane proteins which were confirmed to mediate cesium in plants, pink circles indicate *Arabidopsis* membrane proteins that were confirmed to mediate cesium in heterologous systems and orange circles indicate *Arabidopsis* membrane proteins that were predicted as cesium transporters by modelling or QTL analysis. Moss HAK1 and wheat LCT1 were confirmed to transport cesium in plants whereas barley HAK1 and wheat HKT1 were reported their cesium uptake abilities in heterologous systems. A class of phytohormones, jasmonates (JA) are involved in cesium induced growth inhibition in *Arabidopsis* and other phytohormones such as ethylene, auxins and cytokinins may also participate in cesium induced signalling and/or plant responses

Although it is unlikely that SEC22 functions as a cesium permeable transporter, it may serve as a key regulator in cesium-specific sequestration machinery.

As described above and summarized in Fig. 1, many cation channels and transporters have been suggested as mediating cesium in plants. However, cesium transport capabilities of these proteins are confirmed *in planta* for only a few of them and the rest, directly or indirectly, in heterologous system. Thus further characterization as well as identification of novel cesium transporters will be necessary for understanding the ports of entry of cesium. Only then would it be feasible to apply the knowledge to efficient phytoremediation techniques of radiocesium.

3 Regulation

Cesium has no known nutritional value to plants but plants competitively accumulate cesium via potassium transport machinery. Therefore, applying high levels of potassium fertilizer has been considered in order to limit cesium from entering plants. By contrast, it is well known that high cesium accumulation occurs in potassium starved plants (Sahr et al. 2005a, b; Zhu et al. 2000; Zhu and Smolders 2000). It has been postulated that the response of plants to cesium application would be somewhat similar to the response to potassium starvation. Here the regulation of cesium transport and plant responses to cesium are discussed and compared with regulation and responses in potassium deficient conditions.

3.1 Interaction with Potassium and Other Ions

High cesium accumulation in plants results in growth retardation especially in the shoots (Fig. 2) but potassium deficiency leads to more growth defects in the roots than in the shoots in the short term (Kellermeier et al. 2013; Shin 2014). Shoot growth retardation by cesium treatments may be because cesium cannot replace the biochemical roles of potassium but cesium toxicity is not exclusively derived from potassium starvation (Hampton et al. 2004). Chlorosis of the shoots may be due to decreased chlorophyll a and b contents caused by cesium application, a response which is not remarkable in potassium deficiency alone (Le Lay et al. 2006). The cesium/potassium ratio is an important factor in cesium accumulation as well as tolerance in plants (Hampton et al. 2004; White and Broadley 2000). Increases in potassium concentrations reduce cesium accumulation and *vice versa* (Hampton et al. 2004). Our physiological experiments revealed that *Arabidopsis* seedlings grown in the suboptimal potassium condition (0.5 mM KCl) with 0.3 mM CsCl

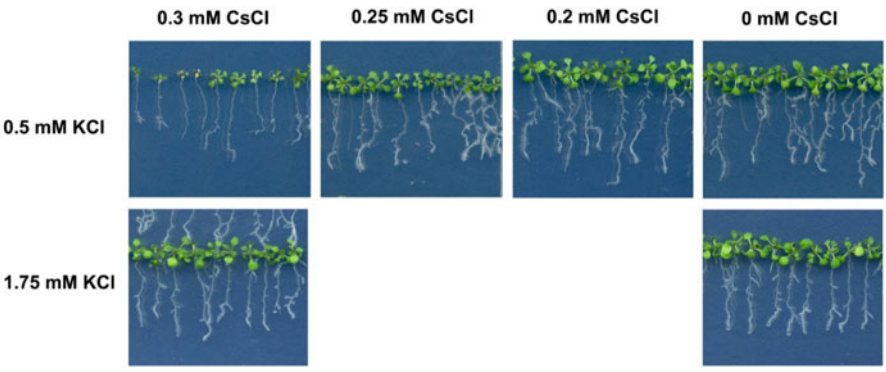

Fig. 2 Phenotype of *Arabidopsis* Col-0 (wild type) grown on suboptimal (0.5 mM) or optimal (1.75 mM) KCl with indicated concentrations of CsCl for 11 days

showed clear growth retardation and chlorosis in the shoots whereas those grown in the optimal potassium condition (1.75 mM KCl) with the same concentration of cesium did not show any growth retardation (Fig. 2). This suggests that the cesium/potassium ratios, rather than absolute concentrations of cesium, are crucial. Upon testing various cesium/potassium ratios in the suboptimal potassium condition, it was found that the cesium/potassium ratios above 0.6 caused severe growth inhibition but the ratio below 0.5 did not, indicating the existence of fine control over the threshold ratio (Kordan 1987; White and Broadley 2000).

It has been reported that cesium dramatically suppresses potassium influx at low (0.0225 mM), intermediate (0.1125 mM) and high (5 mM) potassium conditions in *Arabidopsis* wild type, *atk1*, *hak5* and *hak5;akt1* mutants (Coskun et al. 2013). When ammonium is withdrawn, potassium influx significantly increases possibly due to activation of *HAK5* which is sensitive to ammonium (Qi et al. 2008). However, cesium diminishes the ammonium withdrawal effect on potassium influx. One interesting effect of cesium on potassium influx is that cesium severely inhibits potassium influx at the intermediate potassium levels suggesting that mixed populations of channels and transporters are at work (Coskun et al. 2013).

Cesium influx is affected by other cations as well. Multiple studies have shown that monovalent cations ($Li^+ \leq Na^+ < NH_4^+ < Rb^+ \leq K^+$) and divalent cations ($Ca^{2+} \leq Mg^{2+} < Ba^{2+}$) decrease cesium influx (Bange and Overstreet 1960; Handley and Overstreet 1961; White and Broadley 2000). Cesium influx in potassium sufficient *Arabidopsis* has also been reported to be inhibited by calcium and gadolinium ions which are inhibitors of calcium channels but not by tetrathylammonium and ammonium (Hampton et al. 2004). In contrast, cesium influx in potassium deficient *Arabidopsis* is inhibited by ammonium and gadolinium but not by calcium and tetrathylammonium. More cesium influx has been detected in potassium starved plants. These findings suggest that cesium uptake in potassium sufficient conditions and deficient conditions are differently regulated.

3.2 Hormones

In the case of potassium deficiency signaling, reactive oxygen species (Shin et al. 2005; Shin and Schachtman 2004) and ethylene (Jung et al. 2009; Shin and Schachtman 2004) are key positive regulators. Also, other phytohormones such as jasmonates (Armengaud et al. 2004, 2010), auxins (Shin et al. 2007), cytokinins (Nam et al. 2012) and abscisic acid (Kim et al. 2009) are involved in controlling potassium deficiency response in plants. Since cesium application causes potassium starvation in plants, the response to cesium may be regulated in ways similar to potassium deficiency signaling. One class of these regulators, jasmonates are involved in cesium response in *Arabidopsis* (Adams et al. 2013). A jasmonate biosynthesis mutant, *allene oxide synthase* (*aos*), and a jasmonate-insensitive mutant, *coronatine insensitive1* (*coi1*-16) have been indicated to show tolerance to cesium induced growth inhibition although cesium accumulation in these mutants is not affected.

Also, cesium treatment has induced expression of genes involved in jasmonate signaling and *HAK5* whose expression has been antagonised by methyl jasmonate application. Taken together, the response of plants to cesium, especially growth inhibition, is regulated through jasmonate signaling (Adams et al. 2013). Similarly to potassium starvation (Jung et al. 2009), ethylene seems to be involved in the response of plants to cesium. Three ethylene-insensitive mutants, *ethylene insensive2* (*ein2*-1), *ethylene response sensor2* (*ers2*-1) and *ethylene receptro2* (*etr2*-1) (Hua and Meyerowitz 1998) were less sensitive to cesium treatment compared to the wild type (Fig. 3) (Adams et al. 2013). Furthermore, induction of *HAK5* expression by

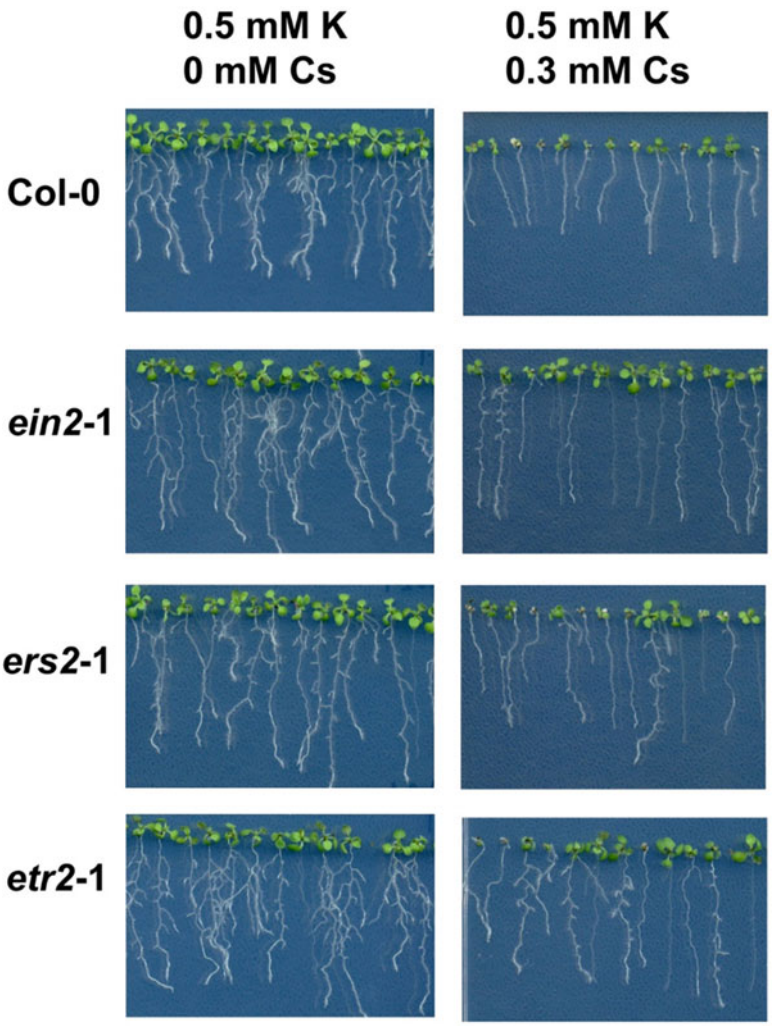

Fig. 3 Phenotype of *Arabidopsis* Col-0 (wild type) and ethylene-insensitive mutants *ein2*-1, *ers1*-1 and *etr2*-1 grown on suboptimal (0.5 mM) KCl with or without 0.3 mM CsCl for 10 days

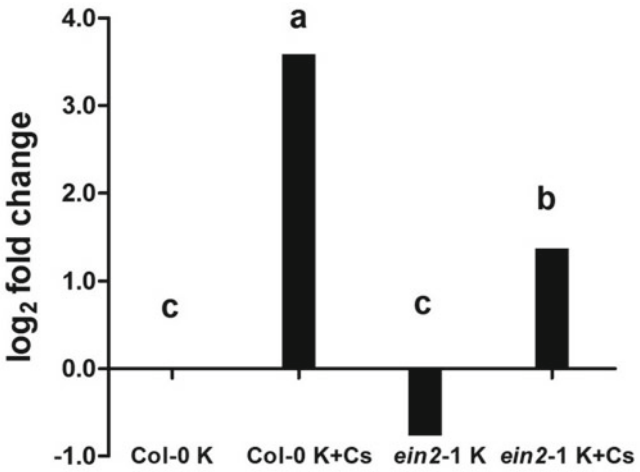

Fig. 4 Gene expression of *HAK5* in *Arabidopsis* Col-0 (wild type) and an ethylene-insensitive mutant *ein2*-1 grown on optimal (1.75 mM) KCl with or without 0.3 mM CsCl for 11 days. Values are $\log_2$ ratios of *HAK5* to *TUB2* (reference gene) expression levels relative to the control. Each sample contained more than 15 seedlings and the experiment was repeated three times, one of which is presented. Statistically significant differences ($p < 0.05$) are indicated with alphabets

cesium in *ein2*-1 was attenuated compared to the wild type (Fig. 4). These data suggest that ethylene plays roles in plant's response to cesium. An auxin-insensitive mutant, *auxin resistant1* (*aux1*-7), a cytokinin biosynthesis quadruple mutant, *isopentneyltransferase1,3,5,7* (*ipt1,3,5,7*), and a cytokinin-insensitive double mutant, *histidine kinase 2;3* (*ahk2;3*) have also been shown as less responsive to cesium than the wild type but less so compared to jasmonate mutants (Adams et al. 2013). Although not fully elucidated, these findings suggest that a similar set of phytohormones controls the plant's response to both potassium deficiency and cesium application.

3.3 Transcriptional Regulation

Sahr and colleagues have analyzed gene expression using low-level radiocesium exposed plants (Sahr et al. 2005b) and non-radioactive cesium treated plants (Sahr et al. 2005a) by suppression subtractive hybridization. Both treatments have altered expression of genes involved in stress response, transport, antioxidants and reactive oxygen species (Sahr et al. 2005a, b). However, alteration in expression of genes related to DNA repair, DNA recombination and cell cycles by radiocesium exposure may be due to only ionizing radiation, not the cesium effect (Sahr et al. 2005b). Since there is not much information available, it may be interesting to perform

microarray analyses to directly compare altered gene expression in plants treated with cesium in potassium sufficient conditions and plants treated with potassium starved conditions in order to distinguish the response of plants to cesium and the response to potassium deficiency.

3.4 Post-Transcriptional and Post-Translational Regulation

MicroRNAs are small, non-coding RNAs with 21–22 nucleotides in length which post-transcriptionally regulate gene expression (Bartel 2004) and many genes involved in mineral nutrient status including phosphate (Xu et al. 2013), sulfate (Liang et al. 2010), copper (Yamasaki et al. 2009), nitrogen (Liang et al. 2012) and heavy metals (Zhou et al. 2012) are known to be controlled by microRNAs. Cesium toxicity is known to result in differential expression of at least 18 microRNAs, out of which ten are irrelevant to high potassium treatment (Jung et al. 2015). Furthermore, transcripts of *AGONOUTE1* (*AGO1*), a key component of microRNA function, have been demonstrated to increase in response to cesium treatment but not to potassium treatment (Jung et al. 2015). However, cesium treatment delays microRNA processing and enhances the proteolytic destabilization of AGO1 protein. Metabolomic and proteomic analyses of *Arabidopsis* cells treated with 1 mM cesium in potassium deficient or sufficient conditions have suggested that the post-translational alteration of antioxidant enzymes such as NADPH:quinone oxidoreductase, NADP-dependent oxidoreductase, cytosolic L-ascorbate peroxidase, superoxide dismutase and catalase is common phenomena for potassium starved and cesium stressed plants (Le Lay et al. 2006). However, it cannot be ruled out that potassium deficiency caused by cesium treatment indirectly induces modification of antioxidant enzymes.

4 Application for Phytoremediation

4.1 Use of Plants to Remediate Radiocesium

Radioactive cesium, which is mainly generated through anthropogenic activities such as in nuclear power reactors and military tests of nuclear weapons, poses a threat to human health and ecosystem due particularly to its high water solubility and strong tendency to bind with soil. Of the various radioactive isotopes of cesium, ^{137}Cs, with a relatively long half-life of 30.17 years, is a major threat when soil and water become contaminated. There is increasing attention being given to the use of plants to remediate soil and water contaminated with radiocesium. The use of plants can be cost effective, eco-friendly and can dramatically reduce the amounts of radioactive waste compared to the other remediation methods such as chemical and

physical methods. The use of plants to remove contaminants from soil and water can be divided into two main methodologies: phytoremediation and biosorption. The former is a technique to remediate contaminated soil and water using living plants and requires the application of knowledge described in the previous section on the mechanism and regulation of cesium uptake in plants to improve efficiency. The latter focuses on the use of biomass as a material to adsorb the contaminants, the efficiency of which depends on physicochemical properties of the biomass, mainly the cell wall in case of plants. The following section summarises the findings on selection of useful plant species for phytoremediation of radiocesium and the factors that affect phytoremediation efficiency. Then follows examples of biosorption using plant materials to remediate radiocesium contamination.

4.2 Search for Plant Species Suitable for Efficient Phytoremediation of Radiocesium

Phytoremediation is classified into multiple categories such as phytoextraction which involves accumulation of the contaminants in the plant body and phytostabilisation which involves immobilisation of the contaminants at the rhizosphere by root adsorption and complex formation (Sharma et al. 2015). Phytoextraction, for example, is preferred over physiochemical methods to remove contaminants since it does not require the removal of surface soil which is fertile thus is appreciated by farmers and results in dramatically reduced amounts of radioactive waste after desiccation and incineration of the plant materials. On the other hand, phytoextraction relies on the plant's ability to absorb the contaminants and the efficiency of this can be low. To overcome this disadvantage, selection of hyperaccumulator plant species has traditionally been the major field of study. The criteria for candidate hyperaccumulators are being the ability to grow well in contaminated sites, able to produce significant biomass, able to tolerate the contaminant and the ability to efficiently absorb and transfer the contaminant to the aerial parts. Many plant species have been tested to determine their efficiency for cesium phytoextraction. Hydroponically grown monocotyledonous napier grass (*Pennisetum purpureum* Schum.) has been reported as producing a large biomass and accumulating high concentrations of cesium in the aerial parts (Kang et al. 2012). Monocotyledonous pothos (*Epipremnum aureum*) and vetiver grass (*Vetiveria zizanoides*) grown in solutions contaminated with ^{137}Cs have also been shown to accumulate significant amounts of cesium, but more is found in the roots than in the shoots (Kamel et al. 2007; Singh et al. 2008). Fukuda and colleagues have selected six strains out of 188 microalgae and aquatic plants which efficiently remove radiocesium from contaminated freshwater media (Fukuda et al. 2014). In addition, dicotyledonous species such as *Catharanthus roseus*, *Chromolaena odorata*, *Calendula alata*, *A. chlorostachys* and *Chenopodium album* (Fulekar et al. 2010; Moogouei et al. 2011; Singh et al. 2009) have been suggested as effective phytoremediation candidates for cesium-contaminated solutions.

However, the results from hydroponic studies need to be carefully analysed if these species are to be considered for soil phytoextraction. Only a handful of experimental data have been reported to assess the effectiveness of various plant species for radiocesium phytoremediation in the soil. These include rape (*Brassica campestris* L.) which has been shown to exhibit high transfer factor and biomass production in ^{137}Cs-contaminated soil (Chou et al. 2005) and arboreal *Chengiopanax sciadophylloides*, grown in contaminated sites after the Fukushima Daiichi Nuclear Power Plant failure which has been found to accumulate high concentrations of cesium one order of magnitude higher than those of other species on the site (Sugiura et al. 2016). It has also been shown that considerable amounts of ^{134}Cs are translocated from roots to fruits which behave as sinks in tomato (*Solanum lycopersicon* Mill.) grown on peat soil (Brambilla et al. 2002). By contrast, some species such as *Calotropis gigantea*, *Calendula alata* and *A. retroflexus* L. have been suggested as exhibiting low cesium uptake efficiency and will require a long period of time to remediate contamination (Borghei et al. 2011; Eapen et al. 2006; Fuhrmann et al. 2002) whereas others, mostly tested in the field studies, have not been recommended for use in radiocesium phytoremediation (Cook et al. 2009; Hoseini et al. 2012; Kobayashi et al. 2010; Yamashita et al. 2014). These findings are nicely summarized elsewhere (Sharma et al. 2015).

Based on the accumulated results from earlier studies on cesium phytoremediation by various plant species, a method to assess the influence of plant taxonomy on relative shoot cesium concentrations using residual maximum likelihood (REML) analysis has been established and the authors have indicated that dicotyledons accumulate more cesium than monocotyledons (Broadley et al. 1999). Willey and colleagues have further developed REML analysis and shown that cesium uptake efficiency by different plant taxa can be predicted through two factors: molecular phylogeny and growth strategy (Willey et al. 2005).

4.3 Biotic Factors Influencing Cesium Phytoremediation Efficiency

The quest for plant species that effectively accumulate radiocesium has long been the focus of attention in the field of cesium phytoremediation as discussed in the previous section. However, the efficiency of remediation of cesium-contaminated soil, even for the "hyperaccumulators", is not high enough to have practical application. In order to improve phytoremediation efficiency, the aid of microorganisms has been considered. Some microorganisms are known to associate with plant roots and play a key role in nutrient/metal uptake at the soil-root interface. Of those, the contribution of arbuscular mycorrhizal (AM) fungi are probably the best studied for radiocesium uptake in plants. Using root-organ culture, Declerck and colleagues have demonstrated that an AM fungus, *Glomus lamellosum*, promotes uptake and accumulation of radiocesium in the roots of transformed carrot (*Dancus carota* L.)

(Declerck et al. 2003) whereas a later study has shown that the ability to absorb cesium is indistinguishable between the mycorrhizal or non-mycorrhizal carrot roots (Dupre de Boulois et al. 2005). Meanwhile, AM fungi have been suggested as enhancing cesium uptake in leek (*Allium porrum*) but not in ryegrass (*Lolium perenne* and *L. multiflorum* Lam.), cucumber (*Cucumis sativus* L.), sunflower (*Helianthus* spp.) or barley (*Hordeum vulgare* L.) (Rosen et al. 2005; Vinichuk et al. 2013). Wiesel and colleagues have offered an explanation for the discrepancy in influence of AM fungi on cesium uptake in plants suggesting that the negative effects of cesium on the growth of AM fungi have been overlooked and this may contribute to the inconsistent results in the previous publications (Wiesel et al. 2015). Another type of fungi, dark septate endophytes including *Pseudosigmoidea ibarakiensis* I.4-2-1, *Veronaeopsis simplex* Y34 and *Helminthosporium velutinum* 41-1 have been shown to enhance cesium accumulation in Chinese cabbage (*B. rapa*) but have been shown to prevent accumulation in tomato (Diene et al. 2014). Alternatively, plant growth promoting rhizobacteria (PGPR) such as *Azospirillum* sp. and *Bacillus pumilus* have been recommended for use in phytoextraction as they improve plant growth as well as cesium uptake in Komatsuna (*B. rapa* var. *perviridis*) (Aung et al. 2015; Djedidi et al. 2014). Further, Tang and colleagues have demonstrated the effectiveness of the combined effects of another PGPR *Burkholderia* sp. and elevated atmospheric CO_2 on plant growth and cesium uptake in *Phytolacca americana* and *A. cruentus* (Tang et al. 2011). The effect of CO_2 on biomass production and increases in cesium uptake has also been shown in *Sorghum vulgare* x *Sorghum vulgare var. sudanense* hybrid and *Trifolium pratense* L. (Wu et al. 2009). It would be intriguing to investigate whether microorganisms could re-programme the regulation of cesium uptake mechanisms in plants, for example, through alteration in the activity of transporters in addition to merely promoting plant growth and root access to the nuclide.

4.4 Abiotic Factors Influencing Cesium Phytoremediation Efficiency

The potential of microorganisms in improving phytoremediation efficiency for radiocesium was described in the previous section. Here, abiotic factors that might affect the efficiency of phytoremediation are discussed. It is well known that the presence of the essential nutrient potassium inhibits cesium uptake due to the chemical similarities between the two ions (Zhu and Smolders 2000). This effect has been tested in the field and shown to be the case with Swiss chard (*Beta vulgaris* var. *cicla* L.) which, when grown with potassium supply, accumulated significantly lower amounts of cesium (Schuller et al. 2005). Soil amendment using bentonite in combination with potassium supply has also been indicated to be effective in reducing cesium transfer to ryegrass (Vandenhove et al. 2005). In addition, application of zeolite and vermiculite as well as potassium to paddy soil has been demonstrated to

reduce cesium concentrations in rice, *Oryza sativa* L. (Ichida et al. 1999). Cesium uptake rates have been shown to decline with the increasing concentrations of sodium in an aquatic floating plant *Riccia fluitans* (Heredia et al. 2002) probably for the same reasons as cesium uptake is affected by potassium. Their results also indicate the influence of pH and calcium on cesium uptake and accumulation. Conversely, potassium deficiency but not phosphorus or nitrogen deficiency is known to increase cesium concentrations in plants as has been reported in *Arabidopsis* cells (Le Lay et al. 2006) and in the shoots of rice (Nobori et al. 2015).

High concentrations of ammonium ions effectively desorb cesium from soil making it more readily available for plant uptake. It has previously been reported that the addition of up to 0.2 M ammonium increases cesium concentrations by two- to twelve-fold in soil-grown cabbage (*B. oleracea* var. *capitata*), tepary bean (*Phaseolus acutifolius*), Indian mustard (*B. juncea*) and reed canarygrass (*Phalaris arundinacea*) (Lasat et al. 1997). Consistent with this, Gommers and colleagues have rationalised that the high cesium concentrations found in willow (*Salix viminalis* L.) grown on a particular type of soil in the radiocesium-contaminated land following the Chernobyl deposition have been affected by soil properties such as relatively low exchangeable potassium and high ammonium concentrations (Gommers et al. 2005). However, inconsistent results have been reported on the effects of ammonium on plant uptake of cesium (Shaw et al. 1992; Smolders et al. 1997; Soudek et al. 2004) and it seems unlikely that natural ammonium concentrations in soil can affect cesium uptake in plants (Zhu and Smolders 2000). It is noteworthy that the highest cesium uptake has been observed at potassium and ammonium concentrations where maximum biomass production has been recorded in sunflower (*H. annuus* L.) (Soudek et al. 2004), suggesting that the influence of ammonium may be on plant growth rather than directly on cesium uptake. In addition, Smolders and colleagues have also argued that increasing concentrations of magnesium and calcium significantly reduce cesium uptake in spinach (*Spinacia oleracea* L.) (Smolders et al. 1997). Increase in low-molecular-weight organic acids exuded from rape roots have been reported in cesium-contaminated soil and the authors suggest that this increase in organic acids contributes to a release of cesium from the soil and, in turn, induces plant uptake of the ion (Chiang et al. 2005).

On another front, soil amendment with chelators and other small compounds has been gathering considerable attention. Soil amendment with one of the chelators, ethylene diamine tetraacetic acid (EDTA), has been shown to increase cesium uptake capacity in Indian mustard by eluting cesium from soil particles and making it available for roots, although EDTA retards plant growth in the long term (Tjahaja et al. 2015). Conversely, CsTolen A, a small compound with an imidazopyridine backbone, has been indicated as reducing cesium accumulation in *Arabidopsis* through selective binding with cesium thus inhibiting its uptake (Adams et al. 2015). This is an example of phytostabilisation and it is possible that a technology could be developed to reduce radiocesium accumulation in agricultural products using the findings on this chemical. Chemical treatments can be very powerful and efficient tools, but the long-term effects of such chemicals on the environment and human health have to be carefully assessed.

4.5 Biosorption of Cesium Using Plant Materials

Biosorption is a technique similar to phytoremediation and offers an alternative eco-friendly and cost-effective way to remediate contaminated sites. However, the crucial difference between this and the methods discussed above is that the biomass used in biosorption is usually not alive. In biosorption, the metal of interest is adsorbed not absorbed as in phytoextraction to the surface of the biomass utilising its physicochemical properties which, in most cases, is chemically modified to provide the optimal surface characteristics (Lo et al. 2014). Therefore, strictly speaking, the key to successful cesium biosorption does not rely on the ability of plants to take up cesium. However, a few examples will be discussed here as an alternative to phytoremediation. Marine algae *Sargassum glaucescens* and *Cystoseira indica* treated with formaldehyde and potassium hexacyanoferrate have been shown to dramatically increase cesium biosorption capacity compared to the intact counterparts (Dabbagh et al. 2008). Similarly, various marine algae treated with potassium hexacyanoferrate have been indicated to attain higher cesium biosorption capacity than native biomass and also desorb cesium effectively by application of sodium hydroxide and potassium hydroxide, rendering the biosorbent available for reuse for several cycles (Jalali-Rad et al. 2004). In an aquatic fern *Azolla filiculoides*, treatment with hydrogen peroxide and magnesium chloride has been reported more effective than potassium hexacyanoferrate treatment for cesium sorption and has been reported to provide for repeated use (Mashkani and Ghazvini 2009). It seems that a large particle size (2-4 mm) is most suitable for efficient biosorption of cesium (Jalali-Rad et al. 2004; Mashkani and Ghazvini 2009). Alternatively, powdered moss (*Funaria hygrometrica*) immobilised in polymer silica matrix in a column has been shown to be capable of adsorbing cesium from low-level radioactive waste solutions and can be regenerated for repeated use after elution with nitric acid (Krishna et al. 2004). Interestingly, mucilaginous seeds of basil (*Ocimum basilicum*) imbibed in water have been demonstrated to show higher cesium adsorption compared to those treated with sodium hydroxide, hydrochloric acid or sodium periodate (Krishna et al. 2004), suggesting that they may serve as a natural alternative to chemically treated immobilised biomass.

5 Conclusion and Future Perspectives

A number of studies have suggested possible cesium transporters and uptake mechanisms in plants, however, most studies on cesium uptake have been performed in heterologous systems and not directly in plants and the detailed molecular regulation of cesium accumulation remains largely unknown. For the practical use of plants in radiocesium remediation, more efficient phytoremediation methods need to be developed. Thus, further elucidation of cesium uptake mechanisms and dissection of the major ports of entry of cesium in plants are required.

Also, the identification and characterization of regulators for cesium uptake and accumulation in plants such as chemicals and microorganisms will be beneficial for developing efficient radiocesium phytoremediation methods.

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Effective Half-Lives of Radiocesium in Terrestrial Plants Observed After Nuclear Power Plant Accidents

Keiko Tagami

Abstract Information on "how fast radiocesium could be removed from plants" is important to decide measures after contamination. To express the decreasing rate, effective half-life (T_{eff}) can be used; T_{eff} is defined as the time required for a 50 % decline of radiocesium in an individual plant or plant population in a natural ecosystem. There are four major radioactivity decrease process in plants when terrestrial environment is contaminated with radiocesium. (1) Removal of radiocesium attached to plant surface by rain and wind (weathering effect), (2) plant mass increase at growing season (dilution effect), (3) removal of radiocesium by shedding leaves and fruits (elimination effect) and (4) decrease of bioavailable radiocesium fraction in soil (aging effect). It is difficult to separate these processes, however, since radiocesium decreasing trend fits well with exponential curves (usually a combination of two exponential curves); effective half-lives (T_{eff}) are reported. This paper summarizes terrestrial plant T_{eff} data in woody and herbaceous plants observed after Fukushima nuclear accident and also compared the data observed after the Chernobyl power plant accident.

Keywords Effective half-life • Fukushima Daiichi Nuclear Power Plant accident • Chernobyl Nuclear Power Plant accident • Herbaceous plants • Woody plants

1 Introduction

The magnitude-9.0 (Richter scale) earthquake on March 11, 2011 triggered a huge tsunami that hit the Pacific Ocean coast of Eastern Japan. The tsunami destroyed the water circulation systems of the Fukushima Daiichi Nuclear Power Plants (FDNPP). Consequently, agricultural crops as well as many other plants became contaminated with radioactive fallout from the accident (Ministry of Health, Labour and Welfare

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(MHLW) 2016; Tagami et al. 2012). Several months later, radiocesium with relatively longer physical half-lives (T_{phy}), i.e., ^{134}Cs ($T_{phy} = 2.06$ years) and ^{137}Cs ($T_{phy} = 30.2$ years) were the only major radiation source to humans remaining in the environment. Although contribution to dose is negligible, it is also necessary to consider ^{135}Cs for its long half-life of 2.3×10^6 years (Zheng et al. 2014).

In the natural environment, decreases of radiocesium concentration in plants with time are usually found after the releases to the atmosphere by nuclear bomb testing and accidental releases (Cline 1981; Komamura et al. 2005; Pröhl et al. 2006; Paller et al. 2014). The radiocesium decreases in plants is usually faster than the physical half-lives of long-lived radiocesium isotopes such as ^{137}Cs and ^{135}Cs. Although the phenomenon is known, if there is not enough numerical information on the decreasing rates in plants, it is difficult to decide measures to recover from contaminated situations. For example, if the decreasing was a slow process then replacement of trees might be considered to decrease dose to humans, but if it was a fast process then it is possible to let the trees be in their original positions without doing any additional remediation acts.

There are four major radioactivity decreasing effects in plants as follows:

Effect-1 (Weathering): Radiocesium released to the environment through the air should directly deposit on plants in particulate, gaseous and/or ionic forms, and then, due to weathering (rain and wind, etc.), some portion of total deposited could be washed off from the plant surface. This process is found in both woody and herbaceous plants.

Effect-2 (Dilution): In a plant growing season, if radiocesium uptake is fast enough to meets plant growth rates, then apparent concentration decrease would not be found. However, generally, decrease in radiocesium concentration due to plant mass increase was found; that is, dilution effect. This process is also found in both wood and herbaceous plants.

Effect-3 (Elimination): Total amount of radiocesium in a plant decreases when the plant removes its tissues such as leaves, fruits, barks, stems and roots because these tissue parts also include radiocesium. This yearly tissue removal event is found in woody and perennial herbaceous plants.

Effect-4 (Soil aging): Soil can retain most radiocesium, but the bioavailability change with time; generally, decreasing trend can be observed after radioactive contaminants were added to soil, which is called as aging effect (declining bioavailable percentage to total radiocesium). It is also necessary to consider radiocesium leaching from surface to deeper layer, and weathering of soil (total radiocesium decrease) affects the radiocesium concentration change with time. Increase of root uptake is the process to slow the T_{eff}, but decrease provide opposite effect.

On the other hand, foliar uptake of re-suspended contaminated soil or dust, radiocesium concentration in plant would increase due to foliar uptake which might slow the radiocesium decrease from the plant; however, the latter effect was inconsequential relative to root uptake process (Hinton et al. 1995). It is also necessary to consider recycle of radiocesium in a plant especially for woody and perennial her-

baceous plants, for example, radiocesium in leaves partially translated to stems/branches before shedding (Tagami and Uchida 2015a), which could slow the decrease of radiocesium in the plant. It is difficult to separate these effects by observing radiocesium concentration in plant in natural ecosystems; thus including all these effects, the environmental half-lives (T_{env}) are observed.

2 Effective Half-life

The effective half-life (T_{eff}), which is defined as the time required for a 50 % decline of radiocesium in an individual plant or plant population in a natural ecosystem. After atmospheric release of radionuclides and their deposition onto plant surfaces, T_{eff} could be calculated by observing concentration decrease with time in plants. For the case of radiocesium, it is reported that the decreasing trend with time in an individual plant or plant population fits well with a combination of two exponential curves as follows when observation time was longer than 1 year (Antonopoulos-Domis et al. 1996).

$$C_t = A\ \exp\left(-\lambda_a t\right) + B\ \exp\left(-\lambda_b t\right) \tag{1}$$

where C_t is the concentration of ^{137}Cs in plant at time t (d), A and B are constants for short and long components, respectively, and λ_a and λ_b are loss rates in a population for short and long components, respectively. Practically, however, a set of T_{eff}s was found when 6–8 years observation results were obtained after the radiocesium release to the atmosphere (Antonopoulos-Domis et al. 1996; Unlü et al. 1995). Indeed, in a short time period, ca. 3–4 years after the atmospheric releases, only the short components have been reported (Antonopoulos-Domis et al. 1991; Tagami and Uchida 2015b). Thus the equation in a short time period is simply used as follows.

$$C_t = A\ \exp\left(-\lambda_a t\right) \tag{2}$$

Thus, T_{eff} is calculated using the following equation:

$$T_{eff} = \ln\left(2\right) / \lambda_a \tag{3}$$

Thus, from fitting of the observation results of radiocesium concentrations in a plant or plant population with time using Eq. 2, effective half-lives (T_{eff}) are firstly calculated using Eq. 2. Relations of T_{eff}, T_{phy} and T_{env} are expressed by the following equation.

$$1 / T_{eff} = 1 / T_{phy} + 1 / T_{env} \tag{4}$$

In this report, T_{eff} are reported using data ^{137}Cs concentrations in several plant species, especially focused on data for several terrestrial plants, i.e., woody and herbaceous plants, observed after Fukushima nuclear accident comparing the data observed after the Chernobyl nuclear accident.

3 Herbaceous Plants

3.1 Very Short-Term Decreasing Rate of Radiocesium in Herbaceous Plants

When radiocesium was released to the atmosphere, direct contamination of aerial part of plant surface should occur, that is, a part of radiocesium deposited on the ground partially intercepted (Carini et al. 2003; IAEA 2010). After the contamination, radiocesium concentration in plants should decrease due to the following two major effects as mentioned above, that is, Effect-1 (weathering) and Effect-2 (dilution). The latter effect is limited when the plant was in mature stage and no additional growing was expected. For the case of the FDNPP accident, heavy radioactive deposition occurred during March, 2011; at that time, most herbaceous plants in the wild was not germinated yet, but only perennial herbaceous plants which have leaves overwinter directly contaminated.

In agricultural fields, winter crops such as leafy vegetables and root vegetables as well as young shoots of wheat were planted in March in Japan, 2011. In order to avoid ingestion of radioactive materials, food monitoring was carried out and the data was applicable to know the radiocesium decreasing rates after the accident. Because the purpose of food monitoring to eliminate foods exceeding standard limit, thus crops measured were suitable size to harvest for markets. For leafy vegetables, such as spinach and lettuce, 2–3 months is necessary to harvest after sowing. Within 60 days after March 11, 2011, therefore, vegetable samples obtained for food monitoring were directly contaminated sometime between at their young to mature stages. Thus the decreasing rates of annual herbaceous plants mainly affected by weathering and mass increase could be calculated. This stage is, however, usually separately reported and not included in the Eq. 1; however, because the information is important, the data are summarized below.

Figure 1 shows, ^{137}Cs concentration change with time in leafy vegetables collected in Fukushima Prefecture and prefectures next to Fukushima Prefectures; the map is shown in Fig. 2. It should be noted that there were two types of reported radioactivity values in March and April 2011: total radiocesium ($^{134}Cs + ^{137}Cs$) or separately determined values (^{134}Cs and ^{137}Cs). If only a total radiocesium concentration was reported, then ^{137}Cs was calculated by subtracting the ^{134}Cs contribution assuming that the ^{134}Cs:^{137}Cs activity ratio was 1:1 on 11 March 2011. From the results, the ^{137}Cs concentration decreases were fitted well using Eq. 2 with p value of <0.001 by *t*-test in both areas. T_{eff} in both areas were calculated and the range was

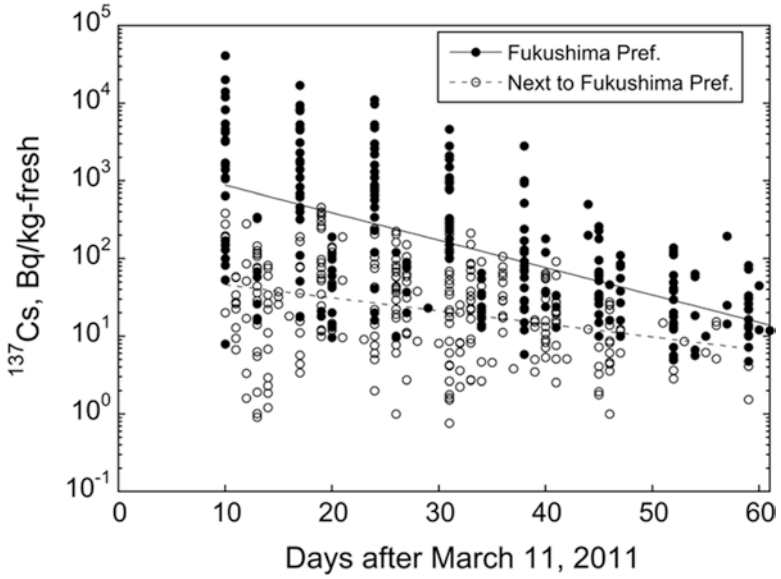

Fig. 1 ^{137}Cs concentration change in leafy vegetables collected in Fukushima Prefecture and prefectures next to Fukushima observed within 60 d after March 11, 2011 (Data adopted from MHLW 2016)

Fig. 2 Map of data collection sites in Japan for Fig. 1

from 8.5 to 17.9 days. For the case of Chernobyl accident, Mück (1997) reported initial T_{eff} values observed after the accident, that is, 4.2 days in lettuce and spinach, and 10.5 days in grass. From these results, it is concluded that weathering and mass increase were the important processes to fast decrease of radiocesium in herbaceous plants. On the other hand, the latter effect could not be found when the plant was in mature stage and no further weight increase was expected.

3.2 Short-Term T_{eff} of ^{137}Cs in Herbaceous Plants

After the contaminated plants were removed from the soil or dead on the soil, then the soil can be the major contamination source for the plants grown on the ground; from this stage, Eq. 1 was applied. For annual plants which were not germinated when the severe depositions occurred after the FDNPP, root uptake was the major radiocesium transfer pathway. It should be noted that only a small portion of total radiocesium in the soil was bioavailable, thus its transfer to plants through roots was generally small compared to the same group elements, e.g., K and Rb (IAEA 2010). Moreover, the bioavailable fraction of radiocesium generally decreases with time after its addition to soil (Cline 1981; Noordijk et al. 1992; Rosén et al. 1996; Tagami and Uchida 1996). Herbaceous plants, especially annual ones are sensitive to this factor; and when only a certain tissue of the plant was collected each year, e.g., cereal grains, beans, etc., then time dependence of the decreasing trend would be found. Unfortunately, however, not many continuous measurement results were found after the FDNPP accident.

Tagami and Uchida (2015b) measured ^{137}Cs in giant butterbur (*Petasites japonicas* (Siebold et Zucc.) Maxim.) and field horsetail (*Equisetum arvense* L.) from 2012 to 2014 grown in wild. Although these two species are perennial herbaceous plants, their above ground parts died every winter so that they can be indicators of radiocesium bioavailability in soil like annual plants. The T_{eff}s observed were ca. 450 (1.2 years) and 360 days (0.99 years), respectively. During this period, total ^{137}Cs concentration in soil and also vertical distribution did not change; thus probably because of the aging effect, bioavailable radiocesium decreased at these T_{eff} values. Smith et al. (1999) reported similar values for grasses with an average T_{eff} value of 1.5 years in UK, probably because grasslands were usually not ploughed so that the measurement conditions were similar to what was observed for wild plants in Japan. For the case of agricultural field, T_{eff} expected to be longer because of soil mixing accelerates the aging effect. Indeed, ^{137}Cs data in soybean (*Glycine max*) and buckwheat (*Fagopyrum esculentum*) grains collected in Fukushima Prefecture in 2012–2014 (detected data only from food monitoring data collated by MHLW 2016) showed longer T_{eff} as shown in Fig. 3. The calculated T_{eff}s were 858 (2.35 years) for soybean and 846 days (2.32 years) for buckwheat. Similar results were reported by Mück (1997) after the Chernobyl accident and the T_{eff} values were 1.4–2.7 years for vegetables and 3.0–3.4 years for cereals in Austria. This soil management difference was also be found in Russia in 1987–1989; T_{eff} for grains, potato and root crops ranged 1.2–2.9 while hay were slightly shorter, 0.9–2.3 (Fesenko et al. 1995).

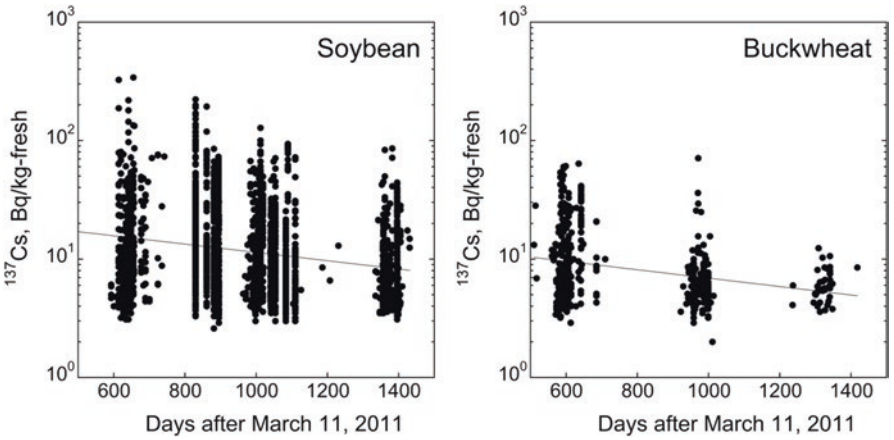

Fig. 3 Short-term ^{137}Cs concentration change with time in soybean and buckwheat collected in Fukushima Prefecture (Data adopted from MHLW 2016)

3.3 Long-Term T_{eff} of ^{137}Cs in Herbaceous Plants

For a longer period of time, leaching to deeper layer and translocation of radiocesium with soil particles need to be considered. Komamura et al. (2001) estimated the contribution of direct contamination from global fallout to the total ^{137}Cs concentration in polished rice grains and found after 1985, the contribution was less than 5 %, thus root uptake pathway was the major contributor after 1985. It should be noted that the effect from the Chernobyl accident was negligible for rice because its planting to the rice paddy field in Japan starts from early May to early June, that is, plant was small or not planted yet so that direct contamination was not important. However, wheat grains were affected because their harvest season is generally around the middle of May to June.

Using the rice grain data reported by Komamura et al. (2005) and additional data from the same group published by Ministry of Education, Culture, Sports, Science and Technology (MEXT 2016), Japan, T_{eff} affected by soil (Effect-4) was calculated. Fifteen sampling sites throughout Japan were used for ^{137}Cs measurements in polished rice grains from 1985 to 2005. For this case, long fraction, λ_b in Eq. 1, was calculated. The results are shown in Fig. 4 and the λ_b was calculated to be 0.0692 year^{-1}; the T_{eff} was 10 years, interestingly, the value was similar to herbaceous plants (T_{eff} = 13.4 years on average) collected in Savanna River site in USA from 1974 to 2011 (Paller et al. 2014).

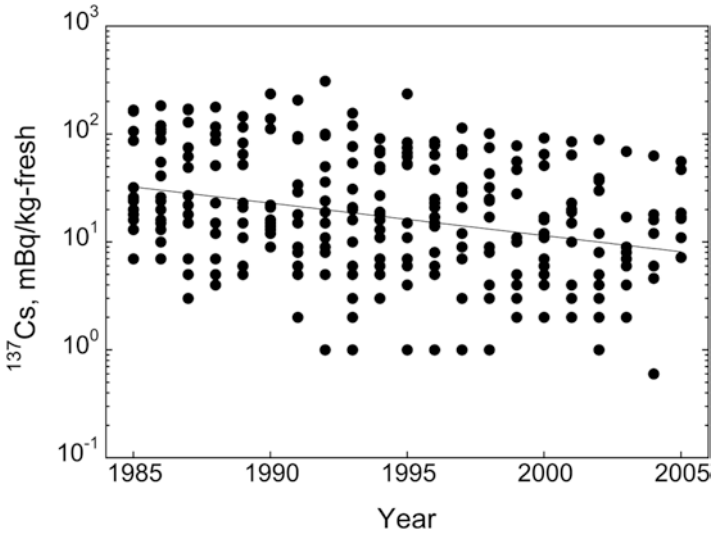

Fig. 4 Long-term ^{137}Cs concentration change in polished rice collected in Japan (Data adopted from Komamura et al. 2005)

4 Woody Plants

4.1 Short-term T_{eff} of ^{137}Cs in Fruit and Tea Plants

Trees are perennial woody plants; when the trees were directly contaminated with radiocesium, the concentration should rapidly decrease with time in a very short period of time as it was found for herbaceous plants, though the data were rarely found. After a certain period of time, radiocesium retained on the above ground parts would be gradually taken up by trees. Evergreen type trees are potentially most effective to take up radiocesium all the year around because leaf is the most important part to take up radiocesium among aerial parts of trees. Topçuoglu et al. (1997) reported that ^{137}Cs from the Chernobyl nuclear accident was stored into the stem of tea trees from old leaves and then translocated to new leaves. Deciduous trees are also possible to take up radiocesium from its aerial parts. Tagami et al. (2012) found radiocesium concentration in new shoots of deciduous trees were between those of evergreen tree and herbaceous plants. There were no emerged leaves of deciduous trees in March 2011 when heavy deposition was observed because it was before the growing season start for the case of the FDNPP accident. Due to the uptake of radiocesium by aboveground parts of trees, radiocesium concentrations in tree tissues in 2011 were highest and then, the concentrations have been decreasing (Kusaba et al. 2015a, b; Sato et al. 2015; Tagami and Uchida 2015c).

After the Chernobyl accident, effective half-lives were reported for trees. Pröhl et al. (2006) summarized T_{env}s of ^{137}Cs in terrestrial environment. T_{env} observed in

pipfruit collected in Germany in 1965–1985 was 5.4 years and that in 1988–1999 was 6.3 years. Thus longer period of time, the T_{env} would be around his range. However, shortly after fallout, ca. within 2–4 years, T_{eff}s were usually shorter than those values. T_{eff} values observed after the Chernobyl accident were reported for fruit trees (Antonopoulos-Domis et al. 1991; Mück 1997) and tea (Unlü et al. 1995); the results are summarized in Table 1. Data obtained after the FDNPP accidents were also listed for comparison (Tagami and Uchida 2015c; Hirono and Nonaka 2016) together with calculated values from literature data (Kusaba et al. 2015a, b; MHLW 2016). In order to calculate T_{eff} from MHLW data, the same criteria reported in Tagami and Uchida (2015c) were used. In brief, the criteria were set that although distribution of radioactivity concentration in a local government area was not uniform, the amount of ^{137}Cs deposited to the land surface was assumed to be the same, and that more than three data per year should be reported in 2011–2013 by the local government. The result in Table 1 shows that the effective half-lives in fruit and tea trees were similar after the Chernobyl and Fukushima accidents. All T_{eff} values ranged from 0.34 to 1.64 years and these values log-normally distributed; thus geometric mean was calculated and the value was 0.86 years.

4.2 *Short-Term T_{eff} of ^{137}Cs in Japanese Trees Obtained After the Chernobyl and Fukushima Accidents*

However, unfortunately, these observations were carried out in different areas which made difficult to compare the T_{eff} values obtained after the Chernobyl and Fukushima accidents, because recent publication mentioned that T_{eff} increased with increasing annual precipitation (Rulík et al. 2014). For statistical analysis, it is necessary to obtain T_{eff} data from the same locations. In the outside of Japan, the effect of Fukushima was small; thus data obtained in Japan were surveyed using environmental radioactivity concentration database compiled by Nuclear Regulation Authority (2016). The database covers wide range of environmental samples, such as agricultural crops, milk, etc.; among them, indicator plants were selected for this analysis. An example is shown in Fig. 5 for ^{137}Cs concentration data in pine needles from 1984 to 2014 collected at one sampling site in Miyagi Prefecture. Using the data 1986–1988 and 2011–2013, T_{eff} in the same sampling site can be compared.

After the data survey, ^{137}Cs data in 2 years old leaves of Japanese black pine (*Pinus thunbergii*) collected at eight sampling sites were identified to be used this analysis. T_{eff} obtained are listed in Table 2. There were no correlations between the T_{eff} between observed after the Chernobyl and Fukushima accident by ANOVA test, and the geometric mean values of T_{eff} were 0.38 and 0.40 years, respectively.

In order to increase the number of T_{eff} values, monitoring sites were selected if the site had a series of ^{137}Cs data in plants in 1986–1988 or 2011–2013. Finally, T_{eff} data for Japanese black pine, tea (*Camellia sinensis*), citrus (*Citrus unshiu*), and Japanese cedar (*Cryptomeria japonica*) were obtained and the summary of the

Table 1 Reported T_{eff} of radiocesium in trees shortly after release

Plant name	Observation period	T_{eff} (in years)	References
Chernobyl			
Apple	1987–1990	0.86	Antonopoulos-Domis et al. (1991)
Apple	1987–1993	1.39	Mück (1997)
Apricot	1987–1990	0.77, 0.84	Antonopoulos-Domis et al. (1991)
Olive	1987–1990	0.83	Antonopoulos-Domis et al. (1991)
Peach	1987–1990	0.84	Antonopoulos-Domis et al. (1991)
Pear	1987–1990	0.69, 0.65	Antonopoulos-Domis et al. (1991)
Pear	1987–1993	1.23	Mück (1997)
Sweet cherry	1987–1990	0.66, 0.68	Antonopoulos-Domis et al. (1991)
Tea	1986–1988	0.34	Unlü et al. (1995)
Fukushima			
Apple	2011–2012	0.61	Renaud and Gonze (2014)
Apple	2011–2013	0.84, 0.96	This study (Data from MHLW 2016)
Blueberry	2011–2013	0.49	This study (Data from Kusaba et al. 2015b)
Chestnut	2011–2013	0.67	This study (Data from Kusaba et al. 2015a)
Chestnut	2011–2013	0.87	This study (Data from MHLW 2016)
Grape	2011–2012	0.57	Renaud and Gonze (2014)
Japanese apricot	2011–2012 2012–2013	0.47 0.76	Renaud and Gonze (2014)
Japanese apricot	2011–2013	0.50, 0.51, 0.54, 0.62, 0.73, 0.83	This study (Data from MHLW 2016)
Peach	2011–2012	0.51	Renaud and Gonze (2014)
Peach	2011–2013	0.63, 0.64, 0.68	This study (Data from MHLW 2016)
Pear	2011–2012	0.72	Renaud and Gonze (2014)
Persimmon	2011–2013	0.63, 0.83, 1.02, 1.13, 1.14, 1.20, 1.30	Tagami and Uchida (2015c)
Sweet cherry	2011–2012	0.52	Renaud and Gonze (2014)
	2012–2013	1.64	
Tea	2011–2013	0.36	Hirono and Nonaka (2016)
Yuzu	2011–2013	0.70, 0.84, 1.30	This study (Data from MHLW 2016)

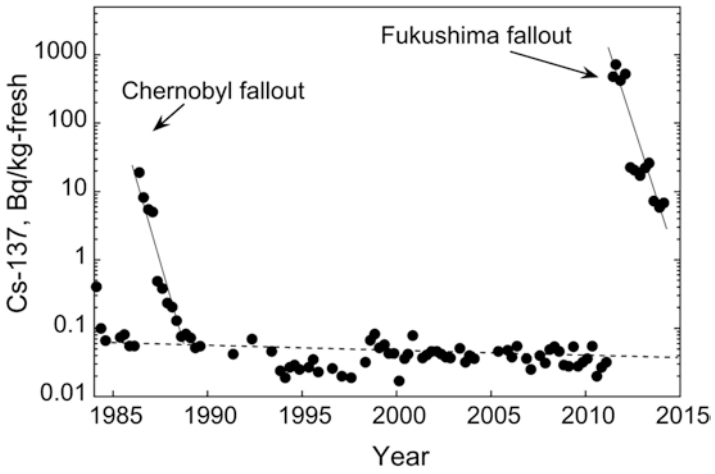

Fig. 5 ^{137}Cs concentrations in Japanese black pine needles collected in Miyagi Prefecture

Table 2 Comparison of T_{eff} of ^{137}Cs in pine leaves originated from the Chernobyl and Fukushima accidents

	After the Chernobyl accident (Observation period: 1986–1988)			After the Fukushima accident (Observation period: 2011–2013)		
Prefecture	Number of ^{137}Cs data	λ (year^{-1})	T_{eff} (years)	Number of ^{137}Cs data	λ (year^{-1})	T_{eff} (years)
Miyagi, A	11	2.34	0.30	11	2.02	0.34
Miyagi, B	22	2.00	0.35	12	1.85	0.37
Fukushima, A	8	1.32	0.52	9	2.74	0.25
Fukushima, B	9	1.22	0.57	11	1.89	0.37
Niigata, A	6	3.20	0.22	6	1.50	0.46
Niigata, B	11	1.98	0.35	11	1.25	0.55
Kyoto	3	1.45	0.48	3	0.98	0.71
Saga	11	1.74	0.40	7	2.15	0.32
Geometric mean		1.82	0.38		1.72	0.40

results are shown in Table 3. Significant difference was not observed among the data observed after the Chernobyl and Fukushima accidents by ANOVA test except for Japanese black pine ($p = 0.023$, with logarithm data of T_{eff} values), although the geometric mean values differed only 0.07 years.

Table 3 Calculated T_{eff} of ^{137}Cs in leaves of Japanese black pine, Japanese cedar and tea trees, and fruit of *Citrus unshiu* collected in Japan

	After the Chernobyl accident (Observation period: 1986–1988)			After the Fukushima accident (Observation period: 2011–2013)		
Plant name	Number of T_{eff} data	λ (year^{-1})	T_{eff} (years)	Number of T_{eff} data	λ (year^{-1})	T_{eff} (years)
Japanese black pine	27	1.53	0.45 (0.22–0.79)	17	1.84	0.38 (0.25–0.71)
Tea	12	1.65	0.42 (0.26–0.76)	10	1.67	0.41 (0.33–0.68)
Citrus unshiu	8	1.28	0.54 (0.39–0.69)	2	1.25	0.54 (0.54–0.56)
Japanese cedar	3	1.38	0.50 (0.45–0.62)	1	1.90	0.36

5 Conclusions

Effective half-lives, T_{eff}, of radiocesium in herbaceous and woody plants were summarized in this paper comparing data obtained after the Chernobyl and FDNPP accidents, because T_{eff} is important to know how fast radiocesium removed from plants. T_{eff}s observed after these two accidents within 2–4 years for herbaceous plants as well as trees were similar. The T_{eff} data obtained after the Chernobyl accident were mostly obtained in temperate regions, that is, Germany, Austria, Greece, etc. The T_{eff} data obtained in Japan after the FDNPP accident were also temperate areas; under similar climate conditions, growing rates of plants would be the same, which caused the same range T_{eff} data. For a longer time period, however, radiocesium decline rates might change among different areas due to different precipitation rates and temperatures. It is also important to measure seasonal change to know main factor to eliminate radiocesium from plants. Thus, continuous measurement of radiocesium in plants is necessary for a long-time period after nuclear accident(s).

