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Preface

Strontium (Sr) is a soft, silvery metallic element generally found in rocks, soil, dust, fossil fuel, water and in oil. Altogether naturally occurring strontium is non-radioactive. Strontium has four stable isotopes, i.e. ^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr. Twenty-eight radioactive strontium isotopes are known, the most relevant being ^{90}Sr, a beta emitter with a half-life period of 28.8 years. It is produced by nuclear fission of ^{235}U or $^{239/241}$Pu in nuclear reactors or during the explosion of nuclear weapons. It is widely used in so-called SNAP devices (Systems for Nuclear Auxiliary Power). Commercially, Sr is mostly used as strontium sulphate and strontium carbonate. Due to the Chernobyl nuclear accident about 30,000 km^2 of land was contaminated with >10 kBq m^{-2} of ^{90}Sr in 1986, and in case of the Fukushima nuclear power plant accident in 2011, ^{90}Sr activity in seawater reached up to 1,500,000 Bq m^{-3} at the Fukushima plant port. During atmospheric nuclear weapons testing, approximately 6×10^{17} Bq of ^{90}Sr was produced and globally distributed until 1980.

Strontium and calcium are homologues, both are alkaline earth metals, and behave similarly in the environment. Hence, strontium is highly plant available, easily enters the food chain and poses a radiological risk to humans. Strontium can migrate rather fast in the environment, particularly in water soluble complexes. Sr is incorporated into the human skeleton by exchanging calcium ions in the hydroxyapatite crystal lattice. There is experimental evidence for low dose of Sr reducing bone resorption (due to cover of the endosteal bone surface with osteoid) and increasing uptake of Ca into bones or stimulating bone formation.

Soil-to-plant transfer is often parameterized by the transfer factor (TF). This is the activity concentration of the radionuclide per unit dry mass in the plants (Bq kg^{-1}) derived by the one in the soil (also given in Bq kg^{-1}). The radionuclide migration and mobility in soil depends on the soil properties (e.g. soil pH, texture, exchangeable calcium and potassium, and organic matter content), weather conditions, plant species and land use practices. Any practical means of reducing the transfer of radiostrontium from soils to plants must be considered. During the first 5 years after the Chernobyl nuclear accident, almost all radionuclides (e.g. ^{137}Cs and ^{90}Sr) were situated in the upper 0–5 cm layer of soil. However, vertical migration during the following decades led to more than 50% of the remaining radionuclides found in horizons below 5 cm depth. However, 85–90% of the activity remains in the 0–20 cm layer of soils and are hence available for root uptake.

On the other hand, exactly the ability of plants to absorb, accumulate and remove ^{90}Sr from contaminated soils provides a dependable and inexpensive method of remediation. It is well documented that plants like cereals are the main sources of foodstuff contamination from ^{90}Sr. Inter-cultivar variation in different plant species may also play roles in Sr uptake from contaminated fields and it may vary between the cultivars with the lowest and highest concentration ratios. According to one estimate after the Chernobyl disaster, alterations (mutation) in plant species due to radioactive fallout resulted in five to tenfold increment in the mobility and bioavailability of ^{90}Sr in the ecosystems where no remediation was implemented. There are numerous methods for the removal of metals from polluted soils, i.e. soil washing, chemical and biological methods including plant-based remediation process called phytoremediation.

Radionuclides do not necessarily enter the plants through the roots but can do so through the stomata (direct deposition from the atmosphere). Usually, this pathway is assumed to contribute only a small fraction of the total amount of radionuclides entering the leaf. When radionuclides enter through the cuticle layer, they are dynamically transported inside the plant cells through both symplastic pathway and with an exchange mechanism between the phloem and the xylem. It is also known that vacuole is a central player in cellular ion homeostasis, due to regulation of cellular pH, ion fluxes and adjustment of the cellular turgor and it is also very important for detoxification processes, due to the removal of toxic components out of the cytoplasm.

The most striking features of this book are related to how Sr enters the ecosystem and its translocation from soil to plants and lastly to humans. Chapters "Strontium in the Ecosystem: Transfer in Plants via Root System", "Factors Influencing the Soil to Plant Transfer of Strontium" and "Distribution of Strontium in Soil: Interception, Weathering, Speciation and Translocation to Plants" deal with bioavailability of Sr in the ecosystem and its translocation via soil to plants. Chapters "Assessment of Entry of ^{90}Sr into Plants in Case of a Heterogeneous Radiation Contamination of Ecosystems" and "Accumulation of ^{90}Sr by Plants of Different Taxonomic Groups from the Soils at the East Ural Radioactive Trace" focus on how at heterogeneous radiation contamination ^{90}Sr is going to enter into plants and how different taxonomic groups of plants influence uptake mechanism. Chapters "Soil-to-Crop Transfer Factors (TFs) of Alkaline Earth Elements and Comparison of TFs of Stable Sr with Those of Global Fallout ^{90}Sr" and "^{90}Sr Distribution in System 'Soil-Scots Pine' (*Pinus sylvestris* L.)" focus on soil to crop transfer factors of alkaline earth elements and ^{90}Sr and also comparison between stable and global fall out of ^{90}Sr, and the case study with ^{90}Sr in Scots pine plant. Chapters "Contamination of the Firewood Taken from the Exclusion Zone of Chernobyl NPP by ^{90}Sr According to Data of 2005–2016" and "The Behaviour of 90Sr in Macrophytes Inhibiting Water Reservoirs in the Belarusian Sector of the Chernobyl NPP Exclusion Zone" focus on how ^{90}Sr is migrating from soil to plants as well as the behaviour of ^{90}Sr in microphytes after the accident at Chernobyl NPP. Last but not least, Chap. "Strontium

Isotopes in Biological Material: A Key Tool for the Geographic Traceability of Foods and Human Beings" discusses the application in geology of different Sr isotopes in geologic fingerprints for geographic traceability in food, forensic and archaeological sciences. To conclude, the information compiled in this volume will bring in-depth knowledge of Sr uptake and translocation and its toxicity in plants after Chernobyl and Fukushima NPP accidents.

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Strontium in the Ecosystem: Transfer in Plants via Root System

Dharmendra K. Gupta, Utsab Deb, Clemens Walther, and Soumya Chatterjee

Introduction

Strontium (symbol Sr, atomic no. 38) is a natural and commonly occurring alkaline earth metal (lying in group IIa, period 5, sandwiched vertically between calcium and barium). The mineral was discovered in 1790 by Adair Crawford and William Cruickshank at Strontian (Scotland). Sr ore is usually found in nature in the form of minerals like celestite ($SrSO_4$), and strontianite ($SrCO_3$); however, it can form a variety of compounds, which are either water soluble or not. Naturally occurring stable (non-radioactive) Sr exists in four stable isotopes, viz.^{84}Sr, ^{86}Sr, ^{87}Sr and ^{88}Sr (ATSDR 2004a; Therapeutic Goods Administration 2014; Qi et al. 2015). Furthermore, some 30 radioactive strontium isotopes are known, the most important one being ^{90}Sr, which is formed during nuclear weapons explosions or in nuclear reactors and was released into the environment by fallouts following accidents like Chernobyl Nuclear Power Plant (NPP) in 1986 and Fukushima NPP in 2011 (Sanzharova et al. 2005; Lipscy et al. 2013). ^{90}Sr is a β-radiation emitter with a radioactive half-life of 28.8 years and hazardous to health as it resembles calcium and creates metabolic imbalance (Merz et al. 2016). The radioactive decay through β emission of ^{90}Sr subsequently produces ^{90}Y (yttrium) and ^{90}Zr (zirconium). Once absorbed inside the body, ^{90}Sr causes bone cancer, leukaemia and softening of tissues situated around the bone and bone marrow and has lasting effects (ATSDR 2004b). Because of high toxicity and capability to be readily involved in geochemical and biological migration processes, ^{90}Sr is a particularly hazardous radionuclide

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(Sanzharova et al. 2005). However, strontium also finds application in treating and preventing diseases like postmenopausal osteoporosis and resultant bone fractures (O'Donnell et al. 2008; Bolland and Grey 2014; Nakano 2016), but the use has been somewhat reduced due to significant safety concerns, particularly the risk of using Sr with patients having a history of ischemic heart disease or venous thromboembolism (Sowa et al. 2014; www.tga.gov.au/alert/strontium-ranelateprotos-and-risk-adverse-events-0; accessed on: 1 Mar 2016). Humans get exposed to radionuclides either directly or indirectly by diverse pathways like consumption of contaminated food or water (Ruttenber et al. 1984; Fowler et al. 1987; Eisenbud and Gesell 1997; Arora et al. 2008). ^{90}Sr, due to rather high solubility in water, once inside the body, gets deposited in bones and teeth (Volkle et al. 1989). Similarly, ^{90}Sr also gets accumulated within plants depending on factors like soil properties, climate, biosphere, and particular species and populations (subspecies, cultivars) etc. (Yablokov et al. 2008).

^{90}Sr in Soil: Its Behaviour and Consequences

Varying meteorological conditions enhance airborne transport of radioactive aerosols from one place to another followed by deposition from atmosphere to soil. After precipitation, the fate of ^{90}Sr is affected by different physico-chemical processes in soil-water-plant compartments (Ehlken and Kirchner 2002). The global precipitation rate varies strongly but has a significant correlation with levels of ^{90}Sr in soil and grass (Bolsunovsky and Dementyev 2011). ^{90}Sr has a greater mobility and migrates faster downward into the soil (especially in mineral (sandy) and organic soils) as compared to other radionuclides like ^{241}Am (Americium), $^{239+240}$Pu (Plutonium) and ^{137}Cs (Cesium) (Gastberger et al. 2000; Solovitch-Vella et al. 2007; Chawla et al. 2010). Soil particles are a very important reservoir of ^{90}Sr in the terrestrial environment (Lee and Clark 2005). Sr is also known to compete with its macrohomologue calcium for exchange sites in minerals (Strebl et al. 2007). Five to tenfold increased mobility and bioavailability of ^{90}Sr has been observed in the non-remediated ecosystems close to Chernobyl (Warner and Harrison 1993). This increase in ^{90}Sr with passage of time may be attributed to the ion-exchange mechanism by soil components and the relatively high concentrations of the radionuclide within the soil (Maskalchuk et al. 2014). Therefore, ^{90}Sr distribution in ecosystems includes transfer into the food chain and entering the human body. Next to ^{137}Cs, ^{90}Sr has the potential to significantly contribute to internal ingestion dose (Gupta et al. 2016a).

The migration of ^{90}Sr in the solid phase of the soil–soil solution–plant system is computed by a concentration factor (CF) defined as CF = $[^{90}\mathrm{Sr}]_p/[^{90}\mathrm{Sr}]_s$, where $[^{90}\mathrm{Sr}]_p$ and $[^{90}\mathrm{Sr}]_s$ are the equilibrium specific activities of ^{90}Sr in plant and soil, respectively, expressed in Bq kg^{-1} (Sanzharova et al. 2005). The CF depends on a number of parameters like the properties of soil, plant and calcium (macrohomologue of Sr) (1983). Maskalchuk et al. (2014) derived an equation to estimate the migration of ^{90}Sr from soil to plants, which includes key characteristics of soil, content of exchangeable Ca^{2+} and fraction of exchangeable $^{90}Sr^{2+}$ in soil and an exchange selectivity coefficient for the $^{90}Sr^{2+}$–Ca^{2+} cation pair. The model suggests reduction of ^{90}Sr migration in the soil–plant system by liming (in case of acidic soils; depending on the cation-exchange capacity of the soil in question) and also by introducing soil sorbents with high sorption potential like organozeolites (Maskalchuk et al. 2014). Thus, long-term soil uptake of ^{90}Sr is strongly influenced by the pedological and agronomic characteristics of the soil and agricultural practices (Koranda and Robison 1978).

Claus et al. (1990) reported a ^{90}Sr transfer factor (TF; mean value 0.18) in ten rye fields with podzolic soils near Bremen, Germany, correlated positively with organic content of soil and negatively with Ca and P content. Further, Rabideau et al. (2005) performed real-time performance studies and consequent modelling of a permeable reactive barrier constructed of a natural zeolite material at the West Valley Demonstration Project in western New York State. Changes in soil temperature and moisture content have a considerable impact on the oxidation states (speciation) and consequently, the geochemical forms of ^{90}Sr in soil (Kovacheva et al. 2014). The concept of "effective half-life" as an indicator of long-term decay kinetics of a radionuclide in an environmental compartment (soil/water/plant) is also being reviewed recently (Corcho-Alvarado et al. 2016). The effective half-life may be expressed as close to ecological half-life and is calculated by using a regression analysis of the specific activity (expressed in mBq kg^{-1} dry weight of soil), assuming that the specific activity in the environmental compartment decreases exponentially with time. The effective half-life integrates all processes that potentially affect the activity and decay of the radionuclides in the compartment including physical radioactive decay (Pröhl et al. 2006; IAEA 2009).

Limited data are available on distribution of artificial radionuclides in meadow soils. However, radionuclide behaviour in the forest environment is becoming an important area of research in radioecology. Pourcelot et al. (2007) pointed out that ^{90}Sr activity concentrations in milk samples from cows that grazed in Alpine pastures are more elevated and show lower variability than that in soil and grass samples from the same area. Lukšienė et al. (2015) studied the spatial distribution of ^{90}Sr and other artificial radionuclide activity and concentration in the top layer (0–10 cm) of undisturbed meadow soils in Chernobyl NPP fallout affected areas of Lithuania. Therefore, milk might be a more sensitive indicator of ^{90}Sr contamination than soil or grass samples (Pourcelot et al. 2007; Kamenova-Totzeva et al. 2017). As early as in 1959, Lee (1959) reported estimated ^{90}Sr in soil, runoff and wheat. Gustafson (1959) reported ^{90}Sr accumulation of radioactivity in the kernels of Thatcher wheat which was about one-tenth of that in leaves and stems. Subsequently, Menzel (1960) found that only a small portion of ^{90}Sr (10%) deposited on cultivated soils were removed in runoff. Other than small plants (crops, etc.) seasonal and multi-year dynamics of Chernobyl-derived ^{90}Sr accumulation in woods are investigated by Shcheglov and Tsvetnova (2004).

Seasonal variation is more regular and predictable than multiyear variation (Fellows et al. 2009). The multi-year dynamics of ^{90}Sr within the plant body depends on type of landscape, kinetics of plant-available forms of ^{90}Sr and irreversible fixation of ^{90}Sr in the root-abundant soil layer (Shcheglov and Tsvetnova 2004).

Mahmoud and El-Hemamy (2005) studied the leakage pattern and consequent hydrological and geological effect of leaking of old (functional since 1961) underground settling tanks at the Egyptian Atomic Energy Authority (EAEA) Inshas site, where the tanks contained ^{90}Sr among other radionuclides. The migration of ^{90}Sr through the unsaturated zone (soil between tank and aquifer) was observed, that depends on soil type, thickness of the unsaturated zone, water velocity and retardation factor, which is a function of the specific distribution coefficient of ^{90}Sr (Mahmoud and El-Hemamy 2005). These studies may be termed as "mini radiological fallouts" as compared to Chernobyl and Fukushima (Maloshtan et al. 2017).

^{90}Sr in Water: Uptake and Interactions

Environmental ^{90}Sr deposited on soil and vegetation likely gets transported into freshwater ecosystems because of washout by spring high waters. Some reports of ^{90}Sr in the food chain and accumulation records are available on Russian, Belorussian and Ukrainian water bodies in post-Chernobyl accident, where the speed of vertical migration of ^{90}Sr was around 2–4 cm year^{-1} (Outola et al. 2009; Saxen and Koskelainen 2002; Fesenko et al. 2011). The vertical migration in soil resulted in higher absorption of ^{90}Sr in plants having deep root systems (Yablokov et al. 2008) finally leading to increased internal irradiation exposures of people in the contaminated territories (Yablokov et al. 2008).

The concentration of radionuclides in non-human biota is usually defined by biota-to-water concentration ratio (CR) (IAEA 2014) and used to subsequently estimate radiation dose to organisms or humans. For aquatic organisms especially for fishes, CR is usually defined as the ratio between the radionuclide concentration in the fish muscle (as mostly consumed) and the concentration in the surrounding water (Yankovich et al. 2010). For environmental risk assessment, CR$^{\text{muscle}}$ values can be converted to CR$^{\text{whole-organism}}$ using conversion coefficients (Yankovich et al. 2010). However, calculating the conversion factor is difficult, hence predicting the elements/radionuclides (like Sr/^{90}Sr), that accumulate in tissues other than muscle. Sr uptake mechanism resembles the ions of Ca (Kryshev 2006; Outola et al. 2009), with Ca^{+2} and Sr^{+2} competing for receptor sites on biological membranes (Van Leeuwen and Koster 2004). According to Saxen and Koskelainen (2002), 95% of ^{90}Sr accumulates in fish skeleton, scales and fins and only 2–5% accumulates in muscles.

Although the CR may ideally be defined as the concentration equilibrium of radionuclides in organisms with its surrounding media, practically, radionuclides in the environment and in organisms vary in a dynamic manner (Batlle et al. 2016). Moreover, to evaluate risk assessment, data on spatio-temporal variation (like data from various sites, longitudinal variation, etc.) must be considered (Avila et al. 2013; Beresford et al. 2016). Hence, it is very difficult accurately predicting risks related to radionuclide contamination from water to aquatic organisms in case of ^{90}Sr. The Fukushima Daiichi NPP (FDNPP) liquid release has given us insight of what may happen to seawater and marine biota after such an accident. Marine mussels (*Septifer virgatus*) provide an important animal system to assess the status of ^{90}Sr uptake and accumulation in marine biota on a spatio-temporal basis (Karube et al. 2016). Nelson (1962) studied ^{90}Sr concentration in freshwater clams and their shells; the study may be used as indicators of the ^{90}Sr contamination of their environment and helpful in predicting the concentrations within the organisms. Studies on the dispersion of ^{90}Sr from FDNPP site along the Pacific coast of eastern Japan (north direction by coastal current) and radiostrontium activity in coastal areas have revealed the environmental aspects related to aquatic ecosystems human safety assessments (Fujimoto et al. 2015; Castrillejo et al. 2016; Konovalenko et al. 2016). Fujimoto et al. (2015) reported correlation of fish contamination by beta emitting radionuclides and their concentration in marine water in and around Fukushima FDNPP harbour. The investigations include Japanese rockfish (*Sebastes cheni*), brown hakeling (*Physiculus maximowiczi*) and fat greenling (*Hexagrammos otakii*).

However, according to the study by Tomilin et al. (1987), the accumulation of radiostrontium in mammals is surprisingly low as evident from experiments being conducted on pigs fed with contaminated mine water to their food ratio as a salt additive. Degteva et al. (1994) accounted the dose received by individuals residing on the adjoining banks of Techa river (Russia) and its flood plains due to release of fission products (primarily ^{90}Sr) from the plutonium production facility “Mayak” from 1949 to 1956. The medium dose to the red bone marrow was about 250 mSv and the mean dose about 400 mSv (Degteva et al. 1994, 1996, 2000). Yablokov et al. (2008) studied the secondary ^{90}Sr contamination of freshwater ecosystems and found that the vertical migration velocity of ^{90}Sr in flood plains, lowland moors, peat bogs, etc. is about 2–4 cm year^{-1}, where plants play a major role in absorption and subsequent taking it to the surface again. This soil to surface transfer leads to a persisting activity in the upper soil horizons and consequently to doses of internal irradiation in the contaminated territories. Delistraty and Van Verst (2009) measured the tissue radiostrontium levels in water bird which are compared to upland birds from two time periods (1971–1990 vs. 1991–2009) and different locations (on-site and off-site). On-site median concentration of ^{90}Sr in bones of water birds was significantly higher than those in on-site upland birds (1991–2009) and on-site median concentrations in 1971–1990 were significantly higher than those in 1991–2009 for ^{90}Sr in water bird muscle (Delistraty and Van Verst 2009). Median concentrations of ^{90}Sr in bone were significantly higher than those in muscle for both avian groups (water and upland) and both locations. This study undoubtedly proves that the absorption mechanism of ^{90}Sr in the body is very similar to calcium (Delistraty and Van Verst 2009).

In soil and water, plants are the first links in the transfer chain of ^{90}Sr from the environment to human food (Moyen and Roblin 2010). Vose and Koontz (1959) have compared the Ca and ^{90}Sr absorption by different species of clover and grass grown in pots in three distinct soil types to distinguish species differences in uptaking strontium from the soil.

Metal Uptake by Plants: General Account

Metals in soil exist as a range of chemical species in a dynamic equilibrium; however, because of various factors like soil properties, solubility, ionic potential, biological constituents, etc. only a fraction of soil metal is readily available for plant uptake (Wenzel et al. 2003; Mitra et al. 2014). Therefore, plant-based remediation of contaminated soil (phytoremediation) can either be direct (where plants take up or absorb the metals through roots and subsequently translocate to the upper part of the plant) and rhizoremediation (where pollutants are confined to the rhizosphere only after transformation of the material either by enzymatic or root specific microbial actions) (Kuiper et al. 2004; Reboreda and Cacador 2008). Secretion of root exudates into the soil matrix plays a crucial role in heavy metal availability and uptake by plants (Mitra et al. 2014). However, the plant cell wall is functionally important in controlling metals uptake into the cytosol by immobilizing ions by providing histidyl groups, and extracellular carbohydrates such as callose and mucilage (Manara 2012). Admission of metals into plant tissue typically occurs through the root cellular membrane, where metals are directed towards the xylem through apoplast (Salt and Rauser 1995). Roots have high affinity chemical receptors to transport the ions into cells (Salt and Kramer 2000).

Effective reduction of heavy metals (HMs) in cytological active areas like cytoplasm and plasmids in the cells is mainly performed by common HM transporters involved in transportation to vacuole or in exclusion at plasma membrane. These HM transporters thereby help in reducing the levels of toxicity exerted by HMs as free radicals or indirect inducers of ROS in the sites (Inouhe et al. 2015). There are substantial evidences that many hyperaccumulator plants for various HMs are prevailed for these transportation mechanisms via xylem transport systems rather than their different detoxification mechanisms in the cells (Hossain et al. 2012; Socha and Guerinot 2014; Inouhe et al. 2015).

Strontium in Plants

Russell (1965) clarified the interactions of a typical vascular plant and radionuclide, which occurs at two levels: the first level is the aerial or shoot portion and the second level is the soil-root zone or rhizosphere of the plant. Accumulation of radionuclides by these two modes is interrelated, because radionuclides reaching the aerial part like foliage and shoot of the plant will ultimately be shed by the plant and eventually reach the soil (Russell 1965). However, two modes of radionuclide entry into plants are usually difficult to separate in practical scenario.

Significant portions of radionuclide solutions deposited on the leaf surfaces may get entry into the edible parts of plants (as for example, more than 40% of the cesium radioisotope in radishes) (Tukey et al. 1961; Oestling et al. 1989; Fortunati et al. 2004). Uptake of ^{90}Sr is also evident on leaf surfaces as it gets precipitated

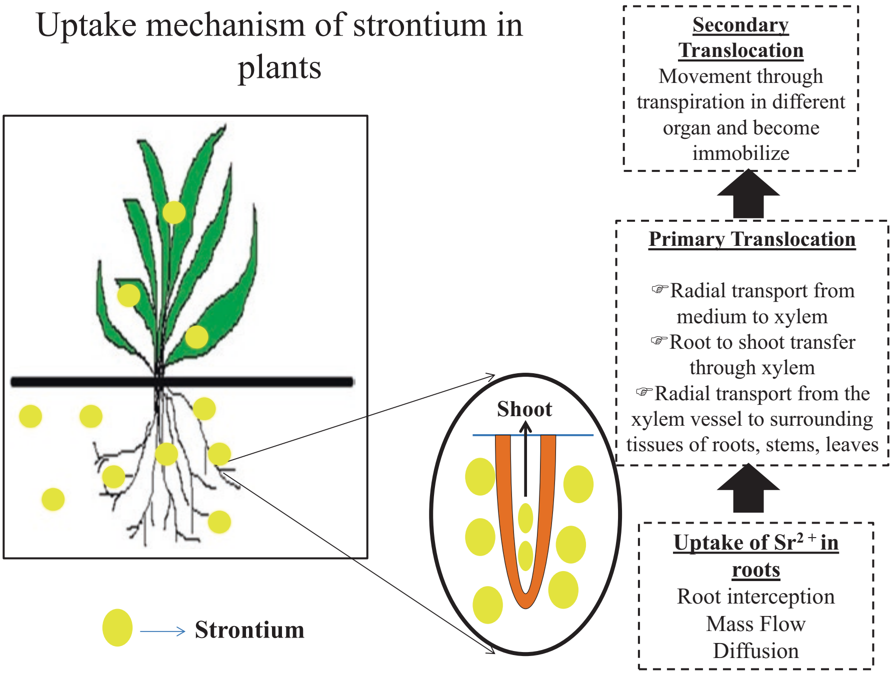

Fig. 1 Overview of uptake mechanism of strontium in plants

from atmospheric mist and contacting the leaf (cuticle and epidermis) (Fortunati et al. 2004). Both active and passive transport occurs for ^{90}Sr uptake via leaf epidermis, where it remains, as translocation from leafs to other plant parts is not common (Liu et al. 2016). According to Comar and Wasserman (1964), foliar uptake of Sr bears a non-specific competition with all the alkaline earth metals as compared to calcium alone for binding to sites on exchange compounds in case of passive transport, or on receptors in case of active transport. ^{90}Sr soil to root uptake depends on various physico-chemical factors of the soil components and the water-soluble form of the radionuclide (Fig. 1). Once the metal enters the body of the plant, it may get accumulated within the leaf tissues, while the accumulation of the metal in seeds and fruits is usually low (Asgari and Cornelis 2015). Even roots show lower activity in ^{90}Sr accumulation and translocation as compared to shoots (Wang et al. 2017). Willey (2014) has reported similar findings where plants showed more Sr in green shoots than woody parts, which is probably due to higher activity and flow of nutrients in green parts of the plants.

In 1950s, a study was conducted to assess the ^{90}Sr absorption in edible plants like wheat, rice, oats and barley. Lee (1959) studied the uptake of ^{90}Sr into wheat (Thatcher variety) and reported that the radioactivity in the kernels was about one-tenth in comparison with that of leaves and stems; milling of those kernels yielded flour with radioactivity lower than in brans. Ichikawa et al. (1962) reported the uptake of ^{90}Sr in rice due to root absorption from soil. Similar finding of root absorption of ^{90}Sr by rice was corroborated by Menzel et al. (1963), showing the definite observed pattern of radionuclide accumulation by field-grown crops.

Rickard and Price (1990) pointed out a positive correlation between ^{90}Sr concentrations in red canary grass grown near the nuclear reactor of the U.S. Department of Energy (Hanford site) in the eggshells of Canadian geese feeding on that red canary grass. This study was an important example of ^{90}Sr bioaccumulation across the food chain. Paasikallio et al. (1994) studied the soil to plant transfer of ^{90}Sr in different cereals and vegetables, where it was found that green leafy vegetables like lettuce and cabbage showed more accumulation, followed by cereals and tuberous vegetables (potato).

Strontium and Rhizosphere

Determining the plant-available fraction of a radionuclide is difficult considering the rhizospheric zone (Gupta et al. 2016a). The plant root-soil region is completely different from the bulk soil. Plants root exudates typically increase the richness of soil microflora (bacterial and fungal communities) by 1–4 orders of magnitude compared to the adjacent bulk soil that aids to degrade diverse pollutants (Anderson et al. 1994). The availability of metal to the plant depends upon the chemistries between soil fraction and variety of substances that a plant root exudes or releases that creates a micro-environment specific to the plants. Root exudates include: diffusates (e.g. organic or amino acids, inorganic ions, sugars, and water), excretions (e.g. bicarbonates, carbon dioxide, and protons), secretions (e.g. siderophores, mucilage, and allelopathic compounds) which help moderating soil microflora community structure to have a better variety of metabolic capabilities with unique gene pool (Hall 2002; LeDuc and Terry 2005; Mitra et al. 2014; Chatterjee et al. 2017a). As for instance, release of H^+ and HCO_3^- at root zone actively influences the pH in their immediate vicinity, thus increasing the accessibility of phosphorus and potassium (Gupta et al. 2016a).

Similarly, ammonium (NH_4^+) mobilizes Sr on soil surface, leading to increased Sr concentration in soil solution, although it inhibits uptake by plants, like that of calcium. Thus, common ammonium fertilizers like diammonium phosphate (DAP) and NPK may act as inhibitor for the soil to plant transfer of ^{90}Sr after a radioactive fallout in agricultural fields, acting as a countermeasure (Guillén et al. 2017).

Plant growth-promoting rhizobacteria (PGPR) perform an important role in development by reducing the physiological stress in plants, when grown in contaminated soils. By stimulating the production of plant growth regulators like indole acetic acid, gibberellic acid, cytokinins and ethylene, PGPR helps in rhizospheric colonization. This microbial colonization contributes to plant health and better response. Rhizobacteria can also secrete antibiotics, hydrocyanic acid, phosphate solubilizing substances, siderophores and 1-aminocyclopropane-1-carboxylic acid (ACC) to increase bioavailability and root absorption of different metals (Meyer 2000; Davies et al. 2001; Gupta et al. 2016a). Burd et al. (1998) reported that after adding of *Kluyvera ascorbata* SUD165/26 a related rhizobacteria, germination and growth of Indian mustard (*Brassica juncea*) seeds was increased by 50–100% in a nickel-contaminated soil.

Number of metal transporter-proteins show significant roles in homeostasis of heavy metals at root cell plasma membranes. Amid diverse group of transporter-proteins, NRAMP (natural resistance-associated macrophage protein), CDF (cation diffusion facilitator) family, ZIP (Zinc importer) families (ZRT, IRT-like Protein; [ZRT—Zinc regulated transporter, IRT—iron regulated transporter]), heavy metal ATPases (HMAs) family like P1B-ATPases, ATP-binding cassette (ABC) transporters, copper transporter (COPT) family proteins, ABC transporters of the mitochondria (ATM), multidrug resistance associated proteins (MRP), Ca^{2+} cation antiporter (CAX), and yellow-stripe-like (YSL) pleiotropic drug resistance (PDR) transporters are well studied (Dubey 2011; Huang et al. 2012; Gupta et al. 2013, 2016a). A ZIP family transporter with histidine-rich domain is important in activated response and uptake to divalent metal ions (Kramer et al. 2007). Nishida et al. (2008) reported that IRT1 is responsible for transportation of metals like Fe^{2+}, Mn^{2+}, Zn^{2+} and Cd^{2+} in root cells of *A. thaliana*. Internal transporters like HMAs family transporters (P1B-type ATPases) help in loading of metals like Cd and Zn from the surrounding tissues into the xylem and performing as an efflux pump (Kramer et al. 2007). Sequestration of heavy metals at vacuole level is mainly carried out by AtHMA3 transporter present in the tonoplast membrane (Manara 2012; Gupta et al. 2013). Plants have their own general tactics that use vacuolar sequestration through proton pumps like vacuolar proton-ATPase (V-ATPase) and vacuolar protonpyrophosphatase (V-PPase) to isolate metals from metabolically active cytosol and cellular organelles (Dalcorso et al. 2010).

Sr Removal from Soil: Phytoremediation Approach

Removal of ^{90}Sr from soil is an interesting study carried out by different workers. As per the field study of Fuhrmann et al. (2002) on redroot pigweed (*Amaranthus retroflexus*), Indian mustard (*Brassica juncea*), and tepary bean (*Phaseolus acutifolius*) to determine the ability of extraction of ^{90}Sr from contaminated soil, it was found that the redroot pigweed was a most potent accumulator (estimated time required for removal of 50% of ^{90}Sr is 7 years, assuming two crops per year). *Calotropis gigantea* plants were also studied to remove ^{90}Sr from soil, where Eapen et al. (2006) demonstrated 90% of the radioactivity can be removed within 24 h. Broadley et al. (2003, 2004) and Willey and Fawcett (2006) identified the efficiency of Caryophyllidae (Eudicot plants, viz. amaranth, buckwheat, chards and beets; most of them being agriculturally important plants) clades for the transfer of alkali earth metals particularly ^{90}Sr from soil to roots.

Zheng et al. (2016) studied angiosperm *Tillandsia usneoides* (Bromeliaceae, known as Spanish moss, an established bio-monitor of air pollution) as a bio-indicator for ^{90}Sr, as this plant can tolerate Sr for an elongated period and has a very high uptake ratio which has a positive correlation with concentration of Sr in solution. It was also reported that high Sr concentration inhibits chlorophyll content due to oxidative stress, while low concentration promotes chlorophyll synthesis in Spanish moss (Zheng et al. 2016). Maxwell et al. (2010) and Amano et al. (2016) reported different methodologies to determine the concentration of ^{90}Sr in edible vegetation samples using liquid scintillation counting (LSC) and inductively coupled plasma (ICP)-mass spectrometry (MS).

Weed plants, in many a times, have been hyperaccumulators of metals or radionuclides (Wenzel et al. 1999; IAEA 2006). The soil-to-plant transfer factor accounts for the uptake of radionuclides via plant roots and represents the activity concentration ratio of the radionuclide per unit dry mass in the plant (Bq kg^{-1}) to that in the soil, designated as Fv (IAEA 2009, 2010). Attar et al. (2016) studied the effect of ageing on the transfer of ^{90}Sr to lettuce (*Lactuca sativa*) and winter wheat (*Triticum aestivum*) with respect to a clayey soil and semi-arid environment in Syria. Moderate accumulation of ^{90}Sr from soil may be compensated by high biomass and fast growth (Hernandez-Allica et al. 2008). Several authors have worked on the suitability of rice, wheat, rye and oat where existing data indicates that oats may be a better accumulator of ^{90}Sr as compared to wheat and rye (Krouglov et al. 1997; Soudek et al. 2006; Schimmack et al. 2007). In case of both rice and oats, it was also found that in the main plant where around 99% of ^{90}Sr is stored is the non-edible overground parts, i.e. leaves and stems as compared to roots because roots have lower biomass (Tsukada et al. 2005; Lin et al. 2015). As per the available reports, the activity concentration in a nutrient solution of 1 Bq L^{-1} of ^{90}Sr or ^{137}Cs corresponds to ca. 2×10^{-15} Moles L^{-1} however, the average concentrations of chemical homologues like K, Ca, and Mg, in soil solution are in the order of 1 mMoles L^{-1} . Uptake and co-precipitation of radionuclides like radioactive strontium by plants is therefore affected by many soil factors, where cation exchanges reactions regulate the concentration of a competitive ion, like Sr, in soil solution. Sorption of radiostrontium is governed by reversible exchange with major cations like Ca^{2+}. Although Sr is exchanged in preference to Ca in minerals, the preference of the same metal may get changed when organic matters are present (Chu et al. 2015).

Strontium Exposure, Plant Responses and Phytoremediation

Irrespective of biological necessity, plants usually take up several cations present in their root region. When plants are exposed to radionuclide and ionizing radiation, cellular and molecular effects take place, involving direct damage of macromolecules or indirect radiolytic reactions producing reactive oxygen species (ROS) (Gupta et al. 2016b, 2017). Ionizing radiation can induce DNA strand breaks, lipid oxidation, or enzyme denaturation (Gupta and Walther 2014; Gupta et al. 2016a). Antioxidative defence system of plants having enzymes (e.g. superoxide dismutase (SOD) and catalase (CAT)) and metabolites (e.g. ascorbate and glutathione) controls the ROS within cellular systems (Gupta and Sandalio 2012). Plants employ complex coordinated mechanism for metal tolerance mechanisms involving both biochemical and physiological processes. To cope with toxic effects of radionuclides, plants may avoid (restricting the metal uptake) or tolerate (survive in the presence of high internal metal concentration) the stress condition (Chatterjee et al. 2017b). Avoidance mechanism of plants involves concentration reduction of metal entering into the cell by extracellular precipitation, biosorption to cell walls, reduced uptake, and/or by increased efflux (Gupta and Walther 2014; Gupta et al. 2016a). Elaborate physiological responses are also present in plants like intracellular chelation by synthesizing organic acids, amino acids, glutathione (GSH), and/or by metal-binding ligands such as phytochelatins (PCs) and metallothioneins (MTs), vacuolar compartmentalization, glyoxalase systems and up-regulation of antioxidant defence to defy the harmful effects rooted by ROS (Gupta and Sandalio 2012; Gupta et al. 2016a).

Sasmaz and Sasmaz (2009) studied Sr phytoremediation potential in *Euphorbia macroclada*, *Verbascum cheiranthifolium* and *Astragalus gummifer*, where they investigated the roots and shoots of the plants for distribution and accumulation of the metal. The Sr enrichment factors for root (ECR) and shoot (ECS) and mean translocation factors (TLF) of these plants were examined and the authors found that the shoots of these plant (*Euphorbia macroclada*, *Verbascum cheiranthifolium* and *Astragalus gummifer*) were competent Sr bioaccumulator (having high TLF) which may be used in amelioration of Sr-contaminated soil (Sasmaz and Sasmaz 2009). The probable use of coyote willow (*Salix exigua*) plants for phytoextraction of the soil Sr through the plant roots and into above-ground shoots is being tested by the Department of Energy (DOE), USA, to protect the environment contaminated with ^{90}Sr (Fellows et al. 2009). The study showed that, as plants take up ^{90}Sr from soil, the soil content of the ^{90}Sr is also being reduced. Further, the study also pointed out that the hazard for detectable transfer of ^{90}Sr through the food chain of herbivorous insects from willow trees grown in the Sr-contaminated soil is minor to nonexistent (Fellows et al. 2009). Wang et al. (2017) studied phytoremediation of Sr-contaminated soil by *Sorghum bicolor*. This study examined soil microbial community-level physiological profiles (CLPPs), where it was found that Sr-spiked soil is having enhanced soil-microbial diversity and activity with considerable augmentation in the height and the stem biomass weight of the plant (with Sr content in tissues decreased in the order of leaves > roots > stems) (Wang et al. 2017).

Conclusion

^{90}Sr is a fission product of uranium (U) and plutonium (Pu). Anthropogenic activities led to discharge and waste disposal of Sr in many places of the world that impacted soil and groundwater. There are certain factors for implementing specific plants for restoration of radionuclide-contaminated sites, which includes relative high levels uptake potential of plants for the radioactive material and growth or high biomass production. Potential plants must amass radionuclide in the above-ground parts at a proportion which significantly exceeds the soil concentration. Several factors limit the application of plant-based techniques that affect the Sr uptake by plants, which include clay/soil particle/minerals properties, and availability of organic matters. However, phytoremediation might be the appropriate and easy technique for restoration of radionuclide-contaminated soil if selected cultivars are used. Therefore, detailed research on plants selection, manipulation of diverse transporters, and explication of anti-stress physiology factors in plants are the area which will be useful to decontaminate Sr-contaminated soil in an eco-friendly manner.

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Factors Influencing the Soil to Plant Transfer of Strontium

Javier Guillén

Introduction

Radiostrontium was released into the environment as consequence of the global fallout due to atmospheric nuclear weapon tests in the 1950–1960s, and accidents involving nuclear material, being Chernobyl (1986) and Fukushima-Daiich (2011) the most important. The quantity of released ^{90}Sr was estimated in 622 PBq for the global fallout, 10 PBq for Chernobyl and 3.3×10^{-3}–0.14 PBq for Fukushima Daiichi (UNSCEAR 2000; IAEA 2015). Strontium is an alkaline earth element, chemically analogous to calcium. The radiotoxicity of ^{90}Sr is higher than that of ^{134,137}Cs (ICRP 2012). These characteristics, along with its long half-life, 28.8 years, make the ^{90}Sr one of the radionuclides to consider in radioprotection. However, the number of studies devoted to this problem is significantly lower than to radiocaesium. One of the reasons for this is the fact that the determination of ^{90}Sr in environmental samples requires radiochemical separation procedures (Guillén et al. 2010), due to the absence of any significant gamma emission in its decay scheme. However, other strontium isotopes, such as ^{85}Sr (γ-emitter), have been used in controlled experiments.

The soil-to-plant transfer is of particular interest, since radiostrontium can enter through this pathway into the human food chain, either by direct consumption or by indirect consumption via animal feeding. International regulations on foodstuff are focused on maximum permitted activity levels after a nuclear emergency (EU 1989a, b; CA 1995, 2011). There is discrepancy between these reference levels, in the actual value of the activity level, 750 or 1000 Bq kg^{-1} f.w., for both ^{89}Sr and ^{90}Sr. In the case of Fukushima Daiichi accident, radiostrontium was not considered in the provisional regulation values (MHLW 2011; Hamada and Ogino 2012), because the radiostrontium deposition was far lower than that of radiocaesium due to its low

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volatility (IAEA 2015). In case of existing exposure scenarios, EU has regulation for foodstuff and feeding stuff from Chernobyl and Fukushima Daiichi contaminated areas, but only regarding radiocaesium (EU 2008, 2014). Therefore, in this chapter, a review of the main factors affecting strontium soil-to-plant transfer was carried out, including its bioavailability in soil, competition with calcium, and remediation techniques.