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Cultivar Difference and Fertilizer Effects on Radioactive Cesium Accumulation in Rice Grown in Fukushima Paddy Field from 2011 to 2014

Nobuhiro Tanaka, Yoshihiro Ohmori, and Toru Fujiwara

Abstract After the Fukushima Daiichi nuclear power plant accident triggered by the Great East Japan Earthquake on March 11, 2011, radioactive cesium (Cs) was released and widely distributed in agricultural field in Tohoku distinct. Radioactive Cs has relatively long half-lives, and thus contamination of agricultural products by radioactive Cs will be long-term serious problem for human health. Therefore, we have observed the levels of radioactive Cs contamination in rice plants grown in Fukushima from 2011 to 2014. Our study demonstrated that there is considerable variation in radioactive Cs concentration among a wide variety of rice cultivars grown in Fukushima paddy field. These results indicated that Cs accumulation levels were genetically regulated in rice plants. In addition, the high nitrogen and low potassium condition produced an increase in the Cs concentration both in straw and brown rice, suggesting that Cs accumulation levels in rice was also controlled by environmental factor.

Keywords Radioactive Cesium • Brown rice • Rice cultivar • Fertilizer effect

1 Introduction

The accident at the Fukushima Dai-ichi Nuclear Power Plant in March 2011 released large amount of radionuclides to the broader area, including the paddy fields. Among the major radionuclides released, the radioactive isotopes of cesium (^{134}Cs and ^{137}Cs) are observable radionuclides because they have relatively long half-lives

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(2.06 years for ^{134}Cs and 30.2 years for ^{137}Cs) (Matsumura et al. 2011). To reduce the Cs accumulation level in rice, we need to understand the mechanism of Cs uptake and transportation in rice plants. Cs accumulation level in rice is thought to be both genetically and environmentally regulated. In this chapter, we measured the radioactive Cs concentration in 85 rice cultivars to identify the genetic factors, and we also investigated the effects of fertilizer on radioactive Cs accumulation in commercial rice cultivars to reveal the environmental factors.

2 Difference in Radioactive Cs Accumulation Among Rice Cultivars Grown in the Paddy Field at Fukushima from 2011 to 2014

Cs belongs to alkali metal group and it is absorbed from the soil and transported to various parts of rice plants, such as the brown rice and straw which are served as foods for human and livestock, respectively. It is believed that both Cs uptake and transport in plants are mediated by potassium (K) transporters. In *Arabidopsis*, one of the KUP/HAK/KT type transporters, AtHAK5, appears to take up non-radioactive Cs (^{133}Cs) under low K conditions (Qi et al. 2008). *AtCNGC1* is also reported as a candidate gene that determines the natural variation of Cs concentrations (Kanter et al. 2010). Understanding of the mechanisms of Cs uptake and transportation in rice plants is required for the reducing Cs accumulation in rice, however, to our knowledge, the gene that control Cs concentration has not been reported to date in rice. To investigate whether there is variation in Cs accumulation levels in plant body among different rice cultivars is useful for the identification of the genes related to Cs absorption. In this section, we report the measurement of the radioactive Cs concentration in selected 85 rice cultivars actually grown in paddy field in Fukushima (Ohmori et al. 2014a).

2.1 Radioactive Cs Accumulation Among Selected 85 Rice Cultivars Grown in Fukushima Paddy Fields in 2011

Tsukada et al. (2002) have reported the concentration of radioactive Cs in rice grown in Aomori prefecture before the Fukushima Dai-ichi Nuclear Power Plant accident, and the difference of ^{133}Cs concentrations in brown rice among different rice cultivars has been also reported (Yamaguchi et al. 2012). However, the amount of radioactive Cs fallout from the Fukushima accident was far above the level observed in past fallout. Thus, we reinvestigate the radioactive Cs concentration in rice grown in the Fukushima area. We have selected 85 rice cultivars from the World Rice Core Collection (WRC), the Japanese Rice Landrace Mini Core Collection (JRC), and other domestic varieties (Table 1). 67 varieties from WRC cover 91 % of

Table 1 List of the 85 rice cultivars tested in this study

No.	Cultivar	No.	Cultivar	No.	Cultivar
1	Karahoushi	30	Co 13	59	Meguro Mochi
2	Houmanshindenine	31	Tachiaoba	60	Senshou
3	Mansaku	32	Bekoaoba	61	Moritawase
4	Himenomochi	33	Kusahonami	62	Momiroman
5	Akage	34	Dango	63	Yamada Baka
6	Hassokuho	35	Tupa 121-3	64	Hetadawee
7	Kahei	36	Hirayama	65	Taichung 65
8	Shinyamadaho 2	37	Naba	66	Mack Kheua
9	Aichiasahi	38	Hinode	67	Rikutou Rikuu 2
10	Hamasari	39	Muha	68	Omachi
11	Hakamuri	40	Touboshi	69	Chinsurah Boro 2
12	Shinriki Mochi	41	Bouzu Mochi	70	Hiyadachitou
13	Raiden	42	Fukuhibiki	71	Okka Mososhi
14	Puluik Arang	43	Okabo	72	Daw Dam
15	Ginbouzu	44	Kabashiko	73	Deng Pao Zhai
16	Vary Futsi	45	Jamaica	74	Basilanon
17	Ishijiro	46	Shichimenchou Mochi	75	Koshihikari
18	Nipponbare	47	Khauk Yoe	76	Badari Dhan
19	Mogumoguaoba	48	Tachisugata	77	Hoshiaoba
20	Bekogonomi	49	Shiroine	78	Asominori
21	Nishiaoba	50	Mizuhochikara	79	Kaneko
22	Nagoyashiro	51	Akamai	80	Oiran
23	Aikoku	52	Kusanohoshi	81	Khau Mac Kho
24	Kameji	53	Sekiyama	82	Joushuu
25	Yumeaoba	54	Fukoku	83	Wataribune
26	Moroberekan	55	Shinriki	84	Iruma Nishiki
27	Hosogara	56	Leaf Star	85	Shinshuu
28	Kasalath	57	Chinya		
29	Kyoutoasahi	58	Gaise n Mochi		

the genetic variation in about 37,000 rice landraces. 50 varieties from JRC cover 87.5% of genetic variation in about 2,000 Japanese rice landraces (Ebana et al. 2008; Kojima et al. 2005). We planted the selected 85 rice cultivars in the Fukushima paddy field, and grown until maturity. Harvested straw and brown rice were separated and radioactive Cs concentrations in them were independently determined (Table 2). The radioactive Cs (^{134}Cs and ^{137}Cs) was determined by using Ge semiconductive detector as described by Mimura et al. (2014) and Tanoi (2013). The ^{134}Cs and ^{137}Cs concentrations in straws distributed in the ranges of 19.4–73.4 and 10.3–100.3 Bq kg^{-1} respectively (Table 2), displaying about four and tenfold variations among the 85 rice cultivars. In addition, the means were 38.9 and 39.0 Bq kg^{-1}, respectively, and the medians were 35.8 and 35.5 Bq kg^{-1} respectively (Table 2).

Table 2 Mean (standard deviation), median, and range for Cs concentration in different rice cultivars grown at Fukushima in 2011

	Mean (SD)	Median	Range
Cs concentration in straw			
^{134}Cs (Bq kg^{-1})	38.9 (13.5)	35.8	19.4–73.4
^{137}Cs (Bq kg^{-1})	39.0 (17.2)	35.5	10.3–100.3
Cs concentration in brown rice			
^{134}Cs (Bq kg^{-1})	8.1 (4.7)	6.7	0.7–20.3
^{137}Cs (Bq kg^{-1})	11.6 (5.8)	10.2	2.7–26.6

In brown rice, ^{134}Cs and ^{137}Cs concentrations ranged from 0.7 to 20.3 Bq kg^{-1} and 2.7 to 26.6 Bq kg^{-1} respectively (Table 2). As in straw, radioactive Cs concentrations showed around 25- and 10-fold variations in brown rice among the 85 rice cultivars (Table 2). The means were 8.1 and 11.6 Bq kg^{-1} respectively, and the medians were 6.7 and 10.2 Bq kg^{-1}, respectively (Table 2). In brown rice, the mean values of ^{134}Cs and ^{137}Cs concentrations were 21 and 30 % of that obtained in straw, respectively (Table 2). There was large variation in radioactive Cs concentration both in straw and grains among selected 85 cultivars. These variations are caused by diversity of gene function among selected cultivars, and the diversity can be used to isolate the genetic factors related to Cs uptake or transport in rice.

Next, we investigated the correlation of radioactive Cs concentrations in straws and brown rice among 85 rice cultivars. We found significant positive correlation between ^{134}Cs and ^{137}Cs concentrations in straw and brown rice, that are $p = 2.2 \times 10^{-12}$, and the $p = 3.4 \times 10^{-29}$, respectively (Fig. 1a, b). The coefficients of determination (R^2) were 0.57 and 0.79 for straw and brown rice, respectively (Fig. 1a, b). Both of ^{134}Cs and ^{137}Cs concentrations showed a significant positive correlation between straws and brown rice, that are $p = 1.2 \times 10^{-6}$ and $p = 4.9 \times 10^{-7}$ for ^{134}Cs and ^{137}Cs, respectively (Fig. 1c, d). The coefficients of determination were 0.33 and 0.35 for ^{134}Cs and ^{137}Cs, respectively (Fig. 1c, d). These results suggested that the Cs concentrations in grains can be approximately estimated from Cs concentrations in straws.

2.2 Radioactive Cs Accumulation Among 15 Selected Rice Cultivars Grown in Fukushima Paddy Field from 2012 to 2014

From the results of the Cs concentration of brown rice in 2011, we selected 15 rice cultivars to test the reproducibility, and planted them at Fukushima paddy field in 2012 and 2013. Deng Pao Zhai, Asominori, Kaneko and Khau Mac Kho (Line number 73, 78, 79 and 81 in Table 1) were selected as high Cs concentration cultivars. Whereas, Mansaku, Akage, Hassokuho, Aichiasahi, Hamasari, Kameji, Kasalath and Wataribune (Line number 3, 5, 6, 9, 10, 24, 28 and 83 in Table 1)

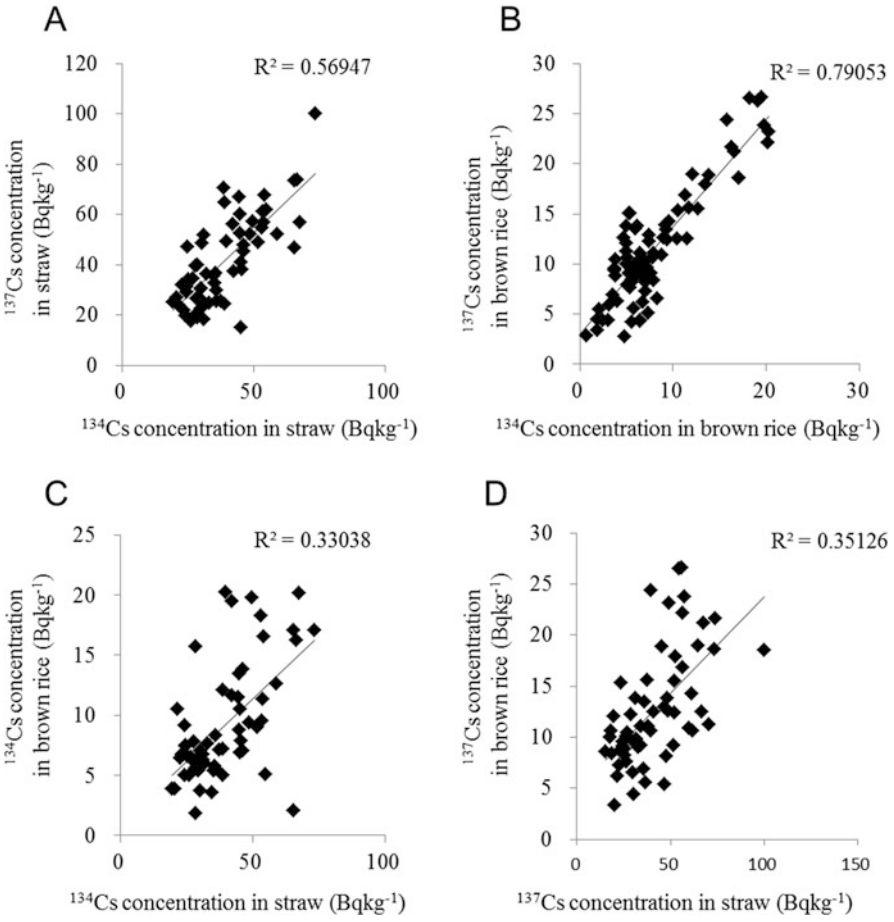

Fig. 1 Correlation diagram for Cs concentration (**a**) Correlation diagram for ^{134}Cs and ^{137}Cs concentration in straw (**b**) Correlation diagram for ^{134}Cs and ^{137}Cs concentrations in brown rice (**c**) Correlation diagram for ^{134}Cs concentration in straw and brown rice (**d**) Correlation diagram for ^{137}Cs concentration in straw and brown rice The corresponding coefficients of determination (R^2) are shown. The *black line* represents the linear regression line corresponding to the least square adjustment of all the data

were selected as low Cs accumulating cultivars. In addition, Nipponbare, Taichung 65 and Koshihikari (Line number 18, 65 and 75 in Table 1) were selected as typical Japanese cultivars. Deng Pao Zhai, Asominori and Khau Mac Kho showed significantly higher concentrations of ^{137}Cs in brown rice among selected 15 rice cultivars (Fig. 2). On the other hand, Mansaku, Aichiasahi, and Hamasari showed relatively lower ^{137}Cs concentrations in brown rice compared with the other cultivars (Fig. 2). We have also planted 15 selected rice cultivars at Fukushima paddy field in 2014. Deng Pao Zhai, Asominori and Khau Mac Kho also showed higher

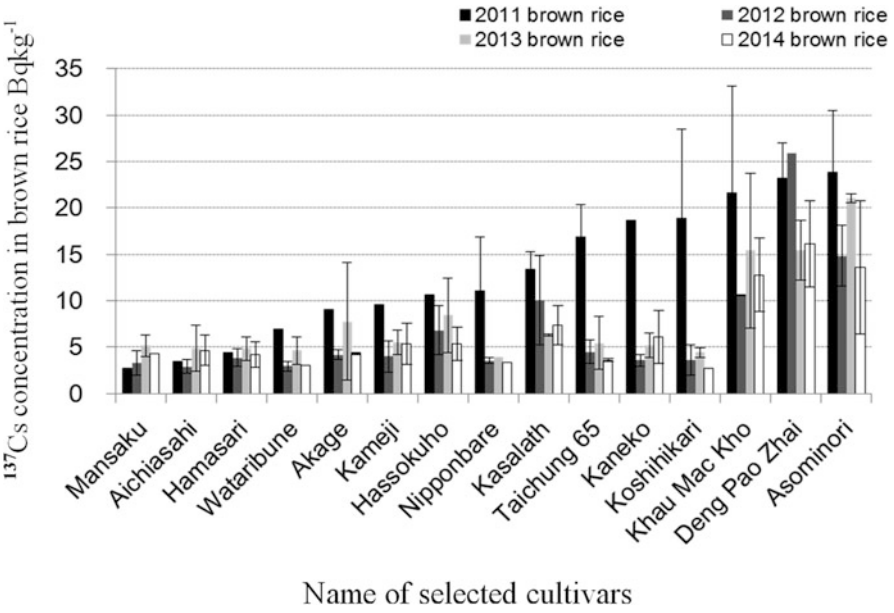

Fig. 2 Comparison of ^{137}Cs concentrations (Bq kg^{-1}) in brown rice from selected cultivars among 2011–2014. *Black, dark gray, gray* and *white boxes* indicate 2011, 2012, 2013 and 2014 data respectively. Means and standard deviations are shown (n = 3)

concentrations of ^{137}Cs in brown rice among selected 15 rice cultivars (Fig. 2), and concentrations of ^{137}Cs in straw were below detection limits in 2014 (data not shown). In conclusion, concentrations of ^{137}Cs among 15 rice cultivars were comparatively conserved in four-year investigations, suggesting that these three cultivars have a low ability in Cs accumulation into brown rice. We believe that our results are important as an actual data of radioactive Cs accumulation levels among the different rice cultivars in the Fukushima paddy field. A molecular genetic approach for these rice cultivars with different Cs accumulation levels may lead to identify the genes that regulate Cs uptake and transportation in rice.

3 Radioactive Cs Concentration of Rice Straw and Brown Rice Grown in Paddy Field Under Four Fertilizer Treatments at Kawamata-cho

In Sect. 2, we described about the identification of genetic factors that determines the Cs uptake or transportation in rice. However, to decrease the Cs concentration in rice, we also need to investigate the effects of cultivation conditions for Cs absorption from soil. Cs uptake and transportation are known to be mediated by

several K transporters because both K and Cs belong to alkali metal group (Qi et al. 2008; Jabnoune et al. 2009). Thus, both K and Cs transportation by K transporters is thought to compete with each other in plants. It has been also reported that Cs uptake is enhanced under low K conditions in various plants (Shaw 1993). For example, K fertilizer applications reduce Cs accumulation in crops such as wheat, barley, rye, and potato under K deficient conditions (Lemmbrechts 1993). In addition, ammonium ion (NH_4^+) is known to affect the elution of Cs ion from soil by replacing NH_4^+ with Cs^+. Therefore, high concentration of NH_4^+ in soil positively induces Cs^+ elution from soil, resulting in enhance of Cs absorption by plant. It has been also reported that the application of N fertilizer enhances Cs accumulation in plant under some kind of soil conditions (Lemmbrechts 1993; Smolders et al. 1997).

To investigate the effect of fertilizer application on radioactive Cs accumulation in rice, we planted and harvested two commercial cultivars of rice, Koshihikari and Hitomebore at Ishidairayama, Yamakiya and Kawamata-cho in Fukushima in 2011 and 2012 under four different fertilizer conditions, i.e., normal, −K, −K +2 N, and no fertilizer conditions (Ohmori et al. 2014b). For normal condition, a commercial fertilizer including 8-18-16 (N-P-K) was applied as a base fertilizer. The amounts of N, P, and K given in normal condition were 6, 9, and 8 kg per 1000 m^2, respectively. In -K condition, total amount of N, P, and K were 6, 9, and 0 kg per 1000 m^2. In -K and +2 N condition, total N, P, and K amounted were 12, 9 and 0 kg per 1000 m^2. For control, no fertilizers were applied.

In 2011, the concentrations of ^{137}Cs in straw under −K +2 N condition were 1.5 and 2.0 times higher than that under normal condition in Koshihikari and Hitomebore, respectively (Fig. 3a). ^{137}Cs concentration in straw was slightly high under –K condition compared with normal condition (Fig. 3a). There was no difference in ^{137}Cs concentration in straw between under normal and no fertilizer condition.

In 2012, we have planted these two cultivars at same paddy field again, and investigated reproducibility of Cs concentrations. ^{137}Cs concentration in straw was the highest under the −K +2 N condition (Fig. 3b). ^{137}Cs concentration in straw was also high under the −K condition (Fig. 3b). Under the no fertilizer and normal conditions, ^{137}Cs concentration in straw was almost indistinguishable. These trends for ^{137}Cs concentrations were similar between Koshihikari and Hitomebore (Fig. 3). In conclusion, the pattern of radioactive Cs concentrations in straw was similar between 2011 and 2012.

We also determined the radioactive Cs concentrations in brown rice grown at Kawamata-cho in 2011 and 2012. In 2011, ^{137}Cs concentration in brown rice was the highest under the −K +2 N condition. It was about twice as high as under the normal condition (Fig. 4a). Under the −K condition, ^{137}Cs concentration in brown rice was also higher than those under the normal condition in Koshihikari (Fig. 4a). ^{137}Cs concentration in brown rice was the lowest under the no fertilizer condition (Fig. 4a).

In 2012, the trends of ^{137}Cs concentrations in brown rice under the four different fertilizer conditions were very similar to those observed in 2011 (Fig. 4b). The ^{137}Cs concentration in brown rice was the highest under the –K +2 N condition, and the lowest under the no fertilizer condition both in 2011 and 2012 (Fig. 4a, b). However, the ^{137}Cs concentration in brown rice under the −K condition in 2012 was no difference from that under the normal condition (Fig. 4b).

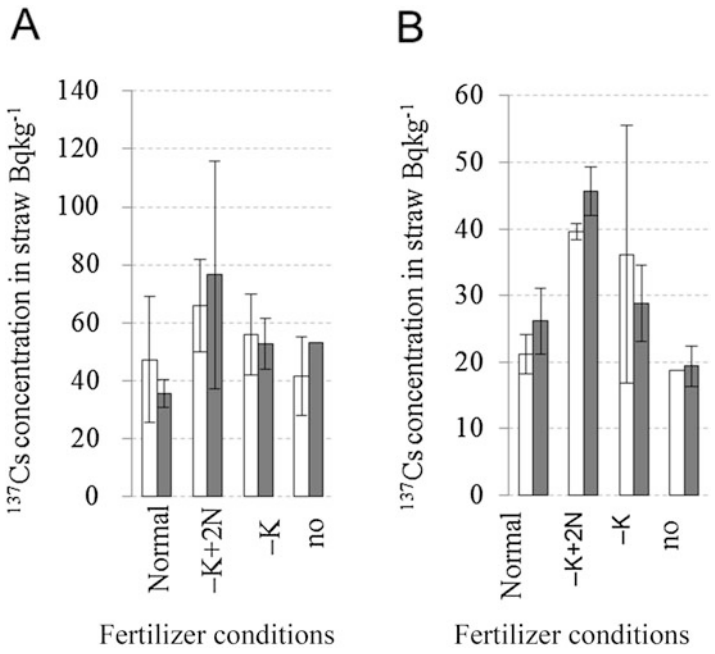

Fig. 3 Cs concentrations in straw under four different fertilizer conditions in 2011 and 2012 (**a**) ^{137}Cs concentrations in straw grown in 2011 (**b**) ^{137}Cs concentrations in straw grown in 2012. *White* and *black boxes* indicate Koshihikari and Hitomebore respectively. Means and standard deviations are shown (n = 3). The concentrations are presented on a dry-weight basis. Normal, the normal fertilizer condition; −K + 2 N, twofold nitrogen without potassium condition; −K, normal N and P but no K condition; and no, no fertilizer condition

In our study, low K conditions have a tendency to promote the radioactive Cs accumulations both in straw and brown rice grown in the paddy field at Kawamata-cho in Fukushima. This result can be explained by chemical competition between K and Cs in K transporters. The fertilizer condition that caused the highest radioactive Cs concentration in rice was low K and high N condition in both 2011 and 2012 (Figs. 3 and 4). These results indicate that N fertilizer also affects radioactive Cs concentrations in both straw and brown rice under low K condition (Figs. 3 and 4). In our hypothesis, N fertilizers promote the elution of radioactive Cs from the soil surface by replacing NH_4^+ with Cs^+. Our results indicate that we need to consider not only K fertilizer but also N fertilizer conditions to reduce harmful-levels of radioactive Cs in rice (over 100 Bq kg^{-1}; the governmental new safety standards for radioactive Cs in food products in Japan).

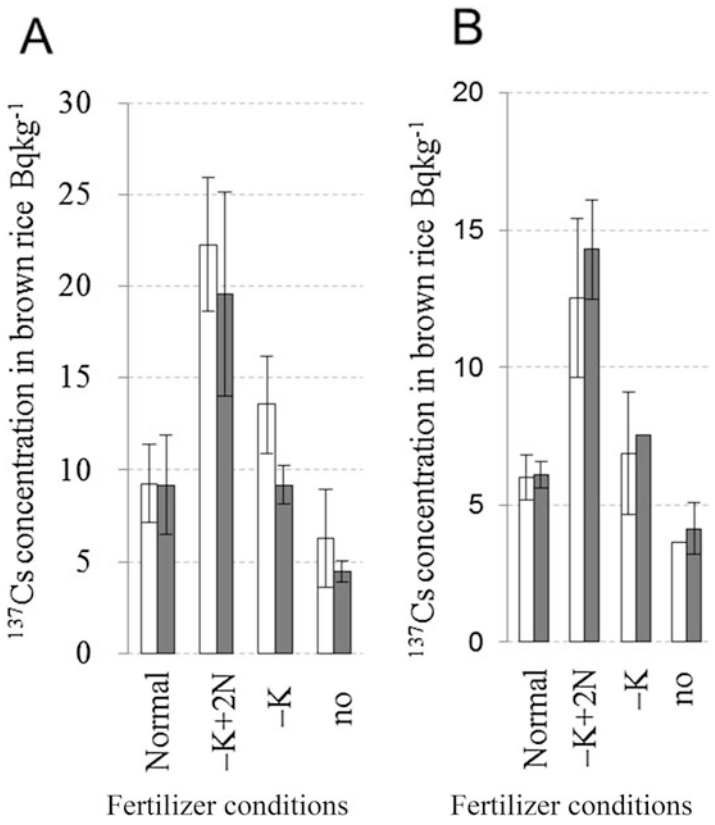

Fig. 4 Cs concentrations in brown rice under four different fertilizer conditions in 2011 and 2012 (**a**) ^{137}Cs concentrations in brown rice grown in 2011 (**b**) ^{137}Cs concentrations in brown rice grown in 2012. *White* and *black boxes* indicate Koshihikari and Hitomebore, respectively. Means and standard deviations are shown (n = 3). Fertilizer conditions are the same as described in the legend of Fig. 3

4 Conclusion

In Sect. 2, we selected several rice cultivars as low Cs concentration cultivars. These cultivars are useful materials for the isolation of genetic factors that regulate Cs uptake and transport in rice. In Sect. 3, we determined the desirable fertilizer condition for rice cultivation to reduce the Cs accumulation in brown rice. Our results demonstrated that radioactive Cs concentration in brown rice is on a declining trend for past 4 years (Fig. 2). Our 4-year investigations of radioactive Cs-contaminated rice show that the rice grown in Fukushima paddy field contains radioactive Cs much less than the governmental safety standards for radioactive Cs in food (100 Bq kg^{-1}).

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Distribution of ^{137}Cs Between the Components of Pine Forest of Chernobyl NPP Exclusion Zone

Maksim Kudzin, Viachaslau Zabrotski, and Dzmitry Harbaruk

Abstract The State nature conservation research institution "Polessye State Radiation Ecological Reserve" was developed on the territory of exclusion zone of Chernobyl NPP in 1988. Since that time up to now the square of pine stands was changed due to opposite factors. It was increased due to natural reforestation and decreased because of water logging, forest fires and arrangement of the fire breaks. Total increase of the square of pine stands was about 0.5 %. Now it is ascertained fact that forest litter of pine stands (mossy, heath, bilberry types of pine forest) remains to be the main accumulator of ^{137}Cs retaining 50–60 % of its inventory. The common peculiarity of distribution of ^{137}Cs through mineral part of soil profile in pine stands was its accumulation by upper 0–5 cm under litter layer with following sharp decreasing in dipper layers. For all types of pine stands the transfer of ^{137}Cs into tree organs and tissues is decreasing in following order: roots > bark > wood. The minimal accumulation of ^{137}Cs by wood is the common feature of the absorption of the ^{137}Cs by dominant species of the understory regardless the forest type. The ability of grassy and semifrutex vegetation of all studied types of the pine forests to absorb ^{137}Cs is decreasing in following order: filices > heath ≥ cowberry ≥ meadow grass > mosses. According to the results obtained the maximal accumulation of ^{137}Cs by the vascular plants regardless of the pine forest type fits their phytomass.

Keywords Chernobyl • Radioactive contamination • Dose rate • Cesium-137 • γ-Spectrometry • Transfer factor • Pine forest

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1 Introduction

The State nature conservation research institution "Polessye State Radiation-Ecological Reserve" (PSRER) began operating on the territory of the Belarusian sector of the exclusion zone of the ChNPP in September 1988. It's superior organization—Department of Liquidation of Consequences of the Catastrophe at the Chernobyl NPP of the Ministry of Emergency Situations of Belarus. PSRER was created in order to perform radiobiological researches and execute radiation-ecological monitoring of the exclusion zone. The primary goals of PSRER's activity includes protection of the forest against fire and afforestation of the lands, primarily those subjected to wind and water erosion. The reserve is located on the South-East of Belarus. It stretches from East to West for 70 km and from North to South for 48 km. At present the area of the reserve is 2,162 km^2. The south border of PSRER coincides with state border of Belarus with Ukraine along about 130 km. The maps of contamination of territory of PSRER by ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239,240}Pu, and ^{241}Am were created only in 2009 due to work of several Belarusian organizations. The levels of contamination of the reserve by ^{137}Cs are presented in Fig. 1.

The significance of the forest ecosystems as for radionuclide migration was realized after the catastrophe at the Chernobyl NPP. The forest is one of the most radiosensitive natural ecosystems that are accounted for by high retentiveness of the

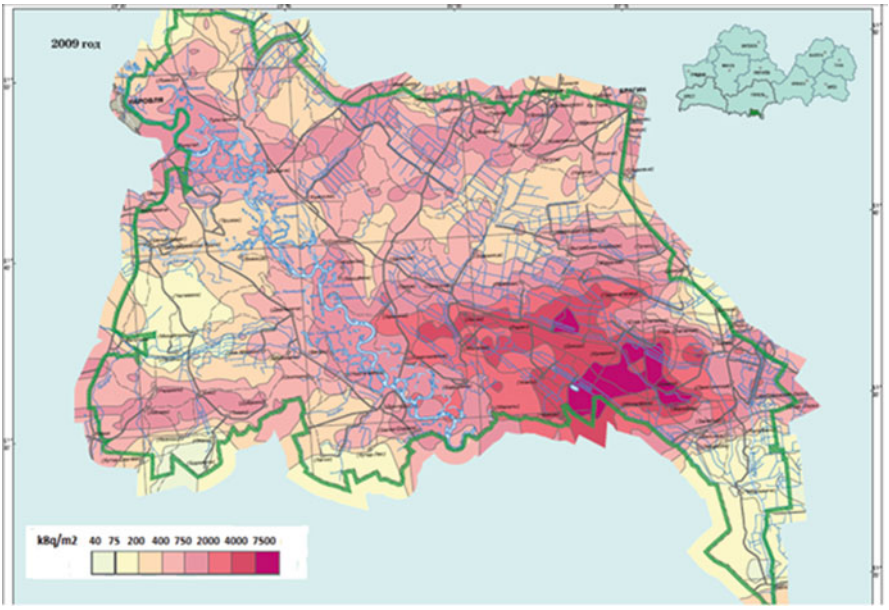

Fig. 1 Contamination (kBq/m^2) of territory of PSRER by ^{137}Cs on 2009 (Reproduced from Izrael and Bogdevich 2009)

radionuclides by tree's crown and low radio resistance of many types of the forest, especially conifers (Kozubov 1987). The large woodlands and even single trees usually participate in filtration of airstreams providing effective deposition of the radioactive particles. At the expense of large surface of the aboveground phytomass—especially for the case of young coniferous growth—deposition of the radionuclides in the forest is 6–12 times higher and depletion of the radioactive cloud occurs 5 times speedier than by open meadows (Perevolotski 2006).

Scotch pine (*Pinus sylvestris*) (further "pine") is the predominant forest-forming type of trees not only in the area of catastrophe but also on the territory of Byelorussian part of exclusion zone of ChNPP. About 43.4 % of area of forest land (527 km^2) of the reserve falls on the plantings of pine family. One of the key peculiarities of forest land in contrast to open meadows is availability of forest litter playing the role of the accumulator of organic substance as well as the main part of the radionuclides. The decreasing of the intensity of the forestry activities in the pine forests of the reserve (because of the radioactive contamination) leads to the increasing of quantity of natural tree waste and depth of forest litter. The litter's depth in pine forests of artificial origin is increasing up to 20 % in comparison with forests where forest exploitation is running (Usenya 2011). Some authors (Kozubov 1987; Perevolotski 2006; Krasnov et al. 2007) investigating the intensity of radionuclide migration in forest lands point out its direct dependence on the types of forest-vegetation conditions. In uniform conditions of forest growing the behavior of the radionuclides in soils covered by the forest differs from that in non-covered soils. The migration of the radionuclides depends also on composition of the stand that is stipulated by different composition and depth of forest litter. Between the ages of 50–60 years the coniferous forest can accumulate from 50 to 90 % of the deposited radionuclides (Kozubov 1987). Taking into account that the mean age of the pine forests of the reserve is equal 49 years, their high radioactive contamination and radiation exposure received from the moment of the catastrophe could be confidently proposed. In that connection it seems actually to continue the researching of the radionuclide behavior in the coniferous forest phytocoenosis. That allows determining the parameters of their migration and accumulation by separate components with a glance of interspecific influence of the synfoliums on each other and in consideration of specific character of the radioactive fall-out on the territory of the exclusion zone of ChNPP.

2 Object and Methods of Research

The ecosystems' components of three different types of pine forest namely; moss, heath, and bilberry-covered (Table 1) were the objective of the research in 2014. A few words about what we understand by the type of forest. This is a wood lot characterized by the same species composition, the same quantity of the story's, close fauna and specifically complex of the forest-vegetation conditions (climatic, soil-ground, and hydrological). The types of forest could be distinguished due to availability of

Table 1 Forestry–radiological characteristic of the plantings on the sample areas

Composition[a]	Forest type[b]	Age, year	Growth class	Forest density, m^3/km^2	Mean		DR, μSvh^{-1}		^{137}Cs contamination density, kBq/m^2
					D[c], cm	H[d], m	3–4 cm above soil	1 m above soil	
10P	mos/A_2	52	I	53,000	21.9	20.2	1.14	0.92	1344.3
10P	mos/A_2	57	I	47,000	21.7	19.9	1.43	1.06	1546.2
10P	mos/A_2	71	I	54,000	27.3	24.9	0.95	0.77	938.9
10P	hea/A_2	73	II	39,000	28.5	21.3	1.45	1.21	2172.6
10P	hea/A_2	64	II	44,000	25.2	23.4	0.89	0.68	980.1
10P	hea/A_2	73	III	30,000	24.0	17.8	0.42	0.37	328.2
9P1B	bil/A_2	64	Ia	45,000	32.9	25.7	0.66	0.59	989.5
9P1B	bil/A_2	61	Ia	49,000	32.5	25.6	1.87	1.37	2318.3
9P1B + A	bil/A_2	64	I	33,000	32.7	23.1	0.37	0.30	481.1

[a]P—pine, B—birch, A—aspen
[b]mos—mossy, hea—heath, bil—bilberry
[c]D—diameter of the trunk
[d]H—tree's height

plant-indicators (specialist species). The type of the dominant plant-indicator depends on such parameters as soil fertility, humidity, structure, etc.

The bilberry type of pine forest differs from other two types by more rich soil-hydrological conditions. Namely these conditions make for the high productivity of all story's including arboreal one as well as biological diversity of vegetation. The bilberry which was exacting to the soil fertility and humidity dominates in a live soil cover. The heath type of pine forest occupies the poor soils. On that reason this type of pine forest exhibits low species diversity of vegetation of all story's. The standing wood of such type was sparse and represented mainly by pine which development is depressed because of poor soil conditions. The understory was developed insufficiently. The mossy pine forest has intermediate position regarding soil fertility. The birch trees begin to appear in these standing woods. The young growth of pine grows at open areas. The soil humidity was enough for abundant development of a live soil cover presented mainly by moss.

In each type of the forest three sample areas (further "SA") were arranged. Preliminarily the gamma-radiation dose rate (further "DR") was measured on the height 1 m in each sample area to be sure that the level of radioactive contamination of the SA is similar. If difference in values of dose rates was higher than 10 % another place for sample area was chosen with more uniform characteristics of radioactive contamination. In accordance with adopted normative documents in and around radiation examination and monitoring of forest resources (Bogdevich 2006; TKP 240-2010; TKP 498-2013) the dimensions of the sample area corresponded 50 × 50 m. Samples of soil with alive soil cover and forest litter were taken in nine points (in the angles and along the diagonals of the sample area) not closer than 1 m from the tree's trunks. The standard sampler with diameter 4 cm and height 20 cm

was used. Dose rate was measured in the points of soil taking at the heights 3–4 cm and 1 m above soil surface. The dose rate at the height 3–4 cm was measured for the rejection of the unrepresentative points (with uncharacteristic local contamination). In the case when the value of dose rate measured at the height 3–4 cm differs from the DR measured at the height 1 m more than two times this place was regarded as uncharacteristic and soil was sampled in another point.

The sampling of forest litter and soil was exempted in undisturbed places between tree's crones not less than 1–1.5 m from the trunk (or stump) avoiding glades, openings, depressions.

To determine the content of ^{137}Cs along the soil profile the following fourfold (quadruplicated) samples were taken:

- Forest litter using pattern (template) 20 × 20 cm from three layers (tree waste (Ol)), enzymatic (Of) and humified (Oh);
- Soil in 5 cm layers on depth 40 cm with use of sampler 20 × 20 cm.

To determine the content of ^{137}Cs in organs and tissues of the plants the following samples were taken:

- Roots and phytomass of the vegetation of a live soil cover.
- Roots, wood and leave of the vegetation of understory.
- Roots, bark and wood of the vegetation of trees of main story.

The samples of the wood without bark were taken on the SA at the height 1.3 m from 20 to 30 growing trees of I-II classes of growth and development according to the Kraft (Craft). The age-specific wood borer was used (the borer reached the center of the trunk). One core sample was taken from each tree. The quantity of the trees from which the core samples were taken depended on the values of the trees trunks diameters:

Diameter in cm	<25	25–30	>35
Quantity of the trees	30	25	20

The trees for the sampling of the bark were chosen from the growing trees of I-II classes of growth and development according to the Kraft. Bark samples were taken from 7 to 10 growing trees from the height 1.3 m (Bulko et al. 2014). The cylindrical borer with diameter 40 mm was used to take the bark with bast (inner bark). Sometimes, after the withdrawal of the borer from the tree the bast remained on the trunk—in that case it was separated using knife and added to the bark sample. The quadruplicated root samples were taken simultaneously with soil through the digging procedures.

The sampling of roots, wood and leaves of the vegetation of understory as well as roots and phytomass of the vegetation of a live soil cover was exercised evenly through all sample areas in quantity enough for the representative sample. The samples selected were packed into the doubled plastic bags providing the presence of the sample's passport. After transportation they were dried on the air until

air-dried condition. Soil was thoroughly mixed; stones, roots and other inclusions were removed. The samples of the forest litter, wood, roots, bark, leaves, grassy plants and mosses were cut into pieces 2–3 cm with help of scissors and pruners. Then they were mixed again until uniform distribution of the matter of the sample was reached.

The dose rate measurements were fulfilled using AT6130 Radiation Monitor. The content of ^{137}Cs in soil samples was determined with help of AT1315 Gamma Beta Radiation Spectrometer. All measurements were fulfilled by the staff of laboratory of spectrometry and radiochemistry which is accredited according to the requirements ISO/IEC 17025 since 2005. Since 2011 laboratory of the reserve regularly participates in worldwide proficiency tests organized by IAEA on the determination of natural and artificial radionuclides in some environmental objects. The laboratory does it to receive the proof of reliability of its analytical data. Transfer factors of ^{137}Cs in tissues and organs of plants (T_{ag}) were calculated to be used as characteristics of their ability to accumulate the ^{137}Cs regardless of density contamination of the soil. T_{ag} was calculated as activity concentration in plant compartments (Bq kg^{-1} dry weight) divided on density of contamination of the soil (Bq m^{-2}).

3 Current State of Pine Forests of Belarusian Part of Exclusion Zone of ChNPP

Since 1988–2014 (for 26 years from the date of formation of PSRER) the square of forest covered lands has been increased on 384 km^2 (on 31.8 %). The intensive development of the forest-forming processes of the small-leaved stands as well as processes of secondary swamping of the formerly reclaimed areas are the reasons of the abovementioned alterations. In first half of twentieth century the square of swamped lands in the borders of contemporary PSRER amounted to about 900 km^2 or 41.7 % of its territory. About 850 km^2 or 39.3 % of the territory of reserve were drained in 1950–1970 years. But in the last decades the gradual silting and overgrowing of the soil-reclamation canals and trenches as well as failure of culverts and other water-controlling hydro engineering constructions result in some troubles in earlier operational hydrological regime of polder systems of former agricultural lands coming to their flooding and waterlogging. Last decades the second growth of the small-leaved types of trees is registered on the open plots of the forest lands where renewal of the pine tree is difficult. That led to the increasing of the forest covered fraction of the Belarusian part of exclusion zone and accordingly to the change of specie's composition of the plantings because of decreasing of pine fraction on 10.6 %.

The pine plantings are widespread in the PSRER. Predominance of their fraction in specie's composition of the forests of exclusion zone remains up to now (Fig. 2). Pine forests occupy 527 km^2 or 43.4 % of the square of the forest resources. During the concerned period the square of pine forests increased on 77 km^2 and their percentage—on 3.5 %. Activities in the framework of maintenance of the

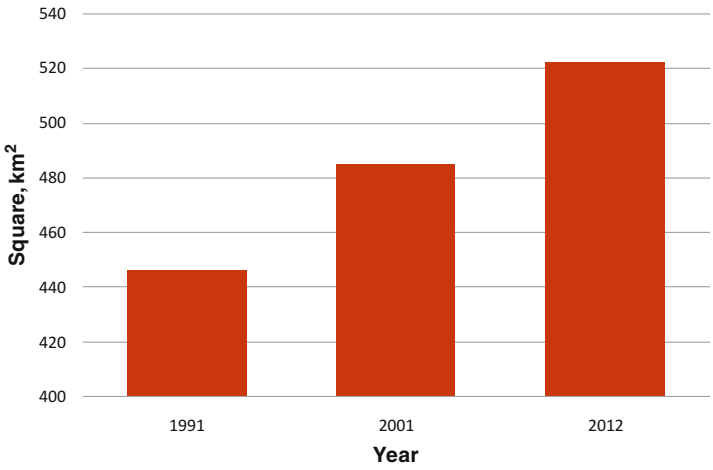

Fig. 2 Dynamics of the square of pine forests in the forest covered lands of Belarusian part of exclusion zone

Table 2 Influence of forest fires and activities in the framework of maintenance of the territory of exclusion zone on the area of pine stand in the period from 1991 to 2014

Factors influencing on forest square	Area, km^2	Percentage of pine forest
Increasing due to reforestation on the former agricultural lands	66.64	12.8
Decreasing due to arrangement of the fire breaks	10.64	2.0
Decreasing due to forest fires	53.89	10.3

territory of exclusion zone as well as forest fires were the main factors determining the changes in the areas of pine stands (Table 2). These factors determine the processes of forest formation and formation of vegetation in a whole until now. The forests of Belarusian part of exclusion zone are characterized by high level of natural fire hazard. The set of fire precaution measures was carried out to decrease the risk of fire incidents. It includes the formation of the fire breaks, arrangement of water ponds, flooding of peaty lands through making of dams on soil-reclamation canals, etc. Arrangement of fire breaks of 12, 20, 40 m width leads to decreasing of the fraction of pine forest on 2 %.

Redistribution of the pine plantings on age groups occurs since 1988: the square of young growth decreased from 67.9 % (302.6 km^2) to 33.4 % (176 km^2), the square of middle-aged plantings increased in more than two times—from 31.9 % (142 km^2) to 66.3 % (347 km^2). The peculiarity of current age structure of pine forests is the predominance of the young growths and middle-aged plantings, occupying 33.4 % (176 km^2) and 65.8 % (347 km^2) accordingly and low fraction of ripening 0.7 % (3.6 km^2), mature and over mature 0.1 % (0.59 km^2) plantings (Scheme 2012).

The natural interrupting and as a result increasing of the wastes and depth of the litter occurs in artificial plantings growing without man control more intensively than in natural plantings. The maximal quantity of tree wastes is forming between 20 and 60 years (Ipat'ev 1999). So, in consideration of the predominance of young growths and middle-aged plantings the high accumulation of the tree wastes could be supposed.

4 Distribution of ^{137}Cs Between Horizons of Forest Litter and Soil in Predominant Types of Pine Forest

The specificity of the radioecological researches from one side and genesis and structure of the forest soils from the other obligate to consider vertical distribution of the radionuclides in soil profile separately in organogenic horizon (forest litter) and mineral layer. The quota of the radionuclides (percentage from the total density of the contamination of the soil profile) presented in separate layers is used usually as indicator of the intensity of the vertical distribution of the radionuclides.

The capability of the forest litter to accumulation of ^{137}Cs depends on its depth, structure and composition, availability of the moss cover and portion of the conifers in the planting as well as availability of the understory's hardwood, bushes and semifrutex. The retention of the radionuclides by the litter is descending in increasing of the fraction of the hardwood in planting and changing of the soil watering from automorphic to hydromorphic type (Perevolotski 2006; Perevolotski and Perevolotskaya 2012a, b). The layer of the forest litter in pine plantings is thick. The slow mineralization of the coniferous-moss's tree waste (6–10 years) (Krasnov et al. 2007) leads to migration of only little part of the radionuclide into humic-eluvial horizon. The content of ^{137}Cs in the forest litter is increasing with increasing of its depth up to 3.5–4.0 cm. There is no change in ^{137}Cs content with further increasing of the litter's depth (Shcheglov 2000).

The distribution of ^{137}Cs between forest litter and soil depends on type of the forest. In the case of pine plantings of PSRER the content of ^{137}Cs in forest litter can reach 48.4 % in mossy pinery, 63.6 % in heath pinery, and 53.9 % in bilberry pinery (Fig. 3).

The percentage (from total content in soil profile) of ^{137}Cs in layer Ol on all sample areas changes insignificantly from 1.2 % in bilberry pinery and 1.3 % in mossy pinery to 2.0 % in heath pinery (Fig. 3). The differences in content of ^{137}Cs in layers Of and Oh are more substantial. In the case of enzymatic layer the percentage of ^{137}Cs changes in mean from 16.2 in heath pinery to 28.9 % in bilberry pinery. In humified layer the ^{137}Cs content changes from 23.8 % in bilberry pinery to 45.4 % in heath pinery (Fig. 3). It should be noted that in bilberry and mossy forest types there is no big difference between content of ^{137}Cs in layers Of and Oh, but in the case of heath pinery its content in layer Oh is almost 3 times higher than in layer Of. So, the redistribution of the ^{137}Cs between layers of forest litter depends on intake of more

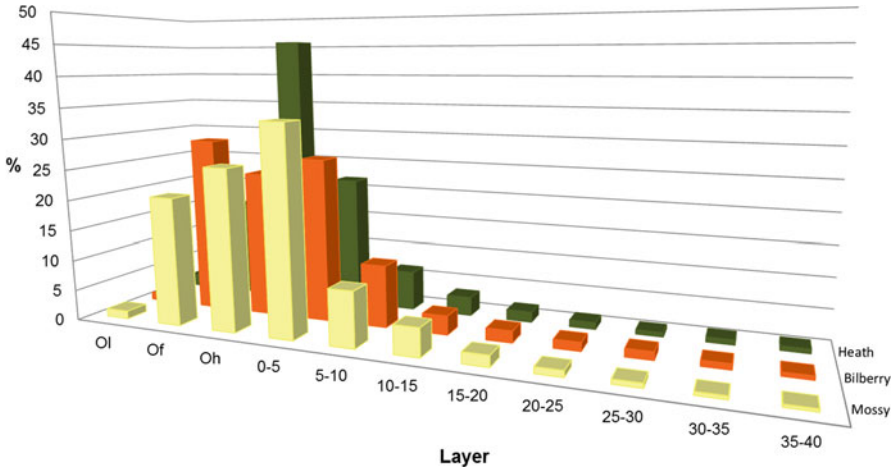

Fig. 3 Distribution of ^{137}Cs in litter and mineral part of forest soils belonging to the bilberry, heath and mossy types of pine forest

pure vegetative tree waste to the soil surface and speed of destruction of organic matter. On that reason the migration of ^{137}Cs between different layers of forest litter is characterized by different intensity. The minimal content of ^{137}Cs in studied forest types is characteristic for the upper layer Ol and seems could be regarded as stable. For pine forests of the reserve it equals 1.5 % that corresponds to the data of other researchers (Shcheglov 2000). The main part of ^{137}Cs is accumulated in the layers Of and Oh. The corresponding values are not stable at the moment. The maximal content of ^{137}Cs is characteristic for the bottom layer (Oh) of the forest litter.

The irregular distribution of ^{137}Cs is observable in mineral part of soil profile. The ^{137}Cs distribution in under litter part of forest soils depends on large quantity of the factors:

- dynamics of accumulation and decomposition of the organic residues
- depth and structure of the forest litter
- mixed character and mosaicity of the soil cover
- the regime of soil watering
- genesis, fertility and acidity of the soil
- granulometric and mineralogical composition of the soil
- relief's element
- age of the planting
- predominant type of the trees and the types of forest-vegetation conditions
- digging activity of wild animals
- vital activity of the soil fauna, mushrooms, natural extracts forming from the leaves
- transfer deep into the soil with atmospheric precipitation and through root system of the plants

– capillary phenomenon, diffusion, forest fires
– climatic and weather conditions (Markina 2005; Perevolotski 2006; Krasnov et al. 2007; Averin 2007; Perevolotskaya 2008).

It is known that the velocity of vertical migration of the radionuclides differs for the different forest types and different layers. According to the present knowledge about 90–98 % of ^{137}Cs is situated in the upper 5 or 10 cm layer (Kozubov 1987; Krasnov et al. 2007). The accumulation of ^{137}Cs by the upper 0–5 cm under litter layer is overall regularity of its distribution through the mineral soil profile of all studied forest types of the reserve. That layer contains 21.8 % of ^{137}Cs in the heath type of the forest, 26.7 % in the bilberry type and 33.6 % in mossy type. The content of ^{137}Cs in deeper soil layers is sharply reduced. The differences in ^{137}Cs content for all types of the forest are insignificant beginning from the depth 15–20 cm where ^{137}Cs content changes from 1.7–1.9 %. The fraction of ^{137}Cs in the layer 35–40 cm is less than 1 % for any forest type (Fig. 3). As a rule the increasing of the soil moisture leads to more intensive radionuclide migration through vertical soil profile. The increasing of the soil fertility at the same soil moisture hinders the penetration of the radionuclide deep down the soil. The inverse relation is valid for the forest litter (Perevolotski 2006; Krasnov et al. 2007).

For the upland pine forests of the reserve the increasing of the soil moisture promotes to the vertical migration of the ^{137}Cs through the mineral part of the soil. For the example the relative content of the ^{137}Cs in soil layer 20–25 cm equals 1 % in mossy and health types of the forest. But the same value of ^{137}Cs content in the bilberry forest type is reached in deeper layer 30–35 cm (Fig. 3). This is because of higher moistening of the soil in bilberry forest type. Present data on distribution of ^{137}Cs through the vertical soil profile points on its approximately equal dividing between organic and mineral soil parts in the cases of bilberry and mossy forest type. This is not the case for the healthy type where main part of the ^{137}Cs (63.6 %) is concentrated in forest litter. According to the available data on distribution of the ^{137}Cs (Fig. 3) between layers of forest litter its main part moved to the under layer. So it can be concluded that the main accumulators of the ^{137}Cs in soils of the upland pine forests of the reserve are the under layer of forest litter and the upper layer (0–5 cm) of the mineral part; that point on the possibility of the further transfer of the ^{137}Cs into mineral part of the soil and next to the forest vegetation.

5 Accumulation of ^{137}Cs by Phytomass of Pine Forests

Up to now the process of secondary radioactive contamination of wood vegetation in forest is continued. Its driving force is entering of the radionuclides through the roots with mineral nutrition. It is worth to note that the less age of the stand the more intensive is the accumulation of radionuclides. This is because more active metabolism of younger trees (Kozubov 1987). The activity concentration (AC) of ^{137}Cs in tissues and organs of pine are widely dispersed (Table 3) under the impact of

Table 3 Activity concentration of ^{137}Cs in tissues and organs of pine, kBq kg^{-1}

Type of pine forest	Tissues and organs	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Mossy	Bark	3	6.24	2.65	4.53	9.29
	Wood	3	1.17	0.60	0.51	1.68
	Roots	3	11.43	5.38	5.84	16.57
Heath	Bark	3	1.85	0.50	1.28	2.23
	Wood	3	0.96	0.25	0.78	1.24
	Roots	3	5.72	1.53	4.00	6.92
Bilberry	Bark	3	2.50	2.04	0.21	4.14
	Wood	3	0.73	0.41	0.26	1.00
	Roots	3	6.66	4.29	1.90	10.22

N: quantity of the sample areas

Table 4 Density of contamination of soil by ^{137}Cs, kBq/m^2

Type of pine forest	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Mossy	3	1276.5	309.3	938.9	1546.2
Heath	3	1321.5	741.9	811.8	2172.6
Bilberry	3	1263.0	948.6	481.1	2318.3

N: quantity of the sample areas

complex of biotic and abiotic factors influencing on its accumulation and redistribution between stand's components. One of the most important is the density contamination of upper soil layers where the roots are situated. According to the data received there is no dependence of activity concentration of ^{137}Cs in the components of the stand on density contamination of the soil. There is large variability of soil contamination by ^{137}Cs, especially in case of bilberry pine forest (Table 4). At the same time when we compare the mean values the difference is not so remarkable.

The differences in the magnitudes characterizing the accumulation of ^{137}Cs by organs and tissues of pine are quite large and reach for wood 6 times, for bark—44, for roots—9. The data received are characterized by great variability because of great quantity of factors influencing the entering of radionuclides into vegetation. Transfer factors of ^{137}Cs in tissues and organs of pine (T_{ag}) were calculated to characterize the ability of the plant to accumulate the ^{137}Cs regardless of density contamination of the soil. Analysis of the transfer factors of ^{137}Cs in tissues and organs of pine (Table 5) shows that irrespective of forest type the ability of the tree components to accumulate the ^{137}Cs decreases in the following order: roots > bark > wood.

The distribution of radionuclides between tree's tissues and organs without taking into account its underground components will be incomplete. It is known (Matsko et al. 1991; Sak 1993) that large quantity of radionuclides is situated in the rhizosphere of the plants. The content of the radionuclide in the roots varies accordingly to depth of the soil profile. It is known also that the quantity of the thin roots of the woody plants is increasing with degradation of soil fertility. On that reason it is understandable the formation of the main root as well as surface root

Table 5 Transfer factors of ^{137}Cs in tissues and organs of pine (T_{ag}, $10^{-3}m^2kg^{-1}$ dry weight)

Type of pine forest	Tissues and organs	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Mossy	Bark	3	4.87	1.33	3.4	6.0
	Wood	3	0.97	0.51	0.40	1.40
	Roots	3	9.23	4.39	4.30	12.70
Heath	Bark	3	1.60	0.79	1.00	2.50
	Wood	3	0.90	0.46	0.40	1.30
	Roots	3	4.97	2.10	2.90	7.10
Bilberry	Bark	3	2.0	1.97	0.40	4.20
	Wood	3	0.63	0.32	0.40	1.0
	Roots	3	5.43	2.14	4.0	7.90

N: quantity of the sample areas

systems by the pine on the poor sand soils and concentration about 80–90 % of the small roots in the upper soil horizons (Shcheglov 2000). The granulometric composition and water regime are the most important factors from the soil-ecological ones in relation to degree of their influence on inhomogeneity of the accumulation of the radionuclides in the plants. The presence of gleization signs in lowered areas of heath pinery (groundwater level 50–100 cm), bilberry pinery (groundwater level 20–30 cm), and mossy pinery (groundwater level 50–100 cm) in different hydrological regimes points on different transfer factors of ^{137}Cs in different forest types (Lovchiy 2012). The root systems of heath and bilberry pine forests accumulate ^{137}Cs approximately 1.8 times less than that of mossy pinery. The decreasing of transfer factor of ^{137}Cs indicates on dominance of processes of its un-exchange fixation by soil (Bulko 2014). Thereupon it could be concluded that in the same hydrological regime the bioavailability of ^{137}Cs is decreasing with increasing of soil fertility.

Understory and young growth belong to the factors determining the development of the forest phytocoenosis. Quantity of types of species which could be regarded as young growth (future forest canopy) on all sample areas is limited and presented by the only specie–oak tree. The absence in young growth of the representatives of the natural renewal of the mother species is not typical for total pine formation of the reserve because the renewal of the good and satisfactory quality is registered on 25 % of square of heath pinery, 55 % moss pinery and 16.6 % bilberry pinery. Such species as Persian berry and mountain ash are growing in the understory under canopy of moss pinery. It should be noted that moss pinery is characterized by the highest diversity of understory species; at the same time its edaphic conditions are more close to that of heath pinery and differ from that of bilberry pinery. It is known that understory is more active in taking of the radionuclides than canopy trees because of peculiarities of structure of its root system and their close to surface arrangement. The type of the soil and its regime (hydrological, temperature, aerial and so on) has the key significance for the vegetation and distribution the radionuclides in it. The pine forests under study are growing on sod-podzol automorphic,

sod-podzol semi-hydromorphic and sod-podzol hydromorphic soils. The heath and bilberry pine forests are characterized by more high levels of ground water in comparison to mossy pine forest; that allows saying about some similarity of edaphic conditions of these forest types.