Transfer Factors

Soil-to-plant transfer processes can be quantified in different ways. The use of transfer factors, F_v, is one of the most common ways, and it is defined as the ratio between the radionuclide concentration in the plant and in the soil (see Eq. 1).

$$F_v = \frac{\text{Bq / kg d.w. plant}}{\text{Bq / kg d.w. soil}} \tag{1}$$

Radionuclide concentration in the plant it is usually considered as the edible part, for humans or intended for animal feeding. It is expressed in dry mass basis to minimise additional variations due to different water content between plant species. Transfer factors expressed in fresh mass can also be found in the literature, especially used in case of direct consumption as is the case of fruit. Typical fresh–dry ratios for different plants are available in IAEA TRS 472, Appendix I (IAEA 2010). Regarding the radionuclide concentration in soil, depth of the layer of soil considered in the calculation is extremely important for those radionuclides deposited on soil and with an inhomogeneous depth distribution, such as ^{90}Sr. In order to reduce the influence of this variable, the International Union of Radioecology (IUR) recommended a standardized root location in soil, assuming that all roots and all radionuclides present in the rooting zone are in that soil layer, 0–10 cm for grass and 0–20 cm for all other crops, including trees (IUR 1992).

Figure 1 shows the worldwide variation of transfer factors for different plants/crops. A range about 4–5 orders of magnitude can be observed, which imply that soil to plant transfer of strontium is influenced by many variables. These variables can be classified in:

- Biological variables: type of crop/plant; biological variability inherent to plants; differences between plant varieties; variations in the nutritional requirements at different development stages; detoxification mechanisms.
- Soil variables: type of radionuclide considered; time and chemical form in which radionuclides are deposited on soil; physicochemical properties of soil (pH, CEC, organic matter, etc.); hydrological conditions within the soil; plant bioavailability of radionuclides.

The F_v values for strontium are usually higher than those for radiocaesium and other radionuclides in the same environment (Al-Oudat et al. 2006; Velasco et al.

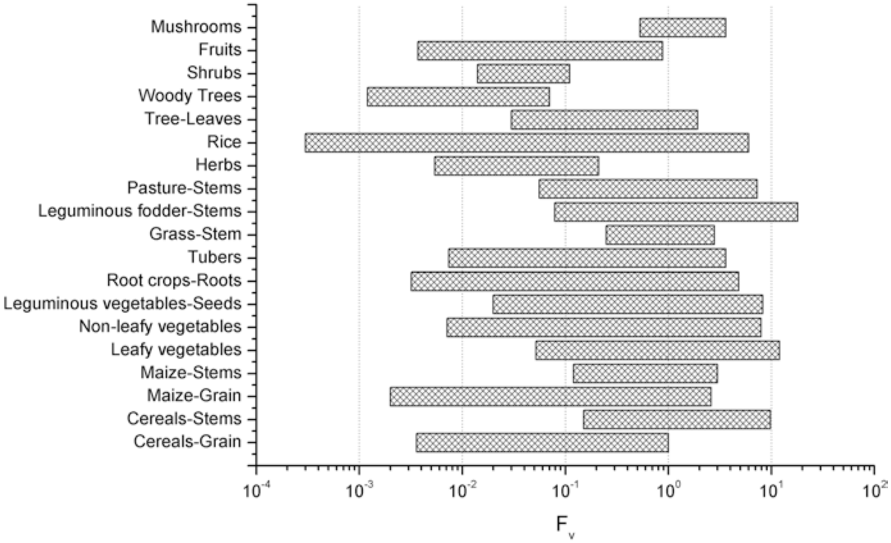

Fig. 1 Worldwide range of variation of transfer factor for strontium (^{90}Sr and stable strontium). Data adopted from Carini (2001), Baeza et al. (2006), Gerstmann and Schimmack (2006), De França et al. (2007), Uchida et al. (2009), IAEA (2010), Aliončik and Butkus (2012), Al Attar et al. (2015), Sarap et al. (2015), Noli and Tsamos (2016)

2008; IAEA 2010; Al Attar et al. 2015), indicating that plant uptake preferentially strontium. Different F_v values for different plant tissues (or parts) can also be observed in Fig. 1, being these maximum values for stems and shoots higher than for grains/seeds for the same plant group, such as cereals, maize, and leguminous vegetables. This was also reported in different studies focusing on transfer to several plant parts:

- Wheat: Roots > Straw > Grain (Al Attar et al. 2015; Sarap et al. 2015; Al Attar et al. 2016)
- Rice: Straw > Hulled Seed (Tsukada et al. 2005; Choi et al. 2007)
- Tree: Leaves > Branches > Fruit > Oil/Seed (Al-Oudat et al. 2006; IAEA 2010)

These F_v values reflect the translocation of strontium within different parts of plant. From a formal point of view, translocation describes the distribution within the plant subsequent to foliar deposition and absorption by leaves (Thiessen et al. 1999). Although foliar deposition is not strictly related to soil-to-plant transfer, it is related to radionuclide mobility within plants. The amount of translocated radionuclide is highly dependent on the stage of development at which deposition occurs (Thiessen et al. 1999), which also reflects the nutritional requirements of the different plant parts in each stage. Strontium was classified as medium mobile element within plant, based on experimental data on cereals and rice (Colle et al. 2009). This degree of mobility may be expected, because strontium is an alkaline earth element, chemically analogue to calcium, which can be considered as a structural element.

Table 1 Range of variation of aggregated transfer factor, T_{agg}, reported in the literature

Plant group	Range T_{agg} ($m^2\ kg^{-1}$ d.w.)	References
Rice-Straw	8.2×10^{-3}–2.1×10^{-2}	Choi et al. (2007)
Rice	1.7×10^{-4}–7.6×10^{-4}	Choi et al. (2009)
Leafy vegetables	4.4×10^{-2}–8.6×10^{-2}	Choi et al. (2009)
Non-leafy vegetables	1.2×10^{-6}–2.8×10^{-6}	Choi et al. (2009)
Root crops	1.7×10^{-4}–8×10^{-4}	Choi et al. (2009)
Tuber	2×10^{-4}–1.6×10^{-3}	Choi et al. (2009)
Tree-Wood	5.7×10^{-4}–1.0×10^{-2}	IAEA (2010)
Tree-Leaves	1.5×10^{-3}–7.8×10^{-2}	IAEA (2010)
Mushrooms	5×10^{-3}–6×10^{-3}	IAEA (2010)

Willey (2014) reported that the radio Ca/Sr was quite stable in plants. This stable ratio value may explain the positive correlation between ^{90}Sr activity concentration in wheat and calcium concentration (Putyatin et al. 2006).

There are also other transfer parameters which are considered in case of radioactive fallout, the aggregated transfer factor, T_{agg}, and the geochemical transfer factor, T_{geo} (see Eqs. 2 and 3, respectively). Radionuclide concentration in soil is expressed in terms of its deposition on surface layer of soil. Table 1 shows the range of variation of aggregated transfer factor, T_{agg}, for different plant groups. Similar differences between plant parts can also be observed (straw > grain).

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

$$T_{geo} = \frac{\text{Bq / m}^2 \text{ in. plant biomass}}{\text{Bq / m}^2 \text{ in soil}} \quad (3)$$

These definitions of transfer factors (F_v, T_{agg} and T_{geo}) are based on the intrinsic assumption that a steady state in the ecosystem is reached and that the transfer process is proportional to the radionuclide concentration in soil. 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).

Factors Affecting the Strontium Soil-to-Plant Transfer

Climate

Climate may influence soil-to-plant transfer, although its direct influence seems to be minimal (IAEA 2010). Table 2 shows, as a way of example, the range of variation for the same plant groups reported in different climates (IAEA 2010). Similar

Table 2 Range of variation of strontium F_v values for different plant groups and climates

	Climate		
Plant group	Temperate	Subtropical	Tropical
Cereals-Grain	3.6×10^{-3}–1	3.6×10^{-2}–6.5×10^{-2}	0.44–0.76
Cereals-Stem	0.15–9.8	1.2×10^{-2}–0.42	–
Leafy vegetables	0.39–7.8	5.2×10^{-2}–5	1.4–12
Root Crops-Roots	3×10^{-2}–4.8	3.2×10^{-2}–0.87	1.2–2.8
Tubers	7.4×10^{-3}–1.6	5.3×10^{-2}–3.6	0.66–0.7

Data adopted from IAEA (2010)

ranges can be observed for climates considered. In some cases, reported ranges in subtropical and tropical climates are narrower than in temperate climate, but it can partly be due to fewer numbers of data available. Clime can also influence the transfer processes in an indirect way, as it influences soil development. In tropical ecosystems, organic material in soil is fast decomposed, and there is a rapid recycling of nutrients and high mineral weathering rates; whereas in temperate ecosystems the organic matter decomposition is slower and it can be accumulated in soil (IAEA 2010).

Biological Variability

As it can be seen in Fig. 1 and Table 1, the type of plant/crop is one of the main factors influencing the transfer process. Ranges were different between for each plant group. Regarding a plant species, the analysis of inter-varietal differences in transfer parameters may seem a good strategy to reduce the dose to general population. Inter-varietal variation can be expressed as the ratio of the maximum to minimum observed concentrations for a given radionuclide and crop (Penrose et al. 2015). For strontium, it ranged from 1.0 to 4.5, being slightly lower than for caesium (Putyatin et al. 2006; Penrose et al. 2015). Gerstmann and Schimmack (2006) also observed a reduction in the variation when considering different cultivars at a given site. Varieties that accumulated low amount of strontium were also found to accumulate low quantities of calcium (Penrose et al. 2015). However, no variety was found to show low accumulation of strontium and caesium at the same time (Gerstmann and Schimmack 2006), and it may not be feasible because the concentrations of these elements are not related in plants (Penrose et al. 2015).

The development stage of the plant can influence the soil-to-plant strontium transfer. Al Attar et al. (2015) reported that lettuces contaminated with ^{90}Sr at earlier stages showed greater F_v values than when it occurred at a later stage. It was attributed to a decrease of physiological activity of plant towards maturation. Transfer of ^{90}Sr to rice plants grown under controlled conditions was reported to be higher when the contamination occurred at booting stage than before transplanting plants to the flooded soil (Choi et al. 2007).

Soil

The association of strontium to different geochemical phases that constitute soil particular is extremely important to soil-to-plant transfer processes. From an experimental point of view, this association is usually determined by the application of sequential extraction procedures using reagents with increasing extraction/replacement power and designed to attack a specific geochemical phase. Many speciation schemes are based on that proposed by Tessier et al. (1979), which considered the following fractions: (1) water soluble, extracted with double-distilled H_2O; (2) exchangeable fraction, extracted with 1 M NH_4OAc; (3) carbonated fraction. extracted with 1 M sodium acetate; (4) easily reducible fraction, extracted with 0.1 M NH_2OHHCl; (5) moderately reducible fraction, extracted with 0.2 M ammonium oxalate; (6) organic/hydrogen-sulfide fraction, extracted with H_2O_2 30% and NH_4OAc; (7) acid fraction, extracted with HNO_3 40%; and (8) residual fraction. This speciation scheme, and its variations, has been used widely to analyse the association of radionuclides to soil particles (Fawaris and Johanson 1995; Riise et al. 1990; Schultz et al. 1998; Guillén et al. 2014). There are also other schemes based on the extraction by different acids and the association with organic matter (Pavlotskaya 1974; Cook et al. 1984; Rigol et al. 1999). However, one of the major handicaps of the use of these sequential extraction procedures is that they are not specific (Schultz et al. 1998). Therefore, it has been suggested to characterise the different fractions as extracted with a given reagent and conditions, instead of the initial geochemical phase named by Tessier and Campbell (1979). Cumulative errors in the speciation procedure can also be another source of uncertainty (Kennedy et al. 1997).

Water soluble and exchangeable fractions are usually considered as readily available for plant uptake. Soil solution consists of the water present in pores within soil particles. There are several procedures to extract it: using porous ceramics cups, adding distilled water; or using specific resins (Nisbet et al. 1993a; Agapkina and Tikhomirov 1994; Jouve et al. 1999). Radionuclide concentration in soil solution has been previously used to describe soil-to-plant transfer processes and the efficiency of agricultural countermeasures (Nisbet et al. 1993a, b; Camps et al. 2004). The K_d values can also be used to derive an estimation of the concentration in soil solution in a mechanistic model (Casadesus et al. 2008). Exchangeable fraction is extracted with NH_4OAc in most speciation schemes, as it is considered a robust extractant for acidic or neutral soils but unsuitable for alkaline soils (Kennedy et al. 1997). It can be buffered into the soil pH to extend the range of pH in which it can be used. Other extractants used are: $MgCl_2$, $CaCl_2$, EDTA, DTPA, NH_4NO_3, KCl, or $NaNO_3$ (Kennedy et al. 1997; Komosa 2002; Rigol et al. 2002).

Strontium is mainly associated with exchangeable fraction in soil in the range 40–70%, but higher values were also reported (Riise et al. 1990; Baeza et al. 1999; Sysoeva et al. 2005; Guillén et al. 2010; Takeda et al. 2010; Jeske and Gworek 2013). Exchangeable fraction is usually higher than water soluble fraction (Guillén et al. 2010; Takeda et al. 2010). These fractions are considered to be very mobile in soil. Therefore, strontium association with them can explain its downwards migration in soil profiles, with higher velocities than those reported for other anthropogenic radionuclides (Arapis et al. 1997; Forsberg et al. 2000; Guillén et al. 2015). Correlation between transfer parameters and ^{90}Sr exchangeable were reported (Sysoeva et al. 2005). The concentration of other elements in the exchangeable fraction can also influence soil-to-plant strontium transfer, in particular calcium, as strontium is its chemical analogue. Exchangeable calcium concentration was reported to be higher than that of strontium for a tropical ecosystem (Bullen and Chadwick 2016). Negative correlation between exchangeable calcium concentration and transfer parameter were also reported (Choi et al. 2007). In fact, an increase in the Ca:Sr ratio in the soil solution was previously suggested as a countermeasure to reduce the strontium transfer (Nisbet et al. 1993a, b).

Ageing effect for strontium is quite low compared to radiocaesium, and it depended on several factors, such as its source term and soil type. Krouglov et al. (1998) reported an increase of exchangeable strontium in soils contaminated by Chernobyl accident as consequence of the weathering of fuel particles deposited in soil. High ^{90}Sr mobility in other nuclear test sites may be related to differences in the chemical form in which it reached the environment, leakage from explosion tunnels along water courses rather than associated with fused silicates (Howard et al. 2004). For other source terms, the available fraction was reported to be almost constant with time (Rigol et al. 1999; Takeda et al. 2010; Al Attar et al. 2016), or it decreased with time but at a lower rate than radiocaesium (Baeza et al. 1999; Rigol et al. 1999; Sysoeva et al. 2005). The F_v values for crops harvested at different times also reflected this variation. They were almost constant with time, taking into account the associated uncertainty (Al Attar et al. 2016), or decreased with time until reaching a pseudo-equilibrium state (Uchida et al. 2009). For Scot pine wood, transfer parameter variations were reported, with a maximum in the period 1986–1991 (probably due to Chernobyl fallout) and a subsequent decrease with time (Aliončik and Butkus 2012).

Organic matter content in soil can have a significant influence on strontium transfer. Soils with high organic matter content, mainly peat, were reported to have low F_v values (Nisbet and Shaw 1994; Carini 2001). This was suggested to be due to the formation of organo-metallic complexes able to retain ^{90}Sr more strongly than ion exchange sites in mineral sites (Nisbet and Shaw 1994). Strontium interaction of with these complexes was considered to be responsible for ageing effect on some soils (Rigol et al. 1999). In speciation schemes, the fraction associated with organic matter is usually extracted with H_2O_2, NaOH, $Na_4P_2O_7$, or NaClO (Tessier and Campbell 1979; Cook et al. 1984; Rigol et al. 1999; Guillén et al. 2015). Organic acids in soil, mainly humic and fulvic acids can be extracted with NaOH, or the combination of NaOH and $Na_4P_2O_7$ (Kononova 1982; Lee and Lee 2000). It is based on the formation of precipitates or soluble complexes with Ca, Fe, Al, and other polyvalent cations, to which humic substances are, bound (Kononova 1982). Then, humic and fulvic acids can be separated by adjusting the solution pH to 2. Humic acids precipitate while fulvic acids remain soluble (Lee and Lee 2000). The fraction extracted with H_2O_2 can be considered as oxidisable organic matter. Strontium association with humic and fulvic acids was lower than the exchangeable fraction, but higher than that extracted with H_2O_2 (Rigol et al. 1999). Strontium association with fulvic acids was higher than with humic acids (Lee and Lee 2000; Guillén et al. 2015).

Evidences on the influence of mycorrhizal fungi on the transfer of strontium are not decisive. Seedlings from two pine species inoculated with ectomycorrhizal fungi were reported to accumulate about ten times more ^{90}Sr than non-mycorrhizal (Entry et al. 1994). This effect was also reported for different grass species (Entry et al. 1999). However, Ladeyn et al. (2008) reported no effect of mycorrhizal association on strontium in *Pinus pinaster* associated with *Rhizopogon roseolus*. Therefore, there is no general trend in the effect of mycorrhizic fungi in symbiotic association with plants and further research is required.

Fractions extracted with inorganic acids are of minor importance for strontium. Different acid concentration can also be considered. Diluted inorganic acids, 1 M HCl, can remove cations from exchange complexes in the soil, and also dissolve oxides, hydroxides, carbonates, and some alkaline earth compounds. In some speciation schemes, this fraction was considered as potentially available. Concentrated mineral acids, 6 M HCl, 7–8 M HNO_3, are able to extract cations from interlayer or structural positions not readily accessed by exchange reactions (Krouglov et al. 1998). Residual fraction represents the fraction really strongly attached to soil particles. Radionuclides in these fractions are considered as unavailable for transfer.

Soil type and its physicochemical parameters can influence the strontium transfer to plants. Soil pH was reported to have a negative correlation with strontium transfer to rice straw (Choi et al. 2007). The association of strontium with clay minerals was weaker than for caesium (Van Bergeijk et al. 1992; Dyer et al. 2006). Strontium can be reversible adsorbed on regular exchange sites (Ehlken and Kirchner 2002; Sysoeva et al. 2005; Zhao and Chen 2006). The mode of strontium adsorption was found to be pH dependent: below pH 4.5 ion exchange predominated; and above that value multilayer adsorption occurred (Atun and Kaplan 1996). Water management can also influence transfer to crops, especially in the case of rice. Rice bran can naturally concentrate alkaline and alkaline earth elements than white rice (milled rice). Choi et al. (2007) suggested than the reductive conditions in flooded soils and the microbial activity can increase the amount of ^{90}Sr released from soil.

Modification of Strontium Transfer

Radionuclide speciation can be modified by the addition of soil amendments. If the objective is to reduce the strontium transfer, it is usually considered as countermeasures. The use of calcium based fertilizers, such as lime, is one of the most usual. It is based on the competition between calcium and strontium in plant uptake. However, its success is more limited than the addition of potassium based fertilizers in the case of radiocaesium, about 20% (Lembrechts 1993). Soils with low calcium content presented the highest reduction for strontium (Shaw 1993). Depending on the soil type, the addition of lime can be beneficial for mineral soils, but deleterious for organic ones (Nisbet et al. 1993a). The limited effectiveness of this countermeasure can be attributed to the fact that the rates of fertilizers used, 1.6–15.6 ton Ca/ha, were not able in increase the calcium concentration in soil solution (Vidal et al. 2001; Camps et al. 2004). Liming in excess may also fail to reduce the strontium uptake by plants (Lembrechts 1993).

The addition of other types of fertilizers may also have some effect on the soil-to-plant transfer. Potassium based fertilizers can increase calcium and strontium concentration in soil solution, but the ratio Ca:^{90}Sr remained unaltered and no effect was reported (Nisbet et al. 1993a, b). The addition of phosphate was considered to be effective due to the potential formation of insoluble strontium phosphate (Shaw 1993). The use of ammonium based fertilizers (diammonium phosphate and NPK type) can increase the radiostrontium uptake by plants and increase the plant calcium concentration (Guillén et al. 2017). The addition of bone to soil can reduce the strontium transfer due to its incorporation into the carbonated fraction, reducing in the same way the exchangeable fraction (Dimović et al. 2013).