Distribution of the components of the species dominating in the understory on the level of ^{137}Cs accumulation showed the close accumulation of the radionuclide by the plants regardless the conditions of forest growing (Tables 6, 7, and 8). The plants absorb from the soil the movable species of ^{137}Cs located near their roots that is in rhizosphere. The degree of radioactive contamination of the soil, biological peculiarities of the plants and value of fraction of movable species of the radionuclide determine the degree of accumulation of ^{137}Cs by the plants (Parfenov et al. 1995). The young growth of English oak has the highest transfer factors of ^{137}Cs. Second place occupies mountain ash; Persian berry shows minimal ability to absorb ^{137}Cs. So the order in which the accumulation of ^{137}Cs decreases for all studied types of pine forest for all components of the plants of understory (wood, roots and leaves) is following:

Wood—English oak > mountain ash > Persian berry
Roots—English oak > mountain ash ≥ Persian berry
Leaves—English oak > mountain ash > Persian berry

Table 6 Transfer factors of ^{137}Cs in tissues and organs of understory plants of mossy pine forest (T_{ag}, $10^{-3}m^2kg^{-1}$ dry weight)

Specie	Tissue/organ	N	Arithmetic mean	Standard deviation	Minimum	Maximum
(Persian berry) *Frangula alnus* Mill.	Wood	3	1.33	0.58	1.00	2.00
	Roots	3	2.40	1.57	0.60	3.50
	Leaves	3	2.80	1.20	1.60	4.00
(Mountain ash) *Sorbus aucuparia* L.	Wood	2	6.1	0.99	5.40	6.80
	Roots	2	15.25	2.19	13.70	16.80
	Leaves	2	9.2	0.99	8.50	9.90
(English oak) *Quercus robur* L.	Wood	2	6.30	0.57	5.90	6.70
	Roots	2	15.45	3.89	12.70	18.20
	Leaves	2	15.85	5.45	12.00	19.70

N: quantity of the sample areas

Table 7 Transfer factors of ^{137}Cs in tissues and organs of understory plants of heath pine forest (T_{ag}, $10^{-3}m^2kg^{-1}$ dry weight)

Specie	Tissue/organ	N	Arithmetic mean	Standard deviation	Minimum	Maximum
(Persian berry) *Frangula alnus* Mill.	Wood	2	0.95	0.21	0.80	1.10
	Roots	2	2.95	0.07	2.90	3.00
	Leaves	2	1.00	0.00	1.00	1.00

N: quantity of the sample areas

Table 8 Transfer factors of ^{137}Cs in tissues and organs of understory plants of bilberry pine forest (T_{ag}, $10^{-3}m^2kg^{-1}$ dry weight)

Specie	Tissue/ organ	N	Arithmetic mean	Standard deviation	Minimum	Maximum
(Persian berry) *Frangula alnus* Mill.	Wood	3	0.72	0.39	0.30	1.06
	Roots	3	8.70	10.14	2.60	20.4
	Leaves	3	3.97	3.75	1.70	8.30
(Mountain ash) *Sorbus aucuparia* L.	Wood	3	1.83	0.50	1.30	2.30
	Roots	3	5.23	1.91	3.80	7.40
	Leaves	3	6.87	2.00	4.80	8.80
(English oak) *Quercus robur* L.	Wood	2	4.05	0.78	3.50	4.60
	Roots	2	10.65	4.03	7.80	13.50
	Leaves	2	12.80	5.23	9.10	16.50

N: quantity of the sample areas

In edaphic conditions of heath type forest all components of mountain ash collect ^{137}Cs more intensively than tissues and organs of Persian berry. Some authors (Parfenov et al. 1995) mark out the mountain ash as specie accumulating ^{137}Cs because of its selective ability to accumulate alkaline-earth elements. The soils of investigated types of the forests are characterized as acidic unsaturated, good drained, weakly retaining different substances. Deficiency of potassium brings about the increasing of entering of the radionuclides to plants (Shcheglov 2000). The ^{137}Cs transfer factors for the components of the tree canopy and understory are decreasing from dry to humid edaphic conditions. The reverse regularity is noted for poor sandy soils (Ipat'ev 1999; Shcheglov 2000; Perevolotski 2006).

Soil particles with dimensions less than 0.01 mm (physical clay), gleization horizon and powerful humic horizon are presented in soil of heath and bilberry pine forests; all of them together stimulate the decreasing of entering of ^{137}Cs into components of the tree canopy and understory.

The ability of the components of understory vegetation to accumulate ^{137}Cs depends on type of forest and specie:

Mossy pine forest; Persian berry—Leaves > Roots > Wood
Bilberry and heath pine forest; Persian berry—Roots > Leaves > Wood
Mossy pine forest; Mountain ash—Roots > Leaves > Wood
Bilberry and heath pine forest; Mountain ash—Leaves ≥ Roots > Wood
Mossy and bilberry pine forest; English oak—Leaves > Roots > Wood

One peculiarity is the same for all species of the understory vegetation; that is minimal ability of the wood to accumulate ^{137}Cs. In moving from dry to more moistened conditions the component characterizing by maximal accumulation of ^{137}Cs changes from leaves to roots. It could be explained by domination of root nutrition and depending of metabolic activity of these organs on growing conditions. The highest ^{137}Cs accumulation by the most active organs regardless of edaphic conditions is observed for English oak that could be specified by domination of root nutrition.

High values of transfer of ^{137}Cs into plants of a live soil cover are characteristic for spore-bearing plants as well as for plants from several families—cowberry, sedge, liliaceae, primrose, heath, carnation, and some others. In ten times more high accumulation of the radionuclides by grassy plants occurs in all type forests, especially pine, in comparison with meadow-swamped lands. The species composition of grassy plants forming the live soil cover in studied forest types differentiates because of differences in soil fertility and its hydrological regime. The vegetation of a live soil cover in heath and bilberry types of pine forest is distinguished by higher species diversity than in mossy type. Mosses represent the original group of the embryophytes. They play the important role in forest ecosystems being dominants or co-dominants of a live soil cover. The development of mosses promotes to the retention of the radionuclides in surface layers. The mosses impact on under layers by its excretions and through the dying off of their bottom parts. Consequently, the environment is forming under the mosses which has increased and more stable humidity, pH value close to 4, less daily interval of temperature variations and favorable conditions for the development of fungi having large mycothallus. In addition more powerful forest litter is forming with higher inventory of organic matter and radionuclides enriched by the elements of mineral nutrition (Krasnov et al. 2007; Mamihin 2012). Mosses are the dominant plants on the sample areas.

The development of moss cover favors to retention of radionuclides in surface soil layers. It could be explained by biological peculiarities of the mosses—from one side by great delaying ability in relation to the aerial depositions; from the other side—by including of already consumed radionuclides into little biological cycling (Perevolotski and Perevolotskaya 2012a, b). The vascular plants growing on the mossy cover of pine forests receive some nutrients from the mossy cover due to trophic connection between mosses and these plants. At the same time the overall regularity for all forest types is higher transfer of ^{137}Cs into plants of grassy-semi frutex story in comparison with mosses. In its turn^{137}Cs transfer factor of *Dicranum* is higher than that of *Pleurozium schreberi*; this fact corresponds to the data of other authors (Krasnov et al. 2007). Specie's selectivity of ^{137}Cs transfer into phytomass and roots of the plants growing in the mossy forest changes according to following order:

Table 9 Transfer factors of ^{137}Cs into components of a live soil cover of mossy pine forest ($T_{ag}(10^{-3}m^2kg^{-1})$)

Specie	Organs and tissues	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Vaccinium myrtillus L.	Phytomass	2	14.10	6.36	9.60	18.60
	Roots	2	7.05	0.92	6.40	7.70
Dicranum polysetum Sw.	Phytomass	3	8.97	2.32	7.20	11.60
Pleurosium schreberi Brid.	Phytomass	3	7.97	2.60	5.30	10.50

N: quantity of the sample areas

Overground part of bilberry>*Dicranum*>*Pleurozium schreberi*>underground part of bilberry (Table 9).

Specie's selectivity of accumulation of ^{137}Cs by plants growing in the heath forest changes according to following order:

Phytomass—Adder-spit>Bilberry>Heath>*Dicranum*>*Pleurozium Schreberi*
Roots—Adder-spit>Bilberry>Heath (Table 10)

Specie's selectivity of accumulation of ^{137}Cs by plants growing in the bilberry pine forest changes according to following order:

Phytomass—Adder-spit>Heath>Blue moor grass>Bilberry>*Dicranum*=*Pleurozium Schreberi*
Roots—Adder-spit>Blue moor grass>Heath>Bilberry (Table 11)

Table 10 Transfer factors of ^{137}Cs into components of alive soil cover of heath pine forest ($T_{ag}(10^{-3}m^2kg^{-1})$)

Specie	Organs /tissues	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Calluna vulgaris L.	Phytomass	3	14.38	9.17	3.60	22.00
	Roots	3	4.87	3.82	0.70	8.20
Pteridium aquilinum L.	Phytomass	2	43.60	22.34	27.80	59.40
	Roots	2	11.00	4.10	8.10	13.90
Dicranum polysetum Sw.	Phytomass	3	6.60	4.84	1.70	10.50
Pleurosium schreberi Brid.	Phytomass	3	5.80	3.38	1.90	7.90

N: quantity of the sample areas

Table 11 Transfer factors of ^{137}Cs into components of alive soil cover of bilberry pine forest ($T_{ag}(10^{-3}m^2kg^{-1})$)

Specie	Organs /tissues	N	Arithmetic mean	Standard deviation	Minimum	Maximum
Vaccinium myrtillus L.	Phytomass	3	10.03	10.20	1.00	21.10
	Roots	3	7.00	5.60	1.50	12.70
Molinia coerulea L.	Phytomass	3	14.37	12.77	5.50	29.00
	Roots	3	11.60	5.31	8.00	17.70
Pteridium aquilinum L.	Phytomass	2	57.60	40.16	29.20	86.00
	Roots	2	25.20	17.25	13.00	37.40
Dicranum polysetum Sw.	Phytomass	3	4.13	2.68	1.90	7.10
Pleurosium schreberi Brid.	Phytomass	3	4.13	1.86	2.40	6.10

N: quantity of the sample areas

The degree of accumulation of ^{137}Cs by grassy plants in meadows depends on kind of specie, soil-landscape conditions, degree of humidification; the same is true for forests. Radioactive contamination of grassy-semi frutex story of pine forests depends on depth of location of the root systems. In particular, ericoides kinds (heath, bilberry) having near surface location of root systems demonstrate higher ability to accumulate ^{137}Cs. It is known also (Tihomirov et al. 1990) that heath has more strong ability to ^{137}Cs than bilberry. The high values of transfer factors of ^{137}Cs are explained also by existence of mycorhiza—a symbiotic association composed of a fungus and roots of a vascular plant.

According to the results of our investigations regardless of forest type the maximal accumulation of ^{137}Cs in the case of vascular plants fits its phytomass. Among species studied the adder-spit (*Pteridium aquilinum*L.) has the highest transfer coefficients that corresponds previous data (Parfenov et al. 1995). Depending on forest type the bilberry (*Vaccinium myrtillus* L.) roots accumulate ^{137}Cs 1.4–2.0 times less than its phytomass; the same figures for heath (*Calluna vulgaris* L.) from 1.7 to 3 times, for adder-spit from 2.3 to 4. Blue moor grass (*Molinia coerulea* L.) is characterized by minimal difference between root and phytomass only 1.2 times.

The grassy and semi frutex vegetation in studied types of the pine forest could be presented in following order corresponding to their ability to ^{137}Cs accumulation: filices > heath ≥ cowberry ≥ meadow grass > mosses.

6 Conclusions

The square of lands covered by the forests was increased since 1988–2014 in the reserve. This tendency is maintained up to now. The square of pine forests decreased on 12.3 % due to influence of the fires and measures devoted to fire safety. The process of secondary waterlogging practically did not affect the pine forests. The additional increasing of the square of pine forests due to reforestation of former agricultural lands (+12.8 %) was not substantial. The preliminary natural restocking under the pinery canopy is not efficient and does not promote increasing of the percentage of pine forests in forest's structure of the reserve. In total, because of waterlogging, fires and arrangement of the fire breaks the increasing of square of pine forests in after Chernobyl period was equal only 0.5 %. The forest litter remains to be the horizon retaining ^{137}Cs. It contains 48.4 % of ^{137}Cs in mossy pinery, 53.9 % in bilberry pinery and 63.6 % in heath pinery. The content of ^{137}Cs in layer Ol of forest litter of pine forest could be regarded as stable and equals 1.5 % from sum of forest litter plus soil profile up to 40 cm. The maximal content of ^{137}Cs is typical for the bottom humified layer (Oh) of forest litter.

The overall peculiarity of ^{137}Cs distribution through the vertical soil profile in heath, bilberry and mossy pine forests is its accumulation in upper 0–5 cm under litter layer with following sharp decreasing in sublayers. On depth 40 cm the ^{137}Cs content in soil is about several tenths of percent. It was shown that intensity of vertical migration of ^{137}Cs in soil of upland pine forest is increased with soil humidity. Regardless of forest type the decreasing of accumulation of ^{137}Cs by stand compo-

nents has following order: roots > bark > wood. It was shown that transfer of ^{137}Cs into wood is higher in fresh sand than in humid sandy loam. The soil-ecological conditions influence on accumulation of ^{137}Cs by woody components in different types of forest.

Distribution of the components of the dominant species in the understory on the ability to accumulate ^{137}Cs shows the same rank regardless the forest type. One common peculiarity for all species of the understory is the minimal accumulation of ^{137}Cs by the wood. The results of our investigations show that for vascular plants namely their phytomass exhibits maximal ability to ^{137}Cs accumulation irrespective of forest type. The representatives of the grassy and semi frutex vegetation in studied types of the pine forest could be arranged in following rank according to their ability to ^{137}Cs accumulation: filices > heath ≥ cowberry ≥ meadow grass > mosses.

The data presented in papers (Stavrovskaya 1981; Martinovich 2001; Baginsky 2007; Perevolotski 2013; Utkin 2016) were used for the calculation of the quantity of the biomass of ecosystem's components of studied forest types. The knowledge of the biomass of components as well as their ^{137}Cs activity concentration allows building the ^{137}Cs distribution through the components of the ecosystems (Fig. 4). According to the (Report of the Chernobyl Forum 2006) which was published to the 20th year of Chernobyl accident in temperate forest ecosystems the stored amount of radiocaesium in the standing biomass of forests is approximately 5 % of the total activity, with the bulk of this activity residing in trees. The same values for studied types of the forest are following: mossy pinery 11.5 %, heath pinery 6.7 %, and bilberry pinery 5.5 %. The observable increasing of the value could be regarded as evidence of the continuing accumulation of the radiocaesium by forest's biomass.

It is known that forest, once radioactively contaminated, represent long term sources of radiation exposure to forest workers and to the general public. This is why the processes of radionuclide transfer in the forest are under the careful IAEA control TRS 472 (2010); TRS 479 (2014). It seems interesting to compare our experimental data on transfer of ^{137}Cs from soil to wood (wood is the most important material from the practical point of view) with already published data TRS 472 (2010); TRS 479 (2014). To do it the data of Table 12 containing the density of soil contamination as well as activity concentration of ^{137}Cs in wood were used. Concentration ratios (CR) were calculated as the ratio of the activity concentration of radionuclide in the wood to that in the soil. The calculations were fulfilled from the suggestion that all radioactive deposition is included into upper 20 cm soil layer.

From the analysis of data of Table 12 it could be concluded that our mean value of CR is sufficiently close to the value presented in international data base TRS 479 (2014). The published values of concentration ratios belong to the vast interval from 1.3×10^{-3} to 1.8. Interval of our values is closer from 0.11 to 0.43. To a large degree it could be stipulated by higher levels of radioactive contamination of the wood. That promotes decreasing of uncertainty of analytical data and diminution of their dispersion. One more advantage of our data is that they could be

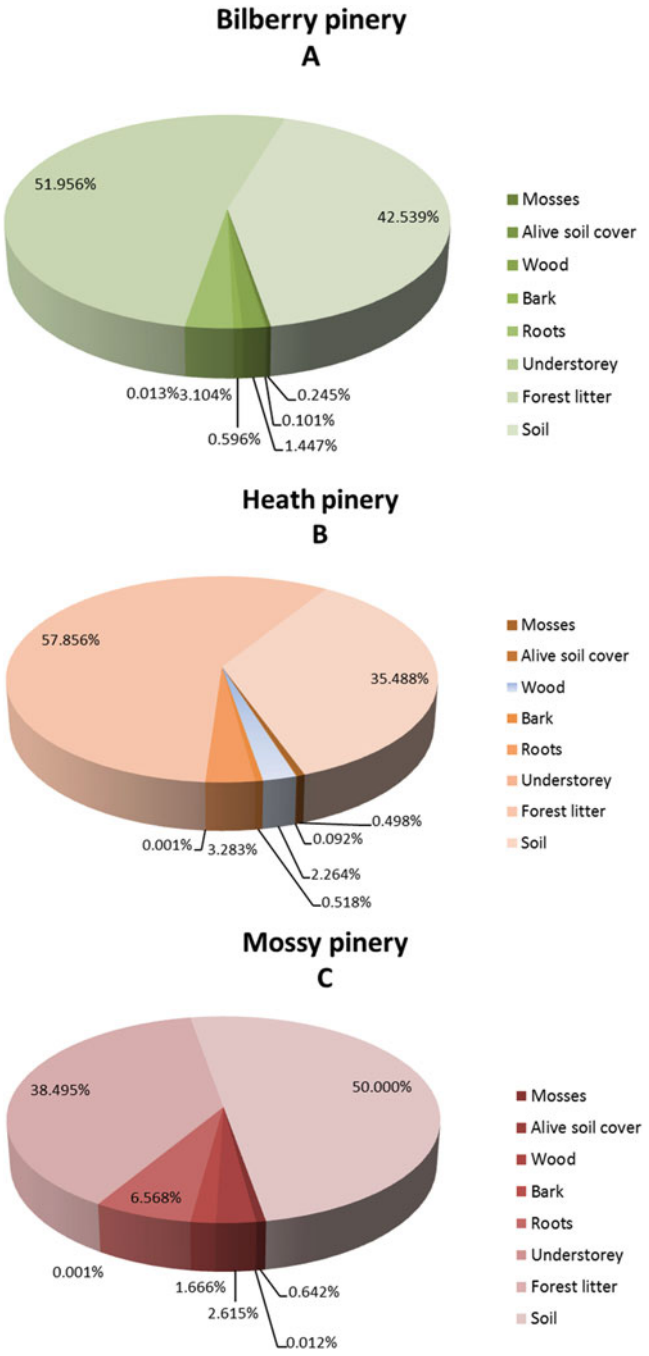

Fig. 4 Distribution of ^{137}Cs between components of ecosystem of bilberry (**a**), heath (**b**) and mossy (**c**) types of pine forest

Table 12 Comparison of our data with already published one

	Our data	Data of TRS 479 (2014) for coniferous trees
Mean value of CR	0.25	0.15
Minimal	0.11	0.0013
Maximal	0.43	1.8
N (sample quantity)	9	235
max/min	3.7	1385

regarded as equilibrium because about 30 years came from the moment of radioactive contamination.

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Accumulation of Cesium by Aquatic Plants and Algae

Koji Iwamoto and Yoshihiro Shiraiwa

Abstract A huge activity of radioactive matter including biophilic elements such as cesium, strontium and iodine were released into the environment from the Fukushima 1st Nuclear Power Plant (F1NPP) which was destroyed by the Great East Japan Earthquake in 2011. It is indispensable for rehabilitation to decontaminate and collect radionuclides spread on soils and water in the huge area. However, the amount and concentration of those radionuclides released were very small and low in comparison with those activities. Chemical processes sometimes require high energy and high cost for removing such low concentration of radionuclides from the environment. On the other hand, biological processes such as bioremediation and phytoremediation are thought to be useful because plants and algae have an ability to absorb and concentrate such radionuclides into bodies and cells. Cesium is reported to be easily absorbed by plants and algal cells via potassium transport systems as an analogue of potassium. On the basis of such knowledge, this study for screening was performed to find organisms possessing very high ability to accumulate radioactive cesium specifically and also radioactive strontium and iodine.

After testing 188 strains of various aquatic plants and algae, several strains were found to show a high ability to accumulate cesium in the cells by the removal of the radioactivity from medium. High cesium-accumulating strains were different from strains which have high ability to remove strontium and iodine, suggesting strains are specific to each element. The best strain for cesium accumulation coded as nak 9 was found to be a eustigmatophycean microalga *Vacuoliviride crystalliferum* which was a novel genus and species. This chapter focuses on the cesium uptake mechanism by plant and algal cells, the screening process how to find most effective strains which are useful for phytoremediation and the analysis of cesium absorption profiles. Morphological properties of the alga are also described.

Keywords Eustigmatophycean alga *Vacuoliviride crystalliferum* • Fukushima 1st Nuclear Power Plant Accident • High cesium-accumulating microalgae • Phytoremediation • Radioactive cesium

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1 Introduction

A huge amount of radionuclides were released due to the F1NPP accident caused by the Great East Japan Earthquake and tsunami of March 11, 2011. The total radioactivity released was estimated 11.6 EBq (exa Bq: 10^{18} Bq) (METI 2011) (Table 1). Most abundant discharged radionuclides released were xenon (^{133}Xe) with ca. 500 PBq (peta Bq: 10^{15} Bq) occupying about a half of total activity, cesium (^{134}Cs and ^{137}Cs), strontium (^{89}Sr, ^{90}Sr), iodine (^{131}I, ^{132}I, ^{133}I and ^{135}I), barium (^{140}Ba), yttrium (^{91}Y) and other elements as minor components.

Table 1 Estimated values of radioactive materials released from the F1NPP Unit 1 to 3

Radionuclide	Half-life period	Unit 1	Unit 2	Unit 3	Total
Xe-133	5.2 d	3.4×10^{18}	3.5×10^{18}	4.4×10^{18}	1.1×10^{19}
Cs-134	2.1 y	7.1×10^{14}	1.6×10^{16}	8.2×10^{14}	1.8×10^{16}
Cs-137	30.0 y	5.9×10^{14}	1.4×10^{16}	7.1×10^{14}	1.5×10^{16}
Sr-89	50.5 d	8.2×10^{13}	6.8×10^{14}	1.2×10^{15}	2.0×10^{15}
Sr-90	29.1 y	6.1×10^{12}	4.8×10^{13}	8.5×10^{13}	1.4×10^{14}
Ba-140	12.7 d	1.3×10^{14}	1.1×10^{15}	1.9×10^{15}	3.2×10^{15}
Te-127m	109.0 d	2.5×10^{14}	7.7×10^{14}	6.9×10^{13}	1.1×10^{15}
Te-129m	33.6 d	7.2×10^{14}	2.4×10^{15}	2.1×10^{14}	3.3×10^{15}
Te-131m	30.0 h	9.5×10^{13}	5.4×10^{10}	1.8×10^{12}	9.7×10^{13}
Te-132	78.2 h	7.4×10^{14}	4.2×10^{11}	1.4×10^{13}	7.6×10^{14}
Ru-103	39.3 d	2.5×10^{9}	1.8×10^{9}	3.2×10^{9}	7.5×10^{9}
Ru-106	368.2 d	7.4×10^{8}	5.1×10^{8}	8.9×10^{8}	2.1×10^{9}
Zr-95	64.0 d	4.6×10^{11}	1.6×10^{13}	2.2×10^{11}	1.7×10^{13}
Ce-141	32.5 d	4.6×10^{11}	1.7×10^{13}	2.2×10^{11}	1.8×10^{13}
Ce-144	284.3 d	3.1×10^{11}	1.1×10^{13}	1.4×10^{11}	1.1×10^{13}
Np-239	2.4 d	3.7×10^{12}	7.1×10^{13}	1.4×10^{12}	7.6×10^{13}
Pu-238	87.7 y	5.8×10^{8}	1.8×10^{10}	2.5×10^{8}	1.9×10^{10}
Pu-239	24,065 y	8.6×10^{7}	3.1×10^{9}	4.0×10^{7}	3.2×10^{9}
Pu-240	6537 y	8.8×10^{7}	3.0×10^{9}	4.0×10^{7}	3.2×10^{9}
Pu-241	14.4 y	3.5×10^{10}	1.2×10^{12}	1.6×10^{10}	1.2×10^{12}
Y-91	58.5 d	3.1×10^{11}	2.7×10^{12}	4.4×10^{11}	3.4×10^{12}
Pr-143	13.6 d	3.6×10^{11}	3.2×10^{12}	5.2×10^{11}	4.1×10^{12}
Nd-147	11.0 d	1.5×10^{11}	1.3×10^{12}	2.2×10^{11}	1.6×10^{12}
Cm-242	162.8 d	1.1×10^{10}	7.7×10^{10}	1.4×10^{10}	1.0×10^{11}
I-131	8.0 d	1.2×10^{16}	1.4×10^{17}	7.0×10^{15}	1.6×10^{17}
I-132	2.3 h	4.5×10^{14}	9.6×10^{11}	1.8×10^{13}	4.7×10^{14}
I-133	20.8 h	6.5×10^{14}	1.4×10^{12}	2.6×10^{13}	6.8×10^{14}
I-135	6.6 h	6.1×10^{14}	1.3×10^{12}	2.4×10^{13}	6.3×10^{14}
Sb-127	3.9 d	1.7×10^{15}	4.2×10^{15}	4.5×10^{14}	6.4×10^{15}
Sb-129	4.3 h	1.6×10^{14}	8.9×10^{10}	3.0×10^{12}	1.6×10^{14}
Mo-99	66.0 h	8.1×10^{7}	1.0×10^{4}	6.7×10^{6}	8.8×10^{7}

d days, *y* years, *h* hours

Among those radionuclides, the most harmful elements were biophilic elements such as cesium, strontium and iodine of which activity were 10–37 PBq, 150 TBq and 90–500 PBq dispersed into air and 1.9 PBq, 90–500 TBq and 2.7 PBq dispersed into the ocean, respectively (Chino et al. 2011; METI 2011; Nuclear Emergency Response Headquarters 2011; Stohl et al. 2012; TEPCO 2012). Such biophilic elements are easily incorporated into human body during taking of air, water, and/or foods, and would increase health risk by internal exposure of radiation (Betsy et al. 2012).

The half-life period of ^{98}Sr, ^{131}I, ^{132}I, ^{133}I and ^{135}I are short enough to decrease radioactivity below detective level rapidly for 50 d, 8 d, 2.3 h, 20.8 h and 6.6 h, respectively. Those radioactive compounds were noticed as very high-risk elements just after the F1NPP accident. However, the attention of inhabitants to risks of strontium and iodine was gradually decreased in comparison with that of cesium. Among radioactive strontium species, ^{90}Sr has rather a long half-life period (50 days), but the attention was not so high because the total amount of released ^{90}Sr was small and the polluted area by ^{90}Sr was limited around nearby the F1NPP for its non- volatility.

On the other hand, both ^{134}Cs and ^{137}Cs with rather a long half-life period of 2.1 and 30.0 years, respectively, are considered to be a threat to health and obstacle of restoration in radio-polluted areas. According to the International Nuclear Event Scale (INES) assessment which is made based on an iodine-converted value of radioactivity, the total radioactivity of Cs is still higher than 400 PBq equivalent to ^{131}I, which is calculated with a conversion factor of ^{137}Cs which is 40 (TEPCO 2012).

The high conversion factor shows a risk to health brought by radioactive cesium as well as that of iodine and strontium. The contamination of radionuclides into the environment and living organisms, such as water and foods, respectively, has carefully been monitored and alerted to the public after the F1NPP accident. Table 2 summarizes the results of inspections on radioactivity levels in livestock products in 4 years after the F1NPP accident based on a provisional regulation value of radio-polluted materials in foods in accordance with the food Sanitation Act determined by the Japanese Ministry of Health, Labour and Welfare (MAFF 2015; MHLW 2011).

As such, radionuclide contamination gives serious influence to the environment, living organisms, and ecosystems for a long period. Development of bioremediation technology will be one of the very important issues for renovating radio-polluted environments. In this Chapter, such possible methods using algae and aquatic plants are introduced according to our studies.

2 Assimilation and Accumulation of Radioactive Cesium by Living Cells

Cesium, strontium, and iodine are easily absorbed by living organisms as biophilic elements. When those radioactive elements are incorporated into animal bodies, internal exposure raises health risk highly. Therefore, the amount of radionuclides, such as ^{137}Cs, ^{90}Sr, and ^{131}I, are continuously monitored in many

Table 2 Results of the inspection on radioactivity level in livestock

Item	March 2011–March 2013[a]			April 2013–March 2014[a]			April 2014–March 2015[a]		
	Total	Above the maximum limit of provisional regulation values	% of above the limit	Total	Above the maximum limit of provisional regulation values	% of Above the limit	Total	Above the maximum limit of provisional regulation values	% of Above the limit
Rice	26,464	592	2.20	11,040,000	28	0.00	10,990,000	0	0
Vegetables	12,671	385	3.00	19,657	0	0	16,712	0	0
Fruits	2732	210	7.70	4243	0	0	3302	0	0
Pulse	689	16	2.30	5167	21	0.40	2586	2	0.10
Tea leaves	2233	192	8.60	447	0	0	206	0	0
Raw milk	1919	8	0.40	2040	0	0	1846	0	0
Beef	78,095	1052	1.30	193,268	0	0	186,937	0	0
Pork, chicken, egg	867	6	0.70	1486	0	0	1180	0	0
Mushrooms and mountain vegetables	3856	779	20.20	7581	194	2.60	8557	103	1.20
Aquatic products	8576	1476	17.20	20,695	302	1.50	20,922	100	0.50

[a]Coverage were 17 prefectures and metropolitan which the Japanese government prescribes to conduct the test, i.e. Aomori, Iwate, Miyagi, Akita, Yamagata, Fukushima, Ibaraki, Tochigi, Gunma, Saitama, Chiba, Tokyo, Kanagawa, Niigata, Yamanashi, Nagano, and Shizuoka. For aquatic products, all prefectures were covered

countries (Betsy et al. 2012). Among radionuclides released from the F1NPP, radioisotopes of cesium, ^{134}Cs and ^{137}Cs, have received the highest attention because of four reasons: namely, (1) their large amounts of discharge (ca.30 PBq), (2) their long half-life period, suggesting long preservation in environment, (3) fairly large contaminated area by radioactive cesium because of their volatility, and (4) the biophilic characters.

Cesium is an analogue of one of the essential elements, potassium, and easily incorporated into cells. However, its bioaccumulation level is at most 100 times higher than that in the environment even in the large fish at the top level of the food chain, and the biological half-life period in an adult human is about 100 days (Suzuki et al. 1973). The assimilation/incorporation of cesium is considered to be mediated by the potassium transport systems such as potassium transporter, sodium/potassium pump, and potassium channels. In animals, potassium is absorbed in the small intestine, and accumulated and utilized in muscles, bone, organs, and brain. In plants, cesium is absorbed via roots and leaves, and affects cell growth and elongation, regulation of stomatal behavior, and signal transduction to the organelle. Plant growth is inhibited by high concentration of cesium, over 200 μM, by competing with potassium and disturbing jasmonate signaling (Adams et al. 2013, 2015). Cesium is also reported to be transported via the transport systems of cation and calcium (White and Broadley 2000). Figure 1 show various transport mechanisms

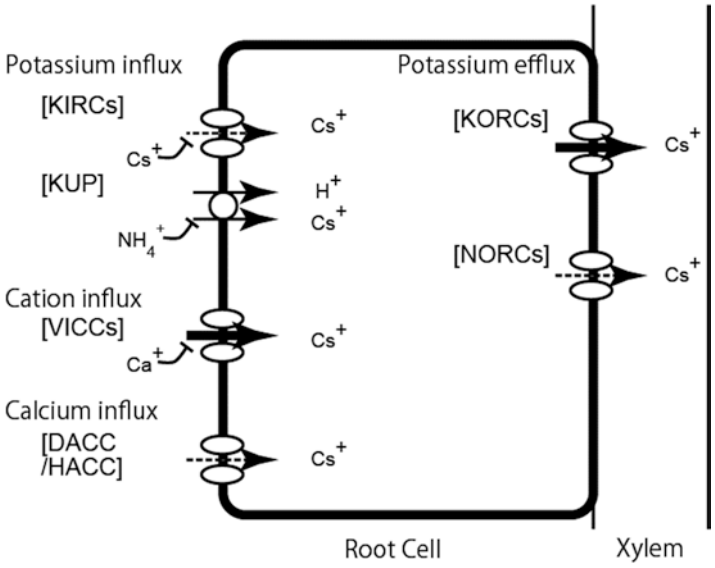

Fig. 1 Cesium influx and efflux system in a plant root cell (Adopted and modified Fig. 8 from White and Broadley 2000). *DACCs*: depolarization-activated Ca^{2+} channel, *HACC*: hyperpolarization-activated Ca^{2+} channel, *KIRC*: inward-rectifying K^+ channels, *KORC*: outward-rectifying K^+ channel, *KUP*: K^+ uptake, *NORC*: nonselective outward-rectifying K^+ channel, *VICC*: voltage independent cation channel

which contribute to cesium influx and efflux across the plasma membrane of plant root cells (Hampton et al. 2005; White and Broadley 2000). The arrows and their width indicate the direction of transport and their relative activity, respectively. For the influx of cesium, two systems are already known, namely the inward-rectifying K^+ channels (KIRCs) and the K^+/H^+ symporters (KUP). High-affinity potassium transporters (HKT), another potassium-transporting system, may also be involved in cesium transport, though the details were not reported so far (Remy et al. 2015).

In these cesium importing systems, the involvement of KUP in cesium uptake was proved by Qi et al. (2008). KUP belongs to the KT/KUP/HAK family that plays various important aspects of plant life including mineral nutrition and developmental regulation. In practice, the phenomenon that expression of AtHAK5 identified from a higher plant *Arabidopsis thaliana* is increased by either K^+ or NH_4^+-deficiency shows the significance of KUP in cesium incorporation at a low level of potassium. Direct evidence of the involvement of KUP in cesium transport was showed by mutants, i.e. AtHAK5 deficient *Arabidopsis* seedlings were depressed cesium accumulation with inhibiting root growth, and AtHAK5 introduced yeast increased cesium uptake (Qi et al. 2008). For the KIRCs system, AtAKT1 and AtKAT3 were known, but their contribution of cesium transport was neglected because their potassium transfer was severely inhibited by cesium with blocking the channel by cesium ion itself.

Besides the potassium transport system, Voltage Independent Cation Channel (VICCs), a cation efflux system, highly contributes to cesium import. AtCNGC (*A. thaliana* cyclic-nucleotide gated channel) and AtGLR (*A. thaliana* glutamate receptor) gene families are involved in VICCs in *A. thaliana* (Hampton et al. 2004). Other cation efflux systems are also involved in the cesium import systems in the cell, which are AtCNGC (*A. thaliana* cyclic-nucleotide gated channel) and AtGLR (*A. thaliana* glutamate receptor) gene family in *A. thaliana.* Other variations reported are HACCs (Hyperpolarization- Activated Ca^{2+} channel for calcium efflux), DACCs (Depolarization-Activated Ca^{2+} channel systems for calcium efflux), KCO (the cation-transporting channel to vacuole), KEAs (K^+ efflux antiporter as potassium-proton antiporters), and AtNHX1 (*A. thaliana* Na^+/H^+ antiporter classified in cation-proton antiporters) (Hampton et al. 2005). For cesium efflux from cells to bundle, KORCs (Outward-Rectifying K Channel) are known to be largely involved in addition to NORCs (Non-selective Outward-Rectifying K Channel) (White and Broadley 2000).

3 Significance of Bioremediation and Phytoremediation for Decontamination of Radionuclides from Polluted Environment

There are variations of methods, such as physical, chemical and biological methods, in environmental remediation engineering. The physical and chemical methods include replacing polluted soil, washing soil with a high amount of water and/or chemicals, neutralizing pollutants by chemicals, immobilizing the soils to avoid

diffusion of pollutants, and so on. Polluted water can be decontaminated using adsorption agent and/or dilution. The biological methods are called as bioremediation which involves methods to degrade and decontaminate pollutants by using the activity of living organisms such as microbes and plants. Bioremediation is further classified into bioaugmentation, biostimulation, and phytoremediation. Among these methods, bioremediation is considered as significant environmental cleanup methods in next-generation because of its high potential to treat various chemical pollutants with low energy costs even at low concentration of pollutants. In the case of the F1NPP accident in 2011, the total amount of radioactive cesium released into the environment were calculated from the total radioactivity released from the nuclear reactor using following formula.

$$W = \frac{B \cdot T_{1/2} \cdot M}{N_A \cdot \log_e 2}$$

where W, total weight (g); B, radioactivity (Bq: counts s^{-1}); $T_{1/2}$, half-life period (s); M, mass number of element ($gmol^{-1}$); N_A, the Avogadro number (6.022×10^{23}); $\log_e 2$, 0.6931.

Total weight of ^{137}Cs released from the F1NPP was calculated as 4.65 kg according to above formula using 15 PBq (B) and 30.17 years ($T_{1/2}$). Such small amount of radioactive cesium with high specific activity was spread over mountains, forests, lakes, rivers, farmland, and cities over 14,000 km^2 in and around Fukushima area, where showed the air radiation levels over 1 Ci/km^2 (3.7×10^{10} Bq; 37,000 Bq/m^2) on November 5, 2011 (MEXT 2012). Under such conditions, phytoremediation, bioremediation using plants and algae, is thought to be one of the effective methods for removing radio-pollutants. The decontamination of radioactive cesium using land plants were examined for low or middle level of polluted soils, though the high level radio-contaminated water stored in and just around the F1NPP was treated by chemical and physical methods using a cesium removal system with zeolite, a reverse osmosis (RO) system, and Advanced Liquid Processing System (ALPS™) system for multi-nuclide removal (Tusa 2014; TEPCO 2016).

Broadley et al. (1999) compared the level of cesium accumulation in plant shoots among 136 taxa in 14 literatures by a residual maximum likelihood (REML) analysis, and then calculated mean relative cesium concentrations among Magnoliophyta taxa. The authors reported that *Amaranthus retroflexus* showed the highest accumulation of radioactive cesium, and Dicotyledons (Magnoliopsida) showed higher activity for accumulation than monocotyledons (Liliopsida). Yamagami et al. (2009) were selected *A. retroflexus* and *A. caudatus* as useful candidates for cesium accumulators from 44 species of wild plants by examining cesium removal activity per unit land area. At Fukushima area, the removal ability of radioactive cesium from contaminated soils was assayed in sunflower (Asteraceae), which has reported as 'hyper-accumulator of radioactive cesium' (Dushenkov et al. 1997), and other 12 plant species in Fabaceae and Poaceae (Kobayashi et al. 2014). However, any plant

species was not found to be effective for decreasing in radioactive cesium from radio-polluted soils remarkably. Further study is required to develop useful plant phytoremediation systems for the decontamination of cesium. For example, screening and the molecular breeding of 'hyper-accumulator of radioactive cesium' are important. In addition, optimization of methods is also important to promote the activity of radioactive cesium accumulation because radioactive cesium content in the soil was reduced by plants cultivated in the fields although the ability was low (Kobayashi et al. 2014).

4 Assimilation and Accumulation of Radioactive Cesium by Aquatic Plants and Microalgae

4.1 Advantage of Microalgae for Decontamination of Water

The advantage of bioremediation is based on the activity which is able to concentrate polluted materials/elements into the cells and bodies, and the possibility to apply for widely spread pollutants even at low levels/amounts/concentrations. Because bioremediation is a method to utilize the ability of living organisms to assimilate and degrade pollutants, it does not require particular industrial levels of facilities/equipment generally, resulting in low cost and low energy. Especially, phytoremediation using microalgae, which can also be called algal bioremediation, is largely advantageous as follows:

1. Microalgae possess high ability to remove pollutants from polluted water because of its high bioaccumulation/bioconcentration activity.
2. Total energy cost for bioremediation is low because microalgae do not require external energy input to grow except sunlight.
3. Volume reduction of polluted material/water is quite easy, which is the most important point for algal bioremediation.

4.2 Screening of High Cesium Accumulating Algae

Fukuda et al. (2014) surveyed the ability to remove the radioactivity from the medium or to accumulate the activity in the cells in 188 strains of algae and aquatic plants including 91 seawater, 86 freshwater, and 11 terrestrial strains. Those strains used in the study covered broad phylogenetic variation involving 45 classes, 21 divisions and 8 super groups such as Cyanophyta, Opistokonta, Excavata, Archaeplastida, Rhizaria, Alveolata, and Stramenopiles (Fig. 2). These organisms show various features in their morphology, physiology, biochemical properties and nutritional properties such as autotrophy and/or heterotrophy. These strains were mainly obtained from the culture collections of the Laboratory of Plant Diversity

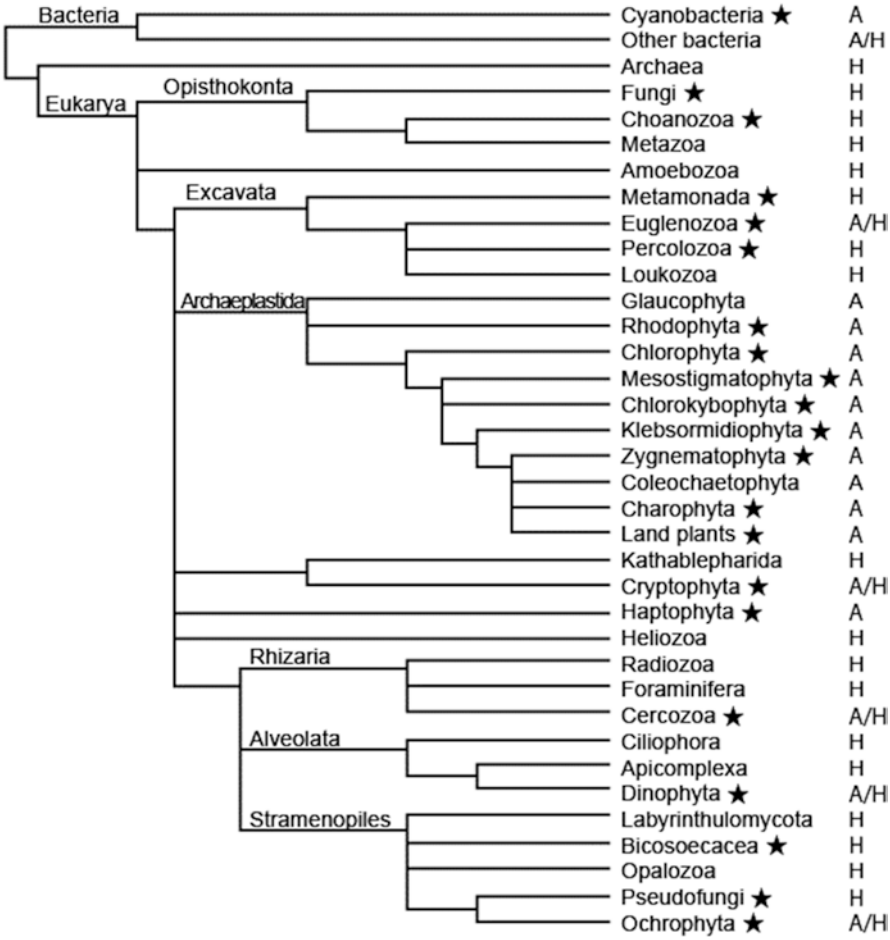

Fig. 2 Phylogenic positions of experimental organisms in the schematic phylogenetic tree of life (phylum-level) (Fukuda et al. 2014). Star, phylum including strains examined in this study; *A, H* and *A/H*, a nutrient condition indicating mostly autotrophic, mostly heterotrophic and a mixture of *A* and *H*, respectively

and Evolutionary Cell biology, University of Tsukuba, Japan, and that of the National Institute for Environmental Studies (NIES) in Tsukuba, Japan.

The strains were maintained in plastic vessels, so-called cell culture flasks, containing respective media for each organism, i.e. marine algal strains were grown in seawater enriched either with ESM, IMK or f/2 medium, and freshwater algae and aquatic plants were grown either in C, CS, or AF-6 medium (Kasai et al. 2004). Heterotrophic algae were organic nutrients supplied AF-6 or seawater medium by addition of glucose, polypeptone and yeast extract (Fukuda et al. 2014). Those flasks were placed under continuous light at an adequate temperature for promoting the growth of microalgae and small aquatic plants. For radionuclide elimination

test, each fresh water medium was prepared without potassium because cesium uptake is known to compete with potassium, an analog of cesium (see Sect. 2 of this chapter). Radioactive cesium used in this study was artificially prepared ^{137}Cs in chloride form (Specific activity: 61.7 GBq mmol^{-1} (3.7 MBq mL^{-1}); Eckert & Ziegler Isotope Products, Valencia, CA, USA). The test for screening was initiated by the addition of radioactive cesium at low concentration (1000 Bq mL^{-1}, 15 nM). After incubation for several days, those living organisms were separated from the culture medium by either centrifugation or filtration, and then both algal cells/plant bodies and the culture medium were brought for the determination of radioactivity using a gamma-ray counter (Aloka Accuflex γ7000, Tokyo, Japan). For microalgae, algal cells were separated by the silicone-oil layer centrifugation method as described in Araie et al. (2011). For aquatic plants, culture medium separated from plant bodies was also subjected to the silicone-oil layer centrifugation method to remove small particles from the media. In selected strains, precise accumulation was assayed in a short time from 15 min to 4 h after addition of ^{137}Cs.

Among 188 strains of microalgae and aquatic plants examined, 137 strains showed radioactive cesium removal activity from the medium in the first screening, although the ability was different among strains (Fig. 3a). After the first screening, 15 strains which showed more than 40 % of removal were selected and subjected to second screening (Fig. 3b). As a result, five strains were selected as highly positive radioactive-cesium eliminators. They were the freshwater eustigmatophycean algae *Vacuoliviride crystalliferum* (nak 9, more than 90 % elimination in two days), the freshwater floridephycean algae *Batrachospermum virgato-decaisneanum* (NIES-1458, ca. 38 % elimination), the chlorophycean algae *Chloroidium saccharophilum* (NIES 2352, ca. 22 % elimination) and two strain of aquatic plants (trachiophytes) *Lemna aoukikusa* (TIR 2 and TIR 3, ca. 45 and 66 % elimination, respectively). The floating grass *L. aoukikusa* would be a good decontaminator because the growth speed is high, and the fronds are easy to be harvested using net.

5 High Cesium Accumulating Alga *V. crystalliferum*

The eustigmatophycean alga *V. crystalliferum* was found to be a new species and new genus (Nakayama et al. 2015). Interestingly, the alga showed distinguished feature that possess large reddish globule and crystalline structure (Fig. 4), and was considered to have an ability to survive under a severe environmental condition because it was originally isolated from unusual greenish sediment in the bottle of glue in 2002 in University of Tsukuba. Actually, *V. crystalliferum* is able to grow at a wide range of temperature, light intensity, pH, and salinity (data not shown). These physiological characteristics of *V. crystalliferum* would contribute to low-cost phytoremediation.

Figure 5 shows the time course of ^{137}Cs elimination from the medium. The radioactive cesium was rapidly eliminated just after the addition of ^{137}Cs. More than 60 % were removed after 15 min, and about 90 % were absorbed in the cell within 1 h. The elimination velocity was calculated as 63 μg ^{137}Cs (mg Chl h^{-1}). This value was

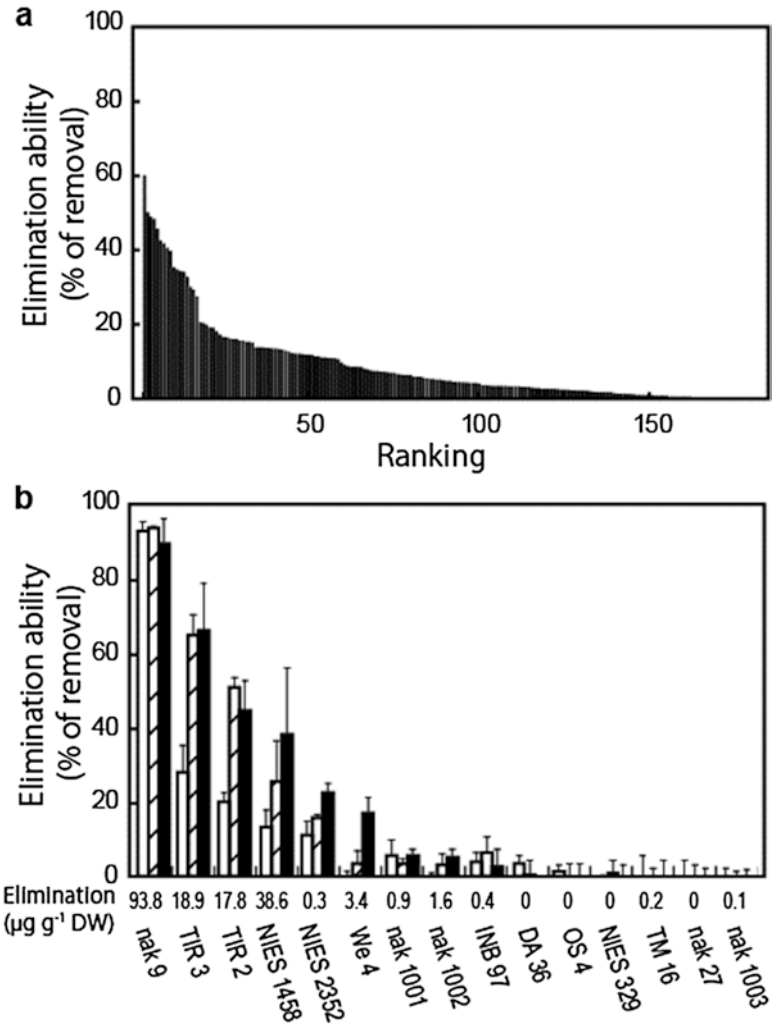

Fig. 3 Screening of highly active species/strains of aquatic plants and microalgae for eliminating radioactive cesium from water. (**a**) First screening. (**b**) Second screening. *White, stripe* and *black bars* indicate the elimination ability at the 2, 4, and 8 days after start testing, respectively. nak 9; *Vacuoliviride crystalliferum*, TIR 3; *Lemna aoukikusa*, NIES-1458; *Batrachospermum virgatodecaisneanum*, NIES-2352; *Chloroidium saccharophilum,* We 4; *Elodea nuttallii*, nak 1001; *Oedogonium* sp., nak 1002; *Rhizochlonium* sp., INB 97; *Coelastrum* sp., DA 36; *Heterosigma akashiwo*, OS 4; *Dixoniella grisea,* NIES 329; *Ulothrix variabilis*, TM 16; *Amphidinium massartii*, nak 27; *Calyptrosphaera sphaeroidea*, nak 1003; *Spirogyra* sp.

higher than that reported in higher plants such as sunflower and vetiver *Vetiveria zizanoides* which were absorbed 150 μg Cs in 100 h and 61 % of ^{137}Cs in 168 h from their medium, respectively (Dushenkov et al. 1997; Singh et al. 2009). The uptake of radioactive cesium was highly suppressed by potassium, suggesting the involve-

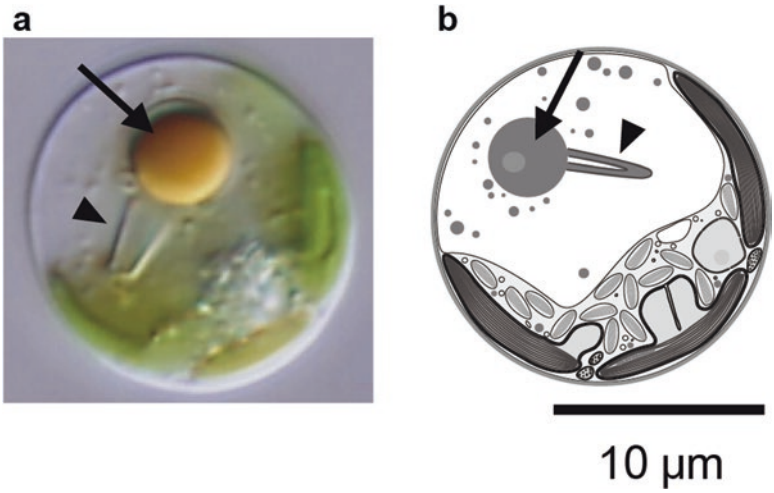

Fig. 4 (**a**) Light micrographs of and (**b**) Drawings of *V. crystalliferum. Arrow;* reddish globule, *Arrow head;* crystalline structure. Figures were provided by Dr. T. Nakayama, University of Tsukuba (also see Nakayama et al. 2015)

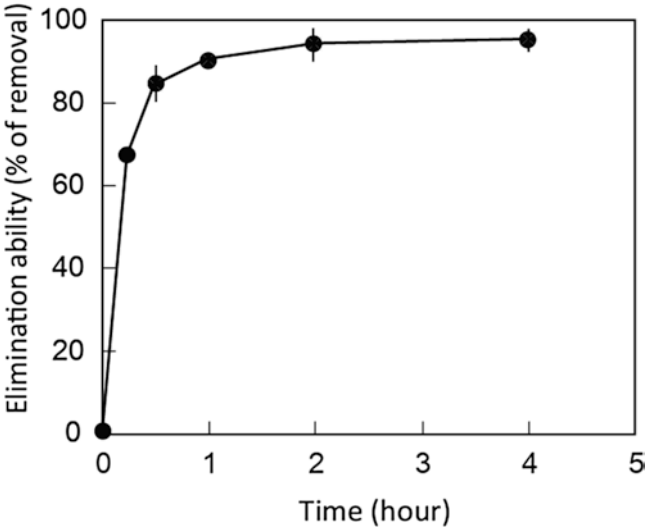

Fig. 5 Elimination of Cs-137 from medium by *V. crystalliferum*

ment of potassium transporters for cesium uptake (Fukuda et al. 2014). Further study is needed to clarify the mechanism and significance of the cesium assimilation in this alga.

The adsorption/absorption coefficient (K_d) in cesium-absorbing microalgal strain (nak 9) was calculated as 4.1×10^5 which is much higher than that of both natural and synthetic zeolites, ferrocyanide and crystalline silicotitrate (CST) (Fig. 6). Although *V. crystalliferum* (strain nak 9) did not show significant absorption of

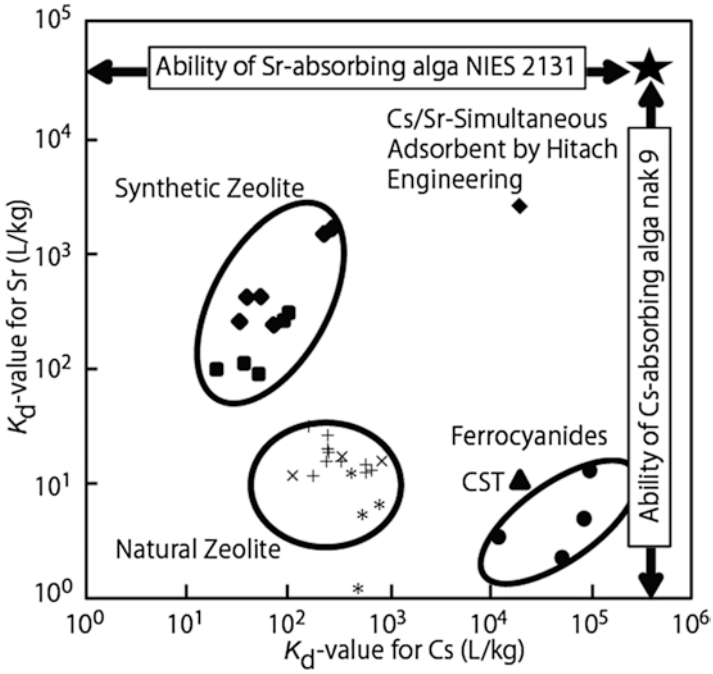

Fig. 6 The highest ability for Cs- and Sr-absorption/adsorption was obtained in the microlagal strain nak9 and NIES2131. CTS: crystalline silicotitrate. Fig. 2 in Kani et al. (2014) was adopted and modified by adding new data obtained in radioactive Cs- and Sr-absorbing algae, nak9 and NIES2131, respectively

strontium, another algal strain *Stigonema ocellatum* NIES 2131 (cyanophyta) showed high activity (Fukuda et al. 2014). Therefore, it could be possible to obtain high removal ability of radioactive cesium and strontium from polluted water by using those two algal strains (Fig. 6). The combination of algae would be higher than the ability of newly developed Cs/Sr-simultaneous adsorbent produced by Hitachi Engineering (Kani et al. 2014). For developing applied technology of algal bioremediation, mass cultivation technology of those microalgal strains and application methods how to use those algal cells for removing a huge amount of radioactive cesium and strontium are needed in further researches.

6 Conclusion and Future Study

High cesium accumulating alga *V. crystalliferum* was found as a new species of eustigmatophytes. This alga was recognized as one of the candidates which can be utilized for phytoremediation of radioactive cesium contamination. The alga has quite unique cell structures such as chop stick-like rods (crystalline structure) and red colored particle (reddish globule) in the vacuole. It is still enigma whether such

unique structure is associated with high radioactive cesium eliminating activity. The alga is also special in its high growth rate even under various environmental conditions which guarantee to produce large biomass throughout a year. Because the elimination ability of the alga is extremely high, the alga would possess a specific cesium uptake system which may contribute to the molecular breeding to produce 'hyper-accumulator of radioactive cesium'. Regarding the F1NPP accident, a huge amount polluted soils still remains in stock bags and, therefore, the development of techniques is strongly required for decontamination of radio-pollutants and reduction of the volume of radio-contaminants. However, radioactive cesium has been reported to bind tightly to fine soil particles (Qin et al. 2012). In particular, radioactive cesium in and around Fukushima area was found to be firmly bind to vermiculitized biotite or called weathered biotite (Mukai et al. 2014). Because these clay minerals are quite common in soils in Japan and sorb cesium far more than the other clay minerals such as fresh biotite, illite, and smectite, it may become a key factor affecting the dynamics and fate of radioactive cesium in Fukushima and Japan (Mukai et al. 2016). Therefore, in order to use microalgae for eliminating radioactive cesium from such soils, the development of another technology is essential to solubilize radioactive cesium from fine soil particles in aqueous solution. Further studies are required under the tight collaboration of scientists from different scientific disciplines.

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Regularities of Accumulation of Cs-137 and Other Radionuclides in Aquatic Vegetation in the Territory of the South-Ural Biogeochemical Province of Techno-genic Radioactive Isotopes

I. Ya. Popova and N.N. Kazachonok

Abstract The aim of this work is to study the regularities of ^{137}Cs and other radionuclides accumulation in the aquatic vegetation in the territory of the South-Ural biogeochemical province of anthropogenic radioactive isotopes. We compared the accumulation of ^{137}Cs and other radionuclides in the aquatic vegetation of lakes, a river and a technical water reservoir—a radioactive waste repository. ^{137}Cs and ^{90}Sr accumulation coefficients in the aquatic vegetation are very variable and can reach several thousands. The accumulation coefficients of ^{137}Cs in the aquatic vegetation are in most cases notably higher than of ^{90}Sr. The hydatophytes and hydrophytes don't differ conceptually in the ability to concentrate radionuclides but in general rooted plants accumulate significantly less ^{90}Sr and ^{137}Cs than unrooted plants. The entry of radionuclides through the roots and the pollution level of bottom silt apparently don't have a great impact on C_a of ^{90}Sr and ^{137}Cs in the aquatic vegetation. However, it can be assumed that as a result of coastline transgression, the degree of hydrophytes submergence and accordingly, the value of radionuclides entry into a plant through its roots can vary. The type of water reservoir apparently doesn't have a great impact on radionuclides accumulation in plants. The most important factors of radionuclides accumulation are apparently weather conditions, sampling point and time that passed after the water reservoir pollution.