Conclusion

An accurate knowledge of the soil-to-plant transfer of radionuclides is necessary in order to assure an adequate level of radioprotection, in particular for radiostrontium. In this study, we have shown that:

- There is a range of variation up to 4–5 orders of magnitude for the F_v values reported in the literature.
- The F_v values are not constant for all plant/crops. There are variations between different plant groups, and also intra-varietal variations within the same species. Other variations regarding plant development stage and different distribution within plant were also reported.
- Strontium in soil is mainly associated with exchangeable and soil solution fraction in soil. Therefore, it is very readily bioavailable for plant uptake.
- Strontium can form complexes with organic matter content in soil and become less bioavailable. Organic soils are reported to present low transfer values. It may also responsible for the ageing effect of radiostrontium.
- Calcium concentration in soil is of major importance to the strontium transfer, especially its concentration in soil solution and exchangeable fractions. An increase in the Ca:Sr ratio in these fractions can reduce the strontium transfer.

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Distribution of Strontium in Soil: Interception, Weathering, Speciation, and Translocation to Plants

Sergiy Dubchak

Introduction

Strontium (Sr) is the element of the II group of the periodic system. Natural strontium belongs to microelements and consists of a mixture of four stable isotopes ^{84}Sr (0.56%), ^{86}Sr (9.96%), ^{87}Sr (7.02%), and ^{88}Sr (82.0%). According to its physical and chemical properties, strontium is analogous to calcium being its companion in geochemical processes. Its estimated clarke content the earth's crust is 340 mg kg^{-1}. Strontium is contained in all plant and animal organisms in an amount of 10^{-2} to 10^{-3}% of dry mass (Annenkov and Yudintseva 2002). The organism of adult human contains about 0.3 g of strontium. Almost all of strontium is localized in the skeleton, while all other organs contain only 3.3 mg of this element. The release of radioactive isotopes of strontium into the environment began in the middle of the last century. The most important strontium radioisotope is long-lived ^{90}Sr ($T_{1/2}$ = 29.12 year). It was widely dispersed to the environment due to fallouts from atmospheric testing of nuclear weapons and accidents at the nuclear fuel cycle facilities and nuclear reactors (Prister 1998).

^{90}Sr is a pure beta emitter with a maximum energy of 0.54 MeV. Upon decay, it forms a daughter radionuclide ^{90}Y with a half-life of 64 h. ^{90}Sr has a long biological half-life in the human body estimated to be about 18 years. Due to its chemical similarity to calcium, it is accumulated in bones and irradiates the bone marrow, causing its high radiotoxicity (Prister 1998). Another radiologically significant strontium radioisotope is beta-emitting ^{89}Sr ($T_{1/2}$ = 51 days). However, due to the shorter half-life, it poses the radiological importance only for relatively short period (about 1 year) till further complete decay.

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Table 1 Distribution of ^{90}Sr contamination in Chernobyl exclusion zone as of year 2007

Soil contamination (kBq m^{-2})	Area (km^2)	Area percentage with level (%)
0–20	422	21.3
20–40	393	19.8
40–75	384	19.4
75–200	321	16.2
200–400	143	7.2
400–750	96	4.8
750–2000	143	7.2
2000–4000	43	2.2
4000–7500	20	1
7500–20,000	11	0.6
>20,000	6.6	0.3
Total	**1982.6**	**100**

Release of Strontium Radioisotopes to the Environment

The total amount of ^{89}Sr and ^{90}Sr released in the atmosphere is estimated about $90 \cdot 10^{18}$ and $600 \cdot 10^{15}$ Bq correspondingly (Pavlockaya 1997). The first significant release of strontium-90 from nuclear facility into the environment (about $2 \cdot 10^{15}$ Bq according to various estimates) occurred in 1957 as a result of a nuclear accident at the "Mayak" factory in the Southern Ural, USSR.

The accident at the Chernobyl and Fukushima nuclear power plants also introduced a large amount of ^{90}Sr into the environment, at that the largest part of the radionuclide was deposited in the relative vicinity of these NPPs. The release of ^{90}Sr after the Chernobyl accident in 1986 is estimated at $8.2 \cdot 10^{15}$ Bq (about 4% of the total inventory). After the accident at the Fukushima nuclear power plant, the relatively small amount of radiostrontium ($0.1 \cdot 10^{15}$ Bq) was released into the environment (Fukushima Daiichi accident: Report 2015).

The contamination of land territories with ^{90}Sr has more local character as compared with ^{137}Cs. Thus, the most of ^{90}Sr released after Chernobyl catastrophe (approximately $3 \cdot 10^{15}$ Bq) was deposited in form of fuel particles within 30-km Exclusion zone (Kashparov et al. 2008). Except of the Chernobyl NPP industrial site and radioactive waste disposal sites, the inventory of radiostrontium in soils of the 30-km Exclusion zone is estimated at about $0.5 \cdot 10^{15}$ Bq. This value corresponds to 0.7–0.8% of ^{90}Sr total activity in the fourth block of Chernobyl NPP. Half of radiostrontium inventory contained in the upper 0–30 cm soil layer is localized within only 2% of the territory of Chernobyl zone, and nearly 80% of its inventory is concentrated within 11% of this area (Table 1, Kashparov et al. 2008).

The analysis of data on the migration ability of ^{90}Sr in the soils of Chernobyl zone demonstrated that three decades after the accident the most of soils have the main inventory of radiostrontium (more than 95% of its activity) in the upper 10–20 cm layer (Kashparov et al. 1999). The intensive vertical migration of ^{90}Sr is

Table 2 Accumulation factors of ^{90}Sr in aboveground parts of wheat (Bq kg^{-1}/kBq m^{-2})

Isotope	Leaves	Stems	Spikelets	Grains
Sr-90	57.70	10.50	5.70	3.40

observed only in weakly humified, unmatted sand, where radiostrontium inventory in the upper 30-cm layer can be only 30–50% (Ivanov et al. 1996).

Speciation and Migration of Strontium in Soil

Usually ^{90}Sr has higher mobility in soils as compared to ^{137}Cs. Like ^{137}Cs, ^{90}Sr can be in water-soluble and water-insoluble forms. Absorption of ^{90}Sr in soils is mainly caused by the ion exchange. The greater part of strontium is retained in the upper soil horizons. The rate of its migration in the soil profile depends on the physico-chemical and mineralogical characteristics of the soil. In the presence of humus horizon in soil profile located under the litter or sod layer in soil profile, ^{90}Sr is concentrated in this horizon. In soils such as sod-podzolic sandy, humus-peaty-gley loam on sand, chernozem-meadow podzolized and leached chernozem, the certain increase of the radionuclide content in the upper part of the illuvial horizon is observed (Kashparov 1998). In saline soils, a second maximum appears which is associated with a lower solubility of strontium sulfate and its mobility. In the upper horizon of this soil ^{90}Sr is efficiently retained by the salt crust. The concentration of strontium in the humus horizon is explained by the high content of humus, the large cation absorption capacity and the formation of inactive compounds with organic matter of soils.

In the model experiments with introduction of ^{90}Sr to different soils in vegetation vessels, it was found that the rate of its migration under the experimental conditions increases with an increase of exchangeable calcium content. An increase of ^{90}Sr migration ability in the soil profile with an increase of the calcium content was also observed in the field conditions. It was also found that migration of strontium increases with raising the acidity and organic matter content (Lazarevich and Chernukha 2007).

The accumulation coefficient of ^{90}Sr by different plant cultures varies in a rather wide range, from 0.02 to 12, while for ^{137}Cs it varies from 0.02 to 1.1 (Ageets 2001). The accumulation of strontium by plant biomass can reach 10–40 kBq per 10 MBq m^{-2}. In particular, ^{90}Sr is accumulated in large quantities in legumes, root crops, and, to a lesser extent (3–7 times less), cereals (Tieplyakov 2010) (Table 2).

According to the classification of radionuclides by type of behavior in the soil–plant system, Sr^{2+} has the following basic characteristics:

- Type of behavior—exchangeable;
- Main mechanism of fixation in the soil—ion exchange;

- The most important factor of migration—the presence of other cations in the solution (Sanzharova 2005).

The fixation and distribution of ^{90}Sr in the soil are mainly determined by the behavior of its isotopic carrier—stable strontium, as well as the chemical analogue of the non-isotope carrier—stable calcium (Ca^{2+}).

When considering ^{90}Sr ions, one can distinguish three following groups:

1. Ions in the soil solution;
2. Exchange ions on the surface of mineral and organic particles;
3. Ions belonging to practically insoluble compounds (Sanzharova 2005).

The rate of strontium migration increases with raising the moistening degree of soils. It was found that the vertical migration of ^{90}Sr proceeds more intensively than ^{137}Cs in the profile of automorphous fallow soils. In some unprocessed lands, the main amount of ^{90}Sr (58–61%) and ^{137}Cs (70–85%) is concentrated in the upper part of 0–5 cm of the root layer. The increased ^{90}Sr transfer to plants is explained by its much greater mobility in the soil than in ^{137}Cs. Thus, about 85–98% of ^{137}Cs is firmly fixed by soil compounds, while for ^{90}Sr this value is only 7–12% (Ministry of Agriculture and Food: Rules 2002).

According to observations data, it was found that the effective half-life of ^{90}Sr in 0–5 cm layer of fallow automorphous sod-podzolic soils is 14.3–15.0 years, while for ^{137}Cs this value is 15.3–21.5 years. With an increase in the hydromorphism degree of soils, the intensity of vertical migration of radionuclides is being increased. Accordingly, for sod-podzolic gleyey sandy-loam soil the effective half-life of radionuclides is reduced (^{90}Sr – to 10.5 years, ^{137}Cs – to 13.8 years; Sanzharova 2005).

^{90}Sr belongs to the group of mobile radionuclides, since it is not involved in the ion exchange reactions with the ions of the clay particles of the soil-absorbing complex and is found in the soil mainly in a mobile state. This radionuclide is characterized with the predominance of easily accessible forms for plants, which amount to 53–87% of its total content and tend to increase with time. ^{90}Sr has an oxidation state +2, and it is found in form of cations in the soil solution. The solubility of ^{90}Sr bicarbonate is higher than that of calcium bicarbonate; therefore strontium is more mobile in the soil than calcium (Kabata-Pendias and Mukherjee 2007). Thus, more than 80% of ^{90}Sr is being in the exchangeable and water-soluble forms. With the lapse of time, the destruction of hot particles containing cesium, strontium and plutonium takes place (Kashparov 1998). With time radiostrontium is being fixed by clay minerals. It is a part of the soil solution in a mobile form, thus increasing the content of water-soluble and exchangeable forms (Lazarevich and Chernukha 2007).

The degree of strontium accumulation by plants from the soil depends on its chemical form, the physiological needs of plants and the physicochemical properties. The stronger the radioisotope is fixed in the soil, the smaller its amount is transferred the plant. Thus, oats cultivated on sand accumulated several times more ^{90}Sr than plants grown on heavy loam. Thus, 8–10% of strontium was transferred to plants from clayey sand, while plants grown on heavy loam accumulated only 1% of the total ^{90}Sr introduced into the soil. Likewise, strontium is accumulated by plants in relatively high degree as compared to other chemical elements according to the following sequence (Kashparov et al. 2005):

Sr > I > Ba > Cs, Ru > Ce > Y, Pm, Zr, Nb > Pu

It was found that ^{90}Sr relatively easily penetrates through the root system into all parts of the plant, while the radioisotopes of cerium, ruthenium, zirconium, yttrium, and plutonium are accumulated mainly in the plant's root system. The essential role in sorption processes of ^{90}Sr by soil is related to the isomorphous replacement in minerals containing calcium and magnesium, in particular calcite and limestone ($CaCO_3$), gypsum ($CaSO_4{\cdot}2H_2O$), and dolomite ($CaMg(CO_3)_2$) (Sanzharova 2005).

The ion exchange is a predominant mechanism of ^{90}Sr absorption by solid phase of the soils that is analogous to the adsorption of stable Sr and Ca. Therefore, the sorption of ^{90}Sr by the solid phase of soils depends on the presence of macro concentrations of cations in the solution. The following sequence of influence of competing cations on sorption of ^{90}Sr by solid phase of soils is revealed: $Al^{3+} > Fe^{3+} > Ba^{2+} > Ca^{2+} > Mg^{2+} > K^{+} > NH_4^{+} > Na^{+}$ (Sanzharova 2005).

The composition and the mineral content of soils have a considerable influence on the state and fixation of ^{90}Sr in the soil-absorbing complex. ^{90}Sr is more firmly fixed in soils with a high content of silt particles. The clayey soil minerals can sorb up to 99% of this radionuclide. ^{90}Sr is sorbed more preferably by such minerals as ascanite, bentonite, vermiculite, phlogopite, and humbrin, and to a much lesser extent—by hydromuscovite and hydrogrogite. Minerals of the montmorillonite group absorb 92.0–99.9% of ^{90}Sr, while minerals of the kaolinite group—40–68%, micas—71–87%, and hydromicas—80–88%. Minerals of the calcite group, feldspars, quartz, and gypsum absorb about 10–50% of ^{90}Sr (Vasilenko and Vasilenko 2002).

The behavior of ^{90}Sr is also affected by the organic matter of the soil. The distribution and mobility of ^{90}Sr in soils is largely determined by the quantity and qualitative composition of humus. The radionuclide is present in soils mainly not in the form of individual compounds with organic substances of a nonspecific nature and actually humic acids, but in the form of complex compounds, which also contain Ca, Fe, and Al (Kashparov et al. 2005). The adsorption mechanisms of ^{90}Sr are different in comparison with ^{137}Cs. Radiostrontium is characterized by a simple and almost complete exchange at surface exchange sites of clay particles. The yield of the radionuclide in the soil solution also increases with the gain of Ca yield, since Sr and Ca are present in the solution in a certain ratio. However, the distribution of ^{90}Sr between the soil solution and the absorbing complex differs from the analogous distribution for Ca. The ^{90}Sr/Ca ratio in the soil solution varies from 0.49 to 0.78 of the ratio of these ions in the soil, which is related to a stronger ^{90}Sr sorption compared to Ca (Sanzharova 2005).

Since ^{90}Sr in transferred to plants from the soil solution, it can be assumed that the concentration of the radionuclide in the plant is directly proportional to its concentration in the soil solution (other things being equal).

The value indicating the change of ^{90}Sr/Ca ratio upon its transfer from soil to plant is called the discrimination coefficient. The effect of ^{90}Sr discrimination is a stronger fixation of this radionuclide in the soil as compared to calcium. Usually, the discrimination coefficient varies depending on the degree of soil saturation with stable calcium, plant species, and the period of plant development. For the most plant species the discrimination coefficient of ^{90}Sr is 0.8–1.0 (Yudintseva and Gulyakin 1998).

$^{90}Sr^{2+}$ is absorbed by plants via transport systems of its macro analogue Ca^{2+}. The transport of Ca^{2+} is mainly conducted in the apoplast by free diffusion accelerated by transpiration in the cell wall, where one part of the ions are in a solution identical to the outer soil solution, and another part of ions is bound by fixed charged centers in the cell walls of the root exchange complex (Kutlakhmetov et al. 2003).

Translocation of Strontium to Plant Species (Factors Influencing the Radionuclide Transfer)

After their absorption, the ions of $^{90}Sr^{2+}$ easily penetrate into the "free space" of the root hair tissues due to the diffusion. Further the ions enter the conductive tissues of the root by active transfer and penetrate into the conductive tissues of the plant. Thus, an upward movement of strontium ions along the vascular tissue is carried out (Ministry of Agriculture and Food: Rules 2002).

Radioactive isotopes of Sr are analogues of Ca, have much similar in their intake into plants and distribution across its various organs. Thus, ^{90}Sr ions after their uptake by roots are accumulated to large extent in aboveground organs of plants (Kashparov et al. 2005).

The regularity of strontium activity distribution in plant organs is that ^{90}Sr, entering the aboveground part of the plant, is mainly concentrated in straw (leaves and stems), less in chaff (spikelets, wisps without grains) and relatively little in grains. The absorption of ^{90}Sr by plants lags behind the increase of aboveground mass. Accordingly, the accumulation of radionuclide per unit of dry weight decreases with the growth of the plant, but it increases during the maturation of plant. The intake of ^{90}Sr into plants increases, as a rule, with an increase in its concentration in solution. The maximum absorption of radiostrontium is observed at a pH close to neutral (Kashparov et al. 2005).

The accumulation of ^{90}Sr by plants is also influenced by their various biological features, among which the evolutionary origin of plants or phylogenesis. Plants of early origin accumulate more radiostrontium than plants originated in later periods (Konoplya 1996). According to the accumulation of strontium, some flora divisions are arranged in the following descending order: lichens > mosses > ferns > gymnosperms > angiosperms (Shkolnik 2012). The differences in the accumulation of radionuclides are revealed within the classes, families and species. The interspecific differences can reach up to 5–100 times. The content of radiostrontium per dry matter of individual crops can differ up to 30 times with the same radionuclide inventory in the soil (Prister 1998). The varietal differences in the accumulation of radiostrontium are much smaller (up to 1.5–3 times), but they must also be taken into account when selecting crops for cultivation under conditions of radioactive contamination (Shkolnik 2012). According to accumulation of ^{90}Sr, plants are classified as high-accumulating (legumes), medium-accumulating (cereals) and weakly accumulating cultures (grains). The accumulation of ^{90}Sr in the commodity part of some plant cultures could be presented in the following descending order: root crops > beans > potatoes > groats > cereals and vegetable crops (Vasilenko and Vasilenko 2002).

The sequence of cultures descending by ^{90}Sr accumulation differs significantly from that of ^{137}Cs. The highest accumulation ^{90}Sr in grain was revealed for the spring rape, followed by descending order: lupine > peas > vetch > barley > spring wheat > oats > winter wheat and winter rye (Lazarevich and Chernukha 2007).

The largest amount of ^{90}Sr is transferred into straw of barley, followed by straw of spring and winter wheat, oats and winter rye. According to the accumulation of radiostrontium in the biomass, the cultures are arranged in the following descending order: clover > lupine > pea > perennial grasses in floodplain lands > perennial cereal–bean mixtures > vetch > spring rape > pea–oat and vetch–oat mixtures >herbs in natural hayfields > grasses on drained lands > grasses on arable lands > corn (Lazarevich and Chernukha 2007).

The transfer factors of ^{90}Sr in plant biomass depend both on the density of pollution and on the soil type, the degree of soil moistening, the granulometric composition and the agrochemical properties. Therefore the transfer factors need periodic rectification. The indexes of soil fertility also have a significant impact on the accumulation of radionuclides by all crops.

The high degree of ^{90}Sr mobility in the soil determines the increased transfer factors of this radioisotope from soil to plants, which are on average an order of magnitude higher than in ^{137}Cs. In general, radiostrontium is transferred to plants from acidic soils more intensively than from weakly acidic, neutral or slightly alkaline soils. The sod-podzolic soils are characterized by high initial acidity and weak alkali saturation. With increase of soil acidity, the fixation strength of ^{90}Sr and ^{137}Cs by soil absorbing complex is reduced and, accordingly, the intensity of their intake to plants is increased. In case when pH is raised, a number of radionuclides are transformed from the ionic form into various hydrolysis complex compounds that reduces their availability for plants (Sanzharova 2005).

Depending on the soil type, the transfer factors of ^{90}Sr can vary for the same inventory of this radionuclide in the soil up to 2–5 times. For example, the accumulation factor of ^{90}Sr for potatoes on sod-podzolic sandy soil is 0.33, while on sod-podzolic loamy soil it is 0.17. Polissya is the region of Ukraine most contaminated with radiostrontium. It has mainly sandy-loamy light sod-podzolic and peaty-bog soils (Kashparov et al. 2005). The transfer coefficients of ^{90}Sr into plants are 4–5 times higher in this region compared to other types of soils, because in case of clay minerals deficiency, the radiostrontium is found in these soils mostly in water-soluble and exchange forms. The accumulation of radionuclides in peaty-bog soils depends on the cultivation of the soil, the mineralization and composition of soil ash, the thickness of the peat layer, the botanical composition of peat-forming plants, the

Table 3 Average values of ^{90}Sr accumulation factor for agricultural crops (Bq kg^{-1}/kBq m^{-2})

Culture/organs	Sandy loam	Medium loam	Chernozem	Heavy loam
Wheat (grain)	0.70	0.20	0.12	0.06
Potato (tubers)	0.35	0.10	0.06	0.03
Table beetroot (root crop)	1.20	0.34	0.20	0.10
Cabbage (head)	0.90	0.22	0.16	0.07
Cucumber (fetus)	0.35	0.10	0.06	0.03
Tomato (fetus)	0.14	0.04	0.02	0.01
Clover (hay)	20.00	6.00	4.00	2.00
Timothy grass (hay)	7.00	2.00	1.20	0.60

Table 4 ^{90}Sr transfer factors of crops depending on the acidity of sod-podzolic loamy soil (Bq kg^{-1} (plant)/Bq kg^{-1} (soil) d.w.)

	Acidity value of soil, pH (KCl)					
Culture	<4.5	4.6–5.0	5.1–5.5	5.6–6.0	6.1–7.0	>7.0
Oats	1.71	1.35	1.25	1.21	1.18	1.10
Barley	–	1.45	1.38	1.32	1.28	1.16
Peas	–	1.31	1.55	1.44	1.37	1.33
Potatoes	0.36	0.27	0.21	0.15	0.13	0.12

acidity of the soil solution, the presence of exchangeable cations, the soil moisture as well as depth and mineralization of groundwater (Kutlakhmetov et al. 2003).

With the same level of contamination, the intake of ^{90}Sr into plants and its accumulation in the crop will be unequal on different soils. The recent studies have shown that reduced intake of ^{90}Sr into plants and, correspondingly, its accumulation in food products is observed on fertile soils characterized by high sorption capacity (Table 3, Klekovkin 2004).

A negative dependence between the content of exchangeable calcium, the acidity level of the soil solution and the intake of ^{90}Sr to plants was found. That is, the higher the concentration of exchangeable calcium in the soil and the lower acidity of the soil solution, the smaller the transfer factors of ^{90}Sr to plants (Kutlakhmetov et al. 2003).

Application of Countermeasures for Reduction of Strontium Transfer to Plants

As the content of exchangeable calcium increases from 550 to 2000 mg CaO per kg of soil, the transfer factors are decreased in 1.5–2 times. The change of acidity of the soil solution from the acidic interval (pH = 4.5–5.0) to the neutral interval (pH = 6.5–7.0) reduces ^{90}Sr transfer to plants by 2–3 times. Further saturation of soil with calcium shifts the pH to the alkaline range, but this is not accompanied with a decrease of transfer factors (Table 4, Konoplya 1996).

The practical application of the above research results is the soil liming. This is one of the most important ways to increase the productivity of agricultural land. When lime is applied to acidic soil, the content of mobile calcium and magnesium increases sharply, which affects the biological availability of some radionuclides, especially ^{90}Sr (Yudintseva and Gulyakin 1998).

When introducing lime fertilizers to acidic soil, the concentration of hydrogen ions decreases in the soil solution, while the content of mobile calcium decreases. This effect strengthens ^{90}Sr binding in the soil, thereby reducing its availability to plants. The liming of acidic soils not only limits ^{90}Sr accumulation in the crops harvest, but also increases the fertility of the soil, but also increases the soil fertility and crop yield, as well as contributes to "dilution" of the radionuclide per unit of plant biomass (Aleksakhin and Korneev 2001).

It was found that the application of lime in a concentration corresponding to complete hydrolytic acidity reduces the content of radiostrontium in crop production by 1.5–3 times (sometimes up to 10 times), depending on the soil type and initial acidity. The intake of ^{90}Sr in the crop of plants can be reduced under the influence of lime depending on the biological features and individual parts of the crop in 3–20 times (Kutlakhmetov et al. 2003). The liming of acid soils reduces the accumulation of ^{90}Sr in the harvest of legumes (peas, vetch, and clover) more than in the harvest of cereals (oats, barley, timothy grass, and meadow grassland). When lime is applied in an amount equivalent to 100% of the cation exchange capacity of the soil, the content of ^{90}Sr in the tops and roots of sugar beet decreases by 7.5 and 20 times respectively, in peas and clover—by 6–8 times, in straw of barley and oats as well as in hay of timothy grass—by 3–4 times (Aleksakhin and Korneev 2001).

It has been found that the greater the soil saturation with exchange alkali, the lower ^{90}Sr transfer factors to plants. The peat bog soils are typical acidic soils poor in potassium, calcium, and magnesium. Therefore ^{90}Sr transfer factors on these soils are 5–20 times higher than on sod-podzolic ones (Prister 2005).

A considerable impact on the ^{90}Sr accumulation by plants has a moistening regime of soils. It is known that the amount of strontium cations displaced from the soil into the solution increases with the gain of humidity (Gerzabek et al. 1999). It is related to the complex nature of the mutual influence of moisture, soil properties and biological characteristics of plants on the migration processes of radiostrontium in the soil-plant chain. With gain of the soil moisture, the fraction of water-soluble and exchangeable ^{90}Sr increases, so the transfer factors and radiostrontium content in vegetation increase as well.

The transfer of Sr to plants is affected by the organic matter of the soil. Humus acids, especially humic acid, form complex compounds with radiostrontium called humates. Therefore the availability of strontium from the organic complexes is reduced a factor of 2–4. The increased bioavailability of ^{90}Sr in peat-bog soils is associated with the ability of organic matter to fix radionuclide ions on the surface of organic colloids (Gudkov et al. 2013). Therefore the strong sorption of radiostrontium is not ensured and its availability to plants is increased. In addition, the acidity of the soil solution in peaty-marsh soils is usually increased, that ensures good solubility of radiostrontium salts and their accessibility to plants. Thus, the factors of soil fertility can have a significant impact on the accumulation of radiostrontium by agricultural crops.

Conclusion

Radioactive strontium is referred to as biologically significant radionuclides being characterized with high toxicity. It role in soil contamination and population exposure has remained considerable due to accidental releases. ^{90}Sr that deposits onto the surface is under the influence of natural factors in the migration processes. After atmosphere, soil is the most important depot of ^{90}Sr. The migration of radiostrontium in the soil is significantly influenced by the physicochemical properties of the radionuclide and the soil, climatic conditions, landscape, soil type, hydrological regime, vegetation, agrochemical features of agriculture, etc.

^{90}Sr is predominantly found in easily accessible forms for plants, and the total content of these forms tends to increase over time. The uptake of radiostrontium from soil is primarily determined by its bioavailability, solubility, agrochemical soil properties, and cultivated plant species. Therefore, the understanding of ^{90}Sr speciation in the environment represents a vital tool for tracing transport mechanisms, distribution pathways, and radionuclide bioavailability.

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Assessment of Entry of ^{90}Sr into Plants in Case of a Heterogeneous Radiation Contamination of Ecosystems

Nina N. Kazachonok

Introduction

Many places of the world are contaminated with radioactive isotopes. According to the recent report (Radiation 2013), there were 107,548 thousand m^2 of land contaminated with radionuclides in the adjoining areas of Rosatom enterprises, till the end of the year 2012. Besides, there were 11,312 settlements in the territory of Russia contaminated because of the Chernobyl accident. According to the State report “On the condition and protection of the environment of the Russian Federation in 2011” (On the condition 2011) a moderately dangerous contamination with metal complex was noted on 9% of examined lands and a dangerous contamination was noted on 3.5% of examined lands in 2011. Often polluted are suburban lands that are most suitable for a high-tech production of fresh vegetables, individual construction, gardening, and homesteading. Residents of settlements located outside the evacuation zone are under the necessity to grow potatoes and vegetables on contaminated soil of household plots or use the received land shares. For example, when the state farm “Bulzinsky” was liquidated, the former workers of the state farm obtained the ownership of land shares (9 ha each) in the territory adjacent to the East-Ural radioactive trace.

For hygienic regulation in radiation accident zone, the forecasting of the developmental programmes for radiation situation under different scenarios of environmental management and rehabilitation is necessary. Modern methods of forecasting are based on statistical analysis of historical data and methods of extrapolation of the discovered dependencies. However, in real conditions, conventional statistical analysis methods may not be suitable. This is connected, in particular, with the heterogeneity of the contamination of the territories.

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Problems of Entry Forecasting of ^{90}Sr and Other Radionuclides into Plants in Case of a Heterogeneous Contamination

The heterogeneity of radioactive contamination, a significant variation in soil and vegetation contamination levels was noted both in the territory of Belarus (Shchur et al. 2015) and in Trans Urals. An example of an area with a strongly heterogeneous radioactive contamination is the East-Ural radioactive trace, formed in 1957 as a result of the precipitation of a radioactive cloud, following the explosion of a waste container at a radiochemical enterprise producing weapons-grade plutonium, which is now known as the Production Association "Mayak." Already at the first stage of research in the area contaminated as a result of activity of "Mayak" PA, it was noted that the distribution of values of radionuclide activity in natural objects and plant products do not comply with Gauss' law (Shamov et al. 2012). During the radiation surveys and other studies, a lot of evidence of local features was obtained to ascertain the distribution of radioactive contamination density associated with the influence of meso and micro landscapes and other natural factors (Beacon 2005). The levels of soil contamination with radionuclides within one settlement can also vary greatly (Kazachonok 2015). In the survey of settlements in the area of influence of the "Mayak" PA, in most cases, the ^{90}Sr and ^{137}Cs activities was found, along with in about 10% samples of garden soil, potatoes showed much higher activity (Popova and Kazachonok 2015). In some cases, the heterogeneity of contamination of agricultural plants was even greater. Figure 1 shows a histogram of ^{90}Sr specific activity distribution in potatoes from the private farms of the town of Kasli in 1965.

When soil is contaminated with ^{137}Cs, a strong heterogeneity also appears. Chegerova (2000) showed that in the Mogilev region the activity of ^{137}Cs

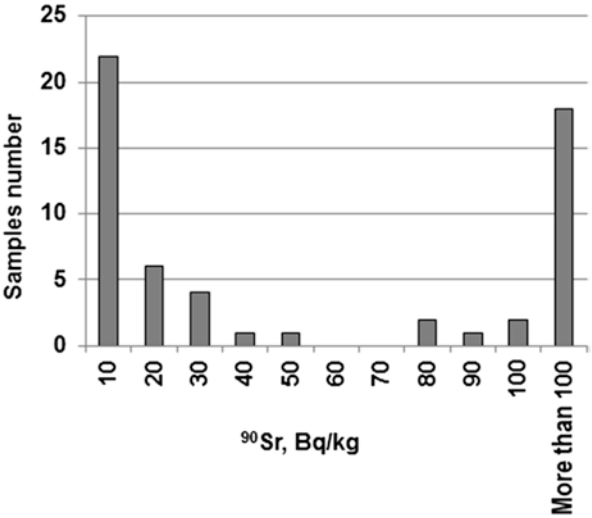

Fig. 1 Histogram of distribution of absolute values of ^{90}Sr specific activity in potatoes in the town of Kasli in 1965

in mushrooms, fresh vegetables, fruits, and berries in 80, 43, and 70% of the samples respectively fits into the first interval of the histogram (Fig. 1), whereas in some samples the activity can be ten times higher than in the first interval (Chegerova 2000).

The reason for the appearance of "dirty samples" is either heterogeneity of territory contamination with radionuclides, or violation of the conditions of protected sanitary zone by some residents of settlements. The violation of the conditions of protected sanitary zones can be episodic or systematic and its causes can be different. Thus, during the initial period of the Techa river pollution with the PA "Mayak" discharges, the reasons for the restrictions were not explained to residents and, above all, the clean water supply was not organized.

Depending on the weather conditions and the yield of herbaceous vegetation, the residents use hay from sites having contamination. As for example in 2010, due to drought conditions, some residents in Southern Urals harvested feed from contaminated areas, although they knew about the sites. Violations may also have economic causes. According to residents of the settlement of Bulzi, in the 1970s many families harvested hay in the territory of the EURT as "it was free" (Popova and Kazachonok 2015).

The heterogeneity of the conditions for the formation of an internal irradiation dose of the population depends also on social factors. For the population of Belarus the tendency is revealed—the smaller are the settlements, the more often the average internal irradiation dose of the surveyed residents exceeds 0.1 mSv. This is due to the fact that in small settlements the infrastructure is less developed, the degree of farming naturalization is more pronounced, the population eats more food products grown in their own garden and produce from the forest. In these settlements there is also a low employment of the population, there are no organized food service areas, trading networks and other conditions that exist in large settlements (Ageeva et al. 2010, 2011; Shchur and Vinogradov 2016).

In our opinion, the Gaussian distribution of the values characterizing the radioactive contamination level (density, specific activity, etc.) can only occur in areas within which the points with different contamination levels are randomly distributed. In real conditions, the heterogeneity of the territory contamination appears often in the form of spots having an epicentre and aureole zones different in terms of area, and their contamination level can vary relatively smoothly and consistently, whereas the area of aureole zones will increase in most cases with radius increasing. In both randomized and systematic sampling, the probability of selecting a sampling point from the spots epicentres will be significantly lower than from aureoles. Therefore, the frequency of soil and vegetation sampling from the least contaminated areas will be much higher than from the most contaminated areas. A multiyear research has shown that, depending on the way how the array is divided into intervals, it is possible to obtain a more or less approximate normal distribution for samples from aureoles and a long "tail" for samples from the epicentre (Popova and Kazachonok 2015). It can be assumed that the distribution in the form of a "springboard" indicates the pollution heterogeneity caused by the natural features of the landscape, and a bimodal or a poly modal distribution indicates the "organizational heterogeneity" associated with an irrational use of natural resources in contaminated landscapes (allocation of heavily contaminated plots for use, noncompliance with restrictions and prohibitions by the population, etc.).

This may lead to underestimation of the overall pollution level of the territory and vegetation. Theoretically, the level of soil contamination, even by a one-dimensional heterogeneity, must be described as the sum of the integrals of the functions of pollution densities reduction from epicentres to background values. In practice, this method is unreasonably labor-consuming.

Assessment of Possibility of Increased Accumulation of ^{90}Sr and Other Radionuclides in Case of a Homogeneous Radiation Contamination

To date, there are many recommendations on agricultural production in the contaminated territories. However, they are designed for largescale farms. In addition, when choosing measures to reduce the product pollution levels in a particular farm for a given period, it is necessary to take into account a large number of conditions that are usually not considered in general regulatory documents and recommendations: market conditions, energy prices, alternative possibilities of products usage etc. It should be taken into account that at the present time there is no legislative basis in Russia for compelling owners of agricultural enterprises to implement any recommendations. Thus, the head or management of a particular farm takes the final decision on the reasonability of applying certain protective measures.

Quite often, the countermeasures can be effective according to one of the parameters, but in general, according to the integral estimation, their application is unreasonable. For example, the combined use of ameliorants (liming, elevated use of organic and phosphate-potassium fertilizers, on light soils—the placement of clay minerals) made it possible to reduce the content of ^{137}Cs in plants up to five times (Sanzharova et al. 2005). However, the placement of a fertilizer complex led to an increase in the yield of cereals and, as a result, to an increase in the collective dose, which nearly neutralized the decrease in the ^{137}Cs specific activity in grain. In addition, production costs increased significantly. Thus, the averted dose, normalized to costs, was only 0.01–0.03 man mSv/million rubles (Prices of the year 1996) Fesenko (1997).

One of the effective methods to reduce the level of pollution of agricultural products is the conversion of the farm (The application 2011). However, this measure requires even greater costs, including costs for a reconversion if it turns out to be unprofitable.

"Guidance on the application of countermeasures in agriculture in the event of an accidental radionuclides release into the environment" issued by the IAEA in 1994 indicates that "the main goal of introducing any countermeasure should be to reduce the population irradiation doses and, thereby reducing the human health risk". In practice, this means choosing such a countermeasure strategy that enables to produce such food products that have contamination levels below intervention levels, as economically as possible and with minimal side effects (The application 2011).

In modern economic conditions, agricultural production in a contaminated area is not always profitable. The increase in economic costs associated with the implementation of recommendations on the agriculture in the territory contaminated with radionuclides is unacceptable for most farms. Therefore, a need arises to assess the reasonability of involving contaminated lands in agricultural production while maintaining its profitability.

For this purpose, it is necessary to predict the entry levels of ^{90}Sr and ^{137}Cs in the marketable part of harvest on the lands affected by radioactive contamination and to estimate the probability of making products which meet modern standards of radiation safety.

Using the radiation pollution mapping data from previous years and the radionuclide half-life values, it is possible to roughly calculate the average contamination density of the territory and the expected level of product contamination. However, the calculation of economic risk of the cultivation of specific crops in specific fields requires more detailed studies. In the initial period after the accident, the pollution density of the territory can be assessed using a direct method. Since radionuclides are located on the surface of the soil, it is sufficient to collect the top layer with a sampler having a known capture area.

In the long term after radioactive fallout, the nature of radionuclide distribution in the soil profile changes. In the forest and virgin soils, the pollution level is determined by the activity of the upper 20 cm of soil. On cultivated soils the distribution pattern in the profile may be different. Thus, on the axis of the East Ural radioactive trace (EURT), 50 years after the accident, in the long-fallow black soil in the lowland, 15–20% of ^{90}Sr was in the 30–85 cm layer. When the soil is treated regularly, especially irrigated, the contamination of the ploughing and subsurface horizon can be almost the same. Thus, the ratio of the densities of contamination with ^{90}Sr of 20–40 cm and 0–20 cm layers of the grey forest heavy loamy soil of “Sovkhoz Beregovoy” LLC in 2007 without irrigation was 0.76 ± 0.21, and with irrigation—1.16 ± 0.29. For ^{137}Cs these ratios were 0.58 ± 0.20 and 0.88 ± 0.43 (Kazachonok and Popova 2014).

It is commonly believed that the bulk of the roots are located in the upper “root-inhabited” soil layer. However, absorption of water and ions occurs in the root fibrils at the root tips, which reach a depth of 2–3 m of agricultural plants. Therefore, to assess the contamination density of the cultivated soil, it is recommended to have data on the radionuclide specific activity in layers having total thickness of at least 40 cm.

The density of soil contamination is defined as the product of the radionuclide specific activity and the weight of soil contaminated with it per unit of area. The soil weight is calculated as the product of its bulk weight, the fixed area and the thickness of the contaminated layer.

It is known that the bulk weight of soils depends on their type, the content of organic matter, the structure and the granulometric composition.

Despite the great variety of agrophysical characteristics of soils, it can be assumed that in most cases the bulk weight increases in the lower horizons, where the content of organic matter is small, the structural properties are less pronounced and the aeration porosity is minimal.

This regularity expresses itself more clearly if not absolute, but relative values of the soil bulk weight are used for the comparison. This method makes it possible to compare soils with different average bulk weight of a meter layer and to reveal the dependence of the soil bulk weight from the depth of sampling. We suggest describing the obtained dependences using monotonic functions, since grey forest soils and, in particular, black soils are characterized by smooth transitions between horizons.

Based on this, according to the values of the bulk weight for different soil layers of the Chelyabinsk region published by Kozachenko (1999) we calculated empiric equations that allow us to define approximately the value of the soil bulk weight at a given depth.

For grey forest soil:

$$Y_x = -4 \cdot 10^{-5}\,\mathrm{x}^2 + 6 \cdot 10^{-3}\,\mathrm{x} + 0.829$$
$$\left(R^2 = 0.986\right)$$

For black soil:

$$Y_x = -4 \cdot 10^{-5}\,\mathrm{x}^2 + 7.5 \cdot 10^{-3}\,\mathrm{x} + 0.747$$
$$\left(R^2 = 0.948\right),$$

where Y_x—relative value of the soil bulk weight at a given depth x,x—Sampling depth in cm.

Accordingly, the absolute value of the bulk weight (V_x) at this depth:

$$V_x = V \cdot Y_x,$$

where V is the average bulk weight of a meter-deep layer of the soil.

By comparing actual and calculated data, the deviation averaged 7 ± 3%, and the deviation is mainly connected with the inaccuracy of the actual data due to sampling layer by layer.

If there is information on the bulk weight of at least the upper layers of the studied soil, it is possible to use not the average tabulated values of V, but to calculate V for a given particular soil, and this will increase the accuracy of bulk weight calculation for all layers.

To do this, it is necessary to divide the actual value of the bulk weight (V_x) in the studied layer by its calculated relative value (Y_x), which can be obtained from Table 3 or, more precisely, calculated using the above formulas.

For calculation of radionuclide contamination density in a given soil layer (P_i) (Bq m^{-2}) the following formula can be used:

$$\mathrm{P}_i = 10 a_i \cdot V_i \cdot h_i,$$

where a_i—radionuclide specific activity (Bq kg^{-1}) in the studied layer i, V_i—soil bulk weight in this layer (g cm^{-3}), h_i—layer thickness (cm), 10—conversion coefficient for the transition from g cm^{-3} to Bq m^{-2}.

For calculation of radionuclide specific activity (a_i) (Bq kg^{-1}) in the soil layer *i* based on known contamination density the following formula is used:

$$a_i = \frac{P_i}{10V_i \cdot h_i}$$

For calculation of radionuclide contamination density in the whole soil profile (P_i) (Bq m^{-2}) this formula can be used:

$$\mathrm{P} = \sum \left(10a_i \cdot V_i \cdot h_i\right),$$

Based on the dependencies, as found, we have created a program to automate calculations of the pollution density with ^{90}Sr and ^{137}Cs of black soil and grey forest soil of the forest-steppe zone of Trans Urals according to the specific activity values of these radionuclides in the soil layers.