Keywords Contamination • South-Ural biogeochemical province • Aquatic vegetation • Accumulation coefficients • Radionuclides • ^{137}Cs • ^{90}Sr

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1 Introduction

Nuclear weapons complex enterprise Production Association "Mayak" was established in the northern part of the Chelyabinsk region immediately after the Second World War for the purposes of nuclear weapons creation for the Soviet Union. Under severe resource and time constraints simplified schemes of radioactive waste (RW) management were accepted. Considering the scale and nature of contamination in the territory surrounding the radiochemical enterprise PA "Mayak" including the East-Ural radioactive trace and the Techa River system, it is arguable that in this territory a powerful anthropogenic biogeochemical anomaly of radioactive isotopes formed, so this area can be considered as one of anthropogenic biogeochemical provinces. This province is probably the oldest and second largest after Chernobyl province. Its feature is an integrated regime of environmental contamination: process emissions from reactors and radiochemical production, radioactive waste tank explosion (formation of the East Ural radioactive trace-EURT), carrying away of Karachay Lake mud bank by the wind, discharges of liquid waste into the Techa River system. It is represented by forest-steppe and forest landscape zones, bogs and floodplains, lakes, settlements and agricultural lands. Radioisotope composition of particular zones varies considerably. Consequently, the South Ural province of radioactive isotopes can be used to simulate various radiation situations.

The aim of this work is to study the regularities of ^{137}Cs and other radionuclides accumulation in aquatic vegetation in the territory of the South-Ural biogeochemical province of anthropogenic radioactive isotopes.

2 Research Methods

In order to study the regularities of radionuclides accumulation in aquatic vegetation we used the materials from the database of Urals Research Centre for Radiation Medicine as well as the materials of modern studies according to the academic research work plan of the Federal Medical and Biological Agency of Russia.

Water sampling was carried out by conventional methods; the bottom silt was collected using bottom sampler. Samples were then dried, crushed and sieved through a 1 mm sieve. Aquatic vegetation was air-dried, the aliquot was selected and burned in a muffle furnace at the t=450–500 °C. Gamma-emitting radionuclides were identified using semiconductor gamma-ray spectrometer Multipurpose Spectrometric Complex "Gamma-plus". ^{90}Sr was identified in the samples based on daughter ^{90}Y by extraction method with flame photometric carrier loss control (stable strontium). Radiochemical separation of ^{137}Cs was carried out from chloride solutions in the form of iodine–antimony salt after concentrating it on nickel ferrocyanide. The ß-activity of the separated radionuclides was measured using low-background radiometric facilities UMF-1500 and UMF-2000. Plutonium isotopes were identified using α-spectrometric method after concentrating and cleaning on the anion exchange resin and the electrolytic separation

on steel targets. ^{242}Pu was used as marking. The studied isotopes were measured and identified in α-spectrometer facility with pulse ionization chamber. The ^{241}Pu content was measured based on the low-energy ß-activity of ^{241}Pu (maximum energy—20.8 keV) and the sum of plutonium isotopes using the liquid scintillation radiometer Quantulus-1220. Tritium in water samples was measured in the ά, ß-radiometric facility Quantulus-1220 by liquid scintillation method after distillation cleaning. Uranium isotopes in water were identified by α-spectrometric method after tributyl phosphate extraction and electrolytic deposition on steel targets. ^{232}U was used as marking. In order to identify physicochemical condition of ^{137}Cs and ^{90}Sr in the bottom silt the conventional method of successive extraction of radionuclide H_2O (water-soluble fraction), 1 N CH_3COONH_4 (exchangeable fraction), 1 N HCl (dynamic fraction) and insoluble residue (fixed fraction) was used. Statistical processing of the results was carried out by conventional methods.

3 Results and Discussion

3.1 The Study of Regularities of Radionuclides Accumulation in Aquatic Vegetation of a Watercourse: The Techa River

The source of Techa River was till the year 1956 the Irtyash Lake with the water transit through the Kyzyltash Lake. From 1956 to 1964 in the river's headwaters a cascade of dams was constructed and this allowed localizing the major part of radioactive substances deposited in the headwaters. Thus the so-called Techa cascade of water bodies (TCW) was formed. Currently the source of the river is formed below the dam of V-11 technical water reservoir due to release of water from the left-bank bypass channel (LBC), the right-bank bypass channel (RBC) and filtration under the V-11 reservoir dam (Mayak 2007).

According to the structure features of the valley and the bed within the Chelyabinsk region, the Techa River can be divided into two parts. First part: source-the village of Muslyumovo. The floodplain is bilateral, the prevailing floodplain width is 2.0–2.5 km, in the guts 0.4–0.8 km, at the end of the part-up to 150–120 m, and the floodplain surface is swampy and tussocky. The floodplain is flooded every year during the high water period with a layer of 0.2–1.0 m and up to 2 m in the highest water period. The River comes out of bogs near the village of Muslyumovo. The river depth in the cripples is 0.3–0.8 m, in the reaches—up to 5 m; the flow velocity is 0.1–0.4 m s^{-1}. Second part: the village of Muslyumovo-the village of Nizhnepetropavlovskoe, with a length of 60 km. The floodplain is bilateral, asymmetric; the average width is 300–400 m. There are width fluctuations from 30 to 35 m near the village of Muslyumovo to 700 m near the village of Nizhnepetropavlovskoe. The floodplain is flooded along the entire length during the ordinary high water periods to a depth of 0.5–2.5 m. The depth in the cripples is

0.4–0.6 m, in the reaches—2–3 m. The flow velocity in the cripples is 1.2 m s^{-1}, and reaches up to 0.4 m s^{-1}. The main tributaries: the Mishelyak River, the Zyuzelga River and the Baskazyk River. The high water period is usually in April. The maximum intensity of water level rise during the high water period is 30 cm s^{-1} the maximum recession intensity is 70 cm day^{-1}. The low water levels are unstable. During the year they fluctuate sharply but insignificantly, from 20 to 50 cm. The rainfall floods are not typical for the Techa River (Basic laws of radionuclide distribution in the system of the Techa River as a result of long-term observations). The water of the river has pH ≈ 7.4. The aqueous run-off at the river mouth is 0.35 $km^3 year^{-1}$. In the area of Brodokalmak and Zatechensky the water discharge in the high water period is approximately the same and is about 300 $m^3 s^{-1}$, in the low water period the run-off decreases up to 1–1.3 $m^3 s^{-1}$. The maximum solid run-off rate was noted in the mouth during the high water period (123.7 kg s^{-1}). During the low water period 1–20 g s^{-1} (Trapeznikov et al. 2007a).

In the years 1949–1956 liquid radioactive waste (LRW) from radiochemical production (nowadays-Production Association "Mayak") was discharged into the Techa River. That's why the floodplain and bottom silt of the Techa River are contaminated with radionuclides, and sludge deposits in the head of river are considered as solid radioactive waste. It is assumed that in the period from 1949 to 1954, at the point of LRW discharge were transferred in particular 47.1 kCi of $^{90}Sr + ^{90}Y$, 30.3 kCi of which-in form of solution, the rest-as a component of natural and anthropogenic suspended particles. 49.9 kCi of $^{137}Cs + ^{137m}Ba$ were transferred, 42.4 of which—as a component of suspended particles, the rest-in form of solution. In the dam site of the village of Muslyumovo the run-off of $^{90}Sr + ^{90}Y$ during this period amounted to 29.3 kCi, $^{137}Cs + ^{137m}Ba$—5.6 kCi (Mokrov 2005). In the year 1956 the dam No. 10 was constructed, in 1964 the dam No. 11 was constructed and formed a new reservoir V-11. There were accumulated about 3.1×10^5 Ci of long-living β-active nuclides in the Techa cascade of water bodies (TCW). Contaminated water comes to open hydrographic system of the Techa River through the right-bank and left-bank bypass channels (RBC, LBC) as well as under the body of dam No.11 (Vakulovsky 2009, 2010, 2011).

According to different researchers, in the year 2000 the yearly average specific activity of ^{90}Sr in the water of the Techa River was about 10 Bq L^{-1}, the activity of other radionuclides was significantly lower than intervention level. According to Baranov et al. (2011), the yearly average activity of ^{90}Sr in the dam site of the Muslyumovo village is about 9 Bq L^{-1} (from 3 Bq L^{-1} in the high water period to 18 Bq L^{-1} in the low water period). According to Drozhko and Mokrov (2008), in the years 1995–2006 the yearly average specific activity of ^{90}Sr in the Techa River water (Muslyumovo) was 10–15 Bq L^{-1} and varied from 2 to 3 Bq L^{-1} during the spring flood to 20–40 Bq L^{-1} during the low water period in summer (Drozhko and Mokrov 2008). It is reported in the annuaires "Radiation situation in the territory of Russia and neighbouring states" that in the dam site of the village of Muslyumovo from 2007 to 2010 the yearly average activity of ^{90}Sr in water increased from 7.7 to 13.6 Bq L^{-1} (according to the information from the Central Laboratory of the Production Association "Mayak") or from 8.9 to 18.5 Bq L^{-1} (according to the

information from the Science and Production Association "Taifun"). The activity of ^{137}Cs decreased slightly in 2010 and amounted accordingly to 0.37 and 0.24 Bq L^{-1} (Vakulovsky 2009, 2010, 2011). In 2005 in the territory of Asanovsky bogs the content of ^{137}Cs in different sampling points (throughout all sampling depth) was 52.8–107.8 MBq m^{-3}, ^{90}Sr—7.9–8.8 MBq m^{-3}. In the territory of Muslyumovsky bogs in 2004 the content of ^{137}Cs amounted to 1.4 MBq m^{-3}, ^{90}Sr—0.3 MBq m^{-3} (Kostyuchenko et al. 2009). Trapeznikov et al. (2000) think that during the high water period from the Techa River to the Iset River as a component of liquid run-off are transferred 1.4×10^6 Bq s^{-1} of ^{90}Sr and 0.02×10^6 Bq s^{-1} of ^{137}Cs. The entry of radionuclides with solid run-off is approximately the same and equals to 0.1×10^6 Bq s^{-1} (Trapeznikov et al. 2007a). Thus, the Techa River is from 1949 till present a source of radioactive contamination of the rivers Iset and Tobol.

Our studies have found that the water of the Techa River is more contaminated in the upstream flow until the village of Muslyumovo (≈54 km away from the dam V-11). In the period from 2000 to 2012 the specific activity of ^{90}Sr in the water in this area ranged from 1.6 to 60.0 Bq L^{-1} and, on the average, based on 121 samples amounted to 17.1 ± 2.2 Bq L^{-1}. The activity of ^{137}Cs varied from 0.06 to 11.5 Bq L^{-1}, on the average- 1.0 ± 0.4 Bq L^{-1}, the activity of ^{3}H varied from 11.2 to 451 Bq L^{-1}, on the average—240 ± 33 Bq L^{-1}, the activity of ^{239,240}Pu did not exceed 0.096 Bq L^{-1}, on the average 0.019 ± 002 Bq L^{-1}.

Figure 1a shows the values of the specific activity of radionuclides in water along the Techa River bed at different distances from the dam V-11, on the average in the years 2000–2012. In general, the water contamination level decreases relatively smooth, the fluctuations of the specific activity of ^{90}Sr and ^{137}Cs are observed. They are connected with the location of big tributaries and the type of the water-collecting areas. The bottom silt is also very contaminated with radionuclides. Figure 1b shows the values of the specific activity of ^{90}Sr, ^{137}Cs and ^{239,240}Pu in the upper layer of bottom silt. The greatest amount of radionuclides is concentrated in the upstream. At the distance of 30 km from the dam V-11 the content of radionuclides in the bottom silt decreases abruptly.

In order to evaluate the ability of aquatic vegetation to concentrate radionuclides the ratio of the radionuclide specific activity in the dry biomass to its specific activity on water is used (C_a). However, the appropriateness of use of such ratio for the rooted plants comes into question because they can receive radionuclides both by roots and foliage. All the more so as the radionuclides content in the bottom silt exceeds their content in the water by hundreds of thousands of times as can be seen from Fig. 1a, b.

In addition, the radionuclides content in the water varies greatly depending on the season and weather conditions. Figures 1c, dshows the values of the radionuclides volumetric activity in the river water sampled at the dam site of the village of Muslyumovo from 1975 to 2012. It is known that if radionuclide activity in water changes, its activity in plants also changes rapidly. In an experiment of Chebotina (1981) after a threefold replacement of radioactive isotopes solution with clear water during 9 days, 10–50 % of absorbed ^{137}Cs and 1–30 % of absorbed ^{90}Sr remain in aquatic vegetation. Trapeznikov et al. (2000, 2007a) have found that the C_a values

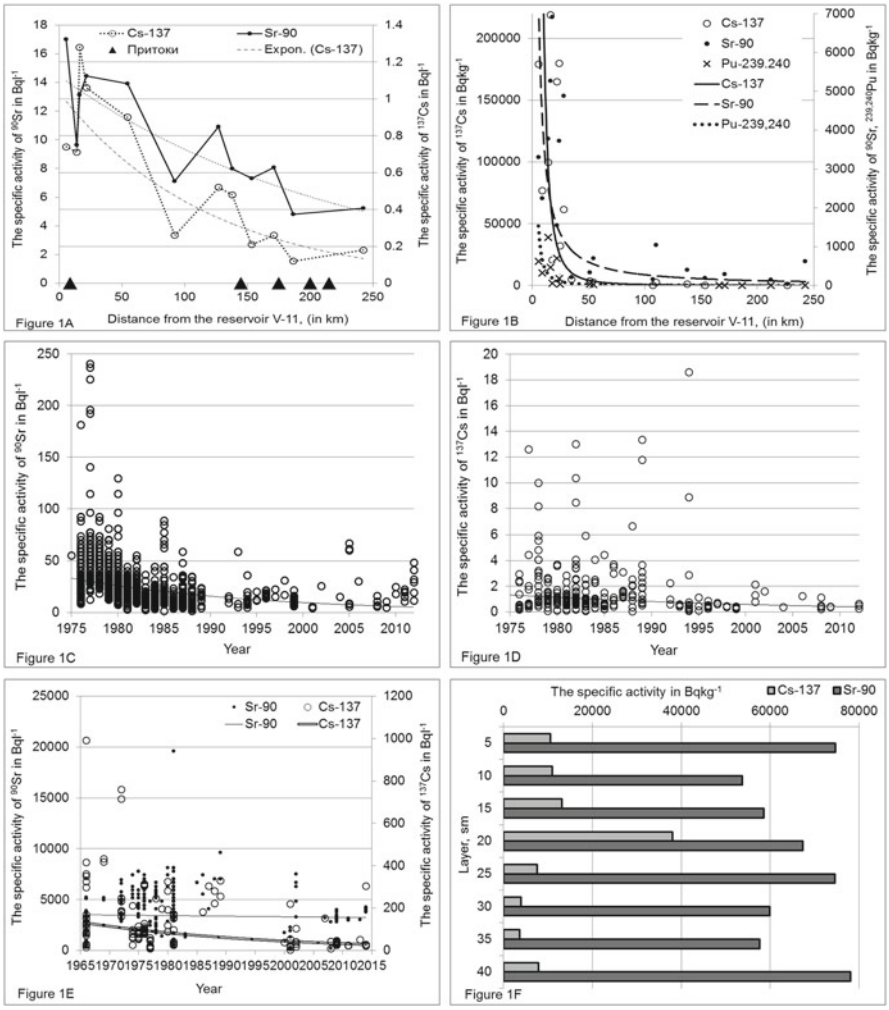

Fig. 1 (**a**) The specific activity of radionuclides in the water of the River Techa. (**b**) Specific activity of the radionuclides in the upper layer of sediments. (**c**) The specific activity of ^{90}Sr in the water near the village of Muslyumovo. (**d**) The specific activity of ^{137}Cs in the water near the village of Muslyumovo. (**e**) The dynamics of the specific activity of radionuclides in the water reservoir V-10. (**f**) The distribution of specific activity of radionuclides in the profile of bottom sediments in 2008

of *Elodea canadensis* and *Ceratophyllum* sp. can change in natural conditions depending on the year season five or sixfold and achieve maximum values in summer months. As a consequence of strong seasonal fluctuations of activity, we calculated the coefficient of ^{90}Sr and ^{137}Cs accumulation in the aquatic vegetation based on the average activity values in the water sampled during the vegetative period (May-October). Over the whole period of studies conducted by the employees of

Urals Research Center for Radiation Medicine were taken 353 samples of 22 plant genera in 20 points along the Techa River stream. The greatest amount of samples was analysed in the years 1977 and 1979. The specific activity of ^{137}Cs in the dry biomass in different samples was from 22 to 44,400 Bq kg^{-1} of dry weight, of ^{90}Sr—from 3 to 262,700 Bq kg^{-1} of dry weight.

Tables 1 and 2 represent the average C_a values for the best studied plant genera. Table 3 shows for comparison the C_a values obtained by Trapeznikov et al. (2007b). It should be noted that the C_a values vary greatly. At the same time the hydatophytes and hydrophytes don't differ conceptually in the ability to concentrate radionuclides. Whereas *Scirpus* sp. and *Typha* sp. have relatively low C_a of ^{137}Cs, these values for *Carex* sp. and *Eriophorum* sp. are virtually identical to C_a of ^{137}Cs in hydatophytes. C_a of ^{90}Sr is relatively low for *Scirpus* sp., *Carex* sp. and *Nuphar lutea*, but for *Typha* sp. and *Eriophorum* sp. they are equal to C_a of hydatophytes. But in general, rooted plants accumulate significantly less ^{90}Sr and ^{137}Cs than unrooted plants, furthermore, any abrupt change of C_a wasn't observed at the distance of 30 km from the dam, although the activity of ^{90}Sr and ^{137}Cs in the bottom silt decreases by a huge ratio (Fig. 1b). Thus, the pollution level of bottom silt apparently doesn't have a great impact on C_a of ^{90}Sr and ^{137}Cs in aquatic vegetation.

Interestingly, the ^{90}Sr and ^{137}Cs activity in water, as shown on the Figure 1a, decreases with the distance nearly synchronous, but the C_a changes in different directions: For most plants except *Ceratophyllum* sp., C_a of ^{137}Cs decreases with the distance and C_a of ^{90}Sr either doesn't depend on distance or increases.

3.2 The Study of Regularities of Radionuclides Accumulation in the Aquatic Vegetation of the Stagnant Reservoir V-10

Techa cascade of water bodies (TCW) was created for the reception and storage of low-level LRW and as the first stage of Techa River's radiation rehabilitation. I order to localize the most heavily polluted areas of headwaters floodplain in 1956 and 1964 soil dams of reservoirs V-10 and V-11, as well as bypass channels for land run-off interception were constructed (Baranov et al. 2011).

The database of Urals Research Center for Radiation Medicine provides information on the radionuclides volumetric activity in the water of reservoir V-10 from 1966 to 2012. The Figure 1a shows the dynamic pattern of the average values of the volumetric activity in the water of reservoir V-10. One can see on the figure that over the past 10 years the volumetric activity of ^{90}Sr and ^{137}Cs decreased slightly but large fluctuation amplitude doesn't allow revealing the trends of the radioactive contamination dynamics of water. It should be noted that the analysis results of samples taken in different parts of the reservoir at different times vary widely. In 2009, we researched the radionuclide composition of water from the reservoir V-10. The results can be found in Table 4.

Table 1 Average values of C_a ^{137}Cs in the dry biomass of the Techa River plant

The distance from the dam reservoir (11 km)	Nearest village or landmark	*Elodea canadensis*	*Myriophyllum verticillatum*	*Cerato phyllum* sp.	*Lemna* sp.	*Scirpus* sp.	*Nuphar lutea*	*Carex* sp.	*Typha* sp.	*Eriophorum* sp.
4.5	Asanov bridge	6166	5781	–	6167	215	1899	6421	416	–
12	Maloye Taskino	–	–	–	2105	–	–	6220	165	–
14	New bridge	–	–	–	11,385	–	–	4466	–	–
15.5	Gerasimovka	–	–	–	2139	112	–	–	73	–
20	Nadyrov bridge	–	–	–	–	–	–	2485	196	–
25	Ibragimovo	–	–	127	2604	124	713	6156	254	1964
32	Farm	–	–	–	2137	60	–	461	37	1350
38	Muslyumovo	4295	–	–	1036	55	382	587	–	–
50	Kumanovo	–	–	–	–	–	–	1717	–	–
103	Nizhnepetropavlovskoe	–	–	–	–	–	–	–	–	–
119	Lobanovo	–	–	532	–	–	740	1110	–	–
141	Verhnyaya Techa	–	–	–	–	–	–	–	–	–
175	Pershinskoe	284	–	–	–	–	–	926	–	–
195	Zatechenskoe	707	–	899	2221	–	–	1050	–	621
	Correlation	−0.98	−1	0.99	−0.26	−0.93	−0.45	−0.58	−0.65	−0.90

Table 2 Average values of C_a ^{90}Sr in the dry biomass of the Techa River plant

The distance from the dam reservoir (11 km)	Nearest village or landmark	*Elodea canadensis*	*Myriophyllum verticillatum*	*Cerato phyllum* sp.	*Lemna* sp.	*Scirpus* sp.	*Nuphar lutea*	*Carex* sp.	*Typha* sp.	*Eriophorum* sp.
4.5	Asanov bridge	291	209	–	208	57	147	109	135	–
12	Maloye Taskino	–	–	–	282	–	–	52	166	–
14	New bridge	–	–	–	225	–	–	94	–	–
15.5	Gerasimovka	–	–	–	263	45	–	–	26	–
20	Nadyrov bridge	–	–	–	–	–	–	19	446	–
25	Ibragimovo	–	200	206	244	87	66	50	177	360
32	Farm	–	–	–	274	60	–	12	192	274
38	Muslyumovo	141	–	–	184	94	44	48	–	–
50	Kumanovo	–	–	–	–	–	–	233	–	–
103	Nizhnepetropavlovskoe	–	–	–	–	–	–	140	–	–
119	Lobanovo	–	–	103	–	–	248	30	–	–
141	Verhnyaya Techa	–	–	–	–	–	–	318	–	–
175	Pershinskoe	644	–	–	–	–	–	197	–	–
195	Zatechenskoe	525	–	263	245	–	–	58	–	–
	Correlation	0.87	−1.00	0.29	0.02	0.67	0.72	0.39	0.26	−0.82

Table 3 Average values of C_a ^{90}Sr, ^{137}Cs in the dry biomass of *Ceratophyllum demersum* the Techa River plant

Nearest village or Landmark	^{90}Sr	^{137}Cs
Nadyrov bridge (Techa River)	240	14,838
Muslyumovo (Techa River)	100	2981
Verhnyaya Techa (Techa River)	906	1290
Zatechenskoe (Techa River)	589	912
Verhni Yar (Iset River)	640[a]	1200[a]
Sorovskoe (Iset River)	371	5491
Mehonskoe (Iset River)	163	4600
Ust-Miasskoe (Iset River)	866	–
Yalutorovsk (Iset River)	907[a]	4000[a]
Yalutorovsk (Tobol River above the confluence of the river Iset)	300[a]	444[a]
Yalutorovsk (Tobol River below the confluence of the Iset River)	1057[a]	767[a]

[a]Adopted from Trapeznikov et al. (2000, 2007a, b)

Table 4 The radionuclide composition of industrial water reservoir V-10

	Activity	
Radionuclide	Bq L^{-1}	%
^{137}Cs	34.8 ± 2.4	0.52
^{90}Sr	3320 ± 160	49.7
^{241}Pu	0.91 ± 0.30	0.014
^{238}Pu	0.14 ± 0.03	0.002
$^{239,240}Pu$	0.11 ± 0.02	0.002
^{234}U	0.51 ± 0.24	0.008
^{238}U	0.36 ± 0.13	0.005
^{3}H	3330 ± 34	49.8

As can be seen in Table 4, ^{90}Sr and ^{3}H contribute greatly to the radioactivity of water in studied reservoir while the correlation $^{90}Sr/^{3}H$ in water equals to 1 and the correlation $^{137}Cs/^{90}Sr$ is equal to 0.01. The contribution of plutonium isotopes to the total activity of water is insignificant and is not greater than 0.02 %. The share of ^{241}Pu in the sum of plutonium isotopes is about 80 %.

The Table 5 shows the dynamics of ^{3}H, ^{90}Sr and ^{137}Cs content in the water of B-10 in the years 2009–2014. During this period the activity of ^{3}H has decreased slightly and the activity of ^{90}Sr has increased. The correlation $^{90}Sr/^{3}H$ has increased up to two times during the last year. ^{3}H activity decrease from 2009 to 2014 corresponds quantitative to its decomposition but it is worth mentioning that in fact there was a sharp decline in activity in the period of 2009–2011 and from 2011 to 2014 the ^{3}H content was relatively stable Kazachonok et al. (2013a). We have previously shown that in 2005 the activity of ^{3}H in the reservoir B-10 amounted to 9700 Bq L^{-1} and that HTO apparently evaporates faster than H_2O, which led to a sharp decrease in activity of ^{3}H from 2005 to 2012, Kazachonok et al. (2013b). Consequently, a relative stability of ^{3}H content in

Table 5 Radionuclide content in the water reservoir V-10 (in Bq L^{-1})

Month and Year of measurement	^{3}H	^{90}Sr	^{137}Cs
07.2009	3426 ± 15	3240 ± 155	41 ± 2.9
08.2009	3225 ± 34	3398 ± 508	28.4 ± 4.2
10.2011	2291	3060 ± 82	22.6
07.2012	2138	–	–
05.2013	1932	3011	51.5
08.2014	–	3972 ± 203	26.8 ± 2.0
09.2014	2103 ± 142	4045 ± 113	23.4 ± 2.3

Table 6 Radionuclide content in sediments of reservoir V-10 (in the year 2009) (in kBq kg^{-1})

Radionuclide	250 m from the dam reservoir V-4	Left bank	Center	Right bank	200 m from the dam reservoir V-10
^{137}Cs	2310 ± 115	205 ± 10	2060 ± 10	216 ± 11	2170 ± 110
^{60}Co	24.3 ± 2.4	1.3 ± 0.8	12.4 ± 1.4	1.6 ± 0.1	12.8 ± 1.7
^{90}Sr	319 ± 50	24.1 ± 3.9	364 ± 55	67.6 ± 9.5	246 ± 40
^{238}Pu	57 ± 11	4.9 ± 1.0	66 ± 13	5.6 ± 1.3	41 ± 8.6
$^{239,240}Pu$	41 ± 9.1	4.1 ± 1.9	56 ± 11	4.3 ± 1.1	32 ± 7.0
^{241}Am	128 ± 18	9.8 ± 2.9	85 ± 13	10.4 ± 3.1	106 ± 15
^{241}Pu	577 ± 125	30 ± 7	302 ± 65	32 ± 7.7	320 ± 67

the years 2011–2014 gives evidence of the continuing contamination of B-10 with ^{3}H and other radionuclides. It should be pointed out that the dynamics of ^{3}H, ^{90}Sr and ^{137}Cs activity in the water of V-10 doesn't correspond. So, in 2013 the activity of ^{3}H and ^{90}Sr was minimal but the activity of ^{137}Cs was maximal. Apparently, the water contamination level fluctuations are subject not only to downflow from the reservoir V-4 but also to other factors influencing the sorption-desorption balance of ^{137}Cs and ^{90}Sr.

The bottom silt represents a complex multicomponent system and plays a crucial role in the formation of hydrochemical regime of water masses and the functioning of ecosystems, water bodies and water courses. They participate actively in the intra-reservoir cycle of matter and energy and are the habitat of numerous groups of living organisms–benthos (Rainin and Vinogradova 2002). Also the bottom silt plays a leading role in the process of self-purification from contaminants of both flowing and stagnant water reservoirs. At the same time, they are also a potential source of secondary contamination of water due to desorption of radionuclides. In the period 2009–2014, we measured the radionuclides content in the bottom silt in 5 stations in the reservoir V-10. The results are shown in Tables 6 and 7.

The main activity is caused by ^{137}Cs (from 64 to 74 %), plutonium isotopes and ^{241}Am account for 16–23 % from the total activity while ^{241}Pu contributes about 12 %. There is a close connection between the content of ^{60}Co and ^{137}Cs in the bottom silt (average ratio—0.007 ± 0.001). The concentration of ^{241}Pu in the bottom soil exceeds the total activity of other plutonium isotopes by 4–6 times. The correlation $^{241}Am/^{137}Cs$ equals at the average to 0.05 ± 0.1. The correlation $^{238}Pu/^{239,240}Pu$ varies from 1.2 to 1.4, and was 1.3 at the average.

Table 7 Radionuclide content in bottom sediments of reservoir V-10 (in the year 2014) (in kBq kg^{-1})

Radionuclide	250 m from the dam reservoir V-4	Left bank	Center	Right bank	200 m from the dam reservoir V-10
^{137}Cs	1840 ± 165	320 ± 30	2770 ± 280	141 ± 14	2140 ± 214
^{60}Co	14.8 ± 2.7	1.0 ± 0.3	14.8 ± 2.2	0.7 ± 0.4	11.5 ± 1.8
^{22}Na	–	–	1.4 ± 0.8	–	1.0 ± 0.7
^{7}Be	–	–	60 ± 42	–	–
^{90}Sr	2220 ± 330	88.6 ± 12.4	1480 ± 220	130 ± 20	960 ± 140
^{238}Pu	73.9 ± 13.3	1.1 ± 0.2	96.6 ± 17.4	0.48 ± 0.09	57.8 ± 10.5
^{239,240}Pu	55.2 ± 9.9	0.8 ± 0.1	83.9 ± 15.1	0.36 ± 0.06	45.2 ± 8.1
^{241}Am	159 ± 29	22 ± 4	159 ± 30	10 ± 2	134 ± 28
^{241}Pu	425	10.7	597	4.5	378

Table 8 Physical and chemical forms of ^{137}Cs and ^{90}Sr in bottom sediments of the reservoir V-10 as a % of the total content

Radionuclide	Soluble in water	Capable to an exchange	Soluble in acid	Insoluble
	Muddy sediments central part of the reservoir			
^{137}Cs	0.4 ± 0.1	1.9 ± 0.1	1.3 ± 0.1	96.6 ± 0.2
^{90}Sr	1.5 ± 0.1	32.1 ± 1.0	34.5 ± 2.5	31.8 ± 2.2
	The sandy sediments of the coastal part of the reservoir			
^{137}Cs	0.3 ± 0.01	3.9 ± 0.1	3.4 ± 0.2	92.4 ± 0.3
^{90}Sr	3.5 ± 0.3	80.4 ± 1.1	8.0 ± 0.8	8.5 ± 0.7

The radionuclides content in the soil depends on the sampling place. The activity of waterside sandy soils covered by a thin layer of silt and dying vegetation is by times lower, than in silt of the reservoir's middle part, which is determined by both different sorption properties of soil and the conditions of primary contamination. According to Baranov et al. 2011, the activity of ^{90}Sr in the bottom silt is relatively evenly distributed and amounts to 240–370 kBq kg^{-1}. The activity of ^{137}Cs in the river's old channel is 30–160 and 5–15 kBq kg^{-1} in the waterside areas. According to our data (Tables 6 and 7), the contamination of the bottom silt with ^{137}Cs is significantly higher. It was found in the samples in 2014 significantly more ^{90}Sr too. The distribution of ^{137}Cs and ^{90}Sr in the bottom silt profile is relatively even (Fig. 1f). The radionuclides entry to plants depends firstly on their bioavailability. Table 8 shows the correlation of physicochemical forms of radionuclides in the bottom silt of the V-10 reservoir. The study results of the content of available for plants radionuclides fractions in the bottom silt indicate that the main amount of ^{137}Cs is in fixed state. The water-soluble fraction makes 0.3–0.4 %, exchangeable and dynamic fraction 1.3–3.9 %, wherein their share is slightly higher in sandy soil than in muddy soil. In waterside sandy soil more than 80 % of ^{90}Sr are in exchangeable state and only 7–9 %—in fixed state. In muddy soil 32.1 % fall to the share of exchangeable ^{90}Sr and 34.5 %—to the share of dynamic ^{90}Sr which may be combined with readily-soluble organic compounds or is in the form of carbonates.

It is worth mentioning that the share of radionuclides in a form available for plants in the bottom silt of V-10 reservoir is significantly lower than in soils formed in the same natural and climatic zone. So, the upper layer (0–5 cm) of gray forest and sod-podzolic soil contained 2.8–9.8 % of water-soluble ^{137}Cs and 2.9–5.0 % of water-soluble ^{90}Sr. The 5–10 cm layer contained 13.7–16.5 % of water-soluble ^{137}Cs and 1.7–3.9 % of water-soluble ^{90}Sr (Kazachonok and Popova 2014). It can be assumed that in conditions of one ecosystem nearly all water-soluble radionuclides fractions get into aquatic environment, that's why the root absorption of ^{90}Sr and ^{137}Cs by aquatic vegetation is significantly smaller than foliar absorption.

The comparison of the bottom absorption coefficients (BAC) for muddy soils of the studied reservoir which we calculated based on the data of 2009 showed that ^{90}Sr is less capable of sorption and the BAC of muddy soils is about 100 L kg^{-1}, of sandy soils 15 L kg^{-1} at the average. The BAC for ^{137}Cs is much higher and equals to 62,640 L kg^{-1} on the average but by the time lower for the waterside soil. The maximum observed BAC values in bottom silt are of plutonium isotopes $4° \times 10^5$ L kg^{-1}. The difference of the BAC values is most probably determined by the features of radionuclides' physicochemical state. In the waterside sandy bottom silt the BAC for all radionuclides is significantly lower than in muddy soil. In 2014, the BAC for ^{137}Cs and ^{90}Sr were slightly higher. For example, the ^{137}Cs BAC of muddy soils was 84,000 L kg^{-1} waterside soils about 8600 L kg^{-1} and ^{90}Sr BAC was 390 L kg^{-1}.

The plant sampling in the V-10 reservoir was carried out in 1965, 1972, 1974, 2009 and 2014. The main representatives of higher aquatic vegetation growing in the V-10 reservoir in the study period were: *Phragmites australis*, *Typha laxmanii*, *Potamogeton pectinatus*, *Potamogeton lucens*, *Polygonum amphibium*, *Scirpus lacustris*, *Lemna* sp., *Ceratophyllum* sp. The activity of ^{137}Cs in the dry phytomass was from 1.1 to 333 kBq kg^{-1}, the activity of ^{90}Sr from 3.3 to 925 kBq kg^{-1}. The total activity of Pu isotopes can reach 9 kBq kg^{-1}. The levels of aquatic vegetation contamination with radionuclides are represented in Table 9.

As the Table 9 shows, in the initial period after the reservoir formation the radionuclides accumulation in plants was relatively small. The high contamination levels were noted in 1972–1974, and in 2009 the levels of plants contamination with ^{90}Sr and especially ^{137}Cs decreased significantly. However, during this period the average content of ^{137}Cs in the water of V-10 decreased too (Fig. 1e). Therefore it was to be expected that C_a of ^{137}Cs will not decrease. The C_a of ^{137}Cs and ^{90}Sr in the vegetation in this period are shown in Tables 10 and 11. Actually, in most cases the C_a increased significantly. In general, the C_a of *Phragmites* was by far lower than of hydatophytes, the same as in the Techa River.

The radionuclides accumulation by hydrobionts proceeds by two basic mechanisms: biological (absorption as a result of vital activity) and physicochemical (concentration on organisms' surface as a result of sorption and coprecipitation with macroelements). While there is a large difference in the content of ^{137}Cs and ^{90}Sr in water, the C_a values of higher aquatic vegetation differ insignificantly (except *Lemna*). This may be due to the belonging to potassiumphilic species as well as the morphological features of the plants. High plutonium isotopes C_a values are most probably determined by the surface adsorption because the adsorptive accumulation

Table 9 The specific activity of radionuclides in selected samples of aquatic vegetation in of the reservoir V-10 (in the year 1965–2009) (in kBq kg^{-1})

Date	Type of plant	^{137}Cs	^{90}Sr	^{241}Pu	^{238}Pu	$^{239,240}Pu$	^{60}Co
01.08.1965	*Potamogeton* sp.	16.0	274	–	–	–	–
02.08.1965	*Myriophyllum verticillatum*	1.5	17.2	–	–	–	–
02.08.1965	*Myriophyllum verticillatum*	1.1	22.6	–	–	–	–
02.08.1965	*Myriophyllum verticillatum*	2.0	16.1	–	–	–	–
02.08.1965	*Lemna* sp.	3.2	3.3	–	–	–	–
02.08.1965	*Lemna* sp.	17.2	17.6	–	–	–	–
06.08.1972	*Phragmites* sp.	2.2	270	–	–	–	–
06.08.1972	*Phragmites* sp.	4.4	237	–	–	–	–
06.08.1972	*Potamogeton* sp.	92.5	629	–	–	–	–
06.08.1972	*Potamogeton* sp.	28.1	666	–	–	–	–
06.08.1972	*Potamogeton* sp.	30.3	481	–	–	–	–
06.08.1972	*Potamogeton* sp.	37.0	660	–	–	–	–
06.08.1972	*Potamogeton* sp.	32.9	925	–	–	–	–
06.08.1972	*Phragmites* sp.	5.9	37.0	–	–	–	–
06.08.1972	*Phragmites* sp.	2.8	59.2	–	–	–	–
06.08.1972	*Phragmites* sp.	5.2	118	–	–	–	–
06.08.1972	*Phragmites* sp.	4.4	107	–	–	–	–
06.08.1972	*Phragmites* sp.	8.9	111	–	–	–	–
01.10.1974	*Myriophyllum verticillatum*	129	555	–	–	–	–
01.10.1974	*Myriophyllum verticillatum*	322	444	–	–	–	–
01.10.1974	*Myriophyllum verticillatum*	333	407	–	–	–	–
01.08.2009	*Potamogeton lucens*	6.2	290	7.9	0.62	0.55	0.74
01.08.2009	*Potamogeton pectinatus*	14.4	401	3.7	0.54	0.55	1.3
01.08.2009	*Phragmites* sp.	1.8	108	0.29	0.015	0. 38	–
01.08.2009	*Lemna* sp.	29.6	346	2.2	0.42	0.29	2.74
01.08.2009	*Polygonum amphibium*	8.4	328	1.7	0.32	0.27	0.41
01.08.2009	*Ceratophyllum* sp.	8.8	677	–	–	–	1.8

Table 10 Average values of C_a ^{137}Cs and ^{90}Sr in the dry biomass of the reservoir V-10 plants (in the year 1965–1974)

Year	*Potamogeton* sp.		*Myriophyllum verticillatum*		*Lemna* sp.		*Phragmites* sp.	
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr
1965	78	89	7.6	6	50	3.4		
1972	223	154					24.5	30.7
1974			29.4	87				

Table 11 Average values of C_a of radionuclides in the dry biomass of the reservoir V-10 plant (in the year 2009)

Types of plant	^{137}Cs	^{90}Sr	^{238}Pu	$^{239,240}Pu$
Phragmites australis	51	33	108	345
Polygonum amphibium	240	99	2250	2440
Potamogeton pectinatus	310	120	4580	5040
Potamogeton lucens	180	90	4410	4960
Lemna sp.	850	110	2980	2610
Ceratophyllum sp.	255	205	–	–

Table 12 The specific activity of radionuclides in aquatic vegetation in of the reservoir V-10 (in the year 2014)

Types of plant	^{137}Cs (kBq kg^{-1})	^{90}Sr (kBq kg^{-1})	^{238}Pu (kBq kg^{-1})	$^{239,240}Pu$ (kBq kg^{-1})	^{241}Pu (kBq kg^{-1})	^{65}Zn (kBq kg^{-1})
Scirpus lacustris	1.5 ± 0.1	229 ± 37	3.8 ± 1.1	6.6 ± 2.1	95.1 ± 12.3	36 ± 25
Typha latifolia	2.0 ± 0.2	255 ± 38	28.8 ± 5.6	38 ± 7.2	27.3 ± 2.2	
Phragmites australis	3.9 ± 0.3	294 ± 44	10.2 ± 2.5	9.6 ± 2.4	42.6 ± 3.4	46 ± 28
Polygonum amphibium	6.7 ± 0.5	590 ± 94	434 ± 54	309 ± 38	2658 ± 301	
Potamogeton pectinatus	10.9 ± 0.9	824 ± 123	558 ± 84	425 ± 64	2704 ± 193	80 ± 40

Table 13 Average values of C_a of radionuclides in the dry biomass of the reservoir V-10 plant (in the year 2014)

Types of plant	^{137}Cs	^{90}Sr
Scirpus lacustris	60	57
Typha latifolia	80	64
Phragmites australis	155	73
Polygonum amphibium	267	147
Potamogeton pectinatus	434	205

mechanism is typical for radionuclides which are in the water environment in the form of suspended matter and colloids and don't have any pronounced biogenic properties (Egorov and Suzdaleva 2005). In 2014, we measured the radionuclides content in 5 taken vegetation species from V-10 (Tables 12 and 13). Unfortunately, the species composition of the phytomass was not constant over the years. Therefore it is impossible to trace the dynamics of accumulation coefficients change for all studied species. We can only say that we observed the increase of C_a ^{137}Cs for *Ceratophyllum* and *Phragmites* over time. Moreover, this increase is due not only to the activity decrease of these radionuclides in water.

3.3 The Study of Regularities of Radionuclides Accumulation in the Aquatic Vegetation of a Stagnant Reservoir: The Uruskul Lake

Aerosol radionuclides discharges from the plants of the Production Association "Mayak" in the 1950–1960s lead to soil contamination in the area of the enterprise to the level of approximately 10^{13} Bq km^{-2} of ^{90}Sr and ^{137}Cs, and 10^{10} Bq km^{-2} of plutonium isotopes. At the same time from radioactive contamination suffered all components of terrestrial and aquatic ecosystems located in the zone of influence of the PA "Mayak" (Stukalov and Rovni 2009).

On September 29, 1957 a waste tank exploded in the radioactive waste repository of the PA "Mayak" and as a result radioactive substances with the total β-activity of about 2 mCi formed a cloud at a height of approximately 1 km. As a consequence of precipitation of isotopic mixture from the cloud, the East Ural radioactive trace (EURT) formed. The axis of maximum contamination passed through the settlements Berdyanish, Saltykova, Galikaeva, Russkaya Karabolka and further down to Tyumen (Akleev and Kiselev 2001). In April 1967 in the studied area the fallouts of radioactive dust were noted (Akleev 2006). The fallouts consisted of ^{137}Cs (48 %), ^{90}Sr+^{90}Y (34 %) Shoigu (2002). The most contaminated settlements were: Metlino (ONIS), Bolshoy Kuyash, Golubinka, Sarykulmyak and Khudayberdinsk (Akleev 2006). Thus, stagnant water reservoirs within a radius of 30–35 km from the PA "Mayak" and in the EURT within a distance of more than 100 km were contaminated too.

In 2008–2011 we studied the samples of water, bottom silt and waterside soil taken from 26 places of an active water use, 24 lakes at a distance of 30–40 km from the industrial site of the PA "Mayak" and from the Elovoe Lake (near the city of Chebarkul) selected for comparison. The ^{137}Cs and ^{90}Sr activity in water correlates with their activity in bottom silt. The activity of ^{137}Cs in water and bottom silt as well as of ^{3}H in water correlates with the ^{137}Cs activity in the 0–10 cm soil layer in the area Kazachonok et al. (2014a). The correlation of the ^{3}H activities in water and the ^{137}Cs activities in the area suggests that the contamination with ^{137}Cs in the area is also mainly due to routine releases, and not to carrying away from the Karachay Lake by the wind. (Kazachonok et al. 2014b).

Many ecologists studied the radionuclides accumulation by the lacustrine vegetation. As far back as in the first years after the EURT formation, Kogotkov and Osipov (2002) found that radioisotope composition of *Phragmites* in different lakes located in the EURT is similar to the radioisotope composition of bottom silt and the radioisotope composition of *Stratoides aloides* to the composition of water. The accumulation coefficients varied over years as well as depending on the water body and plant species. Especially violent accumuation coefficients changes over the monitoring period (1961–1966) were noted in the Bolshoy Igish Lake (up to 14 times), in the Kozhakul Lake (up to 10 times) and the Uruskul Lake (up to 7 times). The spread in accumulation coefficients values is apparently conditional upon by the variability of plants sampling points, although the contamination density diversity on different banks of the lakes is significantly smaller (Kogotkov and Osipov 2002). The Tables 14, 15, and 16 contain the data on C_a of ^{137}Cs and ^{90}Sr by macrophytes from

different lakes according to various researchers. As the Tables 14, 15, and 16 show, the differences of C_a calculated by different researchers are very big. The C_a ^{90}Sr is much more variable than the C_a ^{137}Cs and even for various species of *Typha* sp. they differ more than by eight times, of *Potamogeton* sp. more than two times. The database of Urals Research Center for Radiation Medicine provides the research results of 25 aquatic vegetation species from 25 lakes starting from 1964. The fullest studies were conducted on the Uruskul Lake.

The Uruskul Lake is located in the EURT's head and is therefore contaminated to the maximum extent. The first measurements of the radioactive contamination level of water were performed on October 2, 1957. Initially the total β-activity of water was estimated, and the ^{90}Sr activity in water was measured starting from 1962, the ^{137}Cs activity-from 1968. The dynamic pattern of water contamination level is presented by the Fig. 2a. The study of the radioactive contamination levels

Table 14 Average values of C_a ^{137}Cs in the dry biomass of plants EURT lakes

Lake	Location	Type of plant	C_a
Tygish	East coast	*Scirpus* sp.	821 ± 36
Tygish	East coast	*Stratoides aloides*	493 ± 43
Tygish	East coast	*Juncus* sp.	164 ± 14
Tygish	West coast	*Cladofora aegagrophila*	2814 ± 93
Tygish	West coast	*Chara* sp.	2450 ± 121
Tygish	West coast	*Myriophyllum spicatum*	1557 ± 50
Tygish	West coast	*Stratoides aloides*	386 ± 29
Bolshoi Sungul	West coast	*Cladofora aegagrophila*	4627 ± 264
Bolshoi Sungul	West coast	*Scirpus* sp.	236 ± 27
Chervyanoe	West coast	*Cladofora aegagrophila*	3893 ± 336
Chervyanoe	West coast	*Scirpus* sp.	236 ± 27
Chervyanoe	West coast	*Juncus* sp.	497 ± 21
Chervyanoe	South coast	*Cladofora aegagrophila*	1100 ± 57
Shchuchye	Northeast coast	*Scirpus* sp.	612 ± 37
Shchuchye	Northeast coast	*Stratoides aloides*	1219 ± 181

Adopted from Trapeznikov et al. (2007a, b)

Table 15 Average values of C_a ^{137}Cs and ^{90}Sr in the dry biomass of plants EURT lakes and reservoir V-10

Lakes or Reservoir	Type of plant	Years of measurement	^{90}Sr	^{137}Cs
V-10	*Scirpus* sp.	1991–1992	20	30
V-10	*Lemna* sp.	1991–1992	20	100
Kiziltash	*Scirpus* sp.	1982–1984	200	50
Kiziltash	*Fytoplankton*	1982–1984	700	15
Kogakul	*Scirpus* sp.	1982	60	100
Kogakul	*Potamogeton* sp.	1982	60	200
Alabuga	*Scirpus* sp.	1983	30	570
Alabuga	*Potamogeton* sp.	1983	500	280

Adopted from Smagin (2006)

Table 16 Average values of C_a ^{137}Cs and ^{90}Sr in the dry biomass of plants EURT lakes

Types of plant	Bolshoi Igish		Mali Igish		Kuyanish		Travyanoe		Shablish	
	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs
Typha latifolia	187	69	1917	375	797	457	79	150	502	133
Stratoides aloides	680	261	2434	275	2812	1057	935	100		
Potamogeton lucens	1514	308	1745	175	2642	200	2994	12.5	1281	200
Potamogeton filiformis	–	–	–	–	458	686	–	–	521	167
Potamogeton natans	413	446	823	275	–	–	473	212	–	–
Phragmites australis	63.5	69	553	425	133	143	127	187	88.4	233
Typha angustifolia	196	100	428	125	373	143	197	71.2	156	100
Myriophyllum verticillatum	632	831	–	–	903	629	276	400	500	333
Ceratophyllum demersum	467	692	–	–	236	143	364	175	516	700
Hydrocharis morsus-ranae	214	261	4292	2650	194	171	472	200	–	–
Polygonum amphibium	278	385	923	2500	233	200	–	–	286	300
Scirpus lacustris	–	–	–	–	154	114	131	2.5	97.7	133
Elodea canadensis	112	546	1338	225	3524	1514	1636	262	1445	733
The specific activity of radionuclides in water (in Bq L^{-1})	5.9	0.13	0.53	0.04	0.33	0.035	0.67	0.08	0.43	0.03

Adopted from Kostyuchenko et al. (2012)

of the aquatic vegetation of the Uruskul Lake started in 1964. At first, the total β-activity in plant biomass and ^{137}Cs activity in some samples was identified. In 1966 the studies of ^{90}Sr activity started. Any systematic studies of the radioactive contamination level of certain plant species weren't conducted. Therefore it is impossible to trace exactly the dynamics of the biomass specific activity change. The Figure 2b–d illustrates the summary data on the specific β-activity, ^{90}Sr and ^{137}Cs activity in some plant species. There are also data on radionuclides activity in water over the vegetation sampling period.

As is seen from the Figure 2b–d, the fluctuations of the radionuclides specific activity in water and plant biomass are very big. It is difficult to determine the cause of such fluctuations in a stagnant reservoir at the moment. Accordingly, the C_a in particular cases also varies widely (Table 17). Apparently, for hydrophytes in some cases

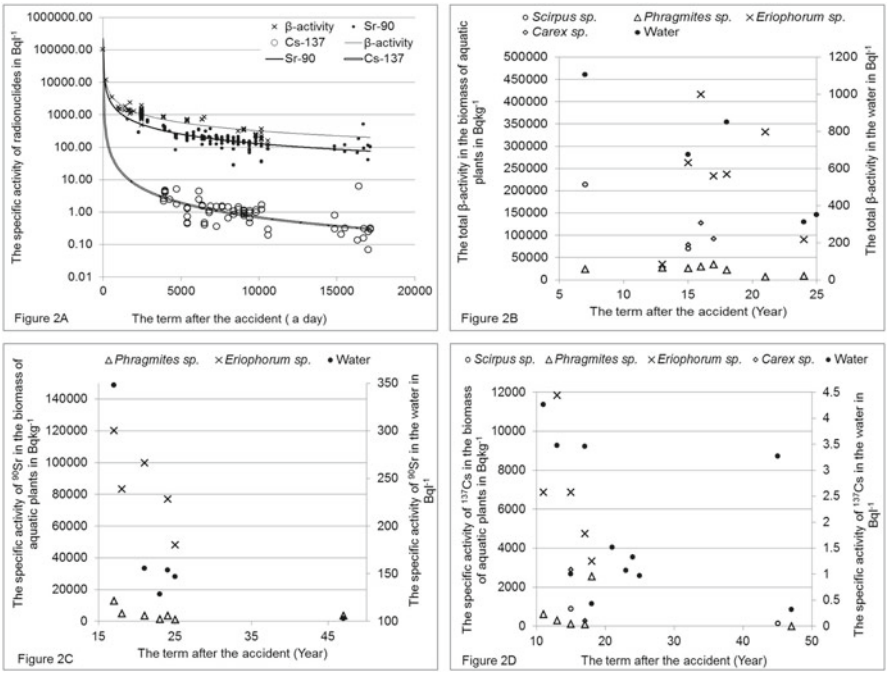

Fig. 2 (**a**) The dynamics of the specific activity of radionuclides in water Lake Uruskul. (**b**) Dynamics of the total β-activity in the water and biomass of aquatic plants in the Lake Uruskul. (**c**) Dynamics of the specific activity of ^{90}Sr in the water and biomass of aquatic plants in the Lake Uruskul. (**d**) Dynamics of the total β-activity in the water and biomass of aquatic plants in the Lake Uruskul

Table 17 Average values of C_a ^{137}Cs and ^{90}Sr in the dry biomass of plants in Lake Uruskul

Year	*Scirpus* sp.		*Phragmites* sp.		*Eriophorum* sp.		*Carex* sp.	
	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	^{90}Sr	^{137}Cs	
1968	–	–	148	–	1613	–	–	
1970	–	–	84	–	3402	–	–	
1972	888	–	130	–	6882	–	–	
1974	–	38	29	346	1374	138	81	
1975	–	26	5981	433	7760	–	–	
1978	–	23	–	643	–	–	–	
1980	–	12	–	–	–	–	–	
1981	–	25	–	501	–	–	–	
1982	–	9	–	328	–	–	–	
2002	46	–	–	–	–	–	–	
2004	–	–	36	–	–	–	–	

the root entry of radionuclides is more important than foliar. It may be assumed that as a result of coastline transgression, the degree of hydrophytes submergence and accordingly, the value of radionuclides entry into a plant through its roots can vary.

It should be pointed out that the accumulation coefficients of ^{137}Cs in the aquatic vegetation are in most cases notably higher than of ^{90}Sr. This differentiates the aquatic vegetation from terrestrial, for which, according to our data, the accumulation coefficients of ^{90}Sr and ^{137}Cs are very close to each other (Kazachonok and Popova 2013). Unfortunately, the species composition of the aquatic vegetation over the sampling period didn't always correspond, which complicates the comparison of the radionuclides accumulation features in different types of aquatic ecosystems. Through the example of *Eriophorum* sp. we can assume that the water body type is of little significance. The ^{137}Cs and ^{90}Sr accumulation coefficients in a watercourse (the Techa River) are virtually identical to the accumulation coefficients in a stagnant reservoir (the Uruskul Lake). The most important factors of radionuclides accumulation are apparently weather conditions, sampling point and time that passed after the water reservoir pollution.

4 Conclusion

1. ^{137}Cs and ^{90}Sr accumulation coefficients in the aquatic vegetation are very variable and can reach several thousands.
2. The accumulation coefficients of ^{137}Cs in the aquatic vegetation are in most cases notably higher than of ^{90}Sr.
3. The hydatophytes and hydrophytes don't differ conceptually in the ability to concentrate radionuclides but in general, rooted plants accumulate significantly less ^{90}Sr and ^{137}Cs than unrooted plants.
4. The entry of radionuclides through the roots and the pollution level of bottom silt apparently don't have any great impact on the C_a of ^{90}Sr and ^{137}Cs in aquatic vegetation. However, it can be assumed that as a result of coastline transgression, the degree of hydrophytes submergence and accordingly, the value of radionuclides entry into a plant through its roots can vary.
5. The type of water reservoir apparently doesn't have any great impact on radionuclides accumulation in plants.
6. The most important factors of radionuclides accumulation are apparently weather conditions, sampling point and time that passed after the water reservoir pollution.

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Radiocesium Phytotoxicity to Single Cell and Higher Plants

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Abstract The chapter presents the results of investigations of transfer, accumulation, translocation of radiocesium and its influence on bioelectrical, vegetative, generative and growth processes in test plants from single cell to whole organism. The aim of our study was to investigate the entering of ^{137}Cs into the hydrophytes and to determine the role of plant cell membrane as a barrier involved in penetration of radionuclides into the cellular compartments. Additionally, we investigated ^{137}Cs effect to the Charophyta *Nitellopsis obtusa* cell membrane bioelectrical properties such as resting potential, K^+ and Cl^- channels activity. This research also determined and compared the toxic effects of ^{137}Cs on seed germination, root (meristematic cells) and shoot (parenchyma cells) growth of *Lepidium sativum* L. as well as the genotoxic impact of this radionuclide to stamen hair cells (generative organ) of *Tradescantia (clone 02)*. The impact of cesium toxicity to plants was evaluated using biological, genetic and electrophysiological methods.

Keywords Toxicity to plants • *Nitellopsis obtusa* • *Lepidium sativum* • *Tradescantia*

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1 Introduction

The start of the twenty-first century is marked by a growing societal concern over human driven impacts on the environment. Widening the scope of radioecology to consider also the radiation protection of non-human biota requires a transition to a truly ecocentric approach in order to address the composite effects of ionizing radiation on man, biota and the ecosystems that they inhabit (Brechignac et al. 2003).

Nowadays a topical radioecological problem is determining the effect of radionuclides on biological objects, especially at low specific activity levels as well as both separately and in conjunction with various environmental factors including the anthropogenic. One of the relatively long-lived radionuclides, and one that most frequently enters environmental systems as a consequence of both nuclear accidents and fallout from nuclear weapons testing, is radiocesium (Povinec et al. 2013; Kumamoto et al. 2015).

Released into natural systems, radiocesium can be transported and fixed by many geological materials, these reactions, eventually, control and reducing damage to ecosystems (Guillaume et al. 2012; Ohno et al. 2012). On the other hand, this radionuclide is highly soluble and can easily be taken up by terrestrial and aquatic organisms due to its chemical characteristics which are similar to those of potassium (Kruyts et al. 2000; Fujimura et al. 2013). Studies of the biological effects of the ionizing radiation of radionuclides are important for the assessment of the radioecological condition of terrestrial and aquatic ecosystems in the surroundings of nuclear power stations, for forecasting the ecotoxicological consequences of radionuclides released into the environment during accidents and for managing the issues of ecological standards in the presence of radionuclides in the environment. The International Union of Radioecology (IUR) points out that in the twenty-first century, special attention should be given to the investigation of the effects of low ionizing radiation doses caused to biota, especially those due to the incorporation of radionuclides into organisms (Brechignac et al. 2003). It is rather difficult to extrapolate experimental data obtained from hydroponically cultured plants which can utilize the soluble (ionic) form of cesium (Soudek et al. 2006) to natural ecosystems (especially terrestrial) because the uptake ability of plants from contaminated soil is extremely small (Kobayashi et al. 2014).

The biological impact of radionuclides depends on their accumulation level and localization in the cells and organism (Gracheva and Korolev 1997; Gudkov 2001). The highest accumulation of ^{137}Cs occurs at the sites in plants with the highest metabolic activity, where cell growth and differentiation is taking place, or in parts actively involved in the transport of water and nutrients (Soudek et al. 2006) and which are relatively sensitive to ionizing radiation. The intensification of the accumulation of ^{137}Cs enhances the internal irradiation doses (Scherhunova et al. 2001). Ionizing radiation can evoke various biological effects in plants, such as the inhibition or activation of DNA synthesis and the synthesis of some metabolites, slackening or acceleration of cell division, growth suppression or stimulation, disturbance of ontogenesis phase dynamics, morphological changes of teratogenic origin, enhanced quantities of cells with increased numbers of chromosomal aberrations, increased number of individuals exhibiting chlorophyll mutations or new traits and reduced seed germination in

progeny generation (Evseeva et al. 2001; Goncharenko and Surkov 2003; Gudkov 2001; Minouflet et al. 2005). Few papers deal with the response of plants to the effect of incorporated ^{137}Cs (Evseeva et al. 2001) and only a few studies have been performed recently on the impact of incorporated ^{137}Cs on plant test-organisms (Butkus et al. 2003; Bystrzejewska-Piotrowska et al. 2005; Korogodina et al. 2004; Lukšienė et al. 2004; Marčiulionienė et al. 2003). After the Chernobyl accident, it was determined that during acute exposure to ionizing radiation, the impact of radionuclides can be two to four times higher (Grodsinsky 2001) in the cell, due to atom decay, than in external irradiation. Radionuclides may enter the inner cell compartments and sometimes bind to the DNA molecule. Genetic effects can be induced by ionizing radiation due to its rather difficult to extrapolate experimental data obtained from hydroponically cultured plants which can utilize the soluble (ionic) form of cesium (Soudek et al. 2006) to natural ecosystems (especially terrestrial) because the uptake ability of plants from contaminated soil is extremely low (Kobayashi et al. 2014), and also radionuclide decay and by transmutation. Transmutation is a change of the chemical nature of decaying atoms and ionizing energy; it affects the site where radioactive decay takes place (Gracheva and Korolev 1997; Marčiulionienė et al. 1992; Gracheva and Shanshiashvili 1983; Nesterov et al. 2001). Internal exposure in plants can increase due to radionuclides accumulated in their tissues, especially in tissues with active cell division (Sokolov et al. 2001; Scherhunova et al. 2001). For example, radiocesium, like its chemical analogue potassium, accumulates in relatively large amounts in both young and meristematic tissues (Gudkov 2001; Tyson et al. 1999). However, the plant response to the action of incorporated ^{137}Cs, particularly at low-level ionizing radiation doses, has not yet been sufficiently investigated (Evseeva et al. 2001).

The aim of the current study was:

1. To determine ^{137}Cs accumulation in compartments of the cells (wall, protoplasm, vacuole) of charophyte algae *Nitellopsis obtusa* (Devs.) J. Groves and in different parts (seeds, roots, shoots) of the plant *Lepidium sativum*.
2. To determine the effect on electrophysiological parameters (rest potential of membrane, its specific resistivity, dynamics of action potential) of the cells of *N. obtusa*.
3. To evaluate the effect of low irradiation doses of ^{137}Cs on vegetative parts of the plant *L. sativum* roots (meristematic cells) and shoots (parenchymal cells) growth – and on the generative organ of *Tradescantia* clone BNL 02 stamen hair cells.

2 Materials and Methods

2.1 Nitellopsis obtusa *as Testing System*

Algae *N. obtusa* collected in different Lithuanian lakes were used for this research. These algae cells have regular cylindrical shape; contain a large central vacuole surrounded by a cytoplasmic membrane the tonoplast. The cytoplasm surrounding the vacuole is separated from the external environment by the plasmalemma and the

cell wall. Experiments relating to ^{137}Cs accumulation in different cell compartments were performed in aquariums containing 1.5 L of filtered lake water with a solution of ^{137}Cs chloride added to reach activity concentration up to 10^2 kBq L^{-1} in six replicates. The water temperature was 24–25 °C. The duration of the experiment was 16 days. The cells of *N. obtusa* were separated manually; because the cells are large (about 21 cm in length and 1.5 mm in diameter) have regular cylindrical shape and pronounced differentiation of compartments. Vacuolar sap was removed by the cutting off one end of the cell. The vacuolar sap usually flows out itself or by a light push of tweezers along the cell. It is important to watch at this moment, that the cytoplasm flow not out together with vacuolar sap. This can be seen in colour and consistency of the preparation. The vacuolar sap is clear and watery; and general fraction of cytoplasm is green and viscous. Further, the cytoplasm was extruded out with tweezers. The admixture of the cytoplasm in the vacuolar sap is not more than 15 %; and the admixture of the residue of the vacuolar sap in the cytoplasm preparation is insignificant. The cell wall was obtained by cutting off both ends of the cell and removing the cytoplasm by strong pressure of the tweezers along the entire cell. The method used for separation of cellular compartments is not very accurate, but has several advantages. It is simple, allows quickly preparing several preparations for measurement, and allows measuring the radioactivity in each compartment of the plant cell (Hampson 1967; Marčiulionienė et al. 2001a, 2015). The ^{137}Cs concentration ratio (CR) was calculated for different cell compartments.

10 cm in length and 1 mm in diameter, cells of *N. obtusa* are ideal material for many physiological observations and manipulations, since they can be considered a single organism-cell maintaining the essential physiological characteristics for a long time following their isolation from the plant and while kept in artificial pond water (APW). Therefore these cells are intensively used for electrophysiology studies and environmental monitoring (Motejuniene et al. 1975; Manusadžianas et al. 1995; Kisnierienė et al. 2009, 2012; Sevriukova et al. 2014a, b). The control solution in the experiments on the electrophysiological properties of *N. obtusa* cells was the APW, containing 0.1 mM KCl, 1 mM NaCl, 0.1 mM $CaCl_2$, 2.5 mM TRIS (tris (hydroxymethyl) aminomethane), adjusted 7.2 pH by HEPES (4-(2-hydroxyethyl) piperazine-1-ethanesulfonic acid) or HCl. Investigated low specific activities 137Cs solutions were prepared from nitratic form in values: 0.005[1], 0.015, 0.03, 0.06 and 0.12 kBq L^{-1}. The experiments were performed at room temperature (20 ± 1 °C) and under daylight conditions (500 ± 10 Lx). The internodal cells were isolated from the neighboring cells and branch lets. The internodes were kept at least overnight in the buffered APW. During the experiments, cells were placed in a Plexiglas chamber and continuously bathed in a flowing solution of APW or test solution at a rate of ~1 mL min^{-1}. As *N. obtusa* is an aquatic plant, the electrophysiological measurement was carried out under their native external condition i.e. in an aquatic solution. The conventional glass-microelectrode technique was used for the registration of the electrical characteristics of internodal cells. The parameters and specifications of experimental techniques are thoroughly described in our previous studies (Kisnierienė et al. 2009, 2012; Sevriukova et al. 2014a, b). The pCLAMP 10 software package was used for data acquisition. All current records, results and analyses are from leak-subtracted data and

normalized to the values obtained under the control conditions. The experiments were performed in current-clamp and voltage-clamp modes. The current-clamp mode was used to record membrane potential (E), while the voltage-clamp mode was used to investigate the potassium and chloride ion transport systems. The clamped region of the cell in the compartment was 0.5 cm long. For the investigation of the transient currents (I), the E of the cells was held at −180 mV (the voltage value close to potassium equilibrium potential, when cell membrane conductance is determined mainly by K^+ transport and electrogenic contribution of metabolic component to membrane potential is negligible). Regarding the outwardly rectifying potassium current, voltage-clamp protocol comprised: holding potential −180 mV; voltage interval for construction of I-V curves [−280 mV ÷ +60 mV]; and electrical stimulation conducted via injection of short (30 ms) rectangular hyperpolarisation or depolarization current pulses every 10 mV step from the holding potential. The value of the peak current when E was in the range from −140 mV ÷ +60 mV was used to evaluate the chloride transport system activity.

2.2 *Lepidium sativum* Bioassay

The test was carried out following by some modificaion (Magone 1989; Montvydienė and Marčiulionienė 2004). In brief, 10 mL of lake water (as control) or a test aqueous solution of ^{137}Cs was pipetted onto three layers of filter paper fitted into a 9-cm glass Petri dish. Twenty-five healthy looking *L. sativum* L. (*Brassicaceae)* seeds of similar size were distributed evenly on the filter paper. The Petri dishes were placed in darkness at 24 ± 1 °C for 48 h afterwards, non-germinated seeds were counted and root lengths were measured. The experimental set of each testing scheme involved three control dishes and three replications for each concentration activity of the radionuclides. The pH of the lake water and test solutions with ^{137}Cs was 7.5. The level of toxic impact on *L. sativum* was assessed by the modified method of Wang (1992). The toxic impact was evaluated according to the percentage of root growth inhibition. The toxic impact of the tested sample solutions on *L. sativum* was classified as very strong (100–60 % inhibition), strong (61–40 % inhibition), moderate (41–20 % inhibition) and weak (lower than 19 % inhibition). The tested concentration was considered non-toxic if the biological parameter of *L. sativum* did not statistically differ from the control, and was considered extremely toxic if the seeds did not germinate. The influence of ^{137}Cs on *L. sativum* shoot growth was tested by growing plants in plastic boxes (100 × 160 × 6 mm) with covers to avoid evaporation. Each box contained 65 mL of water solution and 470 mg (~160) of seeds that were evenly distributed on a glass plate, covered with filter paper. The seeds germinated at 24 ± 1 °C for 24 h in darkness, and the shoots were grown for six days continuous light at a temperature of 23 ± 1 °C. The morphological anatomical studies included all *L. sativum* shoots. The length of each shoot (with straightened leaves) and the weight of all the shoots were measured. A light microscope was used to determine the length, width and area of parenchyma cells.

2.3 Estimation of the RNA-Polymerase II Activity in the Cell Nuclei of L. sativum Shoots

Cell nuclei from the shoots of *L. sativum* plants were isolated using conventional methods (Shevchenko et al. 1999) with modifications (Merkys and Darginavičienė 1997). The isolated nuclei were incubated in a medium suitable to induce RNA-polymerase II activity (Kulaeva et al. 1979; Tautvydas 1971). The incubation medium contained triphosphates GTP, UTP, CTP (all disodium salt, Fluka Chemie AG, Switzerland) and ^{14}C-ATP (0.1 mM, spec. act. 3.1 MBq g^{-1}, Amersham Pharmacia Biotech, UK), Tris (Sigma-Aldrich Chemie GmbH, Steinheim, Germany)-HCl (P.A. Chech Republic) pH 7.6. The enzyme's activity was stopped after 40 min of incubation at +37 °C by adding cooled trichloroacetic acid (final conc. 3 %, Lach:Ner, Neratovice, Czech Republic). The residue was collected on membrane filters (Ø 2.5 μm; Pragopor, PRAGOCHEMA, Czech Republic and washed with trichloroacetic acid and ethanol. A scintillation counter (Beckman LS 1001, USA) was used to measure the activity of incorporated ^{14}C-ATP. The Bradford method (Bradford 1976) was used to determine the protein content.

2.4 Estimation of ^{137}Cs Accumulation and Translocation in Plants

The ^{137}Cs concentration ratio was calculated for different cell compartments in the case of *N. obtusa* and parts of the plant in the case of *L. sativum* using the following formula: $CR = C_1/C_2$ (C_1 is the activity concentration of ^{137}Cs in the respective compartment or part of plant; C_2 is the ^{137}Cs concentration in the tested solution). The translocation factor was calculated using the ratio between ^{137}Cs activity concentration in the shoots and roots of *L. sativum*.

2.5 Tradescantia Clone BNL 02 Bioassay

Experiments with the *Tradescantia* (*Commelinaceae*) clone BNL 02 were performed applying modified methods by Osipova and Shevchenko (1984), as well as Marčiulionienė and co-authors (2003). Four stems of cuttings bearing influorescence were immersed in 200-mL glass flasks containing 150 mL of lake water (as control) or test ^{137}Cs aqueous solution. The flasks were exposed to the 16-h light/8-h dark cycles for 14 days. The genotoxic impact of ^{137}Cs on the *Tradescantia* clone BNL 02 was evaluated by the number of somatic (colorless) mutations and morphological anomalies in the stamen hair (SH), as well as by the amount of non-viable SH (their number indicates lethality when a hair contains less than 12 cells), which were counted using a light microscope. The number of non-viable SH reflects the

ability of cells to divide. In each case, 8000–11200 stamen hairs were counted. The number of somatic mutations in the *Tradescantia* clone BNL 02 stamen hairs in the control (lake water) was 0.62 ± 0.05 %, whereas non-viable SH was not observed. The degree of ^{137}Cs genotoxicity was evaluated according to the methods suggested by Marčiulionienė et al. (1992). A slight genotoxic effect on the *Tradescantia* clone BNL 02 system is observed when the number of somatic mutations and morphologic anomalies do not exceed 1 %, and the ability of stamen hair cells to divide is 100 % (e.g. no non-viable SH). The medium effect was observed when the number of somatic mutations and morphological anomalies was between 1 and 4 % and the ability of SH cells to divide reaches 60 %. A strong genotoxic effect is characterized by the number of somatic mutations and morphologic anomalies exceeding 4 % and the ability of SH cells to divide being less than 60 %.

2.6 *Activity Concentrations and Internal Exposure Doses of ^{137}Cs in Tested Plants*

Investigations into the *N. obtusa* response on ^{137}Cs impact were carried out with working solutions prepared from a standard $^{137}CsNO_3$ solution purchased from the Inspectorate for Ionizing Radiation of Czech Metrology Institute. The activity concentrations of ^{137}Cs (kBq L^{-1}) in tested solutions were as follows: 0.005; 0.015; 0.03; 0.06; 0.12. The ^{137}Cs solutions for testing the impact of ^{137}Cs on *L. sativum* and *Tradescantia* clone BNL 02 were prepared from ^{137}CsCl (Vsesojuznoe objedinenie "Izotop", Leningradskoe otdelenie, FSU). The activity concentrations of ^{137}Cs (kBq L^{-1}) in tested solutions for *L. sativum* were 0.4; 4; 40; 400; and for *Tradescantia* clone BNL 02 were as follows: 0.001; 0.01; 0.125; 1.25. The internal exposure (absorbed) dose to the whole plant (or its part) was calculated according to the methodology suggested by Blaylock et al. (1993):

$$D = 1.6022 \cdot 10^{-13} \sum_i \sum_j \Phi_{ij} \cdot f_{ij} \cdot E_{ij} \int_0^{\tau} C_{0,i}(t)\, dt$$

where: D is the internal exposure α-, β- and γ-radiation dose, Gy; Φ_{ij} is the absorbed fraction of energy E_{ij}; f_{ij} is the yield, $(Bq\ s)^{-1}$, of radiation j per disintegration of the nuclide i; E_{ij} is the energy, MeV, of radiation j for the nuclide i; $C_{0,i}$ is the concentration of the radionuclide i in the organism, Bq kg^{-1} wet weight. Sample preparation for ^{137}Cs measurement was performed in accordance with conventional radioecological methods (Ryndina et al. 1970; Marčiulionienė et al. 1992). ^{137}Cs activity concentrations in the solution and in dry plant biomass were measured by the method of gamma-spectral analysis (Gudelis et al. 2000). The ^{137}Cs activity concentration in the small-volume samples was measured with a gamma spectrometer interfaced with a p-type high purity germanium (HPGe) detector (GWL-series), equipped with a well 40 mm deep and 16 mm in diameter, made by EG&G

ORTEC. The relative efficiency of the detector was 17 % (for ^{137}Cs 661.7 keV radiation). The measurement uncertainty did not exceed 6 %, with a statistical error not larger than 1 % (Gudelis et al. 2000). Internal ^{137}Cs doses were calculated using the method presented in Marčiulionienė et al. (2004).

2.7 Estimation of the External Exposure Doses of ^{137}Cs in L. sativum

For the assessment of the impact of external ^{137}Cs doses, the seeds of *L. sativum* were placed for two days in an irradiation chamber with a ^{137}Cs ionizing radiation source. The chamber (758 mm × 618 mm × 1490 mm) was made of 16 mm steel sheets. The source was placed in a 100 mm thick lead block. The exposure ranged from 0.04 to 5.5 mGy.

2.8 Statistical Analysis

A statistically significant difference between experimental and control samples was assessed by the *t*-test (at $p < 0.05$) using *STATISTICA* Version 7.0 (StatSoft Inc., Tulsa, Oklahoma, USA) software.

3 Results and Discussion

3.1 The Accumulation Process of 137Cs in the Cell of N. obtusa Algae and Effect to the Electrophysiological Properties of these Cells

The experimental data reveals that after 16 days of accumulation, the highest CR of ^{137}Cs occurred at the entirety system of the cell wall and plasmalemma (124 ± 27). The CR values of ^{137}Cs were significantly lower and differed in lesser units in the cytoplasm (14.0 ± 4.1). The CR in vacuolar sap was 2.3 ± 0.3. The obtained data demonstrates that the CR of ^{137}Cs in the cell membranes was nine times higher than in the cytoplasm. In total, 12 % of the ^{137}Cs accumulated in the organism were had accessed the inside of the cells of *N. obtusa* through the cell wall and plasmalemma. The intake of ^{137}Cs through the cell membrane into the cytoplasm may be due to the selective permeability of plasmalemma for K^+ and the ability of protoplasm to concentrate potassium ions whose amount in plasmalemma is 1000 times higher than in lake water (Yu Chitrov and Vorobyov 1971). 14 % of the ^{137}Cs accumulated inside cells are transferred into the vacuole of *N. obtusa* cells through the tonoplast at a ratio 1 g/1 g. However, the vacuole is really large in these cells and its mass is more

than the cytoplasm total mass in the cell. As a result (in total cell tier), it can be argued that inner cytoplasmic membrane is more permeable for ^{137}Cs than the outer cytoplasmic membrane. The analysis of the accumulation levels of ^{137}Cs in the compartments of the *N. obtusa* cells denote that the active transport through outer and inner cytoplasmic membranes is being. Membrane potentials for the K^+ ions, determined mainly by the electrical gradient across the plasma membrane, perform a significant regulatory function in this process (Vorobyov 1979; Rosick et al. 1985).

Investigating the effect of ^{137}Cs on the bioelectrical properties of the plasma membrane of the charophyte cell, we first evaluated how different specific activity affects the steady bioelectrical properties of the plasma membrane, such as membrane resting potential. Examination of the membrane resting potential in *N. obtusa* cells (Fig. 1) showed that differences between the control solution (APW) and the investigated ^{137}Cs concentrations were significant only for concentrations 0.005 and 0.03 kBq L^{-1} (respectively $p=0.0475$ and 0.0499), but differences for all variants of investigated positions are very significant ($p=0.0009$). In all investigated ^{137}Cs specific activity concentrations, the resting potentials were reduced to lower absolute values than compared to the control solution (APW).

Potential −180 mV is the equilibrium potential for the potassium ions channels of cells when the amount of incoming and outgoing K^+ is the same. In lower E than −180 mV values prevails K^+ outgoing current, after prevail incoming K^+ into cell current. Data relating to potassium ions channels for the voltage interval from −60 to −20 mV (Fig. 2) showed that an increase of current impacted all investigated ^{137}Cs specific activities. It has long been known that cesium and potassium are similar in atomic structure. The calculated result for the amount of cesium atoms in the solutions used for experiments are 0.2×10^{-9}, 0.07×10^{-9}, 0.13×10^{-9}, 0.27×10^{-9} and 0.54×10^{-9} mol respectively for concentrations 0.005, 0.015, 0.03, 0.06 and 0.12 kBq L^{-1}. It is such a small amount of cesium that they can be ignored as having

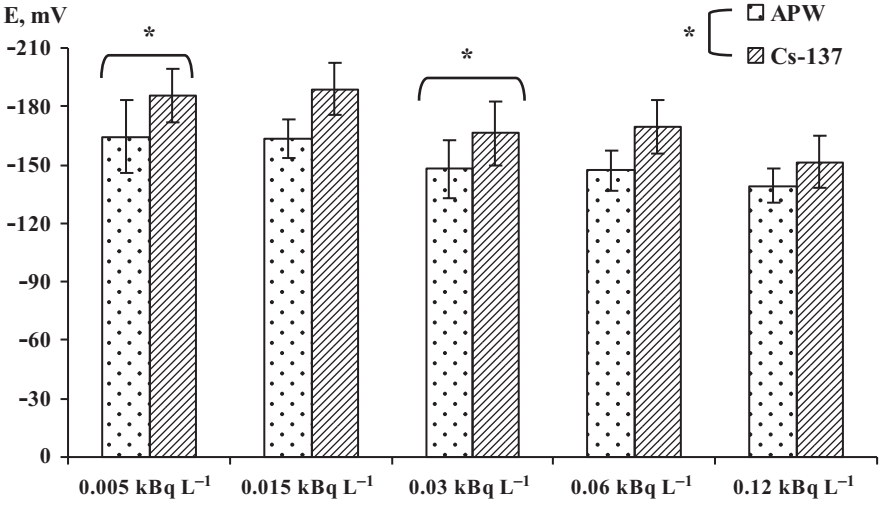

Fig. 1 Effect of different ^{137}Cs specific activity concentrations on the resting potential of *Nitellopsis obtusa* cells (n = 9, *−p < 0.05)

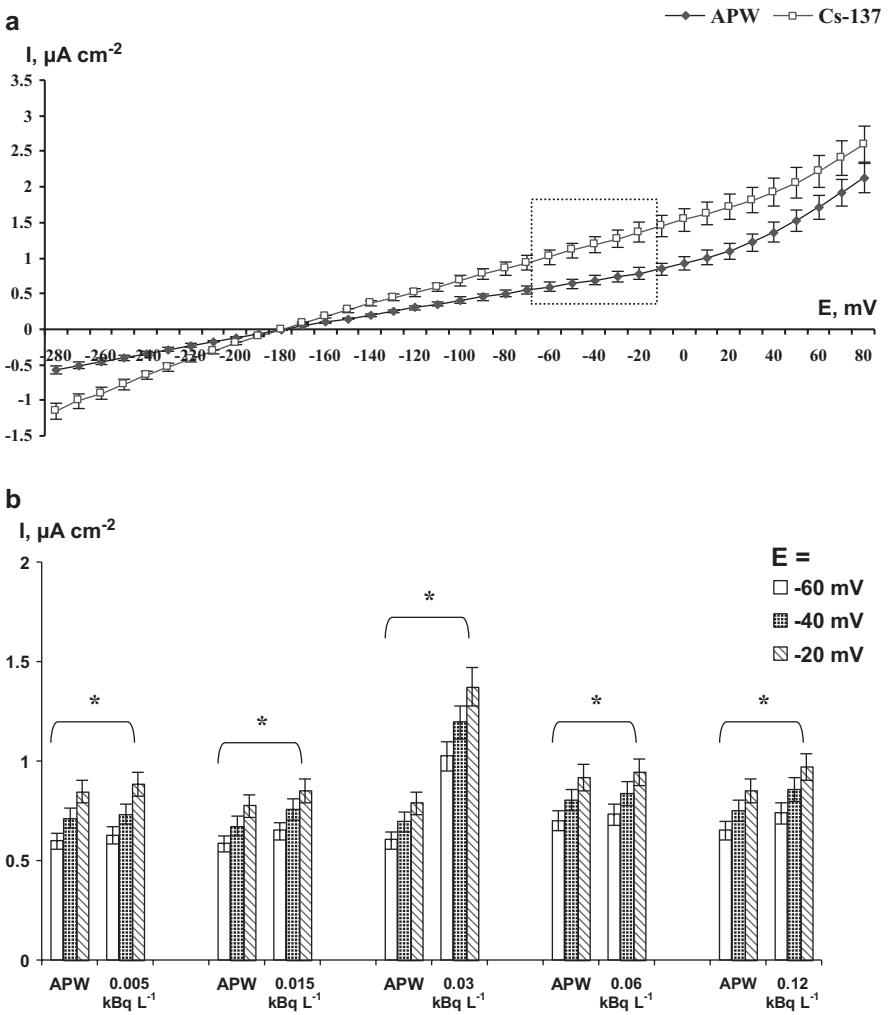

Fig. 2 Effect of ^{137}Cs on the outward on the K^+ transport: (**a**) the K^+ current-voltage relationship for 0.03 kBq L^{-1} specific activity, in square area is interval where the value of the inward K^+ current were evaluated; (**b**) the averaged value of inward K^+ current at E = −60, −40 and −20 mV (APW-control solution, artificial pond water in comparison to investigated ^{137}Cs activity concentrations; all results are presented in values normalized under control condition, n = 9, *–p < 0.05)

a molecular impact on potassium channels in cells. However, it seems that the physical properties of ^{137}Cs in the environment of *N. obtusa* cells significantly increases the K^+ current in the plant cell. It should be noted though that the ΔI between the control solution and investigated concentrations is not directly or inversely proportional to ^{137}Cs specific activities enlargement in the used sample (n = 9).

The range of E from −140 mV ÷ +60 mV was used to evaluate the peak current of chloride in the cell transport system. The magnitudes of the electric current when the outward Cl^- channels in *N. obtusa* cells are maximally permeable are presented

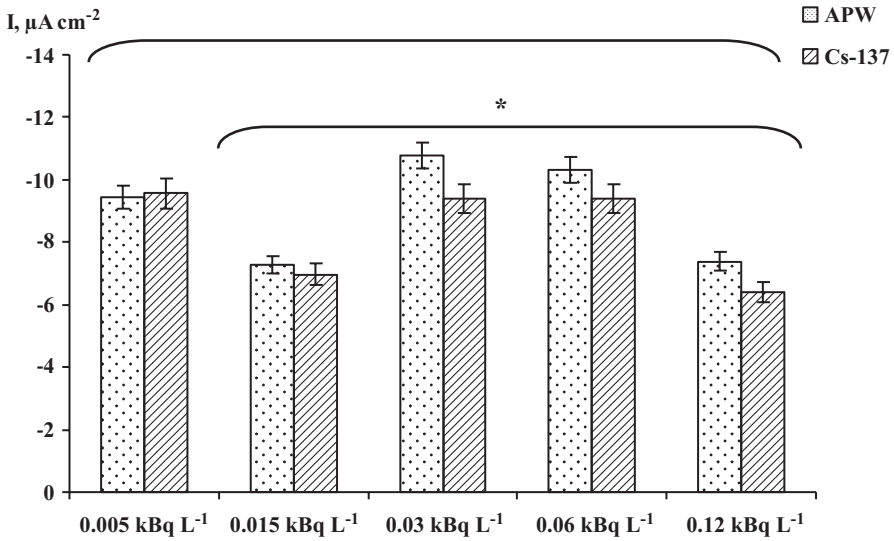

Fig. 3 The magnitude of electric current when outward Cl^- channels in *Nitellopsis obtusa* cells are maximally permeable (APW-control solution, artificial pond water in comparison to investigated ^{137}Cs activity concentrations; all results are presented in values normalized under control condition, n = 9, *–p < 0.05)

in Figure 3. The significance levels are <0.05 for ^{137}Cs specific activities vs. control solutions for concentrations from 0.015 kBq L^{-1}. However, as shown in graphics for the solutions 0.015 kBq L^{-1}, current values correspond in the interval of standard errors, thus it can be argued that the investigated ^{137}Cs solutions impact Cl^- channels in *N. obtusa* cells starting from 0.03 kBq L^{-1} ^{137}Cs specific activity. The effect of stress on the cell is apparent by the fact that the magnitude of the current at which Cl^- channels are maximally permeable decreases.

Tendencies in changes of pattern of bioelectrical response of *N. obtusa,* as well as in transmembrane fluxes of chloride and potassium ions during the action potential, were observed due to exposure of algae cell to ^{137}Cs of comparatively low specific activity. Estimated cesium-induced alterations in electrical activity of the giant alga cell membrane could result in further impairment of electrogenesis, signal transduction and finally cell dysfunction, therefore more extensive electrophysiological investigations into the impact of radionuclides on plant cells are required.

Table 1 Time-dependent accumulation coefficients, translocation factors and distribution of ^{137}Cs in seeds, roots and shoots of *L. sativum* exposed to 40 kBq L^{-1} ^{137}Cs (7 days experiment)

Time	Activity concentration, kBq kg^{-1}			Accumulation coefficient, kg L^{-1}	Translocation factor (roots/shoots)
	Seeds	Roots	Shoots		
5 h	290 ± 20	–	–	7.3	–
1 day	340 ± 20	–	–	8.5	–
2 days	–	500 ± 20	330 ± 40	12.5	0.66
4 days	–	5100 ± 400	1000 ± 70	128.0	0.19
7 days	–	5100 ± 100	1200 ± 80	128.0	0.24

Table 2 The effect of different initial ^{137}Cs activity concentration on seeds germination and root length of *L. sativum* in 2 days experiment

Initial activity concentration, kBq L^{-1}	Activity concentration in root, kBq kg^{-1} (d.w.)	Seed germination, %	Root length, %	Effect
0.4	3.4	96.0 ± 3.6	110.8 ± 2.6	Weak stimulation
4	26	96.7 ± 2.7	111.1 ± 4.5	Weak stimulation
40	340	97.4 ± 2.8	110.7 ± 3.5	Weak stimulation
400	3300	98.0 ± 1.6	111.9 ± 5.3	Weak stimulation

3.2 Accumulation and Impact of ^{137}Cs on *L. sativum*

The study of the dynamics of ^{137}Cs accumulation in seeds, roots and shoots of *L. sativum* was performed on the tested solution where the activity concentration of ^{137}Cs was 40 kBq L^{-1} (Table 1). The highest value of ^{137}Cs activity concentration (5100 kBq kg^{-1}, d.w.) was measured in the roots of the plant (Table 1). The activity concentration of radiocesium in the shoots was 5.1 times lower and in the seeds 15 times lower than in the roots of *L. sativum*. The same tendency of ^{137}Cs accumulation in various plants was noted by other authors (Smolders and Shaw 1995; Zhu et al. 1999; Singh et al. 2009). However, Singh et al. (2009) reported that when the plants were exposed to ^{137}Cs concentrations higher than 10^4 Bq L^{-1}, higher levels of ^{137}Cs were recorded in shoots than roots. The highest value of the CR (0.66 kg L^{-1}) was recorded on the second day of the experiment when the growth of shoots was more intense in comparison to the fourth and seventh day (Table 1). The same tendency was observed in the case of the translocation factor which was higher on the second day when the growth of shoots was more pronounced than on the fourth and seventh days (Table 1). Equilibrium of the ^{137}Cs accumulation in the roots and shoots of *L. sativum* was reached on the fourth day of the experiment (Table 1).

The dependence of the ^{137}Cs accumulation in the roots of *L. sativum* on the activity concentration of that radionuclide in the tested solution was studied on the 2-day experiment. The ^{137}Cs activity concentration in the solution was 0.4, 4, 40 and 400 kBq L^{-1} and the^{137}Cs activity concentration in the roots reached 3.4, 26, 340 and 3300 kBq kg^{-1} d.w. respectively (Table 2). The data revealed a very strong positive

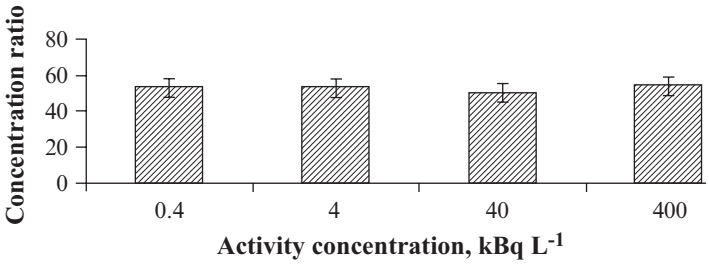

Fig. 4 ^{137}Cs concentration ratio (kg L^{-1}) in roots of *L. sativum* in 2-day experiment

Table 3 Impact of ^{137}Cs on *L. sativum* (7 days experiment) depending on radionuclide activity concentration

Initial activity concentration, kBq L^{-1}	Shoots		Parenchymal cells of shoots		Activity of RNA polymerase II	Effect
	Height, %	Weight, %	Length, %	Width, %		
0.4	98±2	105±3	98±2	100±2	44±4	No effect
4	104±2	103±2	102±2	106±2	66±5	Weak stimulation
40	107±2	115±2	102±2	111±2	57±5	Weak stimulation
400	104±2	110±2	98±2	100±2	34±4	Weak stimulation

correlation between ^{137}Cs activity concentration in the roots of *L. sativum* and ^{137}Cs activity concentration in the tested solution ($r=0.998$, $p<0.000$). However, CR of ^{137}Cs in the roots of *L. sativum* varied only from 50 to 54 and did not depend on ^{137}Cs activity concentration in the tested solution (Fig. 4). The same tendency was noted by other authors (Soudek et al. 2006). Investigations into the impact of different ^{137}Cs activity concentrations (from 0.4 to 400 kBq L^{-1}) on *L. sativum* seed germination and root growth revealed that after two days the seed germination did not statistically differ from the controls (Table 2). However, this radionuclide slightly stimulated (11–12 %) root growth (Table 2).

Analysis of the ^{137}Cs effect on *L. sativum* shoot growth showed that after 7 days of exposure to the solution containing ^{137}Cs, shoot height and particularly weight had been stimulated only by the highest ^{137}Cs concentrations (Table 3). In many cases, shoot parenchyma cell length and width stimulation was observed, which was most pronounced when ^{137}Cs activity concentration was 40 Bq L^{-1} (Table 3). Out of numerous possible indices of cell functional state, RNA-polymerase II activity was selected. It is directly connected with the process of RNA synthesis and, therefore, it is informative of the primary states of protein synthesis in the nucleus. RNA-polymerase II activity was controlled in a model of isolated nuclei in the RNA synthesis system by the amount of tracer ^{14}C-ATP which is dependent on α-aminitine

(mRNA synthesis inhibitor) and in normal growth conditions correlates with cell growth intensity (Darginavičienė et al. 2000).

The model system of the isolated nuclei was formed of the nuclei isolated from cells of *L. sativum* shoots grown in a medium containing ^{137}Cs. All studied ^{137}Cs activity concentrations suppressed the RNA-polymerase II activity (Table 3). It was mostly pronounced at 400 kBq L^{-1} ^{137}Cs activity concentration (by 66 % less than in control). Recalculation of the obtained results for the number of shoots used in the test did not affect the trend observed for this radionuclide. The investigation of the impact of ^{137}Cs on *L. sativum* is widely described in Marčiulionienė et al. (2006a).

The root growth of *L. sativum* was slightly stimulated by all the studied activity concentrations of ^{137}Cs (Table 2). Thus, *L. sativum* root growth dependence on the studied activity concentration of ^{137}Cs was observed, but for seed germination no such dependence was found. Shevchenko et al. (1996) stated that the alterations of meristematic tissues exposed to radiation are related to the morphological alterations of cells, and they can be important for the subsequent plant growth. The reasons for the difference of radiosensitivity of plant meristematic tissues may be related to metabolism disturbances (Evseeva et al. 2001; Shevchenko et al. 1996).

^{137}Cs treatment slightly increased shoot height and weight of *L. sativum,* as well as parenchymal cell length and width (Table 3). The increase was mostly evident when the activity concentration of ^{137}Cs was 40 kBq L^{-1}. At the activity concentration of ^{137}Cs 400 kBq L^{-1}, the increase in shoot height and weight as well as parenchymal cell length and width was much slighter, and this parameter did not statistically differ from the control (Table 3). Naidich (1999) stated that the analysis of morphometric indices which characterize the viability of seedling in the early stages of plant development showed that stimulation of shoot growth by ionizing radiation was observed in parallel with enhanced cytogenetic damages. Consequently, the stimulation of plant growth cannot be considered as useful or at least not a harmful response of plant to the impact of ionizing radiation.

^{137}Cs, at the activity concentrations of 0.4–400 kBq L^{-1}, stimulated the growth of *L. sativum* root and shoots and suppressed RNA-polymerase II activity (Tables 2 and 3). Growth reflects almost all processes that take place in the cell; therefore, for the elucidation of the toxic effects, the internal processes on which growth changes are based are relevant. The effect of ^{137}Cs with respect to cells is stressogenic; therefore, it may influence the processes taking place in the cell nucleus. Studies on RNA-polymerase II activity in isolated nuclei showed that all tested ^{137}Cs activity concentrations (from 0.4 to 400 kBq L^{-1}) inhibited the functioning of RNA-polymerase II. Although ^{137}Cs at test concentrations stimulated plant growth, it suppressed the initial process of protein synthesis that is transcription. The process of enhanced growth in plants can be, by an enhanced functioning of the cell systems, not related to processes controlled by RNA-polymerase II, such as intensified water absorption, stock reserves of seeds and growing cells, etc.

The stimulating effect of radionuclides can cause morphogenetic changes in plants manifested in early developmental stages (Marčiulionienė et al. 1992; Mericle and Mericle 1967). Morphological changes in plants were observed after the Chernobyl NPP accident in the 30 km exclusion zone around the NPP (Grodsinsky 2001). Plant morphological changes due to damaged reproductive organs can also

decrease germination of ripe seeds. It has been found that toxicants at concentrations not exceeding the levels producing toxic effects can stimulate plant metabolism, as well as growth processes in plants and their cells (Adelman et al. 1988; Butkus et al. 2003). Nevertheless, plant enzyme activity can disturbed by metabolic products and the degree of injuries depends on the intensity of metabolism (Adelman et al. 1988; Britt 1996). Gerask'in et al. (2000) noted that the storage and reprocessing of low and intermediate activity waste caused additional environmental contamination, which induced cytogenetic disturbances of both vegetative and reproductive organs in *Pinus sylvestris* L. In fact, exposure to ionizing radiation causes some alterations in plants populations such as chromosomal aberrations, visible mutations, biochemical mutations, changes in genetic structure of population, extinction of sensitive species and, ultimately, degradation of the ecosystem (Shevchenko et al. 1996).

3.3 *Accumulation and Impact of ^{137}Cs on the Tradescantia Clone BNL 02*

It is known that tissues of plant generative organs are more sensitive to the impact of ionizing radiation than the tissues of vegetative organs (Evseeva et al. 2001; Shevchenko et al. 1996). Cytogenetic effects are among the well-defined criteria for estimating the impact of ionizing radiation upon biota (Shevchenko et al. 1996; Gerask'in et al. 2000). The present investigation of the genotoxic impact of the radionuclides on *Tradescantia* showed that on the 14th day of treatment, the lowest activity concentration of ^{137}Cs caused 1.4 % of somatic mutations and morphological anomalies and decreased the ability of cell division by 19 % (Table 4). An increase in the activity concentration of ^{137}Cs from 0.001 to 1.3 kBq L^{-1} caused a decrease in the cell division ability of stamen hair cells of *Tradescantia*, whereas the number of somatic mutations and morphological anomalies showed no significant change (Table 4). Similar results were obtained by other authors who studied radionuclide impact on test-organisms (Lamb et al. 1995). The impact of the studied ^{137}Cs activity concentrations on *L. sativum* root meristematic cells and shoot parenchyma cells, on the activity of RNA-polymerase II and on the *Tradescantia* SH system can be explained by the different metabolism of these radionuclides in the plants. The transport pathway and distribution of ^{137}Cs in plants are similar to that of the stable chemical analogue potassium (Bradford 1976; Evseeva et al. 2001;

Table 4 ^{137}Cs activity concentration in aqueous solution and stems of *Tradescantia* clone BNL 02 and the effect of this radionuclide

Initial activity concentration, kBq L^{-1}	Activity concentration in plant, kBq kg^{-1} d.w.	Somatic mutations, %	Non-viable stamen hairs, %	Effect
0.001	0.1	1.4 ± 0.1	19 ± 1	Moderate
0.01	1.3	1.2 ± 0.4	27 ± 1	Strong
0.13	–	1.6 ± 0.1	79 ± 5	Strong
1.3	–	1.6 ± 0.1	87 ± 4	Strong

White and Broadley 2000; Tagami et al. 2012). The physiological similarities of Cs and K are frequently indicated in radioecological studies (Bradford 1976; White and Broadley 2000; Zhu et al. 2000). ^{137}Cs in the plant cell are localized evenly in cell protoplasm (Grodsinsky et al. 1991; Marčiulionienė 1994). ^{137}Cs in plants accumulates mostly in the zones of cell division and active metabolism (e.g., in plant meristematic and young tissues) (Evseeva et al. 2001). The distribution of ^{137}Cs activity concentration in the same plant species was different. The activity concentration of this radionuclide in some species was higher in the aboveground part of the plant, while in other species it was higher in the underground part of the plant (Marčiulionienė et al. 2001b). Comparison of the ^{137}Cs activity concentration in *L. sativum* roots and aboveground parts demonstrated that after seven days, the ^{137}Cs activity concentration was five times higher in the roots than in the aboveground part of plant when *L. sativum* was cultivated hydroponically with, during the experiment, the roots incubated in a lake water solution of 40 kBq L^{-1} ^{137}Cs activity concentration (Marčiulionienė et al. 2005). The physiological similarity of cesium and potassium is frequently indicated in radioecological and physiological studies (Evseeva et al. 2001). However, in several studies (Evseeva et al. 2001; Urban and Bystrzejewska-Piotrowska 2003), differences between the effects of cesium and potassium on some physiological parameters of *L. sativum* were determined. These distinctions could be due to the much higher atomic weight and ionic radius of cesium than of potassium (Evseeva et al. 2001).

3.4 *Effects of Internal and External Doses of ^{137}Cs on L. sativum and the Tradescantia Clone BNL 02*

Regardless of the dose (from 0.0007 to 0.7 mGy) of a 2-day exposure to internal ^{137}Cs ionizing radiation, *L. sativum* seed germination did not significantly differ from the control, whereas root growth was statistically significantly higher (11–12 %) (Tables 2 and 5). The effect of external^{137}Cs gamma-irradiation on seed germination of *L. sativum* was also insignificant when compared to the control, irrespective of the dose (0.4–5.5 mGy). Roots, however, were 24–33 % longer than

Table 5 ^{137}Cs initial activity concentrations and internal exposure doses of ^{137}Cs in *L. sativum* and *Tradescantia* clone BNL02

L. sativum		*Tradescantia* clone BNL 02	
Initial activity concentration, kBq L^{-1}	Internal exposure dose (2 days experiment), mGy	Initial activity concentration, kBq L^{-1}	Internal exposure dose (14 days experiment), mGy
0.4	0.0007	0.001	0.0003
4	0.006	0.01	0.004
40	0.07	0.125	0.05
400	0.7	1.25	0.5

in control plants (Marčiulionienė et al. 2006b). Non-linear and non-monotonic dose-effect dependence was observed within the studies of the effect of low external doses (6×10^{-4} to 1.2 Gy) on animal biophysical and biochemical properties (Burlakova et al. 1999), as well as in studies of genetic effects in the meristematic cells of *Hordeum vulgare* L. leaves induced by 4–10 cGy irradiation doses (Gerask'in et al. 1996). This diversity in dose-effect dependences due to low irradiation doses has been explained (Burlakova et al. 1999) as a change in the ratio between genetic damage and repair. According to Burlakova et al. (1999), repair systems are not activated at low doses, as it takes longer for them to activate.

After 14 days of exposure, internal doses (0.0003–0.5 mGy) of ^{137}Cs caused 1.2–1.6 % of somatic mutations and yielded 19–87 % of non-viable stamen hairs in *Tradescantia* (Marčiulionienė et al. 2006b). This indicates an inhibition of stamen hair cell reproducibility. Shevchenko and Pomeranceva (1985) conclude that the one percent of somatic mutations induced in *Tradescantia* stamen hair show that genotoxic alterations could cause the disappearance of a species sensitive to ionizing radiation as well as changes in the whole ecosystem. Our data show dependence between the amount of non-viable stamen hairs of *Tradescantia* and low-level exposure to ^{137}Cs, but this dependence was not established for somatic mutations. Similar results were obtained by investigating the impact of low-level radiation doses on test organisms (Lamb et al. 1995). Shevchenko and Pomeranceva (1985) noted direct dose-effect dependence on *Tradescantia* when this plant was exposed to external irradiation of 2 Gy and higher.

The toxic effect of ^{137}Cs internal exposure on meristematic cells and the genotoxic effect on the stamen hair calls can be explained by different radionuclide accumulation in plants and different distribution in plant tissues or cells. It is known that ^{137}Cs, as K analogue, mainly accumulates in the areas of cell division and active metabolism in parenchyma and young tissues. Cellular protoplasm is the site where the largest amount of this radionuclide accumulates (Marčiulionienė 1994; Grodsinsky et al. 1991).

The growth of plant cells involves three different processes: cell division, formation of protoplasm and cell elongation. After the protoplasm has stopped growing, a cell can grow in length 10–50 times the initial size. Low ionizing radiation doses can slow down cell division (Sidorov 1990), which means that it can lead to a more intensive root cell elongation and therefore a longer root.

It is known that radionuclides can stimulate morphogenetic changes that manifest in the early development stages (Marčiulionienė et al. 1992; Mericle and Mericle 1967). Morphological changes in plants were observed after the Chernobyl accident in a 30 km radius from the NPP (Grodsinsky 2001). Using the Scot's pine tree (*P. sylvestris* L.) as bioindicators, it was determined that the storage and reprocessing of low and intermediate activity waste was connected with additional environmental contamination which induced cytogenetic disturbances of both the vegetative and reproductive organs of the pine tree (Gerask'in et al. 2000). Reduced germination of matured seeds was due to damage of the plant's reproductive organs. It was determined that toxicants stimulated plant metabolism and growth. However, the plant enzyme activity can be disturbed by metabolic products, and the more

intensive is the metabolism the higher is the degree of such damage (Adelman et al. 1988). Therefore, the stimulating effect of the investigated internal (0.0007–0.7 mGy) and external (0.04–5.5 mGy) ^{137}Cs exposure on the plant root negatively influences further plant development.

4 Conclusions

^{137}Cs accumulation, time-dependent, activity concentration-dependent and internal/external exposure dose-dependent studies using terrestrial plants (*L. sativum* and *Tradescantia*) and investigations of the electrophysiological parameters of fresh water algae *N. obtusa* have been performed.

The investigation into the ^{137}Cs accumulation process in the plant cell showed that the inner cytoplasmic membrane is more permeable to ^{137}Cs than the outer cytoplasmic membrane, regardless that their structures are similar. Addition of ^{137}Cs into the environment even in small quantities changed the resting potential of plant cells. Furthermore, exposure of *N. obtusa* cells to ^{137}Cs of activity concentrations from 0.005 to 0.12 kBq L^{-1} increased K^+ current through cytoplasmic membrane. Application of ^{137}Cs of very small activity concentration did not evoke alterations in Cl^- outward, but starting from activity concentration of 0.03 kBq L^{-1}, cesium significantly decreased magnitude of chloride efflux during electrical excitation of the cell membrane.

Experimental data showed that the most pronounced ^{137}Cs translocation from roots to shoots of terrestrial plants can be observed in the period of the very beginning of the shoots development. In the case of *L. sativum* it was observed on the second day of plant growth. ^{137}Cs accumulation equilibrium roots/shoots was reached on the fourth day of the *L. sativum* growth. By extrapolating the results of this investigation to the other terrestrial plants, it can be stated that the ^{137}Cs accumulation equilibrium roots/shoots is reached at the stage of the stable growth of plant.

According to the activity concentration-dependent experiments, the impact of accumulated ^{137}Cs from 0.4 to 400 kBq L^{-1} solution, which corresponds to internal doses of 0.0007–0.7 mGy in *L. sativum*, was similar and can be attributed to the impact of slight stimulation. Low external ^{137}Cs exposure (from 0.04 to 5.5 mGy) stimulated the root growth of *L. sativum* as well.

The impact of accumulated ^{137}Cs from 0.001–1.3 kBq L^{-1} solution, corresponding to internal exposure doses of 0.0003–0.5 mGy in *Tradescantia* caused a strong genotoxic effect in the stamen hair system. Accumulated ^{137}Cs reduced the viability of stamen hairs of *Tradescantia* and increased the amount of somatic mutations in them in a dose dependent manner. A large set of data from model experiments has shown that test-plants *N. obtusa*, *L. sativum* and *Tradescantia* are of great significance when studying the impact of ^{137}Cs on specific parameters of these plants.

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Sorbents for Radiocaesium Removal from Natural Water and Soil

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Abstract Sources of contamination of natural water and soil by caesium radionuclides are discussed. Possible levels of radioactive contamination conditioned by various sources accidents are evaluated. The nature of sorption behavior of natural, granulated and modified aluminosilicates is described; a comparison of their physicochemical characteristics and sorption behavior with respect to caesium is also given. It is shown that the sorption characteristics of the natural aluminosilicates depend on their type and deposit; caesium distribution coefficients for the natural aluminosilicates do not exceed 10^3–10^4 L kg^{-1}. Granulated and modified aluminosilicates possess rather better mechanical strength than respective natural ones. Modification of natural aluminosilicates by transition metals ferrocyanides results in increase of both caesium distribution coefficients and capacity up to 10^7 L kg^{-1} and 500 mg g^{-1} respectively. The possibility of granulated and modified aluminosilicates use for radiocaesium removal from natural waters and soils is shown. Decontamination of the natural water from ^{137}Cs by the granulated glauconite under dynamic conditions has shown that the column resource was ≈7000 bed volumes and decontamination factors were at least 100. The highest values of caesium transfer decrease factors from the hydroponics solution to agricultural plants (80 ± 15) were obtained after joint addition of the modified glauconite and a potassium fertilizer into the hydroponics solution.

Keywords Radioactive contamination • Decontamination • Sorbents • Natural waters • Soils deactivation • Rehabilitation • Aluminosilicates • Ferrocyanides

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1 Introduction

Contamination of natural waters and soils by caesium radionuclides occurred as a result of worldwide dispersion after nuclear tests, radiation accidents as well as radionuclides release into the atmosphere and anthropogenic water bodies from enterprises of the nuclear fuel cycle. Nuclear explosions made the major contribution to radioactive contamination of the environment. Thus, the total release of ^{137}Cs into the atmosphere as a result of atmospheric nuclear tests is evaluated to be 910 PBq in contrast to approximately 40 PBq of ^{137}Cs released as a result of irradiated nuclear fuel reprocessing (Myasoyedov 1997). Sapozhnikov et al. (2006) reported that approximately 120 PBq of ^{134}Cs and 83 PBq of ^{137}Cs were released into the environment because of Chernobyl disaster (USSR/Ukraine). In accordance with the assessment of Japan Atomic Energy Agency (JAEA) and the Nuclear Safety Commission of Japan, the Fukushima disaster (Japan) resulted in the release of approximately 11 PBq ^{137}Cs and ^{134}Cs radionuclides. The explosion of the tank with high-level radioactive waste in 1957 at the Mayak PA (Russia) resulted in the release of 74 PBq of radionuclides (Sakharov 2006); contribution of ^{137}Cs to total radioactivity was 0.35 %, i.e. 0.26 PBq (Batorshin and Mokrov 2013). After nuclear explosion, relatively long-lived radionuclides such as ^{137}Cs appear in the stratosphere and transfer through the world during decades resulting in global fallouts. Due to wet and dry fallouts, irregular radionuclide transfer to the earth occurred leading to an increased radioactive contamination in certain zones. For example, after nuclear tests, maximum fallout was at 40–50° northern latitude (Sapozhnikov et al. 2006).

The atmosphere may become a medium for radionuclides transfer to soils and water bodies. Usually, rivers, seas and coastal waters are contaminated more intensive than Open Ocean. The average activity of the Ocean is approximately 1–2 Bq m^{-3}, whereas seas and coastal waters contain 5–20 Bq m^{-3}. A higher level of radioactive contamination may be observed in rivers, lakes and soils if radioactively contaminated water was discharged there or a radiation accident was somewhere near. After the Chernobyl disaster, the contamination level in the 30 km zone exceeded 1500 kBq of ^{137}Cs per square meter (Sapozhnikov et al. 2006). Outside former USSR, the highest levels of ^{137}Cs and ^{134}Cs were observed in Sweden being approximately 100 kBq m^{-2} (Warner and Harrison 1993). Extended areas in Ukraine and Belarus were contaminated stronger than 40 kBq m^{-2}. At some ‘hot points’ in Belarus (near Gomel, 140 km far from the Chernobyl power plant) radioactivity level exceeded 1500 kBq m^{-2} (Warner and Harrison 1993). Bogdevich and Podolyak (2006) reported that in 25 years after the accident the major part of ^{137}Cs (90 %) was located in the upper 25-cm layer of a soil being available for plants.

Approximately 22 % of ^{137}Cs was transferred up to 400 km from Fukushima-1 nuclear power plant (Japan) after the accident (Katsumi 2012). Activity of ^{134}Cs and ^{137}Cs at the most contaminated areas varied from 10 to 1×10^4 kBq m^{-2} (Endo et al. 2012). Activity of soils ranged from 0.1 to 380 kBq kg^{-1} (Endo et al. 2012). Radionuclides may be transferred from a soil to vegetation through a soil solution; for example, Kato et al. (2012) reported that levels of plants contamination in Japan

after the accident were 68–968 kBq kg^{-1}. Thus, a high level of local radioactive contamination of natural waters and soils result in the necessity of deactivation activities, especially in cases of rehabilitation of lands contaminated because of radiation accidents. Sorption method may be used for decontamination of soils and natural water with various salt contents.

2 Sorbents for Decontamination of Natural Media Contaminated by Caesium

Sorbents used for decontamination of radioactively contaminated natural media should be compatible with the environment. They should not negatively affect the environment and should not release hazardous substances more than it is allowed. Large volumes and a complex salt composition are typical for radioactively contaminated natural waters and soil solutions. Therefore, sorbents used for their decontamination should be cheap, effective and 'eco-friendly' and at the same time, they should possess high sorption capacity and selectivity for radionuclides. Sometimes synthetic sorbents are used for decontamination of natural water, especially as sorption filters for drinking water. However, natural sorbent or modified sorbents based on natural minerals are very widely used for these applications. Natural aluminosilicates are used for caesium and strontium separation from aqueous solutions (Vdovina et al. 1976; Sultanov et al. 1976; Chernyavskaya et al. 1983; Tarasevich 1996; Borai et al. 2009; Misaelides 2011). Natural aluminosilicates are quite cheap; however, they possess low sorption capacity, reversible sorption and weak mechanical characteristics. These disadvantages may be decreased due to granulation and physicochemical modification of the natural aluminosilicates. The nature of sorption behavior of natural, granulated and modified aluminosilicates is described in the chapter; a comparison of their physicochemical characteristics and sorption behavior with respect to caesium radionuclides is also given below.

2.1 Natural Aluminosilicates

2.1.1 Structure of Aluminosilicates and Nature of their Sorption Behavior

The silicate tetrahedron $(SiO_4)^{4-}$ containing four oxygen atoms in the vertexes and one silicon atom in the center, is the main element of an aluminosilicate structure. A part of Si^{4+} is replaced by Al^{3+} ions in the crystal lattice. The negative overcharge of this system is compensated by either addition of H^+ ions with further formation of OH^- groups or adsorption of alkaline or alkaline earth cations such as Na^+, K^+, Ca^{2+}, Mg^{2+} from natural water (Kuznetsov et al. 1974). Adsorbed hydrated cations are located in interstices of the crystal lattice. Since these ions are not a part of the crystal lattice, they can be exchanged to other cations after contact with water

solutions. A deeper process of Al^{3+} ions replacing by iron or magnesium ions may be occurred in aluminosilicates. This results in a complex and non-stable chemical composition of aluminosilicates being different for aluminosilicates from various deposits. A deeper Al^{3+} ions replacement requires additional compensation of the tetrahedron overcharge by additional adsorptions of cations from a solution, resulting in the increase of exchange capacity of the aluminosilicate. Therefore, exchange capacity of natural aluminosilicates depends on Al/Si ratio as well as on content of exchangeable ions such as Na^+, K^+, Ca^{2+} and Mg^{2+}. Stability of aluminosilicates increases with the increase of Si/Al ratio (Tarasevich 1981). Besides the degree of ions replacement and Al/Si ratio, the structure of aluminosilicates interstices and channels as well as the ratio between their size and ionic radius affect exchange characteristics of the aluminosilicate. Exchangeability of cations depends on the location of the cation in the crystal lattice where the exchange process occurs. Some cations are bound stronger than other ones. In certain cases, cations compensating overcharge, occurred as a result of the replacement in the tetrahedron, cannot be changed to another ion; the example is non-exchangeable potassium ion in micas (Tarasevich 1981).

As a rule, natural aluminosilicates possess cation exchange features. These features are conditioned by following causes:

1. *Exchange of hydrated Na^+, K^+, Ca^{2+} and Mg^{2+} ions located in the tetrahedral interstices of the aluminosilicate carcass*: This ion exchange is a selective process; the selectivity is conditioned by steric factors. Subject to the crystal structure of the aluminosilicate, the size of the entrance of the tetrahedral interstice varies from 0.4 to 1.1 nm that is very similar to the ionic radius. Therefore, aluminosilicates can adsorb only ions and molecules with size less than the entrance of the tetrahedral interstice. That's why these ion exchangers are called 'molecular sieves'.
2. *Disarrangement of chemical bonds around the edges of the silicate tetrahedrons as a result of destruction of the crystal lattice*: The destruction of the crystal lattice may occur because of the mineral grinding. Lateral surfaces of the crystal have unsaturated valences that may be compensated by ions adsorption from a solution.
3. *Exchange of hydrogen ions in silanol groups Si–OH by a cation*: Some hydroxyl groups may become uncovered as a result of destruction of the crystal lattice. Hydrogen ions in these hydroxyl groups may be replaced by a cation from the solution. This effect explains the exchange capacity of some clay minerals (Kuznetsov et al. 1974).