For podzolic, sodic, solod, and other soils having stark differences of agrophysical properties in different horizons, the dependence of the bulk weight from the horizon and sampling depth will probably be described by more complicated functions. However, such soils are rarely found in the territory of EURT, assessment of their contamination is less important, therefore, making an on-the-fly approximate evaluation for contamination density of podzolic and malt soils is necessary, in our opinion the formula for grey forest soils, and for estimating contamination of sodic soils, the same formula for black soils can be used.

When predicting the level of product contamination in a given field for a planned culture, it is necessary to know the ratio of the radionuclide contamination level of the marketable part of harvest to the soil contamination density (transition coefficient (C_{t})), or the ratio of the radionuclide specific activity in agricultural products to the average specific activity in the root-inhabited horizon (accumulation coefficient (C_{a})).

Both C_{a} and C_{t} differ for different agricultural plants and varieties, depending on different types of soils and their kinds. It is noted that they decrease with time. Therefore, in order to improve the accuracy of the forecast, it is recommended to calculate their average values and standard deviations for specific farms in which a high probability of obtaining products not corresponding to hygienic standards is assumed.

According to the radiochemical analysis data of soil and production, the accumulation coefficients (C_{a}) for each sampling point are calculated:

$C_{\mathrm{a}} = \frac{\mathrm{A_{pr}}}{\mathrm{A_s}}$,where $\mathrm{A_{pr}}$—radionuclide specific activity in plant products, $\mathrm{A_s}$—in soil.

Then, the value of $C_{\mathrm{a}}\left(\bar{x}\right)$, average for all points of the whole field and the standard deviation σ are calculated:

$\sigma = \sqrt{\frac{\sum \left(x_i - \bar{x}\right)^2}{\left(n-1\right)}}$, where x_i—each value of C_{a}, $\bar{x}$ —average value of C_{a}, *n*—measurements number.

The probability of non-exceedance of a given value of *a* (the maximum permissible radionuclide level in the production) will be equal to the value of the distribution function (Gelman 2003):

$$F(x) = \frac{1}{\sqrt{2\pi}\sigma} \int_{-\infty}^{x} e^{\frac{-(a-\bar{x})^2}{2\sigma^2}}$$

Table 1 represents as an example the values of the accumulation coefficients of ^{90}Sr and ^{137}Cs in the marketable part of agriculture sampled by us in “Sovkhoz Beregovoy” LLC, and Table 2, sampled in personal farms in settlements located on the periphery of the EURT and in the 30-km zone of “Mayak” PA.

It can be seen from the tables that the standard deviations are quite large and exceed in some cases $\bar{x}$. This may be due to a deviation from the normal distribution. For example, in the village of Bagaryak in the study of milk samples, the distribution turned out to be bimodal. This is since the pastures on the north-west were contaminated much more than on the south-east, that is, samples of milk from different pastures belonged to different populations. In this case $\bar{x}$, σ, and $F(x)$ are calculated separately for each population.

A lognormal distribution occurs quite often. In the case of a lognormal distribution, when calculating $F(x)$, a is replaced with $\ln(a)$, when calculating $\bar{x}$ and σ, x is replaced with $\ln(x)$.

When C_a for one of the crops and the radionuclide specific activity in soil (A_s) are known, but its standard deviation isn’t known, we can calculate approximate products contamination level (A_{pr}) using the formula:

Table 1 Average values of the accumulation coefficients of ^{90}Sr and ^{137}Cs in the marketable part of agriculture in “Sovkhoz Beregovoy” LLC and their standard deviations (in brackets)

Crop	Product	^{90}Sr	^{137}Cs
Peas	Grains	0.084 (0.029)	0.062 (0.036)
Wheat	Grains	0.036 (0.014)	0.078 (0.060)
Oats	Grains	0.054 (0.031)	0.032 (0.018)
Barley	Grains	0.070 (0.062)	0.039 (0.040)
Onions	Bulb	0.026 (0.009)	0.016 (0.009)
Cabbage	Head	0.029 (0.005)	0.019 (0.008)
Carrots	Root plant	0.237 (0.222)	0.146 (0.165)
Beets	Root plant	0.026 (0.007)	0.016 (0.008)
Potatoes	Tubers	0.020 (0.012)	0.008 (0.003)
Corn	Grass	0.242 (0.189)	0.049 (0.041)
Sudan grass	Grass	0.175 (0.062)	0.083 (0.040)

Table 2 Average values of the accumulation coefficients of ^{137}Cs and ^{90}Sr in the marketable part of agriculture in private farms and their standard deviations (in brackets)

Crop	Product	^{90}Sr	^{137}Cs
Potatoes	Tubers	0.0083 (0.0067)	0.0128 (0.0120)
Carrots	Root plant	0.0193 (0.0140)	0.0379 (0.0786)
Beets	Root plant	0.0172 (0.0099)	0.0156 (0.0124)

$$A_{pr} = A_s \cdot C_a$$

And assume that the probability of non-exceedance of this level is 50% (in case of a normal distribution).

Depending on the purpose of the production and the possibilities of its alternative use (forage, technical raw materials, planting material, etc.), the economic service of the farm determines the probability level of receiving agricultural products that do not meet the radiation and hygienic standards and decides on the introduction of the lands into agriculture.

Assessment of Possibility of Increased Accumulation of ^{90}Sr and Other Radionuclides by Plants in Case of an Inhomogeneous Radiation Contamination

It should be noted that the use of functions of normal distribution to predict the contamination level of agricultural products is permissible only in case of a relatively uniform contamination of agricultural areas. That is, the radionuclide activity values in soil samples or food product samples taken at the same time in one settlement can be referred to one general population. However, practice shows that even within one settlement, the values of the radionuclide specific activity in environmental objects cannot be always related to a single general population. This is especially important for the areas of settlements located on the periphery of the East Ural radioactive trace, where the fallout of radioactive aerosols was nonuniform. In this case, it is necessary to use other methods for analyzing data and predicting pollution of agricultural products.

In our opinion, when processing experimental data for nonuniform radioactive contamination, the application of Bayesian methods and fuzzy logic can be a promising direction. The principles of application of fuzzy logic methods for the analysis of radioactive contamination of soil and agricultural products were outlined by us in 2013 (Kazachonok 2013).

To assess the level of radioactive contamination of a natural or anthropogenic object, the sampling rate should be much greater than the rate of change in the essential characteristics of the object. The number of samples that can be analysed for a particular object during the sampling period is finite, and all these samples either meet or do not meet the specified criterion. The taken samples are not returned back. For example, an object is a group of cows in private farms in the settlement. Their number in the sampling period is finite; the number of milk samples that are taken during the period of research from each cow is determined by the research methodology and is also finite. Therefore, the results of the selection and examination of milk samples can be considered as dependent events.

A dependent event (the result of a sample study) is connected with the truth of one of the incompatible hypotheses H_1, H_2, H_3, …, H_n. For example, the settlement A is:

H_1—not contaminated;
H_2—contaminated.

It is necessary to calculate the probability of the truth of these hypotheses. Prior to the study of the settlement, the a priori probabilities of these hypotheses are defined, relying on the already available data. After receiving the analysis result of the taken sample (event X), the probabilities of the truth of the hypotheses are recounted. For example, if sample X is contaminated, the probability of hypothesis H_1 is increased, and of hypothesis H_2 is decreased.

For example, in a settlement there are 20 plots where potatoes are grown; one sample is taken from each plot at this stage of the study. We will define the settlement as contaminated if at least in one sample the MPL of the studied radionuclide is exceeded. If at least one sample is contaminated, the probability of obtaining a clean sample is 0.947 or less. Let's take the probability of obtaining a clean sample equal to 0.95 as a criterion of referring the settlement to not contaminated, 0.94-to contaminate.

The simplest case is the calculation of the belonging of the studied object to one of two categories, but the Bayesian method makes it possible to calculate the probabilities for more hypotheses.

There are some examples of an algorithm for calculating the probability of three hypotheses using the Bayesian method.

1. The range of values, the probability of which in the study is different from zero, is divided into subranges. The structuring criteria are chosen according to the objectives of the study. For example, the structuring into a subrange of values not exceeding the MPL and a subrange of values exceeding the MPL.
2. The categories of objects to which the studied object can be assigned and the matching criteria with the defined categories of the studied object are specified. For example: object A is classified as "safe" if the probability of taking a sample characterized by a value in the subrange "not exceeding the MPL" = 0.95; object A is classified as "medium dangerous" if the probability of taking a sample characterized by a value in the subrange "not exceeding the MPL" is 0.5; object A is classified as "dangerous" if the probability of taking a sample characterized by a value in the subrange "not exceeding the MPL" = 0.05.
3. The hypotheses of the belonging of the studied object to the specified categories are developed.

   H_1—taken sample X will belong to object A_1 (A is "safe").
   H_2—taken sample X will belong to object A_2 (A is "medium dangerous").
   H_3—taken sample X will belong to object A_3 (A is "dangerous").

The probability that the sample *X* will be "not exceeding the MPL" for samples of A_1

$$\begin{gathered}P_{H1}(X)=0.95\\ \text{for } \textit{samples of } A_2\\ P_{H2}(X)=0.5\\ \text{for } \textit{samples of } A_3\\ P_{H3}(X)=0.05.\end{gathered}$$

The probability that the sample *X′* will be “exceeding the MPL” for samples of A_1

$$\begin{gathered}P_{H1}(X')=0.05\\ \text{for } \textit{samples of } A_2\\ P_{H2}(X')=0.5\\ \text{for } \textit{samples of } A_2\\ P_{H3}(X')=0.95.\end{gathered}$$

The a priori probabilities that the sample is taken from an object classified as “safe” ($P(H_1)$) or as “medium dangerous” ($P(H_2)$) or as “dangerous” ($P(H_3)$) are calculated using available research data. If studies were previously not conducted, then at the first stage the a priori probabilities can be assumed to be equal.

$$P(H_1)=P(H_2)=P(H_3)=1/3$$

4. Taking and analysis of the sample are carried out.

Let the sample *X* be “not exceeding the MPL.”

According to the formula of total probability, the probability that the sample *X* will be “not exceeding the MPL”:

$$P(X)=P(H_1)\cdot P_{H1}(X)+P(H_2)\cdot P_{H2}(X)+P(H_3)\cdot P_{H3}(X)$$

In this case, according to the Bayesian formula, the a posteriori probabilities of the hypotheses are calculated:

$$\begin{gathered}P_X(H_1)=\left(P(H_1)\cdot P_{H1}(X)\right)/P(X)\\ P_X(H_2)=\left(P(H_2)\cdot P_{H2}(X)\right)/P(X)\\ P_X(H_3)=\left(P(H_3)\cdot P_{H3}(X)\right)/P(X)\end{gathered}$$

Let the sample *X′* be “exceeding the MPL.”
In this case, the probability that the sample *X′* will be “exceeding the MPL”:
for samples of A_1

$$\begin{aligned}&P_{H1}(X') = 0.05\\&\text{for } \textit{samples of } A_2\\&P_{H2}(X') = 0.5\\&\text{for } \textit{samples of } A_2\\&P_{H3}(X') = 0.95.\end{aligned}$$

According to the formula of total probability:

$$P(X') = P(H_1)\cdot P_{H1}(X') + P(H_2)\cdot P_{H2}(X') + P(H_3)\cdot P_{H3}(X)$$

According to the Bayesian formula, the a posteriori probabilities of the hypotheses are calculated:

$$\begin{aligned}P_{X'}(H_1) &= (P(H_1)\cdot P_{H1}(X'))/P(X')\\P_{X'}(H_2) &= (P(H_2)\cdot P_{H2}(X'))/P(X')\\P_{X'}(H_3) &= (P(H_3)\cdot P_{H3}(X'))/P(X')\end{aligned}$$

5. The a posteriori probabilities are taken as new a priori ones and the step 4 is repeated.
6. After a certain number of iterations, the a posteriori probabilities corresponding to the study tasks are obtained.

Thus, if it is not possible to calculate the probability of obtaining a sample matching/not matching the given parameters, the Bayesian method makes it possible to calculate the probability that an object belongs to a category for which the probability of interest is characteristic. The Table 3 represents the results of probabilities calculation, according to the given example.

Probabilities of truth of hypotheses of an object's belonging to the specified categories deduced using the Bayesian method are equivalent to the functions of an object's belonging to these categories. Therefore, the results of calculations can be processed using fuzzy logic methods.

In our opinion, when processing experimental data for nonuniform radioactive contamination, the application of fuzzy logic methods can be a promising direction. The principles of application of these methods for the analysis of radioactive contamination of soil and agricultural products were outlined by us in 2013 (Kazachonok 2013).

In fact, we are dealing with the combination of at least two subsets of data: the analysis results of samples obtained in the farms of "law-abiding citizens" (A) and in the farms of "violators" (B). Such a combined set AB will have either a polymodal distribution or a distribution similar to a lognormal. We assume that if the subset A has a symmetric distribution, then the subset of the values x of the set AB for x > M has the function of membership in A equal to $\mu_A(x) = y(2M - x)/y(x)$, and the function

Table 3 An example of probability calculation of the truth of three hypotheses using the Bayesian method

Sample no.	Measurement result	H_1 (object is "safe")		H_2 (object is "medium dangerous")		H_3 (object is "dangerous")	
		$P_{H1}(X)$	$P_X(H_1)$	$P_{H2}(X)$	$P_X(H_2)$	$P_{H3}(X)$	$P_X(H_3)$
	Before the experiment	0.95	0.333333	0.5	0.333333	0.05	0.333333
1	<MPL	0.95	0.633333	0.5	0.333333	0.05	0.033333
2	<MPL	0.95	0.781385	0.5	0.21645	0.05	0.002165
3	>MPL	0.05	0.261594	0.5	0.724638	0.95	0.013768
4	<MPL	0.95	0.406387	0.5	0.592487	0.05	0.001126
5	<MPL	0.95	0.565777	0.5	0.434141	0.05	8.25E-05
6	<MPL	0.95	0.712317	0.5	0.287677	0.05	5.47E-06
7	<MPL	0.95	0.824702	0.5	0.175297	0.05	3.33E-07
8	<MPL	0.95	0.899383	0.5	0.100617	0.05	1.91E-08
9	>MPL	0.05	0.471981	0.5	0.528019	0.95	1.91E-07
10	<MPL	0.95	0.629404	0.5	0.370596	0.05	1.34E-08
11	<MPL	0.95	0.763419	0.5	0.236581	0.05	8.54E-10
12	<MPL	0.95	0.859769	0.5	0.140231	0.05	5.06E-11
13	>MPL	0.05	0.380078	0.5	0.619922	0.95	4.25E-10
14	>MPL	0.05	0.057769	0.5	0.942231	0.95	1.23E-09
15	<MPL	0.95	0.104336	0.5	0.895664	0.05	1.17E-10
16	<MPL	0.95	0.181221	0.5	0.818779	0.05	1.07E-11
17	<MPL	0.95	0.296037	0.5	0.703963	0.05	9.17E-13
18	>MPL	0.05	0.040356	0.5	0.959644	0.95	2.38E-12
19	<MPL	0.95	0.073989	0.5	0.926011	0.05	2.29E-13
20	<MPL	0.95	0.131802	0.5	0.868198	0.05	2.15E-14

of membership in B equal to $\mu_B(x) = (y(x) - y(2M - x))/y(x)$, where M—mode (in case of the polymodal distribution, the first mode), $y(x)$—frequency x, $y(2M - x)$—frequency of a value that is symmetric to x with respect to M (Coffman 1982).

It should be noted that the use of fuzzy models is suggested by other researchers too. In particular, this approach is mentioned in the review paper of Tikhonova and Rylova (2014).

Such an approach, in our opinion, will allow justifying the identification of "violators," as well as conducting a more correct analysis of the dynamics of contamination levels of environmental components in the long term after emergency fallouts.

Conclusions

1. The heterogeneity of radioactive contamination of soil and vegetation causes problems for predicting the probability of increased pollution of plant products with ^{90}Sr and other radionuclides.

2. In case of a homogeneous radioactive contamination, the possibility of non-exceedance of maximum permissible accumulation levels of ^{90}Sr and other radionuclides in plants can be calculated using the function of normal or lognormal distribution.
3. In case of a heterogeneous radioactive contamination, the possibility of non-exceedance of maximum permissible accumulation levels of ^{90}Sr and other radionuclides in plants can be calculated using the Bayesian formula and fuzzy logic methods.

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Accumulation of ^{90}Sr by Plants of Different Taxonomic Groups from the Soils at the East Ural Radioactive Trace

Ludmila N. Mikhailovskaya, Vera N. Pozolotina, and Elena V. Antonova

Introduction

The EURT area is a narrow sector that is oriented in the north-eastern direction with a length of more than 100 km and an area of 23,000 km^2. The EURT was formed in 1957 because of an accident at the "Mayak" PA (Fig. 1), and a key contaminant (among the long-lived radionuclides) was ^{90}Sr. In 1967, the EURT area was contaminated again with airborne radioactive sediments from the shores of Karachay Lake. The "Mayak" PA used this lake for dumping liquid radioactive waste, and the primary contaminant was ^{137}Cs. Many studies have been conducted in the EURT area. Comprehensive knowledge regarding the migration of the key contaminants in the soil–plant cover of this area has been gained, and the long-term dynamics of the radiation situation at the EURT have been recorded (Romanov et al. 1990; Sokolov and Krivolutsky 1993; Tsaturov and Anisimova 1994; Aarkrog et al. 1997; Pozolotina et al. 2008, 2012). The characteristics of the EURT (wide range of soil density contamination, heterogeneity of topographic and ecological conditions and large species diversity of flora) allow unique radioecological studies to be conducted there.

The goal of this research was to study the regularities of the accumulation of ^{90}Sr by plants of different taxonomic groups growing within EURT zone with a wide range of contamination densities and in the territory with background level of contamination.

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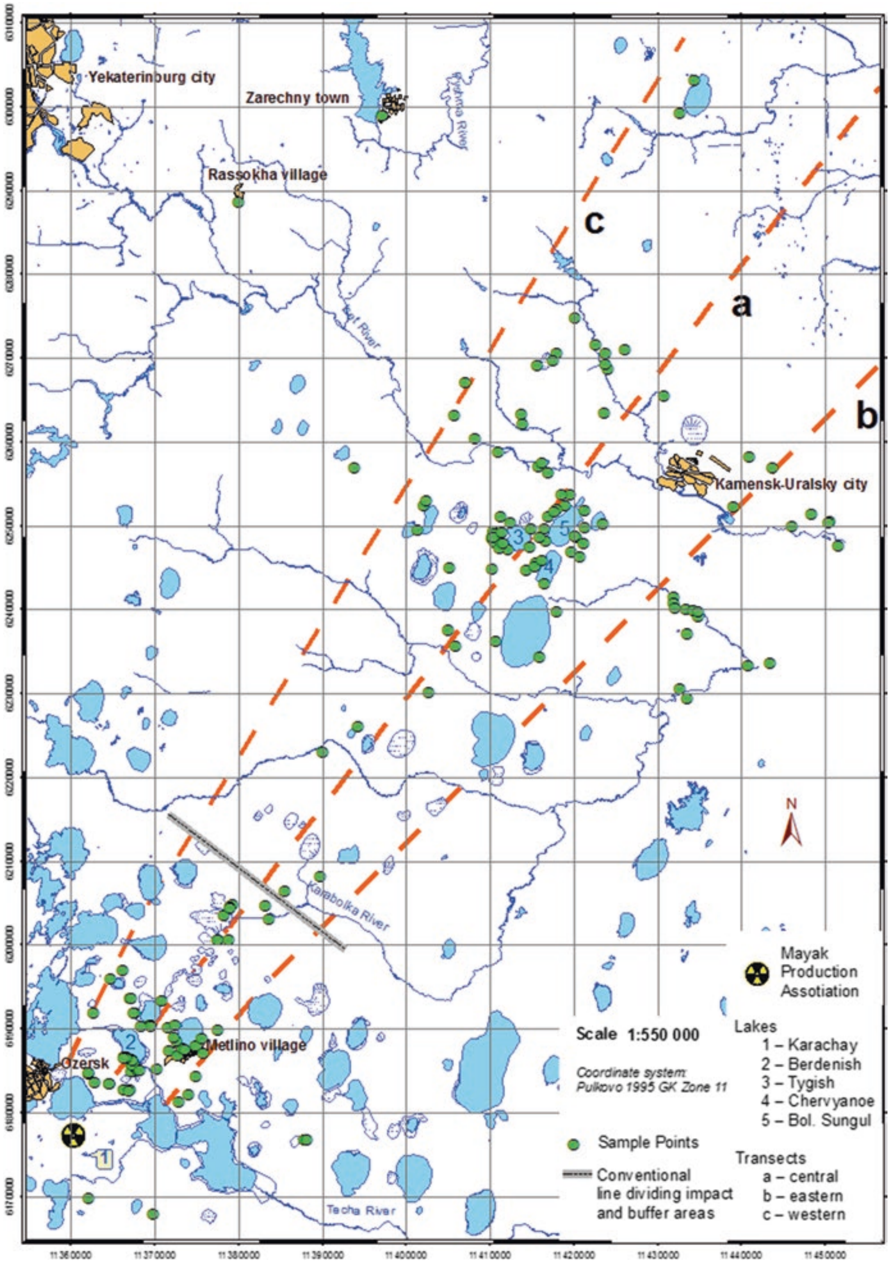


Fig. 1 Study site map. Impact sample points (20) were situated within the central axis of the EURT zone at distances 10–20 km from the epicenter of the accident. Background sample points (20) were selected outside the contaminated area near the Rassokha village

The Study Site

This research was conducted in 2003–2015. The territory of the EURT is located within a forest–steppe zone. The relief is hilly, and the entire area slopes downward from west to east. The soil types are dominated by various subtypes of forest grey soils, chernozem soils of varying thicknesses and degrees of leaching and meadow soils (Pozolotina et al. 2012). Westerly and south-westerly winds prevailing in the region determined the shape of the trace (Teterin 2011). In the forests of the EURT, *Betula pendula* and *Pinus sylvestris* predominate. The species diversity of herbaceous plants includes approximately 300 species. We submitted a floristic description of the EURT communities in a previous paper (Pozolotina et al. 2012).