Sorption sites of natural aluminosilicates may be either selective or nonselective. Selective sorption is conditioned by the fact that the size of caesium ion is similar to the diameter of the entrance of the aluminosilicate tetrahedron. The effective diameter of hydrated caesium ion is 0.3 nm (Peters et al. 1974). Popov et al. (2011) reported that wedge-shaped lateral locations at the lateral cleavages of microcrystals are selective sorption sites. Sorption sites on the basal facets of schistose silicates also show relatively high selectivity for caesium ions. This selectivity is conditioned

by commensurability caesium ion radius and the size of tetrahedral interstice in the interlayer space of the mineral ("trap effect") (Kobets et al. 2014). As Popov et al. (2011) reported hydroxyl groups on the lateral facets of tetrahedrons are nonselective sorption sites. Dissociation of these hydroxyl groups increases within increase of pH value of the solution. Contribution of selective sorption sited to total cation exchange capacity of aluminosilicates is not significant; therefore, reversible sorption of cations by aluminosilicates is usually observed. For example, in the case of schistose silicates approximately 1–3 % of total exchange capacity is conditioned by selective sorption sites; whereas, 30–50 % of capacity is conditioned by nonselective sorption sites on the lateral facets (Kornilovich et al. 2000). Tectosilicates (zeolites) possess rigid crystal structure; they cannot swell or compress after dehydration and almost completely save parameters of their crystal lattice after ion exchange. Due to a weak swell of zeolites, exchangeable ions located in pores of the aluminosilicate possess low mobility. Exchangeable ions and water molecules are located in large intracrystalline interstices and channels of the crystal structure of the aluminosilicate. Heating of a zeolite results in water elimination; free dehydrated interstices may be occupied by other molecules or ions.

In contrast to tectosilicates, fibrous and schistose aluminosilicates are inclined to swell. The parameters of the crystal lattice change because of ions sorption; this makes aluminosilicates to be similar to organic ion exchange resins. After swelling, the structure of layers remains the same; however, the distance between layers changes because of molecules incorporation. Schistose aluminosilicates swelling depends on quantity and nature of exchangeable cations located between layers. For example, kaolin cannot swell because it does not adsorb water into the interlayer space. Weak mechanical characteristics and low stability in acidic solutions are the main disadvantages of natural aluminosilicates. Aluminum elimination from the aluminosilicate carcass may occur in acidic solution resulting in the decrease of mechanical strength and sorption characteristics. Stability of aluminosilicates increases with the increase of Si/Al, i.e. with the decrease of sorption capacity, which is conditioned by aluminum content. The range of pH 5–8 is the optimal for aluminosilicates use. Mordenite and clinoptilolite are relatively stable in acidic solutions.

2.1.2 Sorption Characteristics of the Natural Aluminosilicates with Respect to Caesium

Sorption capacity of natural aluminosilicates is different for various types of minerals and even for the same mineral from various deposits. Static exchange capacity of the glauconite from Karinskoye deposit (Russia) for caesium is 0.11 mmol g^{-1} (Voronina et al. 2015a). Clinoptilolites possess the highest capacity among aluminosilicates. Sorption capacities of clinoptilolites from various deposits are: 0.92 and 0.79 mmol g^{-1} for Dzegvi deposit, Georgia (Chernyavskaya et al. 1983; Voronina et al. 2015a), 1.4 mmol g^{-1} for Dzegvi-Tedzami deposit, Georgia (Bogdanovich et al. 1998), 1.33 mmol g^{-1} for Hekordzula deposit, Georgia (Milyutin et al. 1993), 2.3 mmol g^{-1} for Holinskoye deposit, Russia, 0.16 mmol g^{-1} for Shivertooyskoye

deposit, Russia (Voronina et al. 2015a). Thermal treatment of the clinoptilolite results in the decrease of its exchange capacity (Tsitsishvili et al. 1985). Caesium distribution coefficients (K_d) for some natural aluminosilicates are presented in Table 1. As it might be seen in Table 1, caesium distribution coefficients vary for aluminosilicates from different deposits and depend on the aluminosilicate pretreatment and the composition of the solution. Nevertheless, caesium distribution coefficients do not exceed 10^3–10^4 L kg^{-1}. Clinoptilolite and bentonite from certain deposits show a higher selectivity for caesium. Increase of salt content in the solution results in the decrease of caesium distribution coefficient testifying the low selectivity of natural aluminosilicates. Thermal treatment of the bentonite results in the increase of caesium distribution coefficients. Elimination of adsorbed and capillary water due to heating probably results in the increase of pores quantity and sorption sites unlocking. Increase of the pH value of the solution also results in the increase of caesium distribution coefficients because this promotes dissociation of Si-OH groups and destruction of caesium hydrate environment, making easier Cs^+ cation location in the aluminosilicate structure.

2.2 Granulated Aluminosilicates

Low mechanical strength is typical for natural aluminosilicates; therefore, it is difficult to use them in sorption columns. The glauconite (a schistose aluminosilicate) is a typical example of an aluminosilicate with low mechanical strength. There are rich natural reserves of the glauconite; it is produced in industrial scale since it possesses good radiation stability and satisfactory sorption characteristics. However, low mechanical strength of the glauconite restricts its use as a sorbent. Granulation may be used for improvement of mechanical characteristics of natural aluminosilicates including glauconite. The extrusion method of natural aluminosilicates granulation using hydroxide of a polyvalent metal as a binding material was developed (Betenekov et al. 2009).

2.2.1 Synthesis and Mechanical Characteristics

The glauconite concentrate produced at Karinskoye deposit (Chelyabinsk region, Russia) was used for granulation. Content of the glauconite in the concentrate was 89–96 %, the remainder was mainly quartz. The glauconite concentrate was previously grinded to the size of less than 40 μm (90 % of particles). The method of granulation included three main steps: pretreatment of raw material, extrusion and thermal treatment. The pretreatment was performed in order to obtain a homogeneous and flexible paste. Sol of zirconium hydroxide was used as a binding material. When sol of zirconium hydroxide is transformed to a gel, it binds particles of grinded glauconite keeping its pore structure unchanged. Correct ratio between the aluminosilicate and the binding agent allows obtaining homogeneous paste with the

Table 1 Caesium distribution coefficients (K_d) for natural aluminosilicates

Aluminosilicate	Deposit	Pretreatment	Composition of the solution, pH	K_d, L kg^{-1}	References
Clinoptilolite	Dzegvi (Georgia)		0.1 mol L^{-1} $NaNO_3$	1.2×10^3	Milyutin et al. (1993)
			1.0 mol L^{-1} $NaNO_3$	610	
			0.01 mol L^{-1} $Ca(NO_3)_2$	1.4×10^4	
	Dzegvi-Tedzami (Georgia)		pH 13.7, salinity 1–10 g L^{-1}, Cs concentration: ≤0.02 g L^{-1}	5×10^3	Bogdanovich et al. (1998)
			0.2–1.0 g L^{-1}	2×10^3	
			5.0–10 g L^{-1}	5×10^2	
	Dzegvi-Tedzami (Georgia)		pH 8: $NaNO_3$ 1 g L^{-1}	600	
			$NaNO_3$ 100 g L^{-1}	74	
	Dzegvi-Tedzami (Georgia)	Na-form	pH 8: $NaNO_3$ 1 g Lg L^{-1}	2.7×10^3	
			$NaNO_3$ 10 g Lg L^{-1}	0.9×10^3	
			$NaNO_3$ 100 g L^{-1}	0.1×10^3	
	Dzegvi-Tedzami (Georgia)	Na-form	pH 12: $NaNO_3$ 1 g L^{-1}	4.8×10^3	
			$NaNO_3$ 10 g L^{-1}	1.8×10^3	
			$NaNO_3$ 100 g L^{-1}	2.0×10^3	
	Dzegvi (Georgia)		Drinking water (tap water), pH 8.0	3.9×10^4	Voronina et al. (2015a)
	Holinskoye (Russia)		Drinking water (tap water), pH 8.0	1.3×10^4	
	Shivertooyskoye (Russia)		Drinking water (tap water), pH 8.0	2.5×10^4	
	Shivertooyskoye (Russia)		0.1 mol L^{-1} $NaNO_3$	1.2×10^3	Milyutin et al. (1993)
			1.0 mol L^{-1} $NaNO_3$	120	
			0.01 mol L^{-1} $Ca(NO_3)_2$	1.0×10^4	
	Hekordzula (Georgia)		Distilled water	2.0×10^5	
	Hekordzula (Georgia)		Solid residue was 84 mg L^{-1}	8.5×10^4	
	Hekordzula (Georgia)		Solid residue was 547 mg L^{-1}	1.1×10^4	

(continued)

Table 1 (continued)

Aluminosilicate	Deposit	Pretreatment	Composition of the solution, pH	K_d, L kg^{-1}	References
Clinoptilolite	Ergene (Mongolia)		0.1 mol L^{-1} $NaNO_3$	1.0×10^3	Dyer et al. (2006)
			0.01 mol L^{-1} $NaNO_3$	2.0×10^3	
	Tsagaantsev (Mongolia)		0.1 mol L^{-1} $NaNO_3$	1.8×10^3	
			0.01 mol L^{-1} $NaNO_3$	3.8×10^3	
	Tushleg (Mongolia)		0.1 mol L^{-1} $NaNO_3$	1.7×10^3	
			0.01 mol L^{-1} $NaNO_3$	2.2×10^3	
Natrolite			Caesium concentration was 10^{-5} mol L^{-1}	1.2×10^{4a}	Vdovina et al. (1976)
		300-400 °C and acidic activation	Caesium concentration was 10^{-5} mol L^{-1}	1.8×10^{4a}	Sultanov et al. (1976)
Vermiculite	Booldymskoye (Russia)		Drinking water (tap water), pH 7.8	1.2×10^3	Voronina et al. (2013)
	Kovdorskoye (Russia)		0.1 mol L^{-1} $NaNO_3$	1.0×10^3	Milyutin et al. (1993)
			1.0 mol L^{-1} $NaNO_3$	10^3	
			0.01 mol L^{-1} $Ca(NO_3)_2$	4.4×10^3	
	Grain size of 0.12-0.5 mm	Medium	Laundry wastewater from Beloyarskaya nuclear power plant (Russia)	110	Sharonov and Pogodin (1980)
		Large		360	
	South American	Natural	0.3 mol L^{-1} $NaNO_3$	0.2×10^3	Sinitsyn et al. (1970)
			0.2 mol L^{-1} NaOH		
		Treatment with NH_4^+	0.3 mol L^{-1} $NaNO_3$	90	
			0.2 mol L^{-1} NaOH		
		Treatment with K^+	0.3 mol L^{-1} $NaNO_3$	500	
			0.2 mol L^{-1} NaOH		
Chabazite	Shivertooyskoye (Russia)	Enriched ore containing up to 80 % of the mineral	0.1 mol L^{-1} $NaNO_3$	100	Milyutin et al. (1993)
			1.0 mol L^{-1} $NaNO_3$	9	
			0.01 mol L^{-1} $Ca(NO_3)_2$	3.4×10^3	

Bentonite	Kyshtymskoye (Russia)	Natural	0.1 mol L^{-1} $NaNO_3$	3.5×10^3	Milyutin et al. (1993)
			1.0 mol L^{-1} $NaNO_3$	620	
			0.01 mol L^{-1} $Ca(NO_3)_2$	5.9×10^3	
	–	Treatment with K^+ and heating at 600 °C	0.3 mol L^{-1} $NaNO_3$	2.9×10^3	Sinitsyn et al. (1970)
			0.2 mol L^{-1} NaOH		
	–	–	0.3 mol L^{-1} $NaNO_3$	0.2×10^3	
			0.2 mol L^{-1} NaOH		
Bentonite	Kelesskoye (Russia)	20 °C	–	1.1×10^{3a}	Sultanov et al. (1976)
		2 h, T = 100 °C	–	3.9×10^{3a}	
		200 °C	–	7.3×10^{3a}	
		300 °C	–	9.4×10^{3a}	
		400 °C	–	11.6×10^{4a}	
	Chimeonskoye	20 °C	–	0.6×10^{3a}	
		2 h, T = 100 °C	–	1.9×10^{3a}	
		200 °C	–	2.3×10^{3a}	
		300 °C	–	2.9×10^{3a}	
		400 °C	–	3.1×10^{3a}	
	Azkomarskoye	20 °C	–	0.3×10^{3a}	
		2 h, T = 100 °C	–	0.9×10^{3a}	
		200 °C	–	1.1×10^{3a}	
		300 °C	–	1.3×10^{3a}	
		400 °C	–	1.9×10^{3a}	
	Kamishbashinskoye	20 °C	–	0.3×10^{3a}	
		2 h, T = 100 °C	–	0.4×10^{3a}	
		200 °C	–	0.4×10^{3a}	
		300 °C	–	1.6×10^{3a}	
		400 °C	–	4.4×10^{3a}	

(continued)

Table 1 (continued)

Aluminosilicate	Deposit	Pretreatment	Composition of the solution, pH	K_d, L kg^{-1}	References
Glauconite	Karinskoye (Russia)	Quartz-glauconite concentrate	Drinking water (tap water)	1.3×10^3	Voronina et al. (2015a)
		Glauconite concentrate dried at 115 °C for 6 h	Drinking water	$1.8\bullet10^3$	Tretyakov (2002)
			Water from the cooling pond of Beloyarskaya nuclear power plant (Russia)	$2.0\bullet10^3$	
White alluvial clay	Moldarskoye (Russia)	Dried at 115 °C for 6 h	Drinking water	$1.6\bullet10^3$	Tretyakov (2002)
			Water from the cooling pond of Beloyarskaya nuclear power plant (Russia)	$2.2\bullet10^3$	
Kaolin			Distilled water spiked with ^{137}Cs	0.46×10^{3a}	Rovinskiy et al. (1972)
Hydrated biotite			Laundry wastewater from Beloyarskaya nuclear power plant (Russia)	0.8×10^3	Sharonov and Pogodin (1980)

[a]The value of caesium distribution coefficient was calculated using experimental data given in the publication

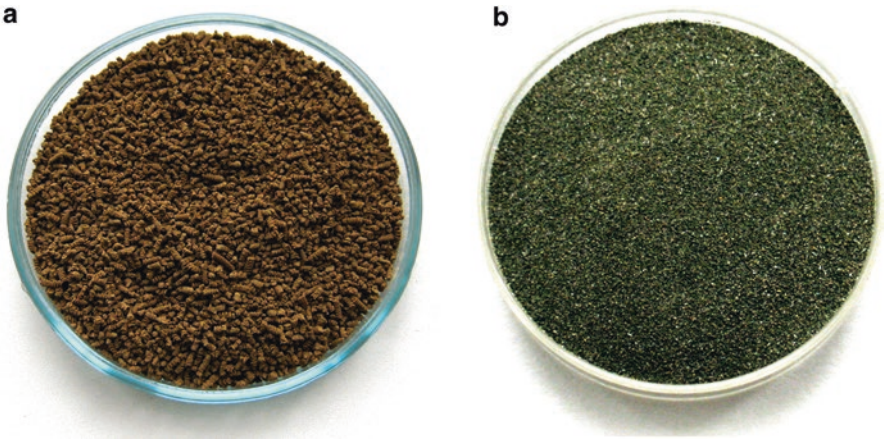

Fig. 1 The appearance of the granulated glauconite (**a**) and the natural glauconite (**b**)

Table 2 Mechanical characteristics of the natural and granulated glauconite

Sorption material	Interparticle porosity (norm > 40 %)	Grindability (norm < 4 %)	Abradability (norm < 0.5 %)	Conventional mechanical strength (Norm < 1 %)
Galuconite concentrate from Karinskoye deposit (Russia)	19.0 ± 2.3	a	a	a
Granulated glauconite	70.3 ± 4.8	0.16 ± 0.05	0.20 ± 0.1	0.9 ± 0.1

[a]The grain size of the glauconite concentrate (0.1 ÷ 0.4 mm) does not allow using the standard methods for determination of mechanical stability

humidity of 32–34 %. During the extrusion step, the paste was mechanically squeezed through a spinneret with a certain form. Thermal treatment of granules was performed at the temperature of 600 °C for 3 h. The glauconite changed colour from dark green to brown after thermal treatment. After all manipulations described above, brown cylindrical pellets with the diameter of 1.0 ± 0.5 mm and the length of 0.5 ÷ 2.8 mm were obtained (Fig. 1).

Combination of a high pressure of extrusion with an intensive mechanical action on the material was the characteristic property of the method. This property provides a high degree of homogenization and satisfactory mechanical stability of pellets; the mechanical characteristics are given in Table 2. Thus, these pellets satisfy the requirements of the Russian State Standard for granulated sorption materials. The mechanical characteristics were determined by the standard method described in the State Standard 51641–2000. The increase of interparticle porosity of the granulated glauconite (65.0 ÷ 75.0 %) as compared with the natural one

(19.0 %) provides the increase of contaminant capacity of the filter as well as the possibility of flow rate increase.

2.2.2 Sorption Characteristics of the Granulated Glauconite

Both natural and granulated glauconites were tested under the same conditions in order to evaluate the influence of granulation on sorption characteristics of the mineral. Determination of sorption characteristics was performed in batch conditions using the limited volume method from tap water (pH 7.8 ± 0.2). In this study a series of solutions with stable caesium concentration of 10^{-7}–1 g L^{-1} (standard solutions of CsCl were used) were prepared and spiked with ^{137}Cs radiotracer. 20 mg of the sorbent was added into 50 mL of a solution; the phases contacted for one week. After that, 0.5 mL of the initial and the final solution was sampled and measured on a semiconductor α-β counter system (UMF-2000 Dosa, Russia). The values of sorption degree *S* were calculated in accordance with the following equation:

$$S = \frac{I_{in} - I_{end}}{I_{in} - I_b} \tag{1}$$

where I_{in}, I_{end} are initial and final count rates of the solution, cpm.

I_b is background count rate, cpm.

Caesium concentrations in a solution after sorption (C_{sol}, g L^{-1}) and in the sorbent after sorption (C_{sorb}, g kg^{-1}) were calculated according to the Eqs. (2 and 3):

$$C_{sol} = C_0 \times (1 - S) \tag{2}$$

where C_0 is initial caesium concentration in a solution, g L^{-1}.

$$C_{sorb} = (C_0 - C_{sol}) \times \frac{V}{m} \tag{3}$$

The results obtained for caesium sorption by the natural and granulated glauconite are presented at Fig. 2 as isotherms of caesium sorption in «lg C_{sol}–lg C_{sorb}» coordinates. Direct proportionality between caesium concentration in the sorbent and in the solution was observed at the initial caesium concentrations range of 10^{-7}–10^{-4} g L^{-1}. The slope ratio of linear sections of the isotherms was near to 1; this indicates that Henry's law may describe sorption and K_d values are invariable. These values determined by mathematical treatment of isotherms were $(1.4 \pm 0.5) \times 10^3$ L kg^{-1} and $(1.6 \pm 0.6) \times 10^4$ L kg^{-1} for the natural and granulated glauconite respectively. The values of static exchange capacity for these sorbents were 14.8 ± 1.5 and 26.9 ± 2.6 mg g^{-1} respectively. It might be seen that the value of caesium distribution coefficient in the Henry region was higher for the granulated glauconite than for the natural one at the same concentration range. The increase of sorption characteristics of

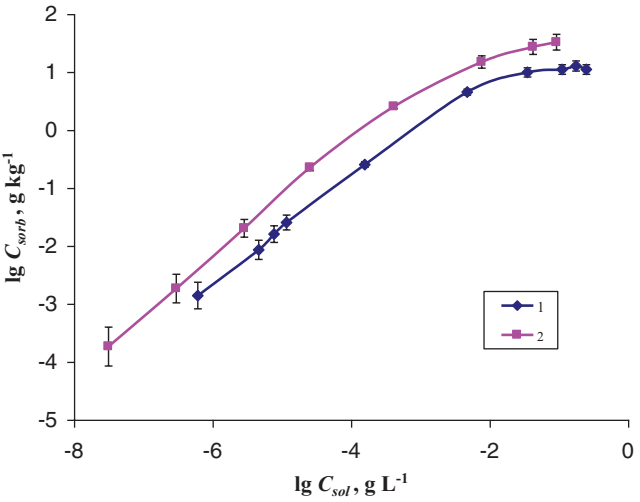

Fig. 2 Isotherms of caesium sorption by the natural (1) and granulated (2) glauconite from tap water with pH 7.8 ± 0.2

the glauconite as a result of granulation might be explained by the fact of addition of zirconium hydroxide (the binding agent) showing some selectivity for caesium. Thus, granulation of the glauconite allows increasing both mechanical and sorption characteristics of this sorption-active material.

2.3 Modified Sorbents Based on the Natural Aluminosilicates

2.3.1 Physicochemical Modification of the Natural Aluminosilicates

Selectivity and capacity of natural aluminosilicates may be increased as a result of their modification by acids, alkalis and salts. Tsitsishvili et al. (1985) reported that the values of exchange capacity of calcium and alkaline clinoptilolites are 1.21 and 1.18 mmol of Cs per gram of the mineral respectively. The most effective sorbents for caesium were synthesized via aluminosilicates chemical modification by the $K_4[Fe(CN)_6]$ solution with or without previous treating by transition metals salts. The methods of synthesis and characteristics of ferrocyanide sorbents based in aluminosilicates are described in a number of publications (Luneva et al. 1994; Rat'ko and Panasyugin 1996; Mimura et al. 1999; Kazemian et al. 2006; Petrova et al. 2008; Milyutin et al. 2010; Voronina et al. 2015a; Vincent et al. 2015).

Table 3 Caesium distribution coefficients (K_d, L kg^{-1}) for some ferrocyanide sorbents based on natural aluminosilicates

Aluminosilicate	Treatment	Composition of the solution, pH value	K_d, L kg^{-1}	References
Clinoptilolite from Shivertooyskoye deposit	$NiSO_4$, $K_4[Fe(CN)_6]$	Tap water, pH 8.0, Cs conc., mg L^{-1} 1×10^{-7}…1×10^{-5}	1.0×10^{7}	Voronina et al. (2015a)
		5×10^{-5}…1	5.0×10^{5}	
		10^{2}…10^{3}	1.6×10^{4}	
Clinoptilolite from Dzegvi deposit	$NiSO_4$, $K_4[Fe(CN)_6]$	Tap water, pH 8.0, Cs conc., mg L^{-1} 10^{-5}…10	1.3×10^{4}	
Clinoptilolite from Holinskoye deposit	$NiSO_4$, $K_4[Fe(CN)_6]$	Tap water, pH 8.0, Cs conc., mg L^{-1} 10^{-5}…10	1.3×10^{4}	
Clinoptilolite from Shivertooyskoye deposit	$Fe_2(SO_4)_3$, $K_4[Fe(CN)_6]$	Tap water, pH 8.0, Cs conc., mg L^{-1} 10^{-7}…1	7.9×10^{4}	
		10^{2}…10^{3}	4.0×10^{3}	
Clinoptilolite in Fe-form	$K_4[Fe(CN)_6]$	–	5.0×10^{3}	Panasyugin et al. (1995)
Clinoptilolite in Cu-form	$K_4[Fe(CN)_6]$	–	1.4×10^{4}	
Clinoptilolite in Ni-form	$K_4[Fe(CN)_6]$	–	8.7×10^{3}	
Glauconite	$NiSO_4$, $K_4[Fe(CN)_6]$	Tap water, pH 8.0, Cs conc., mg L^{-1} 10^{-7}…1	1.0×10^{5}	Voronina et al. (2015a)
		10^{2}…10^{3}	2.5×10^{2}	
Clinoptilolite	Antimony ferrocyanide	0.1 mol L^{-1} HNO_3	4.4×10^{3}	Rat'ko and Panasyugin (1996)
		Na:Ca:Mg:Cl = 1:5:5:25, total salt content 0.8 g L^{-1}	1.3×10^{4}	
Fine dispersed clinoptilolite	Ni^{2+}, $K_4[Fe(CN)_6]$	Tap water, pH 7.8-8.2	2.8×10^{6}	Milyutin et al. (2010)
		Carrier free ^{137}Cs		
Fine dispersed bentonite	Ni^{2+}, $K_4[Fe(CN)_6]$	Tap water, pH 7.8-8.2	1.9×10^{6}	
		Carrier free ^{137}Cs		
Fine dispersed wollastonite	Ni^{2+}, $K_4[Fe(CN)_6]$	Tap water, pH 7.8-8.2	2.6×10^{6}	
		Carrier free ^{137}Cs		

2.3.2 Sorption Characteristics of the Ferrocyanide Sorbents Based on the Natural Aluminosilicates

The increase of sorption capacity of the clinoptilolite as a result of its modification by ferrocyanides is mentioned in publications (Rat'ko and Panasyugin 1996; Voronina et al. 2015a). Luneva et al. (1994) reported that the exchange capacity of the clinoptilolite with hexacyanoferrate groups is 2 mmol of Cs per gram of the sorbent. A significant increase of exchange capacity was observed after surface modification of the clinoptilolite and glauconite by mixed ferrocyanides. For example, the values of exchange capacity were: 1.65 mmol Cs g^{-1} of the mixed

nickel-potassium ferrocyanide based on glauconite, 3.8 mmol Cs g^{-1} of the mixed nickel-potassium ferrocyanide based on clinoptilolite (Shivertooyskoye deposit) and 3.76 mmol Cs g^{-1} of the mixed iron-potassium ferrocyanide based on clinoptilolite (Shivertooyskoye deposit) (Voronina et al. 2015a). Caesium distribution coefficients for some ferrocyanide sorbents based on aluminosilicates are presented in Table 3. In some cases, modification of aluminosilicates by ferrocyanides results in increase of capacity for caesium, but at the same time, their selectivity for caesium does not change. Caesium distribution coefficients for these sorbents are approximately the same as for the respective natural aluminosilicates and do not exceed 10^3–10^4 L kg^{-1}. Petrova et al. (2008) reported that modifying agents were partially stripped into the solution to be decontaminated.

A significant increase of selectivity for caesium is observed after modification of fine-dispersed aluminosilicates resulting in composite sorbents containing 50 % of aluminosilicate and 50 % of nickel-potassium ferrocyanide (Milyutin et al. 2010) or after surface modification of natural aluminosilicates (Voronina et al. 2015a). In the first case, the increase of selectivity is explained by the synthesis of the ferrocyanide phase being stable against peptization on the surface of the aluminosilicate (Milyutin et al. 2010). In the second case, this is explained by formation of nano-sized ferrocyanides with an extended surface area and selective sorption sites on the surface and in pores of the aluminosilicate as a result of surface modification. It is reported that effect of surface modification depends on sorption characteristics of the aluminosilicate, its structure, pore size and porous space. Therefore, sorption characteristics of ferrocyanides based on aluminosilicates will be different even for the same minerals from various deposits. Voronina et al. (2015a) reported that the best results were obtained for the surface modification of the glauconite (Karinskoye deposit) and clinoptilolite (Shivertooyskoye deposit) by nickel-potassium ferrocyanide. The content of the ferrocyanide phase in these sorbents was only 5–10 %; however, both exchange capacity significantly increased and caesium distribution coefficients reached 10^5–10^7 L kg^{-1}. These sorbents possess high effectiveness, chemical stability and environmental safety; therefore, they may be recommended for decontamination of natural waters and soils from radiocaesium.

3 Use of Granulated and Modified Aluminosilicate Sorbents for Deactivation of Natural Water and Soil

3.1 Decontamination of Natural Water

The sorption characteristics of the ferrocyanide sorbents based on natural aluminosilicates (values of static exchange capacity and caesium distribution coefficient given in the Chap. 2.3.2) will be realized if these sorbents will be used for natural water decontamination under static conditions. However, it is not always possible to reach these values in dynamics. Therefore, the study of effectiveness of filters use for natural water decontamination is of interest. The effectiveness of a filter loaded

with granulated glauconite was tested. The study was conducted under dynamic conditions in order to simulate the possible use of this sorption filter at the conditions similar to real. A plastic column with the inner diameter of ≈9 mm was chosen. A sample of the sorbent in the column ($m = 2$ g) provided at least 5 cm of the sorbent layer in the column. The bed volume (B.V.) was ≈3 mL, flow rate was 10–12 B.V. per hour, i.e. approximately 1 mL min^{-1} cm^{-2}. The tap water spiked with ^{137}Cs as a radiotracer containing 1 mg L^{-1} of stable caesium was used as a simulated solution that passed through column. The resource of the filter supposed as the volume (in B.V.) of contaminated water loaded through the filter without caesium breakthrough. The achievement of decontamination factor (D) value of at least 100 and more was the criterion of successful work of the filter.

Dimensionless values of decontamination factor D were calculated in accordance with the Eq. (3):

$$D = \frac{I_0}{I_t} \tag{4}$$

where $I_0 = I_{in} - I_b$ is the initial count rate of the water sample after deduction of background count rate, cpm; $I_t = I_{end} - I_b$ is the count rate of the decontaminated water after deduction of background count rate, cpm.

All samples were measured on the semiconductor α–β counter system UMF-2000. The values of decontamination factor were calculated for each sample of decontaminated water. Since the column test requires operations with a large volume of radioactively contaminated water, there was the risk of a very high activity of the filter even before the end of the experiment. This work would be hazardous and use of a protection shield may be necessary. The method suggested by Betenekov et al. (2006) was used in order to avoid the risk described above. This method consists of alternation of loading water samples containing equal concentrations of stable caesium with and without ^{137}Cs radiotracer. In accordance with the basic principles of the radioactive tracer's method, stable and radioactive isotopes of an element have the same chemical features, including sorption behavior; therefore, saturation of the filter by caesium occurs both in presence and in absence of the radiotracer in an equal degree.

A loading cycle of water through the column with the granulated glauconite consisted of loading of 10 B.V. of spiked simulated solution followed by 700–800 B.V. of tracer-free solution. Eleven loading cycles were carried on the same column with the granulated glauconite. The dependence of D values on the volume of loaded simulated solution is presented in Figure 3. The D values were not less than 100 even after loading of ≈7000 B.V. of the simulated solution; the flow rate did not decrease indicating the good mechanical characteristics of the granulated glauconite. The static exchange capacity of the granulated glauconite for caesium was not exhausted after the column test. Caesium breakthrough was less than 1 % in each tested sample. The achieved value of dynamic exchange capacity was 11.6 mg of Cs per gram of the granulated glauconite; the dynamic caesium distribution coefficient

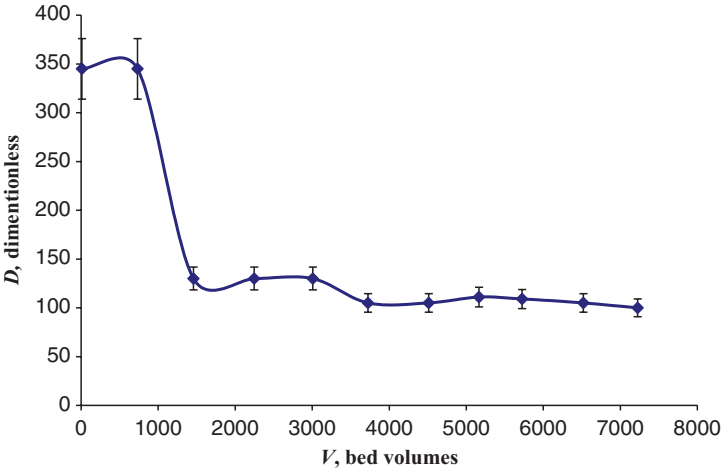

Fig. 3 The dependence of decontamination factor *D* on the volume of the loading solution

was $(1.2 \pm 0.2) \times 10^4$ L kg^{-1}. The further study have shown that sorption of ^{137}Cs was irreversible. It was also determined that the natural glauconite cannot be used in sorption filters because of a very high hydraulic resistance; the flow rate was almost zero at the same conditions. Thus, the results have shown that the granulated glauconite may be used for decontamination of natural water contaminated by caesium radionuclides. In the case of use of a filter containing 1 L of the granulated glauconite, the resource of this filter will be at least 7–10 m^3 of contaminated water.

3.2 Radiocaesium Removal from Soil

A comparative study of the natural and modified aluminosilicates use for ^{137}Cs elimination from soil and soil solutions was performed. The glauconite from Karinskoye deposit (Chelyabinsk region, Russia), the clinoptilolite from Shivertooyskoye (Chita region, Russia) and the mixed nickel-potassium ferrocyanides based on these minerals were used in this study.

The effect of sorbents addition on ^{137}Cs transfer from a radioactively contaminated solution (on hydroponics system) and soils to an agricultural vegetation was studied in order to test the developed sorbents under the conditions similar to those of soils rehabilitation. Feed solutions for hydroponics contained the Knop mixture and were spiked by 10^4 Bq L^{-1} of ^{137}Cs. Chemical composition of the Knop mixture was 1.0 g L^{-1} of Ca $(NO_3)_2$, 0.25 g L^{-1} of KH_2PO_4, 0.125 g L^{-1} of KCl and 0.25 g L^{-1} of $MgSO_4$. Natural or modified aluminosilicates were added to the feed solution or the soil; then oats (*Avéna satíva*) seeds were sowed. Oats vegetation was harvested

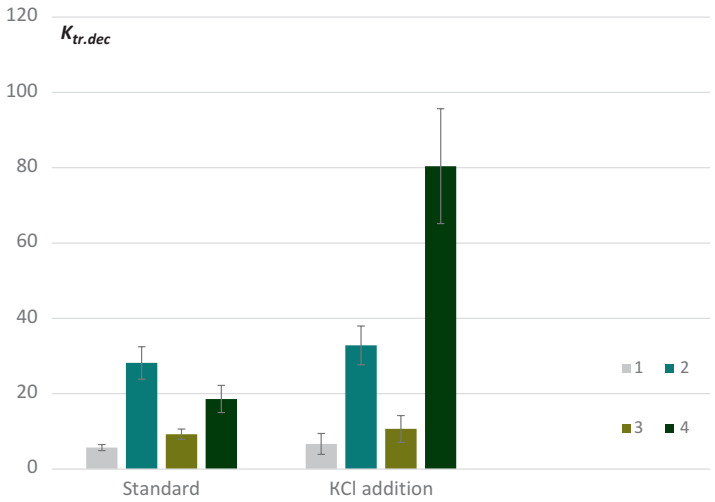

Fig. 4 The values of caesium transfer decrease factor ($K_{tr.\,dec}$) for various sorbents vs. KCl addition: (*1*) the natural clinoptilolite, (*2*) the mixed nickel-potassium ferrocyanide based on clinoptilolite, (*3*) the natural glauconite, (*4*) the mixed nickel-potassium ferrocyanide based on glauconite

during the tillering period in 21 days after seeds sowing. Vegetation samples were dried at 100 °C for 4 h; caesium activity was measured by gamma-spectrometry using scintillation NaI(Tl) gamma-spectrometer Atomtech AT-1315 (Belarus). Oats vegetation grown on solutions (soils) without sorbents under the same conditions was used as control samples. Dimensionless values of concentration ratio (F_V) of caesium in plants and caesium transfer decrease factor ($K_{tr.\,dec}$) from soil to plants in presence of sorbents were calculated according to Eqs. (4 and 5) using experimental data obtained:

$$F_V = \frac{A_v}{A_s} \tag{4}$$

Where, A_v is activity concentration of radiocaesium in the vegetation, Bq g^{-1}, A_s is activity concentration of radiocaesium in the solution (soil), Bq g^{-1}.

$$K_{tr.dec} = \frac{A_{control}}{A_v} \tag{5}$$

Where $A_{control}$ was activity of radiocaesium in plants from control group of plants, grown in soil, contaminated by ^{137}Cs, without addition of sorbents, Bq g^{-1}.

A series of experiments with KCl addition to the feed solution for hydroponics system was performed in order to study the possibility of combined use of the sorbents and potassium fertilizers. The dependences of caesium transfer decrease factors for various sorbents and hydroponics system on potassium concentration are presented at Figure 4. The “standard” potassium concentration at the diagram corresponds to its concentration in the Knop mixture.

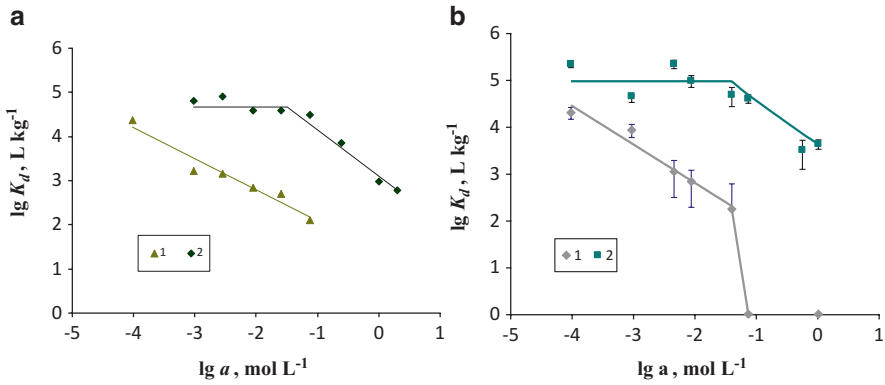

Fig. 5 The dependences of caesium distribution coefficient on potassium active concentration in the solution: (**a**) glauconite (*1*) and the nickel-potassium ferrocyanide based on glauconite (*2*); (**b**) clinoptilolite (*1*) and the nickel-potassium ferrocyanide based on clinoptilolite (*2*)

It is obvious that the addition of ferrocyanide sorbents to the hydroponics solution may result in a stronger decrease of caesium transfer to oats as compared with addition of natural aluminosilicates. The values of $K_{tr.\,dec}$ were 28 ± 4 in case of addition of mixed nickel-potassium ferrocyanide based on glauconite and 19 ± 3 for the sorbent based on clinoptilolite. The combined use of ferrocyanide sorbents and potassium fertilizers resulted in significant increase of $K_{tr.\,dec}$ values. The highest values of $K_{tr.\,dec}$ (80 ± 15) were obtained after combined addition of the modified glauconite and KCl (Voronina et al. 2015b). This effect might be explained by the fact that, on the one hand, ferrocyanide sorbents show a good selectivity for caesium over a wide potassium concentrations range (Fig. 5); therefore, potassium does not suppress caesium sorption at these conditions. On the other hand, relatively high concentrations of potassium suppress caesium uptake by roots. Thus, the combined effect of ferrocyanide sorbents and potassium fertilizers resulted in very high values of $K_{tr.\,dec}$. No any data indicating achievement of the similar effectiveness of rehabilitation activities on radioactively contaminated lands were found in scientific literature. Use of the mixed nickel-potassium ferrocyanides based on glauconite and clinoptilolite for the decrease of caesium transfer from a soil to agricultural vegetation is described in details by Voronina et al. (2015c). The values of caesium transfer decrease factor from the radioactively contaminated soil to oats were as high as 2.4 after addition of the natural glauconite and 21.2 after addition of the mixed nickel-potassium ferrocyanide based on glauconite.

4 Conclusions

Sorbents used for decontamination of natural waters and soils should be relatively cheap, effective (they should possess high sorption capacity and selectivity for radionuclides), ‘eco-friendly’ and should possess satisfactory chemical stability and

mechanical strength. Use of synthetic sorbents for mentioned applications is limited because of the requirements listed above. Natural aluminosilicates traditionally used for caesium removal from natural media have worse sorption and mechanical characteristics than sorbents synthesized by granulation and modification of these aluminosilicates. Granulation with a binding agent results in increase of mechanical characteristics of aluminosilicates; their sorption characteristics for caesium are also slightly increased. Physicochemical modification of aluminosilicates by ferrocyanides results in the increase of their capacity and selectivity for caesium; thus, caesium distribution coefficients the static exchange capacity may reach 10^7 L kg^{-1} and up to 500 mg of Cs per gram of a sorbent respectively.

Nickel-potassium ferrocyanides based on the natural aluminosilicates (clinoptilolite, glauconite, and bentonite) may be recommended for decontamination of natural waters in static conditions. Caesium distribution coefficients for these sorbents are given in Table 3. The granulated glauconite has shown interesting characteristics for caesium separation form natural waters in dynamics. For example, the filter for low mineralized water decontamination containing 1 L of the granulated glauconite should have the resource at least 7–10 m^3 of water.

Mixed nickel-potassium ferrocyanides based on the natural clinoptilolite and glauconite were the prospective sorbents for rehabilitation of radioactively contaminated lands with the aim of their return to farming use. Use of these sorbents resulted in caesium transfer decrease factors from a soil to agricultural plants at least as high as 20; caesium transfer decrease factor of 80 ± 15 was obtained as a result of combined use of the modified glauconite and potassium fertilizers.

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Analysis of Transfer Factor, Anatomical Changes and Growth of Plants During Phytoremediation of Cesium Contaminated Solutions

Roxana Moogouei, Mehdi Borghei, and Golnaz Tajadod

Abstract *Calendula alata*, *Amaranthus chlorostachys* and *Chenopodium album* plants were tested for their potential to uptake and accumulate stable cesium and lead from solutions in a period of 15 days. The plants were grown hydroponically and placed in solutions containing CsCl and $Pb(C_2H_3O_2)_2$ at three different concentrations (0.6, 2 and 5 mg L^{-1}). Cs transfer factors in different solutions were more than 1 with the highest level of 2.45 for 2 mg L^{-1} of CsCl and $Pb(C_2H_3O_2)_2$ in mixed solution. According to its uptake from the environment, cesium effect on anatomical and ontogenical structure of plant was studied. Aerial organs of *Chenopodium album* were investigated for modifications studies. Observation showed that tissue structures of these organs had not changed at all and only xylems are expanded due to fight against tension and cells store cesium in their vacuole in crystalline shape for fighting against toxic effects of cesium. The metal accumulation in *C. alata*, *C. album* and *A. chlorostachys* increased with the increase in metal concentrations in solution, and the metal accumulation in shoots was always significantly higher than those in roots for all three species.

Keywords Cesium • Plants • Anatomy • Transfer factor

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1 Introduction

Interest in cesium distribution in plants organs and the movement of this radionuclide element in ecosystems extends back to the 1950s by the development of technologies that used for nuclear power plants (Cook et al. 2007). The radioisotopes of cesium may be of special concern because of their high solubility in aquatic ecosystems. ^{133}Cs and ^{137}Cs have similar behavior to the necessary element potassium in plants, therefore easily uptake and accumulate by them. Tsukada et al. (2002); Vinichuk et al. (2010) have found strong correlation between distribution of ^{137}Cs and stable Cs in plants. Moreover last studied showed no differences between uptake of radioactive and stable cesium isotopes by *Helianthus annuus* L. (Soudek et al. 2006). Stable cesium solution is phytotoxic in solution culture exceeding 200 μM (Borghei et al. 2011). Plants can uptake and accumulate cesium ions in their shoots and roots. In the study carried by Yang et al. (2016), when bean root exposed to uranium and cesium solution, the roots of the bean accumulated uranium and cesium to concentrations 317–1019 times above the initial concentrations, which ranged from 100 to 700 μg L^{-1} during 72 h. Moreover metals transfer factor (Tf), defined as the ratio of metal concentrations in plant shoots to those in the roots (Gonzaga et al. 2006; Bidar et al. 2007). Transfer factor was calculated to check the effectiveness of plants in translocating metals to their aerial organs (Moogouei et al. 2011; Zabłudowska et al. 2009). Furthermore hyper tolerance is a key property that makes hyper accumulation later. Therefore, it is necessary to consider tolerance of different plants to metal uptake. Several cesium isotopes are radioactive yield of uranium fission and uranium is a parent element which decays to its end product, radiogenic lead. Lead accounts as the most widespread heavy metal contaminant in the environment (Borghei et al. 2011). The plant with scientific name "*Chemopodiu album* L." is an annual plant with stagnant stem and plenty ramifications which almost has pink and purple lines. Its height changing depends on water, weather and soil type. At the begging of growth, it rises from soil perpendicularly and achieving to 3–150 cm height. After sprout and growth (blossoming), plant turns to embowed shape due to seed and bud flower weight. *C. album* can be cultivated in farms with other plants competitively and causes reduction on productions. In some area, it has an edible usage too. Young stems or whole plant can be fed after steaming. It is recommended to consume it cautiously regarding to excessive reserve of oxalic acid in this plant. This plant is seen in most area of Iran such as northern and central part, south eastern, Semnan, Cheshmeh Ali Damghan, Saveh and Arak central plain (Assadi (2001). *C. album* L.can absorbs and purifies heavy metals from environment. Cesium is one of pollutant and basic elements. Due to similarity to potassium, it can be absorbed by plants intensively. Cesium's natural concentrations have not any importance for plants. Phytoremediation is the technique to segregate ^{137}Cs from environment. Plant roots, penetrating through buried position of radioactive wastes, transmit radio activated cesium to shallow part of soil through transpiration system. Finally, this process leads to purge the environment from this toxic element by absorbing them and accumulating it in plant's tissue. In this paper, *C. album* has been studied after hydroponic cultivation for surveying of

cesium absorption ability and it's reservoir in various organs and anatomical changes. Moreover we aim to investigate the tolerance and transfer factor of cesium in plants.

2 Material and Methods

2.1 Hydroponic Cultivating

Hydroponic cultivating system is the soilless cultivated systems. Providing nutrition for plant is done by nutrition solutions in this method. In current research, Hoagland solution was produced base on Hoagland protocol 1950. Separated solutions are produced from macro elements and then appended to 240 L water as major tank. But micro elements are produced in solution with 240 mL volume. In hydroponic system, by means of avoiding light effect on physiologic performance of roots, Hoagland solution is entered to 10 L container with dark colored curtain or containers which cover by dark textile or plastic and they were aerated with air pump through 5 mm pipe. In fact, cap of these containers are coating membrane which seeds budding on them and passing other phase of growth.

2.2 Remediation of Cesium and Lead from Mixed Solution

To study the uptake of cesium and lead in mixed condition, solutions with similar concentration of CsCl and $Pb(C_2H_3O_2)_2$ were added together in 1 L flasks, where the plants were incubated and at the end of the 15 days, samples were analyzed for cesium and lead concentrations. pH of solutions was adjusted to 5.5. The percentage metal uptake was calculated from

$$\%\,\text{remediation} = \left[\left(C_0 - CI\right)/C_0\right] \times 100$$

where C_0 and C_1 are initial and remaining concentrations of metal, respectively in solution (Table 1).

Table 1 Cs transfer factor in *Amaranthus chlorostachys*, *Calendula alata* and *Chenopodium album* from CsCl and $Pb(C_2H_3O_2)_4$ solutions

	Calendula alata			*Chenopodium album*			*Amaranthus chlorostachys*		
Cs (mg L^{-1})	0.47	1.58	3.95	0.47	1.58	3.95	0.47	1.58	3.95
	1.50	2.12	1.43	2.27	1.78	1.47	1.76	2.05	1.59

In Table 1 initial concentrations of CsCl and $Pb(C_2H_3O_2)_4$ in different solution was 0.5, 2 and 5 mg L^{-1} respectively. In all the experiment transfer factors of Cs was more than 1

Table 2 Remediation percent of cesium from mixed solution of CsCl and $Pb(C_2H_3O_2)_4$ in different solution were T_1: 0.5, T_2: 2 and T_3: 5 mg L^{-1}

Plants	T1	T2	T3
C. alata	9.92 ± 1.2	45.56 ± 3.5	46.16 ± 1.4
C. album	14.53 ± 1.6	47.25 ± 0.9	48.01 ± 1.4
A. chlorostachys	59.57 ± 2.1	67.51 ± 4.1	49.11 ± 3.7

2.3 Cesium Absorption and Accumulation

The roots of hydroponically grown plants were washed with distilled water and then plants were incubated with roots immersed in 1500 L solution with three different Cs concentrations. The treatment samples included: (1) control sample free of Cs, samples 2, 3 and 4 containing 0.5, 2 and 5 mg L^{-1} of CsCl, respectively (Moogouei et al. 2011). As a result the concentration of Cs ions in the solutions was 0.47, 1.58 and 3.95 mg L^{-1}. The experiment was arranged with each treatment in triplicate samples. The treatment group was exposed to CsCl solution in 1500 mL flasks for a period of 15 days (Singh et al. 2009). pH of the solution was adjusted to 5.5. Cesium remediation data was showed in Table 2.

2.4 Distribution of Cs in Plants

At the end of the treatment period, plants were thoroughly washed with distilled water and dried in an oven at 60 °C for 48 h. All the experiments conducted for roots and shoot separately. Then the dried samples were digested in HNO_3:$HClO_4$ (5:1, V/V) and analyzed for Cs concentration by flame atomic absorption spectrophotometry. The concentrations of cesium in the samples plants were reported on a dry matter basis (Moogouei et al. 2011).

2.5 Transfer Factor

The Transfer factor, defined as the ratio of metal concentrations in plant shoots to those in the roots (Gonzaga et al. 2006; Bidar et al. 2007). Transfer factor was calculated to check the effectiveness of plants in translocating metals to their aerial organs (Zabłudowska et al. 2009).

2.6 Statistical Analysis

The statistical analysis was done using Statistical Analysis System (SAS) software package. To confirm the variability of results, all the data were subjected to analysis of variance to consider the significance differences. Moreover means comparison between data was obtain using Duncan test.

2.7 Anatomical Studies

After separation, aerial organs of plant including stem and leaf had been laid in alcohol and glycerin for fixation at the same portion. After a week, a handy section was prepared and colored with multiple coloration (Carmen Zuji and methylene blue with ratio 1:10). Then samples were photographed and studied by Nikon microscope.

3 Results

3.1 Absorption and Accumulation of Cesium

In this section, plants were left from hydroponic growth system and washed by distilled water. After studying various part of plant with atomic absorption spectrophotometry, it was specified that cesium accumulates in aerial parts of plant which its highest quantity was estimated at 966.66 mg kg^{-1} by the instrument. Shoots accumulations were 0.56, 204.13, 284.13 and 966.66 mg kg^{-1} in 0, 0.5, 2 and 5 mg L^{-1} cesium solutions respectively. Cesium accumulation in shoots and roots of hydroponically grown *A. chlorostachys*, *C. album* and *C. alata* are shown in Fig. 1. In the

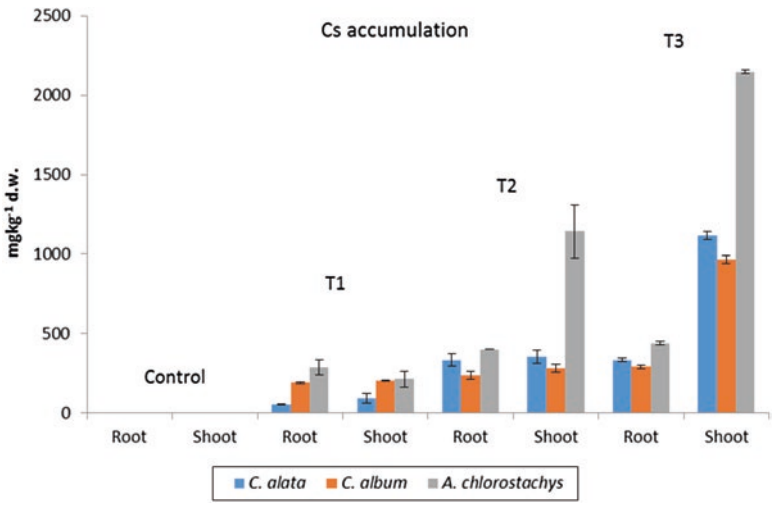

Fig. 1 Cs concentration in roots and shoots of hydroponically grown plants. The values are mean of three replicates ± S.D. T_1: 0.47 mg L^{-1}; T_2: 1.58 mg L^{-1}; T_3: 3.95 mg L^{-1} in Cs solution. Control values are not detectable. $P < 0.05$ data differences are significant

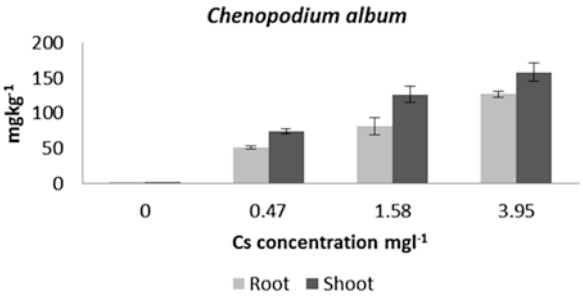

Fig. 2 Cs uptake in roots and shoots of hydroponically grown *Chenopodium album* from mixed CsCl and $Pb(C_2H_3O_2)_4$ solutions. The values are mean of three replicates ± S.D. T_1: 0.47 mg L^{-1}; T_2: 1.58 mg L^{-1}; T_3: 3.95 mg L^{-1} in Cs solution. Control values are not detectable. $P < 0.05$ data differences are significant

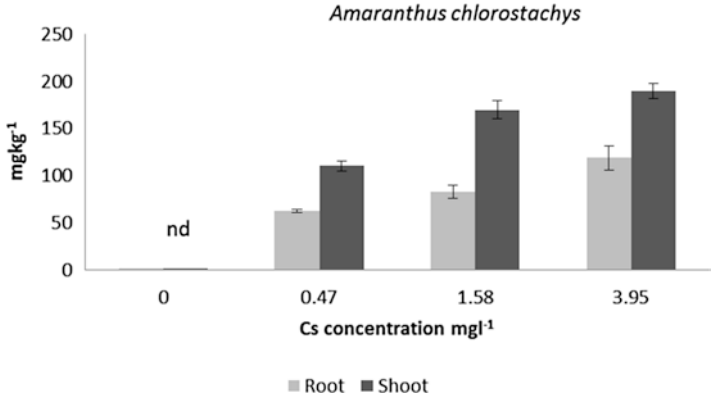

Fig. 3 Cs uptake in roots and shoots of hydroponically grown *Amaranthus chlorostachys* from mixed CSCl and $Pb(C_2H_3O_2)_4$ solutions. The values are mean of three replicates ± S.D. T_1: 0.47 mg L^{-1} in Cs solutions; T_2: 1.58 mg L^{-1} in Cs solution; T_3: 3.95 mg L^{-1} in Cs solution. Control values are not detectable. $P < 0.05$ data differences are significant

mixed solution of Cscl and $Pb(C_2H_3O_2)_4$ cesium accumulation in roots and shoots of plants were shown in Figs. 2 and 3.

3.2 Anatomical Studies

3.2.1 Primary Structure of Stem in Control Plants

Based on previous studies on *C. album*, stem is covered by a cortical epidermis layer. Hypoderm includes several rows of collenchyma cells. Collenchyma is discontinuous and has 8–10 layers. Distance between membrane and Parenchyma is

specified by endoderm and has sclerenchyma texture sparsely. Parenchyma texture cells have oxalate crystals of macle type. Vain groups were sorted in one ring at the center. Regions in each vain group which specified from inside to outside, are including phloem region, pro cambium area and xylem vain region Activity of cambium is definite which result in composing cambium rings which are responsible for composing secondary vain texture.

3.2.2 Secondary Structure of Stem in Control Plants

Study on secondary structure of this specimen showed that the quantity of collenchyma layer have increased under epidermis, moreover the fibers which locating on phloem have increases either (Figs. 4 and 5). Interior cambium gathers together and composes a cambium ring with primary vain groups. This ring unnaturally cuts fiber ligaments at the cambium region Quantity of macle crystals is increasing by producing of secondary structure.

3.2.3 Structure of Control Leaf

Leaves have extensive and thin epidermis. Fuzzes are multi cells and bubble shape, moreover they aren't divergence. Mesophyll has two rows of ladder parenchyma and several rows of spongy parenchyma. Makley crystals are seen in spongy parenchyma cells. Stomata are anisocytic type (Fig. 6).

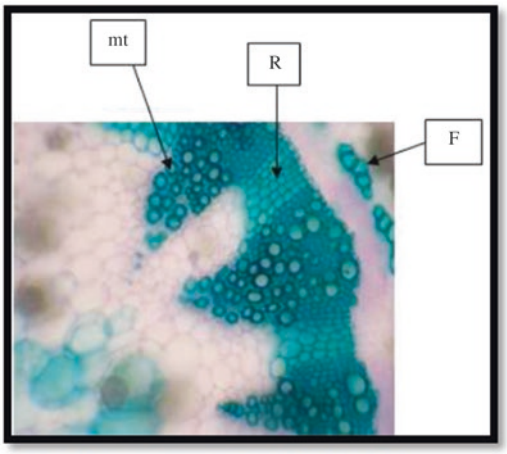

Fig. 4 Secondary structure of stem (Control) (×40)

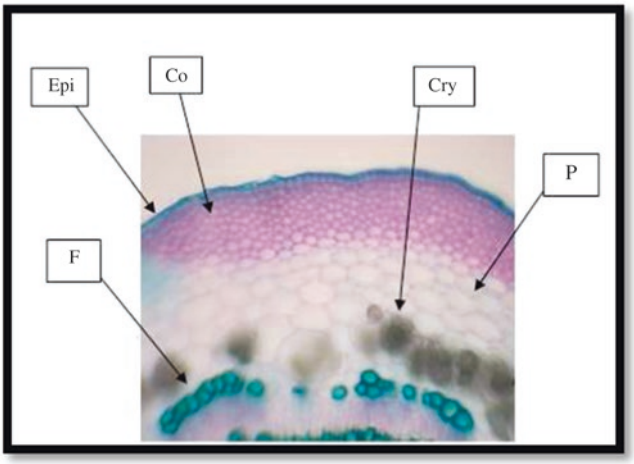

Fig. 5 Increasing crystals quantity by composing secondary structure (×40). *P* parenchyma, *F* fiber, *Epi* epidermis, *Co* collenchyma, *Cry* crystal

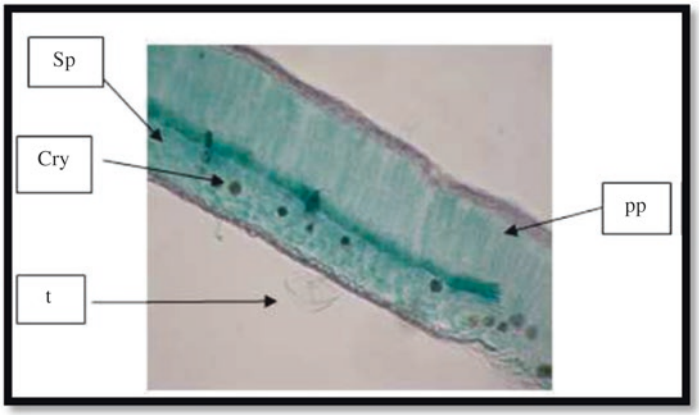

Fig. 6 Leaf cross section (Control) (×20). *Cry* crystal, *pp* ladder parenchyma, *Sp* spongy parenchyma, *t* fuzz

3.2.4 Anatomical Structure of Aerial Organs in Treatment with Cesium

By comparing aerial organs of plant under treatment by controlled plant with same ages, it was specified that cesium absorption by the plant and accumulation of this element in aerial organs (leaf and stem) makes some changes in mentioned organs

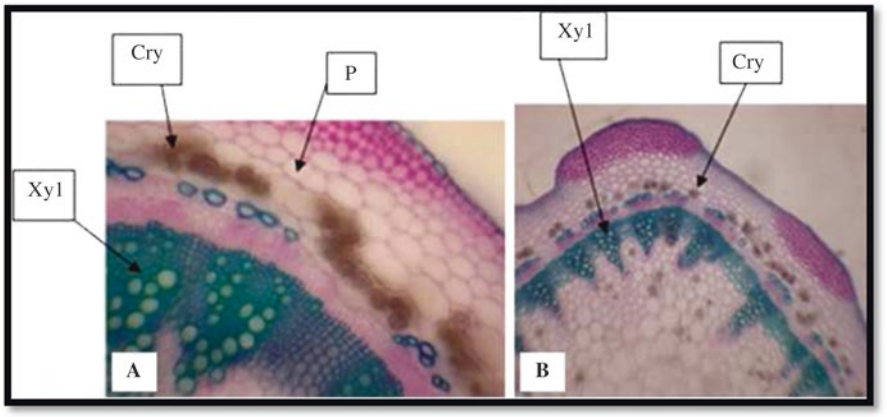

Fig. 7 Stem-treated with cesium (2 ppm). ((**a**) ×20, (**b**) ×10) *Cry* crystal, *P* parenchyma, *Xyl* xylem

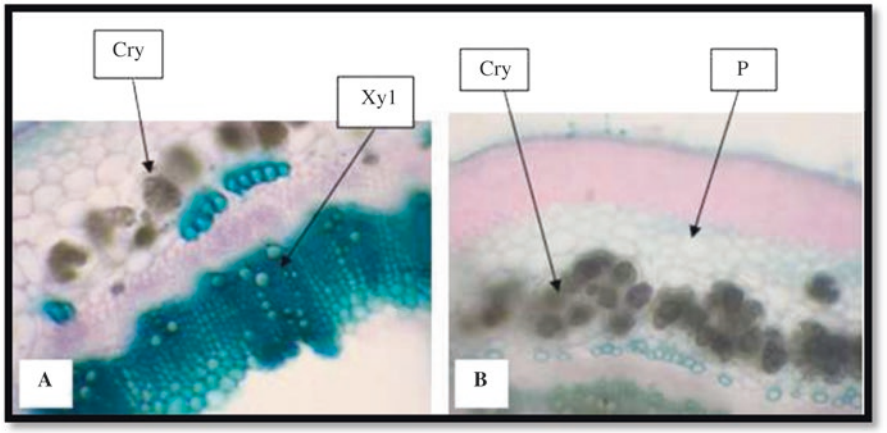

Fig. 8 Stem-treated with cesium (5 ppm) ((**a**) ×20, (**b**) ×40) *Cry* crystal, *P* paranchyma, *Xyl* xylem

which is more intensive by increasing cesium concentration. Available observed changes due to absorb and accumulate of cesium can be classified in three groups:

1. Increase of crystals in stem parenchyma and their color embrace.
2. Decrease in density of xylem and their dimension in stem (Fig. 7).
3. Increasing color embrace property of xylem component in stem (Fig. 8).
4. Decreasing cutaneous parenchyma in plants under treatment.
5. Increasing crystalline density and colored embrace in leaf's parenchyma (Figs. 6 and 9).

There was not seen any other changes in the structure of studied organs.

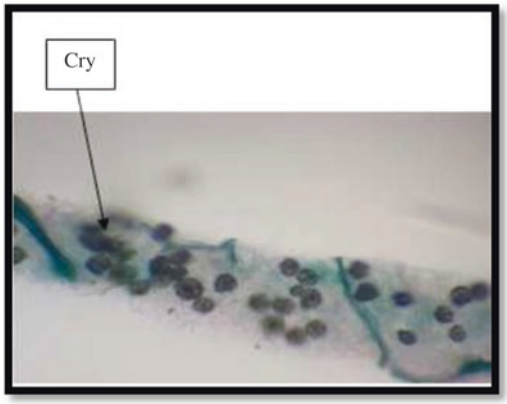

Fig. 9 Leaf cross section-treated with cesium (5 ppm) (×20) *Cry* Crystal

4 Discussion

In all of the control samples (Cs concentration 0 mg L^{-1}) Cs was detected in shoots that shows presence of Cs in the environment. Based on the available data, plants do not have specific transporters for nonessential metal ions, and therefore its transport across a membrane is mediated by systems for essential ions. It has been a common belief that cesium is transferred to plants by means of the same proteins which are mainly involved in the uptake of K^+. A number of transporting proteins (low affinity, inward-rectifying K channel, nonspecific, voltage insensitive cation channel, high affinity K^+–H^+ symporter, voltage-dependent Ca^{2+} channels and outward-rectifying cation channels) facilitate permeation of Cs^+ across the root cell membranes in plants (Bystrzejewska-Piotrowska and Bazala 2008). Important point in phytoremediation is that the plants can be able to survive in polluted environment. Another important point (as applied method for filtrating sewage), is that they must have high performance in refining sewage, beside they not only absorb pollutants, but also show less toxic effects during process (Pivetz Bruce 2001). Plant would be observed as a suitable tool for filtration and environmental management if pollutant concentration in soil and water becomes less than phytotoxic level (Singh et al. 2009). Some weeds were chosen to apply for phytoremediation techniques out of their rooty system (Merkl et al. 2004) *C. album* is one of them. Cesium has chemical activities as similar as potassium. Although there is not seen any nutritional role for cesium in plants, but large amount of this element can be so toxic for plants. Physiologic studies revealed that because of similar entrance mechanism of cesium and potassium to plant, these two element compete with each other login to plant's organs. Ionic transmission through membrane has been focused in recent years (Aprill and SIMS 1990). Ionic channels are activated ways for transmitting charged ion through cell's membrane (Cook et al. 2007). Most of channels are permeable toward other cations such as sodium, lithium, rubidium and cesium. Sizes of these cations are between 0.13 and 0.19 nm which is very similar to potassium's dimension (0.27 nm) and this fact causes their transmission through these channels (Singh

et al. 2009). Respecting these facts, cesium absorption by *C. album* might be related to its molecule's similarity to potassium. Therefore, performances of potassium channels are effective on cesium absorption.

In the experiments are done on algae revealed that cesium is accumulated in plant's cell after absorbing by plant as crystals. Concerting cesium to crystals in algae is a very attractive phenomenon. Cesium is seen as a soluble element in water in algae. This element can be replaced by other first group cations (I) of periodic table and then compose salt with anions (Aprill and SIMS 1990). Cooperating cesium in crystal's structure and its accumulation in vacuoles permit plant to absorb more content of radio activated cesium from environment without any serious incurred damage to plant (Singh et al. 2009). Reducing diameter of vein's spout is a consequence of increasing xylem stiffness rate; demonstrates plant's adaption to tension terms and avoiding water wasting. So, water traverses more rapidly through veins by increasing of diameter of xylem wall's width and decreasing of vein's spout diameter (Singh et al. 2009). It is concluded that expanding subsequent structure is a typical plant's defense against tension and inappropriate conditions of the environment. Color effecting (receipting) of xylem parts is more intensive than control plant which may be due to polymerization enhancing of units which forms xylem. Considering increasing crystals numbers which are formed in the aerial organs of *C. album* and not seeing any damages in these organs and related tissue, it can be concluded that this plant causes to increase absorbed element from environment by enhancing its xylem system. Furthermore this plant fights against destructive effects of cesium by entering this element to crystalline structure. Totally by mentioned process, this plant enhancing its ability to absorb cesium from environment. Regarding to these two occasions:

1. Increasing absorbed amount of cesium due to enhancing xylem tissue.
2. Not observing any damage to plant's tissue which caused by cesium content because of crystal composing this plant, *C. album*, can be introduced as an appropriate purifying tool (phytoremediator) for removing toxic effects of cesium in environment.

5 Conclusions

In the present study *A. Chlorostachys, C. alata* and *C. album* plants used successfully for phytoremediation purpose. It is important that plants can be able to survive in polluted environment. Cesium uptake by *C. album* plants might be related to its molecule's similarity to potassium. Consequently, performances of potassium channels are effective factor on cesium uptake. After phytoremediation cesium is accumulated in plants as crystals. This mechanism permits plant to absorb more content of radionuclide cesium from environment without any serious damage to plant. As a result this plant causes to increase absorbed element from environment by enhancing its xylem system. These plants, *C. album*, *A. chlorostachs* and *C. alata* can be introduced as a suitable purifying tool for removing toxic effects of cesium in the environment.

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Remediation of Areas Contaminated by Caesium: Basic Mechanisms Behind Remedial Options and Experience in Application

Sergey Fesenko, Brenda J. Howard, Natalya Sanzharova, and Miquel Vidal

Abstract There are many areas around the world contaminated with radiocaesium to different extents due to global fallout from nuclear weapons testing, radiation accidents or inadequate waste disposal practices. In recent decades, a wide range of options for remediation of these areas have been developed, tested and implemented to mitigate the potential doses in such areas. A large amount of data on the effectiveness of remediation options has been generated, together with information on ancillary factors such as technical feasibility and side effects. The chapter aims to provide information on available options for remediation of terrestrial and freshwater ecosystems contaminated with radiocaesium. An associated objective is to provide scientific information on the basic mechanisms which impact on effectiveness of the described remedial options.

Keywords Radiocaesium • Terrestrial and freshwater environments • Remediation • Remedial options

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1 Introduction

Remediation (or remedial action) is normally defined as any measure that intended to reduce radiation exposure from existing contamination through actions applied to the contamination itself (the source) or to the exposure pathways to humans (IAEA 2014). A remediation strategy can be defined as a time-dependent sequence of remedial actions which have been justified and are undertaken in a specified area for a defined time. Although the main objective of a remediation action within a remediation strategy is to reduce or to prevent radiation doses to humans, the provision of reassurance to the population of the contaminated areas is also an important objective of remediation as it helps to maintains public confidence in areas affected by radioactive contamination (Fesenko and Howard 2012).

During ^{235}U fission, 13 isotopes of Cs are formed. Among them, the radioactive isotope ^{137}Cs is of greatest importance, as it has a large fission yield (6.2 %) and a relatively long physical half-life (30.17 years). Caesium isotopes are characterised by high biological mobility, because caesium is an alkali element and a chemical analogue of a biochemically important element K (Nisbet and Woodman 2000 2000; Simon et al. 2002). The dominant aqueous species in soil and aquatic systems is thought to be free Cs^+. An important feature of the behaviour of Cs isotopes is their ability for non-exchangeable sorption (fixation) by the soil solid phase (Smolders and Merckx 1993). The Cs^+ ion forms only extremely weak aqueous complexes with SO_4^{-2}, Cl^-, and NO_3^-, and thus the formation of inorganic complexes is not believed to be a major influence on caesium speciation (Yudintseva and Gulyakin 1968). Cs becomes associated with the clay mineral fraction of soils (Sawney 1964) with a high selectivity. Accumulation of ^{137}Cs in plants depends on soil properties with the concentrations of competing cations exerting a modifying effect. Cs mobility is relatively high in peat and sandy soils (Sanzharova et al. 1996). Remediation of areas affected by radiocaesium is focused on reduction of both ingestion dose from the consumption of contaminated foodstuffs or drinking water and external dose from surfaces contaminated by deposited radionuclides. Inhalation of resuspended material is normally a negligible pathway and its importance is much lower than that from ingestion and external exposure in most contamination scenarios.

A large number of remedial actions to be applied in areas affected by radiocaesium have been developed and applied, especially when there has been significant contamination arising from releases such as the Chernobyl accident (Alexakhin 2009) in 1986, the incident in Goiânia, in 1987 in Brazil (Eisenbud, and Gesell 1997) and recently, the Fukushima Daiichi accident (IAEA 2015). Therefore, in recent decades many remedial options have been developed, tested and implemented in the contaminated areas. As a result, a large amount of data on the effectiveness of various remediation options has been generated, together with information on ancillary factors such as the required resources and costs. The experience gained has been invaluable in quantifying the efficiency of remedial actions. In addition, prominence has been given to identifying many other factors which

affect the potential application of various remediation options such as environmental conditions, radionuclide properties, land use of the affected areas and response from the local population and stakeholders which can affect the effectiveness and impact on the suitability of remediation options (Howard 2000; Fesenko et al. 2000a, b, 2012). Information on effectiveness is often presented as a reduction factor i.e. reduction of radiocaesium concentration in the product or reduction of internal and external dose after application of the remedial option. Alternatively, a percentage reduction in radionuclide concentration in the target medium (i.e. soils and crops) after implementation is reported. The latter is most often used for food treatment options and has, therefore, been used here for these options.

2 Basic Mechanisms Behind Soil-Based Remedial Options Affecting Radiocaesium Mobility

Soils constitute the main long-term reservoir of radionuclides in terrestrial ecosystems. Many remediation options are applied to the soil aiming to modify the soil parameters that affect radionuclide mobility. Soil-based remediation options can be divided into those that alter the soil structure (mechanical treatments) and those that directly modify the chemical characteristics of the soil. The application both of these measures has the advantage that they are easy to implement in most areas as the necessary equipment and expertise is already available. The mechanisms behind soil-based remedial options affecting radiocaesium mobility discussed in this section are illustrated based on data obtained for six case studies: two from Belarus (Dublin and Sawichi), two from Russia (VIUA and Rudnuy) and two from Ukraine (Matevki and Christinovka) located in the areas affected by the Chernobyl accident. A full description of these sites is given elsewhere (Vidal et al. 2000).

2.1 Factors Governing Sorption on Cs in Soils

Cs soil sorption properties vary considerably and are commonly quantified using both the Radiocesium Interception Potential (RIP) and the K and NH_4^+ concentrations in the exchange complex of the soil solid phase and soil solution (Sweeck et al. 1990; Vandebroek et al. 2012). The RIP estimates the capacity of a given soil to specifically sorb Cs and can be readily determined based on routine laboratory experiments (Wauters et al. 1996a). The most common protocol to determine the RIP is based on pre-equilibrating the samples with a solution containing 100 mmol L^{-1} of Ca and 0.5 mmol L^{-1} of K (m_K). After pre-equilibrating the samples, these are equilibrated with a solution with the same K and Ca composition, but labelled with radiocaesium. The distribution coefficients (K_d (Cs)) are obtained by measuring the radiocaesium activity concentrations in the supernatant, before and after the equilibration. The calculated product K_d (Cs)$\times m_K$ defines the RIP value (in mmol kg^{-1})

(Wauters et al. 1996a). The RIP value is related to the content and selectivity of expandable clays, especially illite, vermiculite and other 2:1 phyllosilicates, in which Frayed Edge Sites (FES), which are specific sites for Cs sorption, are present (Sweeck et al. 1990). Other exchange sites are of little relevance for Cs sorption (Brouwer et al. 1983; Cremers et al. 1988; Vidal et al. 1995), except for soils with an extremely low clay content (such as soils with a organic matter content over 90 % or highly sandy podzols). In such soils, the role of regular exchange sites (RES) should be taken into account (Vidal et al. 1995; Rigol et al. 1998).

As Cs sorption is controlled by the FES, the Cs solid-liquid distribution coefficient at these sites (K_d^{FES} (Cs)) accounts for more than 80 % of the total sorption process (Vidal et al. 1995). The K_d^{FES} (Cs) can be predicted by dividing the RIP value by the sum of K and NH_4^+ concentrations in the soil solution, the latter amplified by the NH_4-to-K trace selectivity coefficient in the FES (K_c^{FES} (NH_4/K)) (Sweeck et al. 1990). This parameter, which can be easily quantified by laboratory experiments, ranges from 4 to 8 for soils in which specific sites control Cs sorption quantitatively, and down to 2 in those soils where sorption occurs at regular exchange sites (Wauters et al. 1996b; Rigol et al. 1998). For a more accurate prediction of the value of K_d (Cs), a second term must be added to account for Cs sorption at regular exchange sites (RES) (K_d^{RES} (Cs)), by dividing the sum of the exchangeable K and NH_4^+ by the sum of K and NH_4^+ concentrations in the soil solution (in mmol L^{-1}), assuming a selectivity coefficient NH_4/K of approximately 1 at these sites. The overall equation incorporating sorption at specific and regular sites may be written as follows:

$$K_d(Cs) = K_d^{FES}(Cs) + K_d^{RES}(Cs) = \frac{RIP}{K_{ss} + K_c^{FES}(NH_4/K) \bullet NH_{4ss}^+} + \frac{K_{exch} + NH_{4exch}^+}{K_{ss} + NH_{4ss}^+}$$

For the case of highly saline soils, near to marshlands, with high *Na* concentrations in the soil solution, the equation may be slightly modified to include the potential competitive role of Na and its effect on the quantification of K_d^{FES}, correcting the Na concentration by the Na-to-K trace selectivity coefficient in the FES, K_c^{FES} (Na/K). As this coefficient has values of around 0.02 (Wauters et al. 1996b), the role of Na will only have a significant effect when there is an unusually high Na concentrations. The equation may be simplified by considering that Na and NH_4^+ concentrations are generally much lower than K concentrations, as is the case for most agricultural systems with mineral soils. As the value of K_d^{FES} (Cs) is much larger than the value of K_d^{RES} (Cs), K_d (Cs) is reasonably well predicted by the following equation, except for those soil types (upland, peat soils; soils affected by flooding) in which $NH_4^+{}_{ss}$ can be significant:

$$K_d(Cs) = \frac{RIP}{K_{ss}}$$

To date, attempts to predict the RIP value based on soil properties have only been partially successful. Waegeneers et al. (1999) showed that the RIP value depended

not only on the clay content, but also on the type of clay and geological origin of the soil. In their study involving a stepwise regression analysis to two sets of soils, the clay content alone accounted for up to 71 % of the variance of the RIP in the most favourable set of soils, whereas for another set of soils it explained only 13 % of the variance. The regression improved when the silt content and the pH were added to the clay content, depending on the origin of the soils. In more recent work, Gil-García et al. (2009) showed that clay content alone explained 40 % of the variance of the RIP values, and that there was a good correlation between the two variants [$log\ RIP = 1.8 + 1.0 \times log\ clay$; $r = 0.64$; $n = 108$], while the inclusion of the silt content explained the 60 % of the total variance [$log\ RIP = 1.5 + 0.9 \times log\ clay + 0.45 \times log\ silt$]. When organic soils were excluded from these analyses, the clay content alone explained up to 57 % of the variance, with an improved correlation [$log\ RIP = 2.1 + 0.95 \times log\ clay$; $r = 0.76$; $n = 86$], while the inclusion of the silt content increased the explained variance to up to 62 % [$log\ RIP = 1.6 + 0.8 \times log\ clay + 0.5 \times log\ silt$] (Gil-García et al. 2009). Therefore, although the RIP values can be partially predicted from clay and silt contents, limitations to properly predict RIP from soil properties suggests the need to quantify the RIP for a better estimation of the K_d (Cs) (Gil-García et al. 2011). In agreement with the K_d(Cs) dependence, the CR_{ss}(Cs) (soil solution–plant concentration ratio[1]) is expected to vary inversely with K_{ss} and NH_{4ss}. However, for $K_{ss} + NH_{4ss}$ values higher than 0.5–1 mM, the CR_{ss}(Cs) remains reasonably constant (Smolders et al. 1997a, b; Camps et al. 2004). Therefore, a major increase in NH_{4ss} may lead to an increase in F_v(Cs) due to the different mechanisms affecting changes in the K_d (Cs) (the NH_{4ss} is multiplied by the $K_C^{FES}(NH_4/K)$) and in the CR_{ss}(Cs) (in which NH_{4ss} has the same weight as K_{ss}) (Vidal et al. 1995).

2.2 Mechanisms Related to Mechanical Treatments

Radionuclide migration in soil is a relatively slow process in most types of soil and radionuclides deposited on the soil surface typically remain in the upper soil horizons for many decades. Mechanical treatments are intended to decrease the pool of radionuclides in the rooting zone by dilution achieved by mixing the contaminated topsoil layer with deeper soil layers which have lower radionuclide contents. Mechanical treatments such as shallow, deep or skim and burial ploughing, will have the following positive effects:

- dilution, leading to a lower radionuclide activity concentration in crops;
- transfer of radionuclides down to a soil horizon below the crop rooting area;
- decrease in the resuspension of contaminated soil;
- decrease in the adhesion of contaminated soil to plants and
- reduction of external dose rate.

[1] Soil solution–plant concentration ratio, CR_{ss}, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} plant (dw)) to that in the soil solution (Bq L^{-1}).

Mechanical treatments may also modify the capacity of soils to immobilize radionuclides. Examples include situations when the radionuclide-contaminated layer is mixed with a soil layer of different mineralogical composition, or when soil loosening causes a change in the amount of sorbing surface which determines the extent of binding of radionuclides. To illustrate this phenomenon, Table 1 provides RIP values for a ploughed layer, in comparison with *RIP* values of undisturbed 0–10 cm and 10–20 cm layers soils of five sites (Camps et al. 2004). At the VIUA and Rudnuy sites, a decrease in the RIP values in the ploughed soils was noted due to the presence of a 10–20 cm leached horizon with poor radiocaesium sorption properties. This was shown by the lower RIP values in the 10–20 cm layer than those of the 0–10 cm depth layer. The decrease in RIP values suggests there would be a detrimental effect on radiocaesium mobility, since the resulting ploughed soils would have a lower specific sorption capacity in the rooting area. At sites in Dublin and Sawichi there were similar *RIP* values in unploughed control and ploughed soils no secondary detrimental effects on the *RIP* due to ploughing were observed. At the Mateyki site an increase in the radiocaesium sorption properties was observed due to a heterogeneous soil profile in which the topsoil had a lower specific sorption capacity than that of deeper horizons. At this site, the 10–20 cm soil layer in the unploughed control plot was highly heterogeneous in clay content, with a wide range of *RIP* values, which were often greater than those in the topsoil. Thus, ploughing created a soil profile, with higher *RIP* values than the original soil before ploughing in the 0–10 cm. The data show that ploughing not only redistributes radiocaesium in soil but also may change the top soil sorption properties affecting caesium mobility in the soil and root uptake by plants.

Table 1 Changes in RIP after ploughing undisturbed soils

Soils and treatments	*RIP* (mmol kg^{-1})	Soils and treatments	*RIP* (mmol kg^{-1})
Sawichi		*Mateyki*	
C 0–10	240	C 0–10	70
C 10–20	185	C 10–20	15–810
Ploughed layer	150	Ploughed layer	140
Dublin		*VIUA*	
C 0–10	87	C 0–10	90–485
C 10–20	75	C 10–20	27
Ploughed layer	80	Ploughed layer	39
Rudnuy			
C 0–10	545		
C 10–20	39		
Pl	190		

2.3 Mechanisms Related to Changing the Chemical Characteristics of Soils

The radionuclide activity concentration in plants depends on that in the soil solution and on the ion uptake process. The concentration of radionuclides in the soil solution depends on the total concentration in the solid phase, the radionuclide solid–liquid distribution coefficient (K_d)[2], and the reversibly sorbed fraction (Vidal et al. 2009). For many radionuclides, after some time has elapsed since sorption, the reversibly sorbed fraction will be of a similar order of magnitude for soils with similar properties. Thus, the range of variation should be narrower than the range of variation of K_d for all soils, and, therefore, radionuclide availability is often quantified solely in terms of the K_d. The ion uptake process from soil solution to the plant includes plant physiological aspects, related to nutrient uptake and selectivity. Therefore, (CR_{ss})[3] is assumed to depend on the activity concentrations of radionuclide competitive species in the soil solution. Thus, the soil-plant concentration ratio (F_v)[4] may be described by the following relationship: $F_v = CR_{ss}/K_d$. Therefore, the F_v values decrease with increases in the K_d and/or with decreases in the CR_{ss} (Vidal et al. 2009). The same affect is observed also for T_{ag} (aggregated transfer factor defined as the mass activity density (Bq kg^{-1}) in the plant per unit area activity density, A_a (Bq m^{-2}) in the soil m^2 kg^{-1}(ICRU 2001).

2.4 Effects of Commonly Applied Agricultural Treatments on K_d and T_{ag}

The application of potassium to soils is most effective in reducing F_v(Cs) when the K_{exch} is less than 0.5 meq 100 g^{-1} soil, that is, for K_{ss} in the micro-molar range (lower than 0.5–1 mM). Over this range, additional doses of K fertiliser may have a negative effect on T_{ag}(Cs) transfer, because the decrease in the K_d(Cs) is not compensated for by an increase in the dilution effect in the soil solution, and then in the CR_{ss}(Cs) (Smolders et al. 1997a, b; Camps et al. 2004). Liming may be an effective option for reduction of Cs transfer to plants because in soils with an optimum K_{ss} (over 1 mM), the F_v(Cs) may decrease due to competition between Cs and Ca + Mg for exchange sites in the apoplast of the root cortex (Smolders et al. 1997a, b). Also, the increase in $(Ca+Mg)_{ss}$ may increase the K_d(Cs) in soils, due to the masking of RES, an

[2] Distribution coefficient is the ratio of the mass activity density (A_m, in Bq kg^{-1}) of the specified solid phase (usually on a dry mass basis) to the volumetric activity density (A_v, in Bq L^{-1}) of the specified liquid phase.

[3] Soil solution–plant concentration ratio, CR_{ss}, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} plant (dw)) to that in the soil solution (Bq L^{-1}),

[4] Concentration ratio, Fv, is the ratio of the radionuclide activity concentration in the plant (Bq kg^{-1} dw) to that in the soil (Bq kg^{-1} dw).

expansion of clay interlayers, or a decrease in the concentration of monovalent species in the soil solution (Rigol et al. 1999a). The effect of the use of agricultural treatments, combined with ploughing, on changes in the soil properties and related changes in Cs transfer is illustrated in Fig. 1 which shows changes in ^{137}Cs transfer to plants in a set of soils submitted to different agricultural practices, such as ploughing (Pl), NPK fertilization (NPK) and liming (L) (Camps et al. 2004).

In the first year of the experiments, ploughing (Pl) plus reseeding was the most effective treatment in decreasing radiocaesium transfer to plants in most sites (see

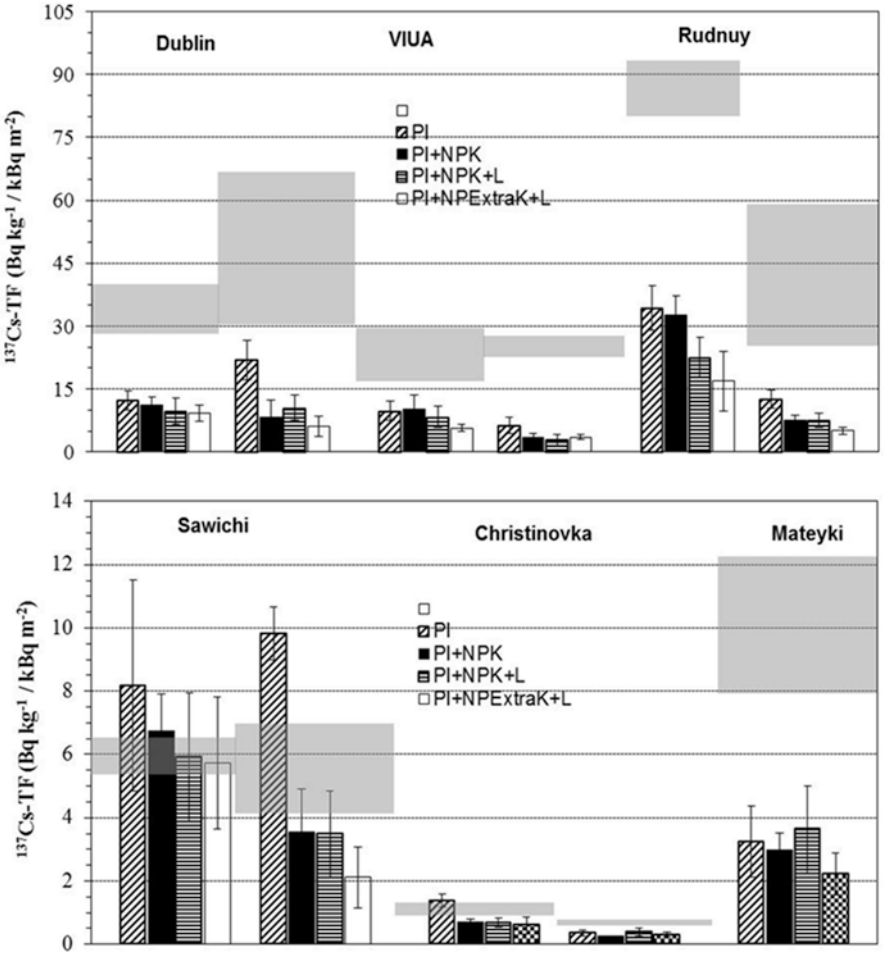

Fig. 1 Effect of agricultural practices on ^{137}Cs aggregated transfer factor (T_{ag}) at the test sites. *Error bars* indicate one standard deviation. Control data with normal application rate of fertilizers are represented by a *hatched area* that includes the mean ± one standard deviation

Mateyki, Dublin, VIUA and Rudnuy sites), while at the Sawichi and Christinovka sites similar or even higher T_{ag} (Cs)[5] values were measured in ploughed (Pl) than in unploughed control plots. The application of NPK fertiliser caused a further decrease in radiocaesium transfer at most sites, especially when applying the extra dose of potassium (Fig. 1), with the exception of the Sawichi site. The effectiveness of liming was minor. Treatment effects became generally more pronounced in the second year. However, the T_{ag}(Cs)s of control soils also decreased with time at some sites (Christinovka and Rudnuy). Key parameters explaining the effect (or lack of) of these treatments on Cs transfer are the potential changes in potassium and NH_4^+ status in the soil exchangeable complex and in the soil solution due to the agricultural treatments. Table 2 summarizes the data related to potassium concentrations in the exchange complex and potassium concentration in the soil solution in the unploughed (control) and the treated plots. As stated above, for potassium concentrations in the soil solution in the micromolar range (lower than 1–2 mM), an increase in the potassium concentration would lead to a decrease in radiocaesium root uptake. Over 1–2 mM, additional application of potassium fertiliser may have a negative effect on radiocaesium transfer, because the decrease in the K_d (Cs) is not compensated for an increase in the dilution effect in the soil solution, and then in the T_{ag}(Cs) (Smolders et al. 1997a, b).

In the first year of the experiments, there was a general increase in the potassium concentrations at several sites, especially in the soil solution, after the addition of extra doses of the NPK fertiliser (see Sawichi, Dublin and VIUA sites), achieving in some cases potassium concentrations in the soil solution in the range of 1–2 mM. At some of the sites, the fertiliser supply was insufficient to increase potassium concentrations over those found in unfertilised ploughed plots, with values remaining in the initial micromolar range (see Christinovka and Mateyki sites). These findings agree with the transfer pattern observed in Fig. 1. The optimum potassium concentrations in soil solution achieved at several sites could not be maintained due to a potassium depletion caused by the increase in nutrient root uptake resulting from the plant growth, or from the increase in biomass in the second year of experiments (compare first and second year at the Dublin and Sawichi sites). The data imply that the application of higher amounts of potassium fertiliser would have maintained an optimum potassium concentration in the soil solution even after a higher plant yield in more mature meadows. The outcome would have been a further decrease in radiocaesium transfer to the crop.

2.5 *Effects of the Application of Organic and Mineral Materials on K_d and T_{ag}*

Application of natural organic and mineral materials to soils is a standard agricultural practice in some regions. The addition of such amendments, usually applied with fertilizers, may improve the agrochemical properties of the soil, leading to an

[5] T_{ag} (Cs) is the ratio of the Cs activity concentration in plant (Bq kg^{-1}) divided by the total deposition on the soil (Bq m^{-2})

Table 2 Effect of agricultural practices on the RIP values, the exchangeable K (K_{exch}, $cmol_c\ kg^{-1}$) and K concentration in the soil solution (K_{ss}, $mmolL^{-1}$)

	$K_{exch,}$ 1st year	2nd year	K_{ss}, 1st year	2nd year
Sawichi				
C 0–10	0.24	0.33	1.7	1.6
C 10–20	0.06	nd	0.3	nd
Pl	0.17	0.07	0.6	nd
Pl + NPK	0.22	0.12	1.0	0.40
Pl + NPK + L	0.30	0.14	1.4	0.47
Pl + NP1.5 K + L[a]	0.30	0.19	1.8	0.60
Dublin				
C 0–10	0.80	0.70	0.8	1.2
C 10–20	0.30	nd	0.4	nd
Pl	0.50	0.50	0.9	nd
Pl + NPK	0.80	0.36	1.4	0.24
Pl + NPK + L	0.68	0.30	1.4	0.35
Pl + NP1.5 K + L	0.82	0.37	1.6	0.43
Christinovka				
C 0–10	0.18	nd	0.10	nd
Pl	0.19	0.18	0.10	0.02
Pl + NPK	0.21	0.24	0.16	0.04
Pl + NPK + L	0.24	0.21	0.13	0.025
Pl + N2P2K + 2 L	0.25	0.20	0.20	0.033
Mateyki				
C 0–10	0.08	na	0.06	na
C 10–20	0.02	na	0.013	na
Pl	0.13	na	0.06	na
Pl + NPK	0.16	na	0.10	na
Pl + NPK + L	0.14	na	0.07	na
Pl + N2P2K + 2 L	0.11	na	0.08	na
VIUA				
C 0–10	0.12	0.08	1.1	0.4
C 10–20	0.06	nd	nd	nd
Pl	0.07	0.09	0.5	0.4
Pl + NPK	0.09	0.07	0.4	0.8
Pl + NPK + L	0.12	0.07	0.4	0.6
Pl + NP1.5 K + L	0.11	0.10	1.1	0.5
Rudnuy				
C 0–10	0.30	0.20	0.9	0.7
C 10-20	0.05	nd	nd	nd
Pl	0.14	0.13	0.4	0.26
Pl + NPK	0.14	0.14	0.4	0.36
Pl + NPK + L	0.17	0.16	0.4	0.5
Pl + NP1.5 K + L	0.17	0.23	0.5	1.1

Notation of Pl + $_xN_xP_yK$ + L means combine application of ploughing mineral fertilizer and lime. x is an increase of application rate in relation to the normal one

nd Not detected, *na* not analysed

increase in crop yields, and possibly also enhancing soil sorption properties (Nisbet 1993; Nisbet et al. 1997). The application of these sorbent materials to the soil modifies the soil solid phase which influences radionuclide uptake in two ways: (1) by increasing the sorbing capacity for radionuclides, and (2) by modifying the composition of the soil solution. To be effective such materials must increase the radionuclide sorption potential (or K_d) of the soils. For Cs, this can readily be achieved by increasing the RIP in the soil. Because only low doses of amendments can be used for an economically justifiable remediation strategy, the K_d of radiocaesium in the added material should exceed the normal values in the soils to be treated by several orders of magnitude to have a significant effect at field level. Therefore, the sorption characteristics of the materials have to be determined in the laboratory before being used at field level (Valcke et al. 1997; Camps et al. 2003). Materials with high mineral matter content, enriched in 2:1 phyllosilicates, are the most suitable materials for sorbing radiocaesium. The addition of organic material would lead to an increase in the (1) organic matter in soil, (2) contents of nutrients and microelements, (3) CEC and (4) $(Ca+Mg)_{exch}$. These increases may lead to a lack of effect of the treatment on radiocaesium sorption or even a detrimental effect in reducing F_v(Cs). A case study, using field plots from Ukraine, Belarus and Russia, is used here to illustrate how the addition of materials does not necessarily lead to a decrease in radiocaesium transfer to plants (Vidal et al. 2001; Camps et al. 2003). Table 3 presents a summary of the main general characteristics of the soils and amendments used in this study.

Figure 2 provides data on the changes of T_{ag}(Cs) for the study sites mentioned earlier, including both the control (hatched area) and amendment plots (vertical

Table 3 Characteristics of soils and amendments tested

Soil amendments	pH	OM	CEC	K_{exch}	NH_{4exch}	Sand	Clay	Texture
Sawichi								
Control	4.4	7.9	25	0.26	0.22	72	5.3	Sandy-loam
Sapropel	4.8	38	86	0.59	0.92	47	27	Sandy-clay loam
Turf	5.9	45	85	0.44	2.1	63	0.7	Sandy-loam
Dublin								
Control	6.3	78	140	0.68	0.50	nd	1.3	–
Mineral soil	6.5	0.1	8.2	0.08	0.08	83	0.6	Loamy-sand
Mateyki								
Control	6.4	44	57	0.24	0.08	nd	12	–
Sandy soil	5.9	0.2	4.4	0.06	0.08	90	1.6	Sandy
Loamy soil	6.1	0.8	10	0.14	0.08	55.9	12.9	Sandy-loam
VIUA								
Control	6.7	3.5	9.7	0.12	0.20	79.8	3.4	Loamy-sand
Rudnuy								
Control	5.6	13	15	0.13	0.11	90.2	3.1	Sandy
Phosphorite	6.8	2.4	8.6	0.40	0.09	26.9	20.1	Loamy-silt
Polygorskite	7.7	4.3	12	0.41	0.08	1.4	81.1	Clay

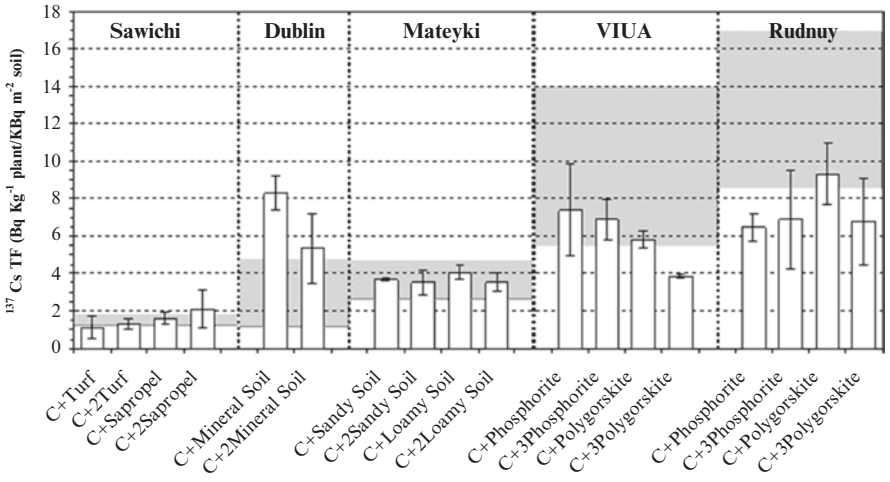

Fig. 2 Effect of soil amendment addition on ^{137}Cs aggregated transfer factor for plants Tag. *Error bars* indicate one standard deviation. Control treatments are represented by a *hatched area* that includes the mean ± one standard deviation

bars) (Vidal et al. 2001). The addition of amendments did not lead to a decrease in transfer in most of the treatments. A positive effect of phosphorite and especially polygorskite amendments was determined at only at a few sites. Radiocaesium transfer to the plants was reduced by around 1.5–2-fold at the VIUA site, where the highest amount of polygorskite was used, and at the Rudnuy site, where two amendments were applied. At the Dublin site, a negative effect was observed when using the mineral amendment, whereas at the Mateyki site a negligible effect was observed for both amendments tested.

RIP values were determined in control and amended soils (Table 4). Although RIP values were highly variable, the RIP values of the amended soils seemed to increase in only a few scenarios. This was the case at the Mateyki site when adding the highest amount of loamy soil, and at the Rudnuy site when adding polygorskite clay and phosphorite. The improvement in the RIP values was related to the much higher values of the RIP of the amendment related to the untreated soils. The low doses prevented a significant, positive effect in increasing the RIP and then reducing radiocaesium transfer in most cases, although the increase in RIP values at the Rudnuy site when phosphorite and polygorskite were applied corresponded to the decrease in radiocaesium transfer observed at field level. The Dublin site constituted a different scenario because the RIP values decreased when adding the mineral material, as the mineral soil added had a sandy texture, with lower clay content than the Dublin soil. This amendment also had a lower affinity for radiocaesium, with a lower RIP than that of the Dublin control soil. The resulting decrease in RIP may explain the increase in radiocaesium transfer to the plants observed at this site. The effect of the amendment application on the soil solution can also be evaluated by quantifying the concentration of K and NH_4^+ of the soil solution for all control and

Table 4 RIP (mmol kg^{-1}), and K and NH_4^+ in soil solution (mmol L^{-1}) of field samples and distribution coefficients (K_d (Cs); L kg^{-1})) of laboratory samples

Soils amendments	Appl. doses (%)	Initial RIP	Mixture RIP	K	NH_4^+	Initial K_d	Mixture K_d
Sawichi							
Control	–	400	–	1.2	0.8	45	–
Turf	1.4	65	460	0.9	0.4	36	45
	3.0	–	390	0.9	0.6	–	47
Sapropel	1.3	332	400	0.9	0.7	116	40
	2.9	–	360	1.1	0.6	–	39
Dublin							
Control	–	161	–	0.9	0.5	40	–
Mineral soil	17	15	119	1.2	0.6	10	29
	33	–	121	1.3	0.6	–	21
VIUA							
Control	–	24	–	1.0	2.0	25	–
Phosphorite	1.0	810	29	0.6	–	73	26
	3.1	–	20	0.5	1.5	–	26
Polygorskite	0.5	7700	23	0.7	–	376	26
	1.4	–	17	0.5	0.4	–	29
Rudnuy							
Control	–	130	–	0.6	1.7	99	–
Phosphorite	1.4	810	210	0.3	2.2	173	104
	4.2	–	250	0.4	2.0	–	104
Polygorskite	0.7	7700	180	0.4	2.0	902	101
	1.9	–	200	0.3	1.9	–	133
Mateyki							
Control	–	207	–	0.06	0.18	577	–
Sandy soil	6.3	31	170	0.03	0.18	137	546
	12.6	–	250	0.05	0.17	–	523
Loamy soil	6.3	455	260	0.04	0.20	–	–
	12.6	–	320	0.05	0.15	–	–

treated sites (see Table 4). Although a high variability was observed for field samples, the addition of the amendments did not lead to a significant variation in potassium concentrations in the amended soils. Considering the low *potassium* concentrations in some of the control plots (see VIUA, Rudnuy and Mateyki sites), it would have been interesting to increase the fertilizer rate along with the use of the amendments as for this range of K concentration (around 0.5–1 mM), an increase in the K concentration in the soil solution would lead to a decrease in radiocaesium root uptake (Smolders et al. 1997a, b). A significant decrease in the NH_4^+ concentration occurred at the VIUA site when amended with polygorskite. This outcome suggested a beneficial secondary effect when using this amendment, explaining the

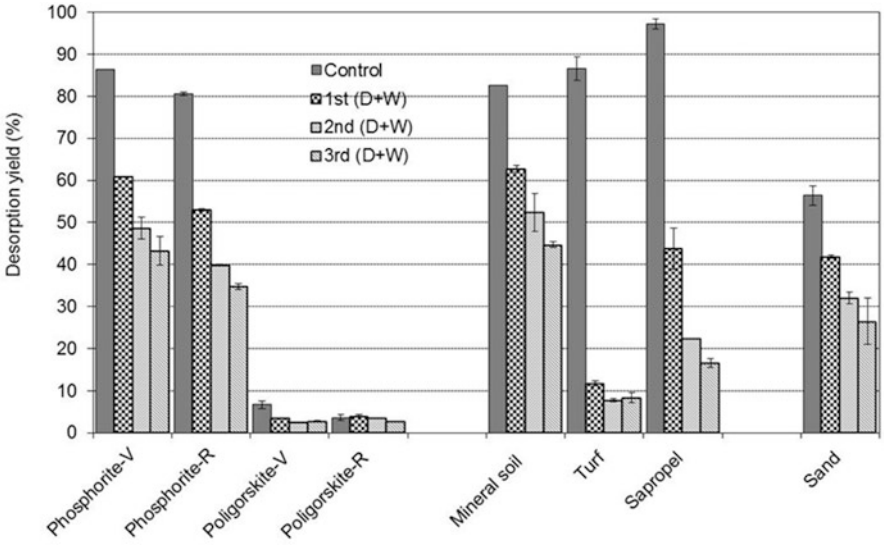

Fig. 3 Variation of radiocaesium desorption yields in amendments exposed to 1, 2 and 3 drying-wetting (D+W) cycles. *Error bars* indicate one standard deviation

decrease in the radiocaesium transfer to plants at the VIUA site when using polygorskite, since both RIP and K^+ concentrations remained constant.

As a way to further characterize the materials (and as a suggested strategy to previously characterize materials at laboratory level before being applied at field). Table 4 summarizes the K_d values obtained for soils, amendments and mixture in laboratory conditions, in which the K_d was calculated in an ionic media representative for each soil solution. Only in the case of polygorskite clay at the Rudnuy sites the K_d (Cs) was one order of magnitude higher than in the soil. In a few cases, the amendments had a much lower sorption capacity than that of the corresponding soil, as was the case of the mineral soil at the Dublin site and the sandy soil at the Mateyki site. The K_d of the soil+amendment mixtures confirmed the lack of the suitability of the materials and administration rates tested. Promising materials, such as polygorskite, were used at such low doses that their effect was diminished. Other materials, such as the mineral soil at the Dublin site, were used in higher doses, but their low K_d led to a decrease in the K_d of the mixture with respect to the initial soil.

Additionally, there may be a significant decrease in the reversibility of the Cs sorption in the medium and long term, which is responsible for a decrease in the Cs root uptake with time and that must be considered for a better assessment of the eventual amendment performance (Absalom et al. 1999; Rigol et al. 1999b). Figure 3 shows the variation of radiocaesium extraction yields in amendment samples exposed to drying-wetting cycles, which has previously been shown to be a good laboratory approach to simulate dynamics at field level (Rigol et al. 1999b). The initial extraction yields ranged from 3.6 % in the polygorskite, to 97 % in the sapropel. Therefore, initial extraction yields were generally high in all the amend-

ments tested, showing that the initial sorption was almost reversible, with the exception of the polygorskite clay, which showed an extremely low radiocaesium extraction yield. Application of such cycles leads to decreases in the extraction yields proportional to the number of cycles applied, indicating that the amount of radiocaesium fixed by the amendment could increase with time. Therefore, interaction dynamics were significant for all samples; with a decrease in the extraction yields of up to one order of magnitude (see the turf material). Again, the polygorskite clay was an exception to this pattern as its final extraction yield was only slightly lower than the initial low yield.

2.6 Prediction of the Effect of the Materials from Sorption and Desorption Data

The extent of the effect of the addition of amendments, in terms of changes in the sorption-desorption pattern of the soil, can be rationalized based not only on the changes in the RIP in the soil-amendment mixture, but also by correcting the fraction of radiocaesium reversibly sorbed at a given time. As stated before, changes in the radionuclide sorption-desorption pattern in the soil will be the predominant effect affecting root uptake, considering that only small changes in the exchangeable complex and in the soil solution can be anticipated due to the low amendment doses used. The equation for a numerical estimation may be written as follows for the case of radiocaesium:

$$Effect = \frac{RIP_s \cdot \left(1 - w_{amend}\right) / f_{rev,s} + RIP_{amend} \cdot w_{amend} / f_{rev,amend}}{RIP_s / f_{rev,s}},$$

in which RIP_s refers to the radiocaesium sorption potential of the soil, RIP_{amend} refers to the radiocaesium sorption potential of the amendment, w_{amend} is the dose in grams of amendment per gram of soil-amendment mixture, $f_{rev,s}$ is the fraction of radionuclide reversibly sorbed in the soil, and $f_{rev,amend}$ is the fraction of radionuclide reversibly sorbed in the amendment. This equation is time dependent since radiocaesium reversibly sorbed fraction will significantly change with time. This equation is useful to calculate the amendment dose to achieve a given effect, or to predict an effect with an affordable administration rate, besides also considering significant secondary effects that may affect radionuclide root uptake and eventual concentration in plants (such as changes in the exchangeable complex and in the soil solution composition and plant biomass). Besides, the term RIP can be substituted by the K_d (*Cs*), if the radiocaesium distribution coefficient is calculated at the same soil solution scenario for both the soil and the amendment. Unless the sorption capacity of the materials is at least 2–3 orders of magnitude higher than that of the untreated soils no effects will be observed in Cs mobility (such as root uptake), considering the low administration rate (often lower than 5 %) that can be applied at field level. From the materials tested in the study described above only polygorskite could be considered as an adequate

material, although the low administration rate (<1.9 %) made it difficult to observe an unequivocal beneficial effect on reducing Cs transfer.

3 Remedial Options for Agriculture

The ingestion pathway dominates in many contamination scenarios. Therefore, the application of remedial options to intensively and extensively farmed areas is a critical part of many remediation strategies. Subsequently, remedial options need to be targeted on various media (soil, plants, and water) and contamination pathways from the media to crops. The remedial options for agricultural systems should not only be aimed at addressing dose concerns, but also a wide range of other issues such as maintaining the local economy, promoting/upholding consumer trust and ensuring appropriate disposal of wastes (Howard 2012). The soil crop pathway includes (1) arable land used for the production of crops intended for the human food chain (including cereals, vegetables and horticultural crops, and fruit) and for non-food crops for industry (such as flax, bioenergy or biofuel crops) and (2) grassland[6] used for the production of fodder crops such as hay and silage intended as feed for animal consumption. The various available remedial options that intervene along the soil-crop pathways can be grouped according to two main tasks, namely:

- removing or burying most of the contamination, which also reduces external and potential inhalation doses to workers and
- reduction of the soil-to-plant transfer of radionuclides, through a variety of techniques including forms of ploughing, crop selection and soil treatments.

3.1 Selection of Crops and Varieties with Low Accumulation of Radiocaesium

Radiocaesium transfer to plants is highly affected by its physical and chemical properties, soil properties, and the characteristics of different plant species and varieties (Table 5) (Sanzharova et al. 1996). The selection of crops and/or varieties with lower accumulation of radiocaesium is a remedial option is relatively easy to implement and acceptable to farmers as it requires little additional cost and can be implemented within routine farming practice. The decision on which crop or variety to grow should take into account the yield that would be obtained and the relative difference in radiocaesium accumulation between the former and new crop. The main

[6] The term grassland is used here to refer to both cultivated and uncultivated land used as either pasture for grazing animals or for growing fodder. Pasture is used in the report when referring to land used for grazing animals. The term meadow is used in the report for case studies in the FSU and refers to uncultivated grassland used for grazing animals or to grow some fodder crops.

Table 5 Radiocaesium transfer factors for different plant species, based on IAEA TECDOC 1616 (Adopted from Sanzharova et al. 1996)

Plant group	Plant compartment	N	Mean	Min	Max
Cereals	Grain	470	2.9×10^{-2}	2.0×10^{-4}	9.0×10^{-1}
	Stems and shoots	130	1.5×10^{-1}	4.3×10^{-3}	3.7
Maize	Grain	67	3.3×10^{-2}	3.0×10^{-3}	2.6×10^{-1}
	Stems and shoots	101	7.3×10^{-2}	3.0×10^{-3}	4.9×10^{-1}
Leafy Vegetables	Leaves	290	6.0×10^{-2}	3.0×10^{-4}	9.8×10^{-1}
Non-leafy Vegetables	Fruits, heads, berries, buds	38	2.1×10^{-2}	7.0×10^{-4}	7.3×10^{-1}
Leguminous Vegetables	Seeds and pods	126	4.0×10^{-2}	1.0×10^{-3}	7.1×10^{-1}
Root crops	Roots	81	4.2×10^{-2}	1.0×10^{-3}	8.8×10^{-1}
	Leaves	12	3.5×10^{-2}	6.0×10^{-3}	4.5×10^{-1}
Tubers	Tubers	138	5.6×10^{-2}	4.0×10^{-3}	6.0×10^{-1}
Grasses	Stems and shoots	64	6.3×10^{-2}	4.8×10^{-3}	9.9×10^{-1}
Fodder Leguminous	Stems and shoots	85	1.6×10^{-1}	1.0×10^{-2}	1.8
Pasture	Stems and shoots	401	2.5×10^{-1}	1.0×10^{-2}	5.0
Herbs	Stems, leaves	4	6.6×10^{-2}	4.8×10^{-3}	2.8

Table 6 Variety-specific differences in ^{137}Cs accumulation in different agricultural plants (Adopted from Alexakhin 1993)

Plants	No of varieties studied	Range of transfer factor (B_v) values specific for varieties, dimensionless	Ratio of maximum B_v to minimum B_v
Spring wheat, grain	15	0.015–0.046	3.1
Winter rye, grain	11	0.015–0.068	4.5
Barley, grain	11	0.013–0.041	3.2
Oats, grain	4	0.018–0.072	4.0
Potato, tubers	5	0.029–0.058	2.0
Beetroots, roots	4	0.065–0.089	1.4
Tomato, fruit	5	0.01–0.029	2.9
Lupine	16	0.032-0.112	3.5
Clover	8	0.016–0.057	3.5
Vetch	8	0.023–0.065	2.8

reasons for the plant related variations in radiocaesium transfer factors is that crop differ in many factors such as their requirement for certain nutrients such as K, P etc.; in detoxification and exclusion mechanisms; specific patterns of distribution of roots in the soil and rhizosphere properties such as the presence of mycorrhiza (Smolders and Merckx 1993).

Crops selection for cultivation using normal agricultural practices needs to take into account the deposition density of soils and reported F_v values for different crops/cultivars. For example, crops with high F_v values for the type of soil are not

grown in areas with a high deposition density of radiocaesium. Evaluation of the major varieties of the farm plants in areas affected by the Chernobyl accident demonstrates more than tenfold inter-crop differences and up to 4.5-fold for inter-variety differences (Table 6). When new varieties of crops are included into an established crop rotation there may need to be a reassessment of fertilisation schemes due to a modification of nutrient requirements. Evaluation of relevant information and dialogue with farmers or other operators will help to identify which crop variety substitutions are appropriate and ensure suitable modified agricultural practices are adopted. A market for alternative crop varieties should exist or would need to be created. Differences in accumulation of radiocaesium by different varieties provides a possibility for selection of plants and plant cultivars with low accumulation of radionuclides to be used in the contaminated areas if other requirements are met such as productivity, resistance to insects and plant susceptibility to diseases. The effectiveness of this approach, in terms of the crop contamination reduction, can vary on average from five- to tenfold, but could be higher if new varieties with low accumulation of radiocaesium are identified.

3.2 Ploughing and Soil Removal

There four different ploughing options that can be used for remediation of the agricultural lands contaminated with radiocaesium. Ordinary ploughing and rotary cultivation or disking are part of routine agricultural practice and can be used in arable lands annually or as a part of amelioration (radical improvement) of meadows or pastures. In the latter case, the use of land is restricted for one or two years until the grass has re-established. Other options considered in this section are beyond normal farming operations and require specialised ploughing equipment to be delivered to the remediation sites. All ploughing options are based on mechanical redistribution of radionuclides in the soil profile through dilution or removal of radiocaesium from the top soil. These options can reduce both external and internal exposures. External exposure is reduced because the more contaminated deeper layers are placed lower in the soil profile and the upper less contaminated soil layer serves as a shielding media against irradiation. Internal exposure is reduced because the transfer of radiocaesium from soil to plant is lowered as the plant roots are in less contaminated soil (Table 7). To be effective, ploughing should only to be applied once and should not be carried out again because the buried radionuclides can then be placed higher up the soil column and in the rooting zone once more.

An ordinary single-furrow mouldboard plough can be used to mix radiocaesium in the top 18–30 cm of the soil profile. Much of the contamination at the surface will be buried more deeply in the soil thereby reducing plant uptake by up to threefold, with an average reduction factor of 2.0. External dose may be reduced from two- to fivefold, depending on the depth of ploughing (RIARAE 1987, 1988). An average reduction factor for root uptake of three- to fivefold may be achieved from deep ploughing with a maximum reduction factor of 10 (Alexakhin 1993;

Table 7 Reduction factors for different types of ploughing options implemented for remediation after the Chernobyl accident (Adopted from Alexakhin 1993; Bogdevich 2002; Fesenko et al. 2007)

Remedial options	Ploughing depth, cm	^{137}Cs average reduction factor in plants	Reduction factor of external dose rate
Soil removal	5	10–20	10–100
Ordinary ploughing (first application)	18–25	2.0 (1.5–3)	2–4
Deep ploughing	30–50	3.0 (2.0–7.0)	2.0–20.0
Skim and burial ploughing	50–70	>20	>20
Rotary cultivation, disking (first application)	10	1.7 (1.5–2.0)	1.0–1.5

Bogdevich 2002; Fesenko et al. 2007; Maubert et al. 1993; Vovk et al. 1993). The external dose may be reduced by 2–20 folds with the highest reduction factors achieved for complete inversion of soil (Alexakhin 1993; Bogdevich 2002; Fesenko et al. 2007). The measure would not be suitable in regions with thin topsoils as soil fertility and structure would be detrimentally affected. There may be resistance to topsoil burial because of the associated impact on soil-dwelling and other flora and fauna. Future restriction on deep tilling may be imposed to ensure that the radiocaesium is not returned to the rooting zone although subsequent normal ploughing (to ca. 25 cm depth) will not bring much contamination back to the surface. The effectiveness of rotary cultivation, disking is not as effective as other ploughing options with an average reduction of radiocaesium transfer to plants up to two times (Table 7). The option can only be applied to natural, non-arable soils as part of meadow amelioration techniques.