Sampling Method

We chose 20 reference sites within the head part of the EURT zone at distances 10–20 km from the epicenter of the accident with uniform relief, soil cover and vegetation. Sampling points are shown in Fig. 1. Background sites (20) with identical soil and vegetation characteristics were selected outside the contaminated area near the Rassokha village. Within each plot we made three soil sections of 100 m^2 at each site. Soil profiles were located at the vertices of an equilateral triangle with a side length of 10 m. Soil samples were taken as layers with a 5–10-cm capacity to a depth of 50 cm (sample areas were 0.01–0.04 m^2). The total content of radionuclides in the 50-cm soil layer was normalized to a unit area (i.e., 1 m^2), which was then considered the contamination density. We averaged the samples of three individual profiles located at the vertices of the triangle to create a representative sample. Samples of the aboveground mass of herbaceous plants, leaves and the large branches of trees as well as samples of the biomass of both mosses and epiphytic lichens near the profiles were collected. We also collected fruiting bodies of basidiomycetes and wood-destroying fungi (mushrooms).

Method for ^{90}Sr Determination

All samples were dried. Vegetable samples were crushed, and soils were triturated and passed through a sieve (cell diameter was 1 mm). The samples were then incinerated at 450 °C. All analytical methods were performed in accredited Institute of Plant & Animal Ecology laboratories (accreditation certificate CAPK RU.0001.441492). The content of ^{90}Sr was determined using radiochemical methods (Tsvetaeva et al. 1984). The procedures were based on radionuclide leaching by 6N HCl solutions from prepared preliminary samples. Precipitation of ^{90}Sr occurred in the oxalate form, separating the ^{90}Sr that remained in balance with the ^{90}Y daughter product of decay. The measurement of ^{90}Y was performed with the use of a UMF-2000 alpha-beta radiometer (Russia), whose detection precision was 0.2 Bq. All results for the samples were calculated on an air dry-weight basis.

Results and Discussion

Assessment of Contamination Density

The soil contamination densities of ^{90}Sr within the background sites ranged from 0.5 to 2.9 kBq m^{-2} .This level does not exceed background values for the Urals region on the whole (Izrael 2013; Molchanova et al. 2014b, 2016). Most of the ^{90}Sr (253–316 TBq) is concentrated in the central part of the EURT (Molchanova et al. 2009, 2014a). The maximum contamination of ^{90}Sr (6 km from the epicenter of the accident) reached 70,000 kBq m^{-2} but decreased by more than an order of magnitude along the central axis at a distance of 20 km (Fig. 2). At a similar section of the western boundary of the EURT, the contamination density was well below 110–12 kBq m^{-2}, and at the eastern boundary it was 117–25 kBq m^{-2}. The spatial distribution of ^{90}Sr along the central axis with increasing distance from the accident site is approximated by an exponential function ($y = a \cdot e^{bx}$) with a high degree of reliability (Pozolotina et al. 2008, 2010, 2012; Molchanova et al. 2011).

Vertical Distribution of ^{90}Sr in Soil Profiles

The vertical distribution of ^{90}Sr in soils of the same type has a similar character, both at the central axis of the trace and beyond this zone (Pozolotina et al. 2008). As an example, Table 1 shows a typical distribution of ^{90}Sr in brown forest soil at the southern tip of the central axis of the EURT (N55° 45′E 60° 50′) and at the control plot (N56° 42′ E 61° 02′). Radionuclide contents decrease with decreasing soil profile depth. We recorded the maximum concentrations of radionuclides in forest litter, and the highest density of contamination was recorded in the 0–5-cm soil layer. More than 90% of the radionuclides were retained in the 0–20-cm soil layer; the ^{90}Sr inventory in this layer ranges from 70 to 98% of the total content of the soil profile in the EURT (Pozolotina et al. 2008, 2010; Molchanova et al. 2009; Deryagin et al. 2015; Tarasov et al. 2016).

In the forest litter within the contaminated territory, the inventory of ^{90}Sr was only 2%; beyond the EURT zone, this value reached 9%. Herbs that grow on brown forest soils of the EURT retained only 0.09% of this radionuclide from its total deposition in the soil profile; in the background area, this value was 0.3%. The transition coefficients (TC) for the EURT and background sites were 1.8 and 21.7 respectively, calculated according to the following formula:

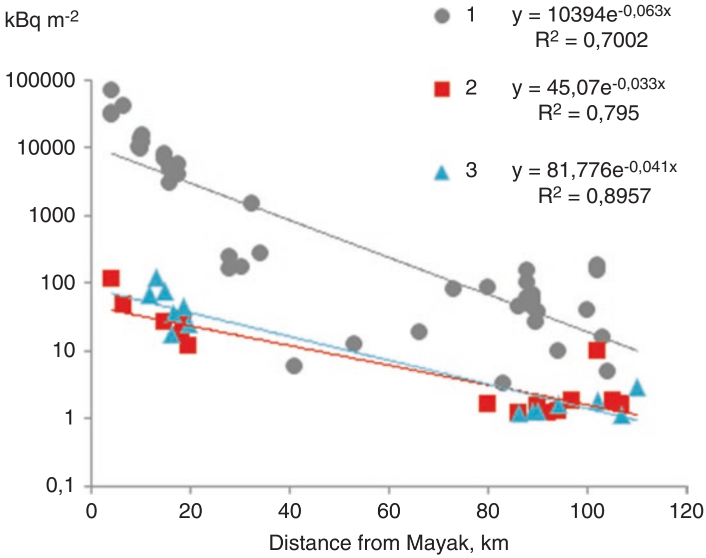


Fig. 2 The soil contamination densities by ^{90}Sr with increasing distance from the epicenter of the accident is satisfactorily ($p < 0.05$) approximated by an exponential function. (1) Central axis; (2) Western boundary; (3) Eastern boundary

Table 1 Example of typical vertical distribution of ^{90}Sr in brown forest soil at the central axis of the EURT and at the control plot

	Central axis of EURT			Background site		
Sample, (depth in cm)	Bq kg^{-1}	kBq m^{-2}	% from total deposition	Bq kg^{-1}	kBq m^{-2}	% from total deposition
Grass	72,896	36.4	0.09	47.7	0.006	0.3
Litter	913,673	804.0	2.0	129	0.2	9.0
Soil: 0–5	279,700	15,291.2	38.4	19.8	1,1	50.0
5–10	198,500	13,631.0	34.2	9.1	0.3	13.6
10–15	87,090	6096.3	15.2	5.4	0.3	13.6
15–20	38,460	2794.9	7.0	1.8	0.2	9.0
20–25	11,549	962.4	2.4	1.4	0.1	4.5
25–30	3070	202,6	0.5	BLD	BLD	BLD
30–35	983	56.4	0.14	BLD	BLD	BLD
35–40	298	17.0	0.04	BLD	BLD	BLD
40–50	60	6.8	0.02	BLD	BLD	BLD
Total deposition		39,862.6			2.2	

BLD below limit of determination

Table 2 Inventory of ^{90}Sr in different components of forest ecosystems (min-max)

Component	Dry mass, kg m^{-2}	^{90}Sr, %from total deposition in soil	
		EURT	Background site
Trees branches	1.0–2.2	0.7–5.7	3.0–3.4
Dying leaves	0.1–0.5	0.5–0.6	0.8–1.9
Grass	0.2–2.0	0.04–0.4	0.06–0.4
Forest litter	0.3–4.6	1.9–12.2	0.8–10.0
Mosses	0.2–1.2	0.1–0.4	0.6–2.8
Lichens	0.08–1.0	0.06–0.14	0.4–1.9

$$\mathrm{TC} = \frac{\text{Activity concentration of}\,^{90}\text{Sr in the organisms}\left(\text{Bq kg}^{-1}\ \text{dry weight}\right)}{\text{Soil contamination densities of}\,^{90}\text{Sr}\left(\text{kBq m}^{-2}\ \text{dry weight}\right)}$$

The thickness of the soil layer is 50 cm. Unit of measurement of the TC is n 10^{-3} kg^{-1} m^2. We previously reported a decrease in the TC of plants in the EURT territory (Pozolotina et al. 2008, 2010, 2012; Molchanova et al. 2009, 2011).

Inventory of ^{90}Sr in the Different Components of Forest Ecosystems

We evaluated the involvement of ^{90}Sr in biological cycles using the components of forest ecosystems located within the EURT and beyond. In forest ecosystems, woody plants form the majority of biomass (1.0–2.2 kg m^{-2}). The dried leaves of trees, mosses and lichens (when the projected coverage is near 100%) form 0.1 to 0.5 kg m^{-2} of biomass annually. Forest litter and both tree trunks and branches accumulated greatest (in per cent) amounts of ^{90}Sr regardless of the level of soil contamination (Table 2).

The properties of the various components of ecosystems determine the role of these components in the involvement of ^{90}Sr in the biological cycle. Dry, dead leaves and grasses fall to the soil surface annually, forming forest litter. The litter contains ^{90}Sr that was accumulated by plants. The decomposition of litter occurs slowly in the forests of the middle latitudes; therefore, radionuclides remain in the litter for many years. The axial organs of woody plants accumulate radionuclides from the soil and retain it for the entire lifespan of the tree (tens and hundreds of years). Moss–lichen cover accumulates amounts of radionuclides that are comparable with those deposited in the forest litter, and mosses and lichens can retain these radionuclides for many years. Relative deposits of ^{90}Sr in the leaves, mosses, and lichens were lower in the contaminated zone than in the background area (see Table 2). This may be due to the contribution of atmospheric fallout, as this fallout primarily pollutes these components of ecosystems (Shcheglov et al. 2001; Mikhailovskaya et al. 2017). At present, the observed differences cannot be explained by the ^{90}Sr bioavailability. The proportion of bioavailable forms of ^{90}Sr in the EURT soils is 85–87% (Tarasov et al. 2016), and in background areas this share reaches only 67% (Karavaeva et al. 2009).

High variability of ^{90}Sr deposition was observed for all the components of forest ecosystems. This high variability could be due to the peculiarities of the geochemical properties of soils, the variability of weather conditions and the properties of different plant species in the surveyed areas. The deposition capacity of different taxonomic groups of plants is determined not only by the amount of biomass they produce but also by their different abilities to accumulate radionuclides (Shcheglov et al. 2001).

Content of ^{90}Sr in the Plants of Different Taxa

Both grey forest and chernozem soils had similar agrochemical properties and contamination levels at the EURT sites. The inventories of ^{90}Sr in the soils of this zone were 3000–5000 kBq m^{-2}, which are three orders of magnitude higher than the background values (0.5–2.9 kBq m^{-2}) (Izrael 2013; Molchanova et al. 2014b, 2016). We evaluated the accumulation capacities for ^{90}Sr of the following species of woody plants: *Betula pendula* Roth., *Pinus sylvestris* L., and *Salix caprea* L. Herbaceous plants were represented by many species. In forests, *Trifolium medium*, *Polygonatum odoratum*, *Galium spurium*, *Prunella vulgaris*, *Angelica sylvestris*, *Pyrola rotundifolia*, *Thalictrum simplex*, *Thalictrum minus*, and *Rubus saxatilis* were abundant. In meadows, *Artemisia vulgaris*, *Vicia cracca*, *Lathyrus pratensis*, *Bromus inermis*, *Rumex confertus*, *Bunias orientalis*, *Galium boreale*, *Galium verum*, *Galium mollugo*, *Lithospermum officinale*, *Stellaria graminea*, and *Geum rivale* were abundant. There were several ruderal species, including *Urtica dioica*, *Cirsium setosum*, *Plantago media*, and *Tussilago farfara*. We also investigated several species of mosses, including *Hylocomium splendens*, *Polytrichum commune*, and *Pleurozium schreberi*, and one lichen, *Hypogymnia physodes*. Specific morphophysiological properties, life spans, life forms, and ecological features characterize these plants of different taxa.

The greatest concentrations of ^{90}Sr were found in ground mosses: 41,500 Bq kg^{-1} in the EURT zone and 83 Bq kg^{-1} in the background area (Table 3). The radionuclide content was lowest in the large branches of trees; in the aboveground biomass of herbaceous plants; and in the thallus of epiphytic lichens, although the differences between these taxa were not statistically significant. For comparison, we present in Table 3 the data on ^{90}Sr concentrations in the fruiting bodies of fungi of the orders Aphyllophorales (*Fomes fomentarius* L. Fr.) and Agaricales (*Lactarius resimus* Fr., *Russula foetens* Fr., and *Russula cyanoxantha* Fr.). The ^{90}Sr content in fungi was clearly minimal.

The content of ^{90}Sr in plants in the background area was therefore 140–800 times lower than that of plants in the EURT territory (for mushrooms, the content was 90–400 times lower). At the same time, the levels of soil contamination in these areas differed by a factor of approximately 2500. The coefficients of variation (CVs)

Table 3 Concentrations of ^{90}Sr in plants of different taxa and fungi

Taxa, life form	Part of plant	*n*	EURT *M* ± SE, Bq kg^{-1}	EURT CV,%	*n*	Background site *M* ± SE Bq kg^{-1}	Background site CV,%
Higher angiosperms							
Trees	Leaves	20	30,870 ± 22,936	74.3	20	44 ± 10	22.4
	Axial organs	20	14,220 ± 5204	36.6	20	28 ± 2	7.6
Shrubs	Aboveground mass	9	22,405 ± 5534	24.7	6	28 ± 6	20.2
Herbs	Aboveground mass	20	13,970 ± 4638	33.2	20	51 ± 5	9.8
Higher spore							
Mosses, turfs	Stems and leaves	20	41,500 ± 9047	21.8	20	83 ± 28	34.0
Leafy lichens	Thallus	10	9166 ± 2942	32.1	9	66 ± 17	26.0
Fungi							
Order Aphyllophorales	Fruit bodies	6	3420 ± 1210	35.4	6	10 ± 4	35.7
Order Agaricales	Fruit bodies	3	1500 ± 712	47.5	6	16 ± 9	56.3

M ± *SE* average value ± standard error, *CV* the coefficient of variation

of the radionuclide content in the representatives of different taxa varied from 7 to 75%. The range of values within each taxon may be due to differences in the accumulation capacity of the species.

The specific activities of ^{90}Sr in the aboveground biomass of plants of different taxa increased with increasing soil contamination density, and a power function ($y = a \cdot x^b$) satisfactorily approximated this dependence (Fig. 3). The absorption of ^{137}Cs from the soil by plants is often described by a non-linear function (Beresford and Wright 2005; Pozolotina et al. 2008, 2010, 2012; Molchanova et al. 2009). Our results show that the differences in the content of ^{90}Sr in plants of different taxa are small. Most of the values are located in a relatively narrow range of approximately one order of magnitude. Apparently, soil contamination levels mainly determine the absorption of ^{90}Sr by plants. In practice, the accumulation of ^{90}Sr by species of different taxa may be similar. Furthermore, we studied the ^{90}Sr accumulation in the aboveground biomass of several species from the Fabaceae family (Table 4).

All species grew at sites with a relatively homogeneous soil contamination density. The concentrations of ^{90}Sr in plants varied within one order of magnitude. We recorded the greatest content of ^{90}Sr in the aboveground biomass of *Cytisus ruthenicus*; the lowest, in *Melilotus officinalis*. The *CV*s of the specific activity of radionuclide in some cases reached 60–80%. TC differed slightly for plants from the head part of the EURT zone (1.0–5.9), and TC was higher in the background area (13–47). This phenomenon can be caused by the individual variability of the accumulative capacity of plants and the heterogeneity of soil contamination at the level of micro relief. In addition, the heterogeneity of radionuclide distribution in the soil arises as

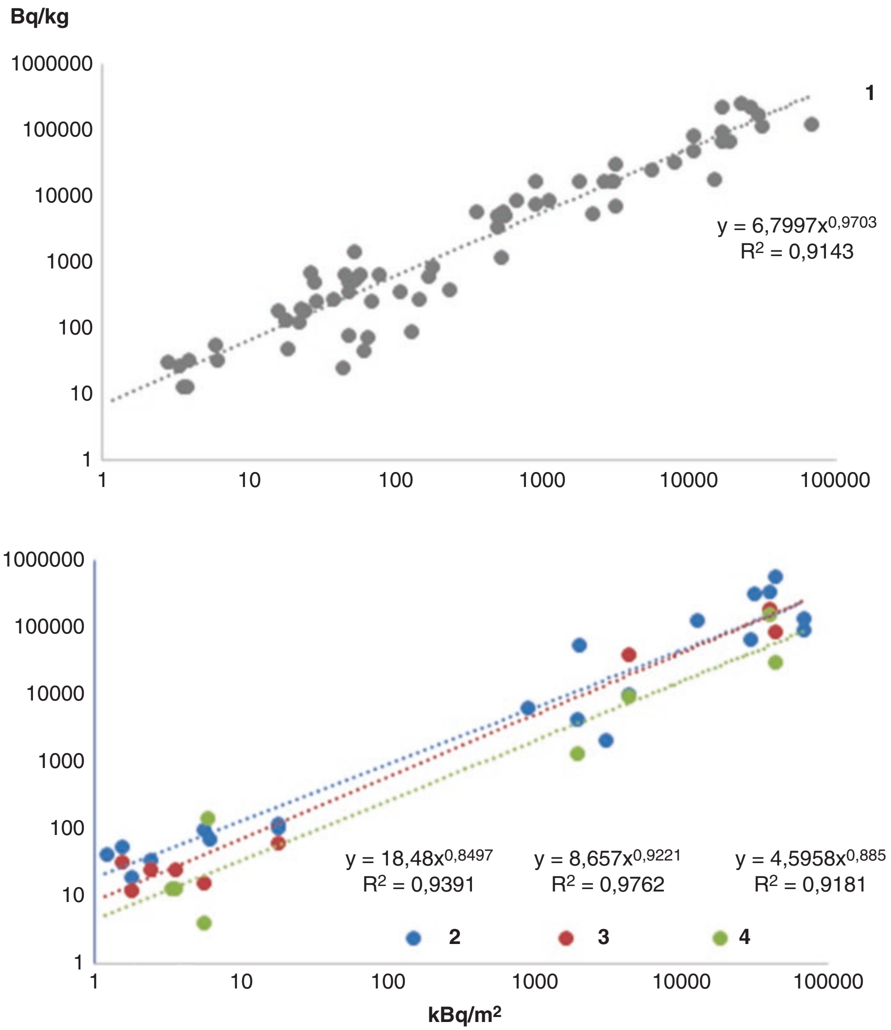


Fig. 3 The ^{90}Sr concentrations (Bq kg^{-1}) in the aboveground mass of plants of different taxa in dependence on the soil contamination density (Bq m^{-2}). (1) Herbs; (2) Trees; (3) Mosses; (4) Lichens; significance level $p < 0.01$

a result of uneven fallout of radionuclide from the atmosphere. Elevated TC in the territory with a background level of contamination in comparison with EURT zone may be due to a relatively large contribution (percentage, %) of aerial fallout (Table 5). Additional studies of precise parameters of aerial pollution are needed.