Although, all these options were used after the Chernobyl accident (Alexakhin 2009) their applications were restricted mainly to the regions with fertile clay soil. The limited application of this option can also be explained by the environmental constraints in its application, since most of the soils in the most contaminated areas have a thin fertile layer of soil. If no plants are present, a specialist plough with two ploughshares can be used for skim and burial ploughing. The first ploughshare skims off a thin layer of contaminated topsoil (ca. 5 cm; but adjustable) and buries it at a depth of about 45 cm. The deeper soil layer (ca. 5–50 cm) is lifted by the second ploughshare and placed at the top without inverting the 5–45 cm horizon. Therefore, much of the contamination at the surface will be buried deeply in the soil profile. This procedure reduces both external exposure and root uptake from the contaminants, the negative effect on soil fertility which occurs in deep ploughing is minimised and resuspension is also reduced. More than a tenfold reduction in the contamination of the upper soil layers may occur, if the skim and burial ploughing is optimised according to the contaminant distribution in the soil. An associated reduction in soil-to-plant transfer of 10 and in external dose of around 20-fold may be achieved (Roed et al. 1996).

Skim and burial ploughs need to be available for this remedial option to be applied at a large scale and this may be a problem in the initial stages of remediation as these ploughs are not normally in widespread use. The side effects are similar to those above for deep ploughing. In areas affected by the Chernobyl accident the option was used on a limited scale because of similar environmental constraints to those outlined for deep ploughing (namely thin top-soils as soil fertility and detrimental effect on soil structure). Ploughing may substantially change the landscape and the soil properties. Soil fertility may also be reduced requiring additional fertilisation if the land is used for crop production. There may also be negative effects on biodiversity, particularly for soil dwelling organisms, and therefore soil functioning such as decomposition rates. Field drainage systems may be destroyed leading to waterlogging. This remedial option may result in resuspension of radionuclides associated with small soil particles during and after ploughing until the formation of a new root mat in the upper soil layer.

The most contaminated top soil layer can be removed using road construction equipment such as graders, bulldozers, front end loaders, excavators and scrapers or a turf harvester. Removal of the upper soil will take away much of the contamination. The depth of the soil layer which can be removed depends on the thickness of the fertile layer of contaminated soil if applied for agricultural soils. If the original depth of fertile soil layer is not greater than that removed then additional fertile soil would need to be added with acceptably low radionuclide activity concentrations (topsoil replacement). The removal of much of the contamination at the surface will greatly reduce (1) radionuclide uptake by plant roots; (2) external exposure, and (3) resuspension of radionuclides from the soil (Alexakhin 2009). Topsoil removal (or replacement) was used as a remedial option in the FSU following the Chernobyl accident, in Spain after Palomares incident, in Brazil after the Goiania incident and most recently and extensively in the areas affected by Fukushima Dai-ichi accident (IAEA 2015).

After top soil removal, the reduction in radiocaesium contamination of the top soil may reach 10–100 folds, whilst that for soil-to-plant transfer may be 10–20 folds (IAEA 2006, 2015; Alexakhin 2009). In some cases it may be advantageous to remove part of the vegetation cover before removing the layer of soil. The efficiency of removal of the surface layer may be affected by the degree of optimisation achieved in a wide range of factors. These include: the thickness of the removed layer, surface unevenness, presence of rock and stones, soil texture and moisture content and vertical radionuclide distribution relative to the depth of the soil removed. Soil removal may generate a large volume of contaminated waste and issues related to disposal of this waste should be carefully planned before remediation starts.

3.3 Arable Soil Fertilization

Application of mineral fertilizers is routinely used in agriculture to enhance crop yields. The fertilizer is normally comprised of nitrogen, phosphate and potassium fertilisers which are mixed into the soil by harrowing or ploughing before planting/

sowing of arable crops. Mineral fertilizer application rates are adjusted according to crop requirements based on the soil properties, recommended crop cultivation technologies and existing farming practice (Alexakhin 1993; Alexakhin et al. 1996; Nisbet 1993). Application of mineral fertilisers as a remedial option involves a change in both the ratio and application rates of the individual elements (i.e. N, P and K) in the NPK mix applied onto contaminated lands. The use of fertilisers to reduce plant root uptake of radiocaesium are based on decreasing the Cs:K ratio in the soil solution whilst maintaining optimal growth conditions for plants (Fesenko et al. 2007). Mineral fertilisers have been used extensively in all three FSU countries to reduce uptake to plants from contaminated soil in areas affected by the Chernobyl accident. The numerous studies performed after the Chernobyl accident has made it possible to identify the optimal ratios of nutrients for land contaminated by radiocaesium in the contaminated areas (Table 8). In particular, because potassium is a chemical analogue for caesium its application in elevated rates was identified as an effective remedial option to reduce the accumulation of radiocaesium in crops. The optimum ratio of minerals to achieve the maximum reduction in root uptake of radiocaesium was determined to be a N:P:K ratio of 1:1.5:2 (RIARAE 1988). The ratios given are the relative amounts used for each element (N, P and K) for remediation compared with that used for normal application rates. Thus, N:P:K of 1:1.5:2 means the same amount as for normal practice is applied for *N*, 1.5-fold higher for P and twofold higher for K.

Table 8 Reduction factor for radiocaesium accumulation in farm crops for different combinations of mineral fertilisers on leached chernozem

Application rate of fertilisers	Winter wheat	Spring wheat	Oats	Barley	Mean
$P_{60}K_{60}$[a]	1.0	1.0	1.0	1.0	1.0
$N_{90}P_{60}$	1.0	1.0	1.0	1.0	1.0
$N_{90}K_{90}$	1.0	1.0	1.0	1.0	1.0
$N_{60}P_{60}K_{60}$[b]	1.06	1.1	0.9	1.0	1.0
$N_{90}P_{60}\hat{E}_{60}$	1	1.1	0.75	1.1	1.0
$N_{120}P_{60}K_{60}$	0.8	0.7	1.0	0.8	0.8
$N_{150}P_{60}K_{60}$	0.7	0.8	0.6	0.6	0.7
$N_{90}P_{30}K_{60}$	1.2	1.2	1.0	1.0	1.1
$N_{90}P_{60}K_{60}$	1.2	1.0	1.2	1.4	1.2
$N_{90}P_{90}K_{60}$	1.2	1.1	1.2	1.3	1.2
$N_{90}P_{120}K_{60}$	1.3	1.4	1.3	1.5	1.4
$N_{90}P_{120}K_{120}$	1.2	1.5	1.3	1.7	1.4
$N_{60}P_{60}K_{90}$	1.5	1.8	1.6	2.3	1.8
$N_{60}P_{60}K_{120}$	1.7	1.9	2.0	2.0	1.9
$N_{60}P_{60}K_{150}$	1.8	2.0	2.0	2.3	2.0
$N_{60}P_{90}K_{120}$[c]	1.9	2.0	2.0	2.0	2.0

[a]Notation of $N_xP_yK_z$ means mineral fertilizer application rate where x kg of active N per ha, y kg of active P per ha, z kg of active K per ha are applied

[b]Application rate under normal conditions

[c]Optimal combination

During crop cultivation on contaminated soils, *N* fertilizer application rates need to take into account the expected yield. Increasing *N* application can increase radiocaesium transfer to plants due to soil acidification and its effect on K_d(Cs) (RIARAE 2006). Ammonium fertilizers should, therefore, be avoided, and N fertilizer should be applied in the form of nitrate (Nisbet 1993). Supplementary Mg fertilisation and liming may be required to maintain an optimal ionic equilibrium in both soil and plants. On acid and weakly acid soils, mineral fertilizers are only applied after liming, since the use of mineral fertilizers alone, especially in acidic forms, may increase the acidity of the soil solution and increase radiocaesium transfer to crops (Fesenko et al. 2012). The maximum effect of mineral fertilisers' application can be achieved on pure soddy podzolic soils. Application of mineral fertilizers as recommended in the first post-Chernobyl recommendations for the long-term period (RIARAE 1988) may reduce radiocaesium transfer to plants by two to fivefolds (Table 9). Enhancing the rate of P fertilizer application within an NPK fertilizer reduces radiocaesium uptake by plants on mineral soils by 1.5–3-fold. Factors influencing the effectiveness of the option for radiocaesium are potassium status of the soil/soil solution (see above) and the type of crops (Fesenko et al. 2012).

Table 9 Summary of data for radiocaesium on the effectiveness of mineral fertilizers with recommended N:P:K ratio (Adopted from Brown et al. 1995)

Soils	Country	Plants	Average reduction of ^{137}Cs transfer to crops
Soddy podzolic	Belarus	Winter rye	2.8
		Barley	1.1
		Oats	1.8
Soddy podzolic	Russia	Barley	1.5
		0ats	1.5
		Winter rye	1.5
		Maize silage	1.7
		Potato	1.8
		Beetroots	2.1
		Vegetable	2.2
Soddy podzolic	Ukraine	Cabbage	1.4
		Carrots	1.4
		Beetroots	2
		Oats	1.1
Sandy soils		Maize silage	1.5
		Beetroots	1.8
		Lupin	1.1
		Pears	1.6
Peaty		Cabbage	4
		Maize silage	5
		Beetroots	3
		Rape	2–3
		Grass	1.5

Although there may be changes in nutrient status etc., the impact is likely to be small for intensively managed arable soil where mineral fertilisers are routinely applied at normal rates. K and P are the key elements which improve soil fertility and, therefore, increases in their application rates often leads to higher crop yields. Assuming that this remedial option is carried out for soils where the exchangeable K is sub-optimal for the crop, there will be a potential increase in crop yield and quality. Application of phosphates may reduce the availability of essential micronutrients which, when complexed with phosphates, are also of low solubility.

3.4 Liming of Arable Soils

Liming of soil is part of conventional agricultural practice for acidic soils which are present in some areas that are most affected by the Chernobyl accident. The effectiveness of liming is mainly based on neutralization of soil solution acidity, displacement of hydrogen ions from the soil sorbing complex and calcium saturation in the exchangeable complex. Thus, application of lime to maintain a neutral pH in the soil solution may substantially reduce radiocaesium transfer to plants since radiocaesium accumulation in plants is enhanced in acidic soil solutions (Nisbet 1993).

Table 10 Summary of data for radiocaesium on the effectiveness of liming with recommended application rates (Adopted from Brown et al. 1995)

Country	Soil	Plants	Reduction in Cs transfer to plants
Belarus	Mineral	Cereals	1.5–3
	Organic	Cereals	1.5–3
Russia	Mineral	Barley	1.9
		Oats	2.1
		Winter rye	1.8
		Potato	2.3
		Maize	1.6
		Beetroot	2.1
Ukraine	Mineral	Maize silage	1.8
		Beetroot	2.0
		Lupin	2.7
		Peas	2.4
	Organic	Cabbage	2
		Maize	1
		Beetroot	1

Based on the post Chernobyl research in the contaminated areas the application rates of lime for these regions was recommended to be increased on average by 1.5 times from the normal application rate. To maintain the effect, the liming should

be applied every four-five years with an application rate from 2 to 10 tonnes per ha (Table 10). Liming can be applied in different forms such as: dolomite powder, calcareous tuffs (travertine[7]) and marlstone[8] (Gulyakin et al. 1978; Ratnikov et al. 1992). The amount of lime used depends on pH and other soil properties (CEC, calcium status, granulometric composition, organic content). Lime is normally applied as an ameliorant[9] to soils of a low pH or low Ca status, but the application rate and the frequency of application is determined by the soil fertility (Table 10). It is normally ploughed into the soil before arable crops are planted or sown. When the total amount intended to be applied over a growing season exceeds 8 t ha^{-1} then lime is applied on two occasions: half during ploughing and half during the plant growth period as this has a greater sustained impact on soil fertility (Bogdevich 2002).

In European countries (with higher levels of soil fertility compared with the FSU countries) maintenance liming normally takes place every 5 years (0.5–2 t $CaCO_3$ ha^{-1}; depending on the soil pH) with the aim of reaching pH 7 in mineral soils and pH 6 in organic soils (Woodman and Nisbet 1999). Liming may reduce radiocaesium transfer to farm products by 1.5–4.0 folds (Table 11) respectively depending on factors such as the initial soil pH, CEC and calcium status, hydrological regime of the soil, productivity and type of crops. The effectiveness is usually higher on organic soils than on mineral soils (Fesenko et al. 2012) (Table 10). Liming can change the microbiological status of the soil and may affects water quality. There is a potential secondary negative effect on radiocaesium transfer in soils with a low potassium

Table 11 Summary of data for radiocaesium on the effectiveness of organic fertilizers (Adopted from Brown et al. 1995)

Country	Fertiliser type	Plants	Reduction in plant uptake
Belarus	Manure, 60 t ha^{-1}	Potato	1.5–2.8
	Sapropel, 60 t ha^{-1}	Various crops	1.7
Russia	Manure	Maize silage	2.4
		Barley	1.6
		Oats	1.3
		Winter rye	1.4
		Potato	2.9
		Beetroot	2.2
		Vegetables	2.9
Ukraine	Manure, 50 t ha^{-1}	Maize silage	2
		Beetroots	4
		Lupin	1.3
		Pears	2
	Sapropel, 100 t ha^{-1}	Beetroot	4–5
		Peas	2
		Lupin	3
		Maize silage	2

[7] Travertine is a terrestrial sedimentary rock, formed by the precipitation of carbonate minerals from solution in ground and surface waters.

[8] Marlstone is a calcium carbonate or lime-rich mud or mudstone which contains variable amounts of clays and aragonite

[9] A substance added to soil to improve the growing conditions for plant roots

adsorption ratio (much higher Ca than K concentrations) and low K concentration (lower than 0.5 mM) in the soil solution. In these cases, there may be a partial loading of Ca at the FES, leading to a lower K_d(Cs) values (Fesenko et al. 2012). Therefore, liming should be accompanied by K fertilisation to prevent this process. Liming may also induce microelement deficiencies in crops (in particular, Mn, Zn) and additional application of micro fertilizer[10] may be necessary.

3.5 Application of Organic Materials to Arable Soils

The application of organic materials may reduce radiocaesium transfer to plants by 1.3–5-folds (Alexakhin 1993, 2009; Brown et al. 1995). The effectiveness of organic material fertilizer for radiocaesium accumulation in plants is higher on low fertile light sandy soil compared with loamy soils, due to the large difference in the CEC values (Table 11).

Application of organic materials is a conventional farming practice which should be slightly changed to provide a reduction of radiocaesium transfer to plants. Organic material applied specifically to decrease radionuclide transfer to plants may be of different origins and can include manure, straw and plant derived fertilizers (species such as lupin and serradella) (Belova et al. 2004; RIARAE 2006). Peat and sapropel may also be used as soil ameliorants (RIARAE 2006; Fesenko et al. 2012). Sapropel is formed from bottom sediments in natural lakes and consists of plant and animal residues decomposed in anaerobic conditions. The main advantages of sapropel are a high content of organic matter (up to 70 %), with a high content of humic acids and nitrogen, high CEC, the presence of mineral matter and mobile forms of nutrients (Brown et al. 1995). However, sapropel may be acidic (pH 4.5–6.5) leading to reductions in the pH of soil solution. Depending on the prevailing soil acidity this may lead to some increase in radionuclide transfer to plants (Fesenko et al. 2012).

Organic materials are normally applied to soils with a low organic content and of light granulometric texture. They are easy to apply and increase plant production by enhancing the nutrient and microelement content of treated soils. The conventional application rate of organic material depends on soil properties including the organic content, CEC and granulometric composition as well as the type of crop. Increased application rates of organic fertilisers were used widely in the FSU following the Chernobyl accident on arable land with deposition densities above 185 kBq m^{-2} (Fesenko et al. 2007). The application rates recommended were 1.5–2-folds higher than those used normally (RIARAE 2006; Fesenko et al. 2012). The timing of organic fertilization is crop dependent. For example, peat-manure composts are applied in spring for a planned yield of medium and late cultivars of potato, whereas for early potato it is applied in autumn during winter ploughing. Because organic

[10] Microfertilisers are fertilisers containing microelement, i.e. chemical elements which the plant requires in microquantities.

fertilizers are routinely applied on intensively managed arable soils the side effects are minimal. Crop yield may be increased by up to twofold and an associated improvement in soil fertility can be expected. Application of organic fertilizers such as manure and acid peat may increase the uptake of radiocaesium by plants in the first year after application because of changes of soil acidity. However, in the second year after application, organic fertilizers may produce a decrease in radiocaesium uptake by plants due to mineralization in soil leading to an increase in the content of potassium in the soil solution.

3.6 Application of Mineral Sorbents to Arable Soils

Mineral sorbents used for remediation may have different origins. The most commonly used materials are clays (such as bentonites and palygorskite) and zeolites (such as clinoptilolite), because these materials have a high sorption affinity for certain radionuclides. Application of mineral sorbents is not normally part of normal farming practices in most areas. Mineral sorbents added to soil enhance the sorption capacity of the soil so they should have a much higher sorption capacity for the target radionuclide than that of untreated soils (Sawney 1964; Nishita et al. 1968; Bakunov and Yudintseva 1989). A recommended particle size for mineral sorbents added to soil is 1 mm or lower to maximize sorption capacity. Zeolites can be used as a substitute for lime, due to its high pH, and it can be also added to peat-manure compost. The dry sorbent material should be uniformly spread on the soil surface before planting, and then the soil is re-ploughed. The mass of the applied material depends on the contamination level and soil properties. Application rates vary from 5 to 30 t ha^{-1}, the upper value was recommended for light sandy soil following the Chernobyl accident. When zeolite is applied, increased doses of mineral fertilisers and microelements should also be used because of their additional sorption by the sorbents implemented for remediation.

The use of mineral sorbents may reduce radiocaesium transfer to crops up to 2.5-fold depending on soil texture (RIARAE 2006). The maximum effectiveness may be observed for light sandy soil with low fertility and low K_d values such as sandy soils; the effect is low for clay soils with a high fertility (and high CEC status). The effectiveness of mineral sorbent addition tends to increase with time. Positive effects from the use of zeolites, for instance, appear from the second or third year after application when there has been an adequate amount of time for the clay to form a sorbing complex with radiocaesium in the soil (Wilkins et al. 1996). Mineral sorbents for soils need to be available in large amounts to be applicable at field level. Application of mineral sorbents can change the nutrient status and CEC of soil. The use of sorbents can stabilize the sorption properties of soil and improve its fertility providing positive effects for crop planting. One detrimental aspect of the application of the mineral sorbents as a remedial option is its relatively high cost. If there

Table 12 Summary of the data on effectiveness of combined application of mineral fertilisers and lime on radiocaesium accumulation in plants

Combinations of options	Reduction in radiocaesium accumulation in plants
$N_{60}P_{90}K_{120}$	2.0
$N_{60}P_{90}K60$	1.4
$CaCO_3$ (1Hg)	1.6
$CaCO_3$ (1.5Hg)	1.9
$N60P_{90}K_{60}+CaCO_3$ (1Hg)	1.9
$N_{90}P_{60}K_{90}+CaCO_3$ (1Hg)	2.0
$N_{60}P_{60}K_{90}+CaCO_3$ (1.5Hg)	2.2
$N_{60}P_{90}K_{120}+CaCO_3$ (1Hg)	2.6
$N_{60}P_{60}K_{90}+CaCO_3$ (1.5Hg)	2.5
$N_{60}P_{90}K_{120}+CaCO_3$ (1.5Hg)	3.2

are local deposits of mineral raw materials close to the contaminated areas these costs may be reduced.

3.7 Combination of Soil Based Options

Most of the remedial options discussed earlier can be applied individually and in combination. A combined use of mineral, organic fertilizers and liming is the most effective way to reduce radionuclide accumulation in farm crops. The combined use of lime and organic matter may reduce radiocaesium transfer to plants by up to three- to fivefold and the effects persists into the second and third years after the application. The use of increased application rates of P-K fertilizers combined with liming reduces transfer of radiocaesium by three- to fivefolds respectively. As an example Table 12 provides summary data on effectiveness of combined application of mineral fertilizers and liming.

A combined use of liming and increased rates of P-K fertilizers together with normal application rates of N can decrease radionuclide accumulation in farm crops up to two to fourfolds more than after liming alone. Many crops are sensitive to a deficiency of microelements, therefore, the additional application of micro fertilizer[11] is essential for remedial options such as liming and use of increased doses of P fertilizers.

[11] Microfertilisers are fertilisers containing microelement, i.e. chemical elements which the plant requires in micro quantities.

3.8 Amelioration of Grasslands

Remedial options for meadow soils normally combines some options already mentioned such as disking, ploughing and crop selection with additional procedures which are often used within conventional farming practices. These options involve adapting standard operations to achieve a maximal reduction in radiocaesium transfer to fodder plants in certain site specific conditions. There are two basic techniques of amelioration applied after the Chernobyl accident, namely radical and surface improvement. As contamination of milk and milk products is the main contributor to internal exposure of the affected population, radical improvement of meadows and pastures was a key element of the remediation policy applied in the contaminated regions. As well as reducing radiocaesium transfer to plants, both options have the additional benefit of increasing the productivity of lands used for fodder production and can be used for both uncultivated and cultivated grassland.

Surface improvement includes land improvement (extraction of shrubs, small hillocks of grass, weed control), application of soil mechanical treatment techniques (disking of the root mat which involves disrupting the root mat using heavy discs in 2–3 cuts) and enhanced mineral fertilization. In addition grasses are re-sown in the third year following the commencement of the procedure, thereby improving productivity of plants and ensuring appropriate ratios amongst the different species in the grass mix (RIARAE 1988; Sanzharova et al. 1996; Vidal et al. 2000). The grass mix is includes plant varieties with low accumulation of radiocaesium. The option includes application of lime materials (on acid soils) and increased amounts of P and K within an NPK fertilizer. The approach to select ratios between application rates of NPK fertilisers (or nitrogen, phosphorus and potassium) recommended for remediation of grasslands were similar to that of suggested for arable soils, namely, 1:1.5:2 compared with normal rates of fertilizer application (RIARAE 1988; Sanzharova et al. 1996). The optimal administration rates (active substance per ha) designed to minimise radiocaesium transfer to fodder plants are: (1) $N_{120}P_{90}K_{120}$[12] for dry grassland on mineral soils, (2) $N_{120}P_{90}K_{120}$ for dry grassland on floodplain soils or (3) $N_{180}P_{120}K_{180}$ for wet grassland on floodplain soils. Liming is obligatory for acid soils with application rates which are 1.5–2 folds higher than those normally estimated for the soil solution acidity for fodder land (Sanzharova et al. 1996).

Cereal grasses are preferable on radioactively contaminated lands; since accumulation of radionuclides by cereal grasses is c. two- to threefold lower than that by legume grasses. The grass composition used for surface improvement may include up to 20 % of legume grasses (white clover, vetch). A proportion of land may be sown with perennial grasses which are suitable for both hay production and animals grazing in early spring (in Russia 20 % brome grass was recommendation after the Chernobyl accident (RIARAE 2006). This remedial option was used extensively in the FSU after the Chernobyl accident (Fesenko et al. 2007).

[12] Notation of $N_xP_yK_z$ means mineral fertilizer application rate where x kg of active N per ha, y kg of active P per ha, z kg of active K per ha are applied.

Radical improvement consists of similar options to those of surface improvement with additional soil ploughing. Thus, radical improvement includes removal of shrubs, small hillocks of grass, root mat destruction, ploughing, disking, rototilling and chiselling, liming of acid soil (if necessary), application of increased amounts of P-K within NPK fertilizers and the selection of grass mixtures with the minimum possible accumulation of radionuclides. As above, drainage can also be included for wet soil. The effectiveness of these types of remedial option depends on the state and type of land used for fodder production and varies depending on the type of meadow, the hydrological regime, soil type, nutrient status and pH. The appropriate selection of plant species for reseeding is also important as transfer of radionuclides to different species can vary substantially.

Since the first year after the Chernobyl accident radical improvement has been a key remediation measure carried out extensively in practically all contaminated areas of Belarus, Russia and Ukraine. Radical improvement was effective in field conditions achieving a 3.0–4.0-fold reduction in root uptake of radiocaesium (Table 13). For organic soil the effectiveness increases to 3.0–5.0-fold, with maximum effectiveness (with drainage) of 10–20-fold in wet peat soil. The mechanism of action and effectiveness of radical improvement of meadows strongly depends on the types of meadow and soil properties (Sanzharova et al. 1996; Vidal et al. 2001; Bogdevich 2002). One limitation is that radical improvement (as other agricultural actions involving ploughing) cannot be carried out on sandy soils (due to risk of destruction very shallow fertile layer), steep slopes and river valleys due to the risk of erosion (RIARAE 1991; Brown et al. 1995). Surface improvement is around 20 % cheaper than that of radical improvement. Furthermore, surface improvement may be implemented on erosion-prone sites and low-productivity grassland such as those in floodplain areas where application of radical improvement is restricted. For the case of first application, surface improvement provides reduction of the soil-plant transfer

Table 13 Summary of the reduction factors of different remedial options for grasslands used in the FSU countries (Adopted from Alexakhin 1991, Sanzharova et al. 1996; Bogdevich 2002; RIARAE 1998)

Remedial option	Soils	Reduction of ^{137}Cs transfer to plants
Radical improvement		
First application	Mineral	3.0-4.0
Further applications	Mineral	2.0–3.0
First application	Organic	3.0-20[a]
Further applications	Organic	3.0–8.0
Surface improvement		
First application	Mineral	2.0–3.0
Further applications	Mineral	1.3–2.0
First application	Organic	1.5–6.0
Further applications	Organic	1.5–3.5

For wet peat with drainage

of radiocaesium in a range from 2.0 to 3.0 and 1.5–6.0 for mineral and organic (peat) soils respectively. For further application the effectiveness of this option is about 1.5–2.0 lower. On peaty soils the reduction factor is on average two- to fivefold higher than on mineral soils FSU (Sanzharova et al. 1996). Surface improvement of wet peat soil with drainage (where required) may reduce radiocaesium accumulation in grass by up to a factor of 10 (RIARAE 2006).

Radical improvement may decrease the transfer of radiocaesium to fodder plants by 3–20 folds. Reduction factors for soil-plant transfer of radiocaesium following radical improvement were in the range for mineral soils, from two- to fourfold, and for organic soils from three- to sixfold. A combined option such as drainage and radical improvement of fodder lands may reduce accumulation in grass of radiocaesium by up to a factor of 10. If applied to wet peat soil the reduction factor is greater at up to 20-fold. The option remains effective over 3–5 years and should then be reapplied although it will then have a lower effectiveness. In repeated radical improvement the reduction factors for transfer to plants are 2.0–3.0.

The above options can have potentially high environmental side effects because of the change of ecosystem from natural to cultivated grassland. Disking, application of lime and fertilisers and reseeding will change the ecological characteristics of the land with possible reductions in biodiversity. Grasslands are often the habitat of endangered species and a change in nutrient status may be harmful to these species. A significant increase in NPK application can lead to pollution of ground and surface waters by these elements. When applied on floodplain grassland, water body contamination by fertilizers may occur. Higher productivity of grassland should be anticipated since surface improvement of haylands and pastures increases their productivity by 25–50 % at a minimal cost which can be recovered within 1–2 years. If improvement is carried out under a rolling programme there should be no significant loss of grazing. There may be disruption to farming and other related activities, although there are also benefits to the farmer who will have more improved pastures in the long term. The availability of additional improved grazing can reduce wintering costs and result in higher prices for improved stock. The application of amelioration of meadow is only possible in areas where the soil structure and landscape are suitable. The option cannot be applied on grassland located on waterlogged soils, or on sites where the upper organic horizon is less than 10 cm deep. In the Chernobyl affected areas surface improvement is only applied to dry grassland on soddy-podzolic soils if the resulting plant species stand contains at least 50–60 % of valuable fodder comprising of cereal grasses and 25–30 % of legumes grasses.

3.9 Change in Land Use and in Crop Composition Grown on Contaminated Land

Different types of the land use are associated with different planting regimes. As demonstrated in Section 3.1 (see Table 5), the difference in radiocaesium accumulation by plants can be as great as a factor of 100 dependent on soil and plant properties.

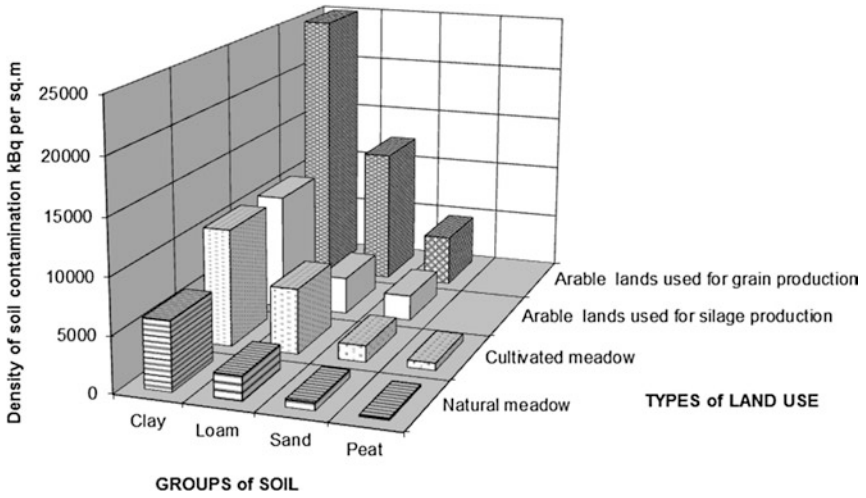

Fig. 4 Densities of soil contamination by ^{137}Cs which would result in exceeding the Chernobyl related temporary permissible (or action) levels (TPLs) for 1994 in various products produced in Russia for different types of land use and soil groups (Adopted from Fesenko et al. 2000a, b)

Therefore, the contamination of agricultural products is specific to every land use, and depends on the soil type and agricultural products produced on contaminated lands. Exclusion of agricultural products vulnerable to the contamination may be an effective method to retain the economic sustainability of affected regions. In particular, contaminated lands may be used for crop production instead of grazing animals or even for non-food produce, such as cotton/flax for fibre; rapeseed for bio-diesel; sugar beet for bio-ethanol; perennial grasses or coppice for biofuel. Agricultural land may also be used for the production of leather and wool. In situations where the land is highly contaminated it may be used for forestry or the placement of suitable industrial enterprises. Such land-use related variation needs to be considered when selecting a possible alternate land use for contaminated regions, together with any variation in permissible levels (TPL) for different agricultural products.

To demonstrate the effect of different land use options Fesenko et al. (2000a, b) used data from the monitoring programme to estimate the ^{137}Cs deposition of soil contamination which would result in exceeding TPLs in various products from different types of land use in 1994 (Fig. 4). There was a marked difference in the amount of ^{137}Cs deposition in soil which would limit the suitability of different types of land use. Using such analyses it was possible to decide whether areas with a defined land use and soil types require remediation, or whether a change in land use might be appropriate. Options such as the conversion of arable land into meadow, converting agricultural land to forestry were implemented in some of the most contaminated areas.

In Belarus, a land use change to rape seed production was applied in contaminated areas with the aim of producing two products: edible oil and protein cake as an animal fodder (Bogdevich 2002). The varieties of rape seed grown had a two- to threefold lower ^{137}Cs and ^{90}Sr uptake rate than many other varieties. Additional fer-

tilisers (Liming 6 t ha^{-1} and fertilization with $N_{90}P_{90}K_{180}$) are used to reduce radiocaesium and radiostrontium uptake into the plant by a factor of about two. This reduces contamination of the seed, which is used for the protein cake. During processing of the rapeseed, radiocaesium is effectively removed, and negligible amounts remain in the final product. The production of rapeseed oil in this way has proved to be an effective, economically viable way to use contaminated land and is profitable for both the farmer and processing industry. From 1992 to 2002 the area under rape seed cultivation in Belarus has increased fourfold to 22000 ha (Bogdevich 2002). The option was widely used in the areas most affected after the Chernobyl accident and was proven to be an effective way to remediate contaminated areas.

4 Remedial Options for Freshwater Ecosystems

Freshwater ecosystems are rather diverse environment and include lakes, rivers and groundwater. Each of these water bodies is unique in terms of the factors governing flows into, within and out of the water body. The effectiveness of remedial options for freshwater ecosystems is dependent on many parameters which are highly site specific and which can substantially constrain potential remediation options that can be applied. The doses arising from freshwater pathways are normally lower than that from terrestrial food. In terms of collective doses to humans, the irrigation and water supply pathways represent the most important pathways. Nevertheless, freshwater-related ingestion pathways may be important contributors to individual doses for people living in settlements surrounding closed lakes, which do not have an outflow. For example, the Kozhany settlement in the Bryansk region, Russia, is located on the shore of a closed lake in an area with peaty soil that was highly contaminated after the Chernobyl accident in 1986. The ^{137}Cs activity concentrations in lake water and fish were two orders of magnitude higher than that from local rivers and open lakes, and remain relatively high for 10–20 years after the accident Travnikova et al. (2004) at 10–20 kBq kg^{-1} fw of ^{137}Cs in lake fish which and exceeded the temporary Russian permissible levels for the fish by a factor of 20–40. The consumption of the fish was up to 50 % of the internal doses to residents of the settlement (Travnikova et al. 2004).

Exposure of the public from contaminated bottom sediments or from sand on the beach can also can occur in highly contaminated areas. However due to the self-shielding of water, external doses from recreational use of contaminated lakes and rivers are normally relatively low (Fesenko et al. 2012). Thus, the focus of implementation of remedial options for aquatic environments is reduction of radiocaesium transfer from contaminated watersheds to water bodies and various options to decrease contamination of edible freshwater species. Relevant remedial options for freshwater can be divided into two main groups: (1) preventing contamination of water bodies by the use of physical or chemical barriers, and/or (2) treatment of water (blending and/or purification). These options are more effective if combined with dietary advice, the provision of monitoring kits, food labelling and compensation schemes (Fesenko et al. 2012).

The first option involves the addition of these barriers to the boundaries of water bodies and therefore can have some features in common with chemical additives used in the second option. Dredging of canal-bed traps to intercept suspended particles in contaminated rivers was carried out after the Chernobyl accident (Voitsekhovitch et al. 1988). These canal-bed traps were highly inefficient for two reasons: (1) flow rates were too high to trap small suspended particles which had adsorbed much of the radioactivity; (2) a significant proportion of the radioactivity (and most of the "available" activity) was in dissolved forms and could not have been intercepted by sediment traps.

4.1 Prevention of Water Body Contamination

The concept behind this option is to prevent remobilisation of runoff radiocaesium from the catchment by installing dykes thereby decreasing long-term transfer to freshwater ecosystems (Smith et al. 2001; Onishi et al. 2007). This measure was widely used after the Chernobyl accident mainly around the Pripyat river floodplain to prevent the secondary contamination of freshwater ecosystems from highly contaminated areas surrounding the river and floodplain. The dyke led to a substantial reduction of radiocaesium transfer to downstream of the Kiev Reservoir and Dnieper River after Chernobyl (Onishi et al. 2007). However, the effectiveness of the option was low for large scale applications since only a small fraction of the total amount of radioactive material deposited in aquatic ecosystems and their catchments retained.

Zeolite containing dykes were constructed on smaller rivers and streams around Chernobyl to intercept dissolved radionuclides. These were ineffective since only 5–10 % of ^{137}Cs in the small rivers and streams were adsorbed (Voitsekhovitch et al. 1997). Buffer strips around rivers can be used to retain radiocaesium contained in eroded particles or in dissolved form and encourage its infiltration and sorption in the soil. This option has been in widespread use within different environment protection schemes to reduce runoff of fertilisers and pesticides from agricultural land to the water (Smith et al. 2001). Removal of radioactivity from water by "filtering" through reed beds is also possible, though probably not feasible on a large scale.

4.2 Fertilization of Water Bodies

The fertilisation of lakes can only be applied to low productivity lakes with low potassium and phosphorus concentrations in the water and low pH. As for agricultural systems, the uptake of radiocaesium by freshwater species from water may be affected by competition with potassium ions, water pH and the presence of other microelements such as phosphorus in the water. Studies on both weapons-test-derived and Chernobyl radiocaesium demonstrates that the uptake to fish (or the

concentration factor (*CF*) of radiocaesium in fish) is inversely proportional to the potassium (Fleishman 1973; Blaylock 1982; Smith et al. 2000, 2003). An empirical model for the prediction of radiocaesium concentration factor (CF) values in fish (Rowan and Rasmussen 1994) in the form:

$$CF = \frac{A}{\left[K^{+}\right]^{B}},$$

where CF is measured in l kg^{-1} and $[K^+]$ is in mg L^{-1}, *A* and *B* are the model constants.

The approach was tested against measurements made between 1992 and 1997 in 10 lakes in Russia, Belarus and Ukraine. The model validations, based on measurements of K^+ and suspended solids concentrations in the lake water, were in good agreement with measured values. Therefore, addition of potassium based materials may have the potential to be effectively used to reduce radiocaesium concentrations in fish and other freshwater species. The effectiveness of potassium application strongly depends on the water chemistry at the time of application (e.g. potassium concentration, pH and total phosphorus concentration), the amount and type of K applied and the water retention time. For lakes with rapid inflows and outflows of water, many repeated applications would be necessary to achieve a consistent stable effect (Fesenko et al. 2012). A low effectiveness of this option with a reduction of ^{137}Cs in perch of only 1.1-fold was reported for Swedish lakes affected by the Chernobyl accident (Håkanson and Andersson 1992). Addition of potassium chloride to Lake Svyatoe, Belarus (a lake with a very low natural potassium content), was much more efficient with a tenfold increase in the potassium content of the lake water giving a two– threefold reduction in radiocaesium activity concentration in fish (Smith et al. 2001). For lakes, the effectiveness of potassium application strongly depends on the initial calcium concentration, pH, total phosphorus concentration in the water, amount and type of liming applied and the water retention time. Some modelling assessments have shown that a reduction of about 1.3–1.7-fold in fish can theoretically be obtained (Smith et al. 2001). There may be negative side-effects of the application of various elements to water bodies. Application of mixed K, NH_4, and phosphorus fertilizer may cause eutrophication whilst potassium applied as KOH may increase the pH in lake water. The potential importance of these possible impacts has not been assessed and may be minor in some cases. In one experiment the application of potassium led to a threefold increase in radiocaesium activity concentration in water due to competition with K in sediments (Smith et al. 2000). This side effect would make the option unacceptable if the water is to be used for drinking or irrigation.

Fertilization of lakes with lime and/or phosphorous may be useful as a remedial option for a variety of toxic contaminants in lakes with high productivity, where transfer rates of radionuclides, other metals or organic pollutants may be quite high. Different methods of fertilisation can be used to reduce transfer to freshwater spe-

cies. Lime or lime mixed with phosphorus can be directly added to the water (or to the ice over the winter period) or used for fertilisation of the catchment of affected water bodies, providing a more gradual source of these substances to the lake water. Different commercial fertilisers as well as phosphorus containing effluents from fish farms can be also used for such fertilisation.

For lakes, effective remediation would need to considerably raise the Ca level, pH and alkalinity and sustain the elevated levels for a long period of time; however, the duration of water chemical responses may be relatively short, depending on the water residence time of the lake. Catchment application may avoid a "liming spike" and considerably prolong the duration of an effect. There is a considerable experience in application of lime in relation to acidification but not for radionuclides. Although, this measure was tested in Scandinavian countries after the Chernobyl accident (Håkanson and Andersson 1992; Outola and Rask 2011), the measure has not been implemented extensively as a remedial option. The effectiveness of potassium application strongly depends on the initial *Ca* concentration, pH and P concentration in the water, amount and type of liming applied and the water retention time. Some assessments have shown that a reduction of about 1.3–1.7-fold in fish can be obtained (Smith et al. 2001). However, in Swedish lakes contaminated after the Chernobyl accident, a low reduction factor of 1.1 was reported for perch fry (Håkanson and Andersson 1992). One negative possible side effect is that liming of naturally acid systems can lead to profound, structural changes in the ecosystem. As a result some changes in the wetland flora may be expected such as the replacement of bog moss by leaf mosses and sedges and damage to sensitive species (e.g. lichens) due to wind drift of lime to adjacent areas (especially if using helicopters) (Smith et al. 2001). The buffering capacity of lakes can be temporarily improved with liming, which has a stabilizing effect on a lake, and is therefore beneficial for lakes (and food chains) that are susceptible to acidification. Catchment liming may also lead to improved conditions for animals and plants in streams and rivers due to reduced transport of metals such as *Fe* and *Al* into the lake from the catchment area (Smith et al. 2001).

5 Forest Ecosystems

Forests are extensive natural resources which provide economic, nutritional, recreational and social benefits to people in many countries so any restrictions applied to the forest due to contamination may result in high economic losses and have severe social and psychological consequences. In many areas subjected to radioactive contamination, forests and forest products were, and still are, an important source of internal and external exposure of the population (IAEA 2006). For example, in the initial years after the Chernobyl accident, the contribution of agricultural products (mainly milk) to the internal dose was much higher than that of forest products. However, the contributions of forest exposure pathways increased with time (Tikhomirov and Shcheglov 1994) from 10 to 15 % in 1987 to

40-45 % in 1996 (Fesenko et al. 2000a, b). Unfortunately, there are only a few practical options that can be implemented in forest ecosystems. The imposition of restrictions on access to forests and consumption of forest products may be effective for a few years but is unlikely to be sustainable in the long term. Therefore, the provision of advice about possible doses from the forest on which products are likely to be contaminated combined with access to monitoring stations to measure contamination is probably a more practically realistic way to reduce such doses (Fesenko et al. 2012).

5.1 Forest Soil Treatment with Fertilizer or Liming

As for agricultural soil, surface application of fertilizers (NPK or PK) or liming in case of acid forest soils in the forest can reduce root uptake of radiocaesium by forest plants and enhance their growth (Fesenko et al. 2000a, b). Such options were tested in Belarus, Russia and Nordic countries after the Chernobyl accidents, but were not used on a large scale (Ipatyev et al. 1999) (see Fig. 5). A good example of the effect of fertilisation with potassium of the forest soils is presented in Rosén et al. (2011) who studied the long-term effects of a single application of potassium fertilizer (100 kg K ha^{-1}) on ^{137}Cs uptake in some understory and fungal species (Figs. 6). The uptake of ^{137}Cs by plants and fungi growing on K-fertilized plots 17 years after application was significantly lower than in similar species collected at non-fertilized sites. The ^{137}Cs activity concentration was 21–58 % lower in fungal sporocarps and 40–61 % lower in plants in the K-fertilized area compared with the control sites. The reductions in ^{137}Cs activity concentration in both understory species and mushroom fruiting bodies were statistically significant for most studied years. The data demonstrates that application of K fertilizer to forests might be effective long-term measure to decrease contamination of plants and fungi, although this option is expensive and cannot be applied in a large scale.

The application of dolomite powder leads to a rise in Ca^{+2} concentrations in soil solution. Bivalent Ca^{+2} cations are not capable of penetrating inward to the wedge-shaped zone of clay minerals, but can stabilize it in a dilated state, during which more ^{137}Cs cations can occupy FES sites. If there is a decline in Ca^{+2} concentration in soil solution, due to vertical migration and other processes in soil, layer collapse of clay minerals ("delayed collapse") is intensified and, consequently, ^{137}Cs fixation is increased in the crystal lattice. Typically reduction factors of 1.5–2 have been achieved by application of dolomite powder in forests, however, the effectiveness is dependent on forest soil fertility and the type of contaminated forest (Ipatyev et al. 1999; Spiridonov et al. 2005). Fertilizer application to the forest soil is generally more efficient if applied in boreal forest than in Mediterranean forest, and fairly efficient in deciduous forest (Ipatyev et al. 1999).

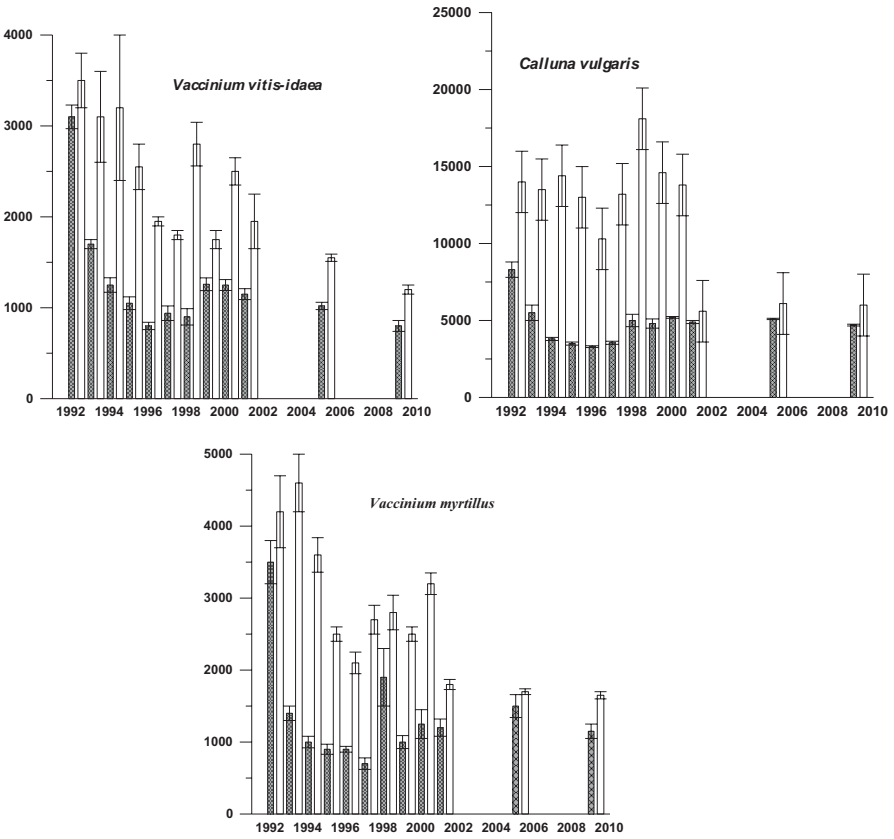

Fig. 5 ^{137}Cs activity concentrations (Bq kg^{-1} d.w.) in heather (*Calluna vulgaris*), bilberry (*Vaccinium myrtillus*) and lingonberry (*Vaccinium vitis-idaea*) in potassium treated and control plots in 1992–2009 (Adopted from Rosén et al. 2011)

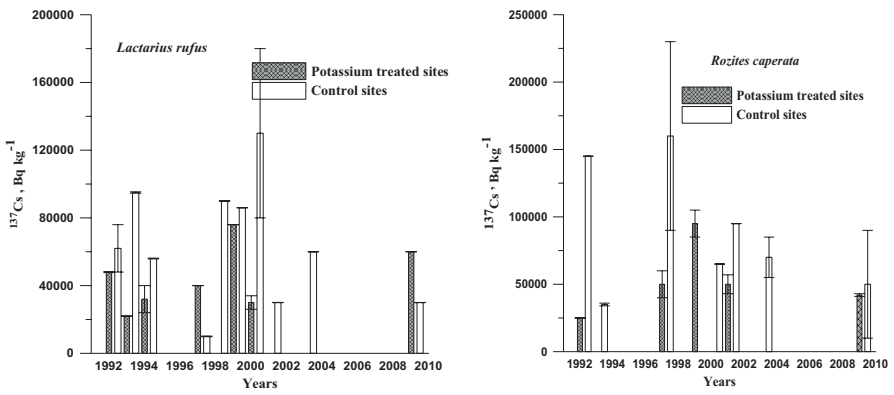

Fig. 6 ^{137}Cs activity concentrations (Bq kg^{-1} d.w.) in some fungal species in potassium treated and control plots in *Lactarius rufus* and *Rozites caperata*

5.2 Modification of Tree Felling Schedules

The remedial option is based on changes in the timing of tree harvesting, intermediate felling and other forest services with the aim to minimize root uptake of radiocaesium in stem wood, or to delay wood harvesting so that processes such as soil immobilisation, vertical migration down the soil profile or physical decay of radionuclides reduce the contamination in the wood. Early felling is most suitable for mature or nearly mature trees soon after the time of contamination whilst delayed felling is most suitable for young trees reaching maturity about 20 years after contamination (Fesenko et al. 2012). The effectiveness depends on factors such as which radionuclides dominate in the long term, their radiocaesium activity concentrations in the forest, and forest characteristics such as age of trees, productivity, forest soil, type of understory (Frissel et al. 1996). Overall, reduction factors predicted from the model approach for early felling can reach two- to tenfold, whilst delayed felling can lead to reduction as high as 1.5–2-fold compared with the normal felling programme. However, changing standard forestry practices can lead to economic losses due to delays, or additional costs in harvesting the timber. Increased transport costs are also expected if nearby areas cannot be used as sources of wood for industry. Forestry operation plans may need to be approved by regional forestry administration. Markets may experience seasonal gluts and shortages so alternative wood sources should be available to compensate for the delay in harvesting. If large forest areas are clear-felled, the hydrology of the site must be considered especially with respect to possible erosion in the catchment (Nisbet et al. 2009).

5.3 Change of Hunting Season

Due to seasonal variation in diet of wild animals, the radiocaesium contamination of some game species varies significantly with season in Western and Eastern Europe. In particular, radiocaesium activity concentrations in muscle of game animals from areas where fungi can be abundant in certain years can be much higher than the average annual values (Hove et al. 1990; Johanson 1994). Hunting is normally restricted to certain periods of the year. By changing or restricting the hunting season to the time of year when the contamination levels in the game meat are not enhanced due to dietary preferences, the internal dose to humans consuming game meat can be reduced. A change of hunting season was applied in the FSU and some Nordic countries following the Chernobyl accident (IAEA 2006). Varying hunting times achieved a 2–3-fold reduction of radiocaesium activity concentrations in moose meat with even higher reduction of up to four to sixfolds for meat from roe deer, wild boar and wild reindeer. The effectiveness is greater if large quantities of mushrooms are available which occasionally happens in the autumn leading to potentially higher seasonal increases in wild meat contamination (Fesenko et al. 2012). The impact of changes in hunting season on breeding should be considered

for each species. The continuous management of large game animals through hunting licenses is an integral part of controlling the populations of game animals at sustainable levels. It is thus important to continue hunting, or recommence hunting, as soon as possible after contamination occurs.

Some detrimental side effects may result from this measure. Increased grazing on adjacent agricultural lands may occur if the hunting season is delayed. If hunting is performed earlier than normal then lower slaughter weights may be anticipated. If hunting is restricted to seasons with bad weather, harsh climates may make hunting less attractive in some countries. The hunting season must not be changed to coincide with breeding and other seasons which are important for maintaining animal populations (Johanson 1994).

5.4 Optimisation of Forest Management

Logging may be transferred to areas with low external doses and to areas with lower radionuclide activity concentrations in wood. The same approach can be used for other forest products (Fesenko et al. 2012). Recommendations on suitable areas for tree felling and gathering of mushrooms or berries in forests of the FSU affected by the Chernobyl accident were given to local people and led to lower external doses to foresters or other people gathering food in the forest as well as substantial reductions in internal dose due to radiocaesium. The effectiveness depends on the contamination densities in forested areas. A reduction of the external dose of two- to threefolds may be achieved. For forest products (harvested wood, berries or mushrooms), reductions of two- to fivefold in internal doses are achievable (Fesenko et al. 2012).

Conclusions: Selecting Remedial Options in Areas Affected by Radiocaesium

The effectiveness of the available techniques used to reduce radionuclide activity concentrations in agricultural products varies depending on factors such as: application rates and duration of the application of remedial actions, the type of materials used, extent of adherence to required procedures (compliance), acceptability of the remedial options for the stakeholders and many other site specific factors to be taken into account within the remediation planning phase. Implementation of some remediation options may require specific equipment or resources which are not in normal usage in the production systems requiring remediation. Given the high variability in the effectiveness of remediation options, it is advisable to test their impact in relevant practical conditions in contaminated areas before using them on a wide scale (Howard et al. 2004, 2005; RIARAE 2006; Nisbet et al. 2009).

There are normally costs associated with implementation of remediation options which are readily defined for labour, consumables, equipment, transport and waste disposal. However, there are also other indirect costs of remediation such as the loss of production and retail sales through disruption and/or closure of businesses, loss of market share and regional impacts on tourism. Such indirect costs are often just as important as the direct costs, but more difficult to quantify partly because they can impact over much broader areas and populations than the contaminated area itself (Howard et al. 2005). Conversely, some remediation options can have direct monetary benefits such as additional crop yield due to mineral fertiliser application.

Some remediation options may produce contaminated by-products or so called "remediation waste". For example, if soil removal is implemented, a large quantity of contaminated soil may be generated even from relatively small areas. This may require long term disposal of the resulting waste and the cost of disposal may be much higher than that of implementing the remediation measure (Howard 2012). Waste disposal schemes for contaminated waste should preferably be planned before the waste is generated, with potential disposal sites and resources identified where possible (IAEA 2015). After the Fukushima Daiichi accident the removal of contaminated topsoil was widely used (IAEA 2015). There were many discussions regarding the planning and disposal of remediation waste to decide whether on-site or off-site disposal was preferable. As well as technical issues, aspects related to social considerations were important. There was a particular focus on perceptions of whether a "dilute and disperse" or "contain and concentrate" strategy was more justified (Oughton and Forsberg 2009).

To be a part of a remediation strategy the application of remedial options should be considered from many different aspects and be shown to be sustainable. This means that they need to sustain normal socio-economic activities and have no significant disadvantages such as unacceptable cost, waste generation or detrimental side effects. Some options require more rigorous analysis or additional experimental studies at selected sites and situations to that their applicability in a specific context can be defined before inclusion in a remediation strategy.

Phytoremediation represents an example of options which require additional consideration before application as a part of remediation strategy. The potential use of phytoremediation is based on the premise that some plants have a particularly high uptake of certain radionuclides. Therefore, if these plants are periodically harvested and disposed of in an appropriate manner, this option may provide the potential to remove radionuclides from the main environmental sink, the soil. However, currently there has been no small or large scale adoption of the methods at existing sites for radionuclides for three main reasons, namely: (1), the total amount of radionuclide removed from soil is a very small fraction of the total radionuclide content present, even for those radionuclides with a comparatively high transfer from soil to plant (2) the process would need to continue for many decades before the soil became adequately decontaminated to use for food production, and (3) the option generates waste which would then have to be disposed of appropriately generating additional costs.

Implementation of soil-based agricultural actions can change soil properties leading to direct environmental impacts including changes in biodiversity, soil fer-

tility and structure and enhanced soil erosion. There can also be associated effects which reduce the quality of air and aquatic ecosystems. Conversely, some side effects of the application of agricultural remediation options can be beneficial. A good example is the increase in crop yield associated with enhanced fertilizer application. The increase in mass, apart from being a benefit to the farmer, may also dilute radionuclide activity concentrations in crops. Enhancing the productivity of grassland may also improve the condition of livestock and the effect may be particularly pronounced in previously less intensively managed systems.

Some *in-situ* biological and chemical leaching techniques could be applied to soil to remove radiocaesium such as soil washing, flotation, chemical extraction and bioleaching. Implementation of such options may require large quantities of the leaching substance, and can substantially contaminate surface and groundwater. Such options may have a substantial environmental detrimental effect, may prevent re-establishment of vegetation, and would not normally be a part of a sustainable remediation strategy for releases of radionuclides (IAEA 2015).

The imposition of restrictions on utilising forests can lead to negative side effects. If all access by the public is restricted, or limited, local residents may have insufficient access to resources such as wood collected for cooking, fires and other domestic and industrial purposes; berries and mushrooms; and access to forest meadows used for livestock. The cost of forestry may also be increased due to limitations on the time which forest workers can spend in contaminated forests which, in turn, can lead to deterioration in the maintenance of forests with negative ecological consequences. Application of restrictive remediation options such as limited access to forest in the areas affected by the Chernobyl accident had negative psychological and sociological consequences. The economic losses were also large (Salt and Rafferty 2001).

For many remediation scenarios, the application of remedial options can substantially change the ecological status of an area. The impacts, and hence acceptability, of deep-ploughing in an intensively agricultural area are high, but this will not also be the case for many semi natural ecosystems, even though the effectiveness may be much higher for the latter situation. In such cases, environmental legislation must also be taken into account (Nisbet et al. 2009).

Studies on effectiveness of remediation implementation in many areas affected by the nuclear accidents at the Chernobyl and Fukushima Daiichi accidents have identified many factors that had a highly detrimental impact on the economic and social activities on contaminated areas. Furthermore, some remediation actions may impose a stigma on the areas with contaminated environments (Oughton et al. 2004, 2009; IAEA 2015). The type of information disseminated about remediation should be targeted to meet a variety of needs of different stakeholders. The form of communication should be adapted to different levels of understanding and the prevailing circumstances to address the relevant issues, and should be implemented at the same time as the development of restoration strategies (IAEA 2015). Provision of local counting equipment may allow the public to monitor their own food, and improve autonomy and empowerment. The application of many remediation options can be limited by the wide range of factors discussed above which should be evaluated beforehand. Such evaluation needs to also consider administrative and regulatory issues such as legal constraints, environ-

mental protection criteria; and cultural or heritage protection. In some cases there may be positive impacts in these areas. For instance, forest ecosystems may benefit from the absence of human exploitation (IAEA 2006).

Wide-scale implementation of remediation must take into account social and ethical issues when considering justification of possible options (Oughton et al. 2004; Oughton and Forsberg 2009). The consequences of implementation of remedial actions need to be evaluated with respect to potential impacts such as the effect on current and future generations, sustainability, and the relative harm to the environment compared with benefits to humans. If social aspects are respected, there is a greater likelihood that the implementation of remedial actions will be acceptable to both the public and operators (IAEA 2006; Fesenko et al. 2007; ICRP 2009). Thus, the impact on both individuals and communities needs to be considered. In particular, some remedial options may affect society by causing disruption (e.g. through restricting access or activities); anxiety and stress (e.g. by causing panic, upheaval); and stigma (e.g. by affecting businesses or tourism). Involvement of the affected communities in self-help options can reinforce liberty and dignity and be highly beneficial. These options were widely used in some regions of Belarus within the ETHOS project (Dubreuil et al. 1999). Remedial options implemented by the population themselves are normally ethically robust and often efficient and cost effective. Some of them do not require specific experience and can be carried out by local people with minimal training and advice (ICRP 2009). The involvement of affected population in implementation of remediation can give a feeling of control of the situation, which also prevents undue anxiety (ICRP 2009; IAEA 2015).

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