Thus, within a single family, there are species with the same accumulation capacity and species whose capacities significantly differ from each other. We obtained similar data for other taxa growing on homogeneous contaminated plots in the EURT

Table 4 The accumulation of ^{90}Sr by several species of the Fabaceae family

Species	EURT			Background area		
	M ± SE Bq kg^{-1}	CV, %	TC	M ± SE, Bq kg^{-1}	CV, %	TC
Cytisus ruthenicus	40,235 ± 2816	7.0	4.9	43.6 ± 3.4	8.0	25.5
Lathyrus pratensis	38,689 ± 18,454	47.7	3.1	*	*	*
Medicago sativa	39,623 ± 4160	10.5	2.0	50.6 ± 20.6	40.7	20.8
Melilotus officinalis	4979 ± 528	10.6	1.0	36.2 ± 4.0	11.0	13.4
Trifolium medium	16,655 ± 7545	45.3	5.9	64.7 ± 31.9	49.3	47.2
Vicia cracca	8750 ± 88	1.0	2.0	54.1 ± 16.7	30.9	23.4

M ± *SE* average value ± standard error, *CV* the coefficient of variation, *TC* transition coefficient, *n* 10^{-3} kg^{-1} m^2, * not detected

Table 5 The contribution of atmospheric fallout for the contamination of grasses

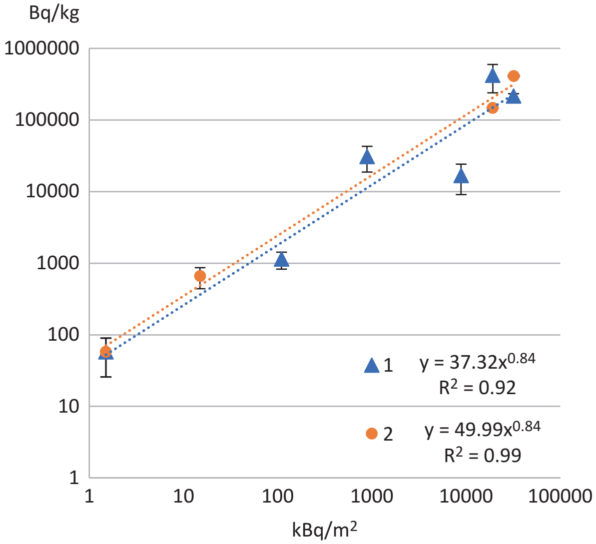
Site	Atmospheric fallout, Bq m^{-2} per year	The contribution of atmospheric fallout to the grasses contamination (depending on the biomass of grasses)	
		Bq kg^{-1} air-dry mass (min-max)	% from total content in the above-ground mass (min-max)
EURT	50	8.25–82.5	0.0015–0.168
Background	3	0.495–4.95	0.5–38.0

territory and beyond (Molchanova et al. 2014b; Mikhailovskaya et al. 2015). In general, the TCs of herbs were higher in the background area than in the EURT zone.

The relatively greater accumulation of ^{90}Sr by plants in the background area could be due, in part, to the contribution of fallout from the atmosphere. We therefore assessed the surface contamination for this. The fallout of ^{90}Sr from the atmosphere within 100 km of the Mayak PA varies from 5 to 50 Bq m^{-2} year^{-1} (Vakulovsky 2011; Shershakov et al. 2015). These depositions include global fallout and staff releases of nuclear fuel cycle enterprises. At the background sites in the Urals region, the global fallout is approximately 1–3 Bq m^{-2} year^{-1} (Vakulovsky 2011; Shershakov et al. 2015). The vegetative period for herbs in the Urals does not exceed 4 months (0.33 years), and the biomass of herbs is approximately 0.2–2.0 kg m^{-2}. The results show that the level of contamination of herbaceous plants from ^{90}Sr fallout can reach 8.25–82.50 Bq kg^{-1} in the most polluted part of the EURT (depending on the biomass of grasses) and 0.50–4.95 Bq kg^{-1} in the background area (Table 5).

The contribution of atmospheric fallout to the general contamination of grasses did not exceed one-tenth of a per cent of the total radionuclide content in the EURT grasses, and in the background area, this relative content reached several tens of per cents; this is the maximum possible value. In reality, the contribution of ^{90}Sr from the atmosphere to plant pollution is less. Even if 100% of ^{90}Sr falls on the surface of plants, immediately the cleansing processes are included (washing off by rainfall, falling leaves, etc.) Processes of pollution and purification of plants occur constantly

Fig. 4 The content of ^{90}Sr ($\mathrm{Bq\ kg^{-1}}$) in the aboveground mass (1) and seeds (2) of *Trifolium medium*, depending on the density of soil contamination ($\mathrm{Bq\ m^{-2}}$), significance level $p < 0.01$



and simultaneously under natural conditions. Note that the content of ^{90}Sr in the surface air within the EURT zone is *n* 10^{-5} Bq $\mathrm{m^{-3}}$, and in the background areas is n 10^{-7} Bq $\mathrm{m^{-3}}$ (Shershakov et al. 2015). Therefore the contribution of soil particles to plant contamination will also be higher as a percentage in the background areas compared to impact sites. We assume that the relatively greater contribution of the non-root intake of ^{90}Sr to the plants can cause high TC in the background areas.

In addition, fresh fallout, which pollutes the annual soil–plant cover, may be more accessible to plants than are radionuclides that have accumulated in the soil for many years. This phenomenon was investigated under experimental conditions (Karavaeva et al. 2008). The evaluation of the contribution of fresh aerial fallout to plant pollution in natural conditions is a complex task and requires special study. Using the species *Trifolium medium* as example, we showed that both the aboveground biomass and seeds accumulate ^{90}Sr at approximately the same rate (Fig. 4). The concentrations of ^{90}Sr in the leaves and seeds of *Trifolium medium* increased with increasing density of soil contamination, and this dependence was strongly approximated by a power function.

Conclusions

- The range of ^{90}Sr soil contamination density varies in the territory of the EURT from 10 to 70,000 kBq $\mathrm{m^{-2}}$. The spatial distribution of this radionuclide with increasing distance from the epicenter of the accident is approximated satisfactorily by an exponential function ($y = a \cdot e^{bx}$). Currently (55 years after the accident) more than 70% of ^{90}Sr remains in the root layer of soils (at a depth of 0–20 cm).

- The accumulation capacity of mosses, lichens and vascular plants with respect to ^{90}Sr was similar. Most of the values are located within a relatively narrow range of approximately one order of magnitude. Thus, soil contamination density mainly determines ^{90}Sr accumulation by plants. This dependence is strongly approximated by a power function ($y = a \cdot x^b$) in gradient of contamination.
- The results showed that the maximum concentrations of ^{90}Sr were found in surface mosses and in the leaves of woody plants; the large branches of trees, herbaceous plants and epiphytic lichens accumulated 2–4 times less ^{90}Sr, and the content of ^{90}Sr in fungi was minimal. Within each large taxon, the accumulation of ^{90}Sr in the aboveground biomass of plants was determined by their specific features.

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Soil-to-Crop Transfer Factors (TFs) of Alkaline Earth Elements and Comparison of TFs of Stable Sr with Those of Global Fallout ^{90}Sr

Keiko Tagami and Shigeo Uchida

Introduction

The abundance of strontium (Sr) in the continental crust is 333 mg kg^{-1}, ranking Sr as 17th among 77 indigenous elements (Wedepohl 1995). In the environment, biogeochemical properties of Sr are similar to those of calcium (Ca). Because Ca^{2+} is a crucial regulator of plant growth and development, Ca is recognized as an essential element in plants, and the element is taken up through plant roots. Sr is also taken up by plants through Ca channels (Ehlken and Kirchner 2002), although Sr is a nonessential element. Therefore, compared to other nonessential elements, Sr shows high values of the soil-to-crop transfer factor (TF), which is defined as the crop–soil concentration ratio (dry weight basis). For example, a summary of TFs of radioactive elements in leafy vegetables compiled by the IAEA (2010) is given in Table 1.

Strontium is recognized as one of the minor metals used in industrial products in Japan (e.g., ferrite magnets, flares). Due to human activities, some releases of Sr to the environment are expected, and, consequently, mass of Sr ingestion by humans would increase in the future. The toxicity to human is not much found and ingestion of Sr of 1–4 mg day^{-1} is no risk as summarized by Kabata-Pendias and Mukherjee (2007). Due to its low toxicity to humans, the element concentration does not need to be monitored in foods; however, by simply applying TFs obtained under equilibrium conditions, the Sr mass ingested by humans can easily be estimated using soil Sr concentrations if Sr increments are found in soil samples.

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Table 1 Classification of elements by using GM values of soil-to-crop transfer factor for leafy vegetables compiled by the IAEA (2010)

10^0	10^{-1}	10^{-2}	10^{-3}	$\leqq 10^{-4}$
K, P, Zn	Co, Mn, Mo, Rb, **Sr,** Te	Cs, Na, Nb, Np, Pb, Pr, Ra, Ru, U	Ba, Ce, Cm, Cr, Fe, I, La, Po, Th, Y, Zr	Ag, Am, Pu, Sb

Considering the radiation toxicity, ^{90}Sr, a pure-beta emitter with a long half-life of 28.8 years, is known to be one of the highly toxic radionuclides to humans. Strontium-90 is a nuclear fission product and it is found in the environment as global fallout due to the atmospheric nuclear weapon testing's, mainly in the 1950s and 1960s. The radionuclide also has been released in accidents such as the Chernobyl nuclear accident in 1986. As mentioned above, TFs of Sr are usually high, so that ^{90}Sr intake from agricultural products can occur. Therefore, many TFs of radiostrontium have been reported and these data were compiled by the IAEA (2010); it should be noted that the TF data were mainly obtained in European and North American countries. Because the critical foods in Japan and other Asian countries differ from those in European and North American countries, it is necessary to provide more TF data for more practical assessment use in Japan. In our previous reports (Uchida et al. 2007a, b; Ishikawa et al. 2009), we focused on rice and vegetables because these are critical foods in Japan. We provided TFs of Sr (TF_{Sr}) using stable Sr data in the edible part of crops and associated soil samples from agricultural fields throughout Japan to obtain TF_{Sr} under equilibrium conditions. In this report, since we could add some more TF_{Sr} data to the previous values we reported, we summarize all these data and compared them with those of three other alkaline earth elements, i.e., Mg, Ca, and Ba. We also carried out data mining of global fallout ^{90}Sr in crop and soil samples to provide values of TFs of ^{90}Sr (TF_{90Sr}) and compared them with TF_{Sr} values.

Strontium, Magnesium, Calcium, and Barium Concentrations in Japanese Agricultural Field Soils

Paddy field and upland field soils (plowed soil layer: up to ca. 20 cm depth) were collected nationwide from 68 and 99 sampling sites, respectively, in 2002–2011. Data were partially reported in our previous publications (Uchida et al. 2007a, b). Soils were classified as Andosols (49 sites), Cambisols (26 sites), Fluvisols (83 sites), and other soil types (nine sites) according to the FAO/UNESCO soil taxonomy (Food and Agriculture Organization 2005). Sample collection and pretreatment methods were as follows. From one sampling field, five sub-samples, approximately 1 kg on fresh weight basis each, were collected in the harvest season and these sub-samples were mixed well. Each soil sample was air-dried and passed through a 2-mm mesh sieve. A portion of each air-dried and sieved soil sample was thoroughly ground into fine powder. The fine powdered sample was used to measure the element concentrations according to the methods reported elsewhere (Uchida et al. 2007a, b). Briefly, each soil sample was digested with a mixture of HNO_3 and HF using a microwave digester (CEM, Mars 5). Following their digestion, soil samples were evaporated to near dryness at 140 °C. Then $HClO_4$ was added and evaporated to remove Si. The residue was dissolved with HNO_3 and H_2O_2, and evaporated again. Finally, the residue was dissolved with HNO_3 and diluted with deionized water to adjust 2% HNO_3. The digestion samples were made in duplicate. All the acids used were in the ultrapure analytical grade (Tama Chemicals, AA-100). Water (>18.1 MΩ) which was treated using a Milli-Q water system (Millipore Co.) was used throughout the work. After diluting the sample solutions to a suitable concentration, Mg, Ca, Sr and Ba in dissolved samples were measured using ICP-optical emission spectrometry (Seiko, Vista Pro). Typical analytical errors between two replicates were within 10%. All concentration data reported are on dry weight basis; hereafter, concentrations of Sr, Mg, Ca, and Ba in soils (mg kg^{-1} dry) are described as S_{Sr}, S_{Mg}, S_{Ca}, and S_{Ba}, respectively.

Figure 1a shows probability distributions of the S_{Sr}, S_{Mg}, S_{Ca}, and S_{Ba} in fluvisol samples, and they were a log-normal type. Andosol and cambisol samples also showed a log-normal type distribution (data not shown), thus we calculated geometric mean (GM) values for each soil type and they are shown in Table 2. No difference was found for Sr concentrations in any soil types. For Mg, Ca, and Ba, their concentrations in Andosol were significantly higher than in Cambisol and Fluvisol by *t*-test ($p < 0.01$), however, according to the GM values, their difference was just within a factor of two so the difference was small compared to the concentration variation of each element in each soil type. In Table 2, we also provided some soil characteristics. Concentrations of acid oxalate extractable Al and Fe, shown as act. Al and act. Fe, respectively in the table was slightly higher in Andosol than in other soil types; Andosol is a volcanic ash soil and widely distributed in Japan.

The GM values of S_{Sr}, S_{Mg}, S_{Ca}, and S_{Ba} were then compared with reported data in soils (Reimann and De Caritat 1998). For Sr, the world average concentration was 240 mg kg^{-1}, and agricultural soil samples from 0 to 25 cm depths collected in Finland had an average concentration of 22.9 mg kg^{-1}. The GM of S_{Sr} was 77 mg kg^{-1}, thus Sr concentration in Japanese agricultural field soils was within the range of these reported data. For Mg, Ca, and Ba, their world average concentrations were 9000, 14,000, and 500 mg kg^{-1}, respectively, and those in agricultural soil samples from 0 to 25 cm depths collected in Finland were 2220, 3280, and 43.2 mg kg^{-1}, respectively. Thus, GMs of S_{Mg}, S_{Ca}, and S_{Ba} in Japan, 6060, 10,900, and 250 mg kg^{-1}, respectively, were within these values. From these results, we can generalize that agricultural crops in Japan are grown under the similar alkaline earth element concentrations as found elsewhere in the world. We previously reported that the concentrations of major and trace elements in Japanese upland field soils did not differ too much amongst soils in which plants grow easily (Tagami and Uchida 2010), and thus other element concentrations, as total, were almost the same as for alkaline earth elements.

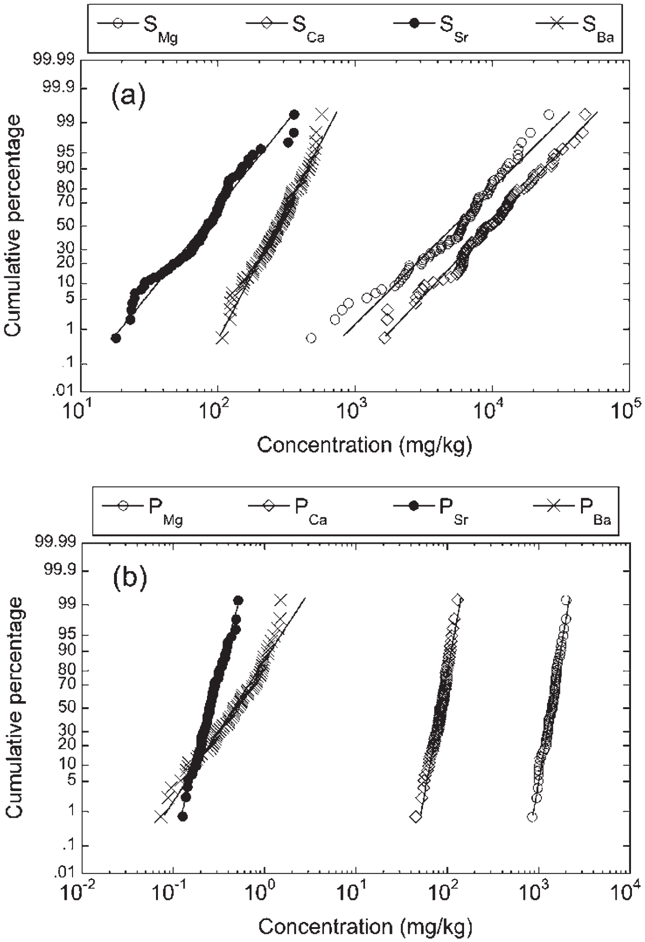

Fig. 1 Probability distributions of concentrations of Mg, Ca, Sr and Ba in (**a**) fluvisol (S_{Mg}, S_{Ca}, S_{Sr} and S_{Ba}) and (**b**) brown rice (P_{Mg}, P_{Ca}, P_{Sr} and P_{Ba}) samples collected throughout Japan

Strontium, Magnesium, Calcium, and Barium Concentrations in Plants

For brown rice, 68 samples were collected from the same sampling sites as the paddy field soils. Dust and soil particles were removed from the rice heads and then air-dried before they were hulled to obtain brown rice. From upland fields, wheat, soybean, leafy vegetables (cabbage, Chinese cabbage, lettuce, spinach, Welsh onion, etc.), fruit vegetables (cucumber, tomato, eggplant, etc.), tubers (sweet potato, potato), and root crops (Japanese radish, carrot, onion) were collected. Data were partially reported in our previous publications (Uchida et al. 2007a, b; Tagami and Uchida 2009). Edible parts of the samples were washed with deionized water at least three times to remove dust and soil particles. The washed parts were paper-towel dried, chopped and

Table 2 Characteristics of agricultural field soils in Japan and Mg, Ca, Sr and Ba concentrations on dry weight basis

Soil type	*N*		pH	CEC meq 100 g^{-1}	ex. Ca mg kg^{-1}	ex. K mg kg^{-1}	act. Al g kg^{-1}	act. Fe g kg^{-1}	Mg mg kg^{-1}	Ca mg kg^{-1}	Sr mg kg^{-1}	Ba mg kg^{-1}
Andosol	49	**GM**	**6.2**	**18**	**2910**	**360**	**27**	**12**	**8340**	**14,500**	**77**	**190**
		Min	5.0	9.0	560	76	1.9	4.0	1220	3050	17	56
		Max	7.4	45	8160	2000	96	33	25,000	59,500	250	440
Cambisol	26	**GM**	**6.0**	**14**	**1950**	**380**	**3.0**	**6.1**	**5020**	**8970**	**73**	**300**
		Min	4.6	6.0	600	110	1.0	1.8	360	630	12	100
		Max	7.3	24	4510	3200	66	28	18,000	29,000	240	560
Fluvisol	83	**GM**	**6.0**	**13**	**1920**	**260**	**2.7**	**5.8**	**5270**	**9540**	**78**	**270**
		Min	4.3	5.0	620	74	0.7	1.5	480	1650	18	110
		Max	8.1	27	8380	1890	41	22	25,800	47,600	360	570
Other soils	9	**GM**	**7.2**	**17**	**3500**	**310**	**2.4**	**6.1**	**6760**	**12,900**	**76**	**250**
		Min	5.7	13	440	59	0.5	1.1	2890	3670	26	150
		Max	8.5	26	8300	960	30	12	14,300	36,000	210	520
All	167	**GM**	**6.1**	**15**	**2250**	**300**	**5.3**	**7.3**	**6060**	**10,900**	**77**	**250**
		Min	4.3	5.0	440	59	0.5	1.1	360	630	12	56
		Max	8.5	45	8380	3200	96	33	25,800	59,500	360	570

CEC cation exchange capacity, *ex. Ca and ex. K* exchangeable Ca and K, act. Al and act. Fe, active Al and Fe extracted with acid oxalate solution freeze-dried. A portion of each freeze-dried sample was thoroughly ground into fine powder. The powdered samples were used for element concentration measurements after the same microwave digestion method used for soil samples; before this step, however, 10 h heating was carried out with HNO_3 at 80°C to decompose organic matter. After diluting the sample solutions to a suitable concentration, Mg, Ca and Ba in dissolved samples were measured using the ICP-optical emission spectrometry, and Sr was measured by ICP-mass spectrometry (Yokogawa, Agilent 7500c). Dry weight basis data are reported here; hereafter, concentrations of Sr, Mg, Ca, and Ba in crops (mg kg^{-1} dry) are described as P_{Sr}, P_{Mg}, P_{Ca}, and P_{Ba}, respectively. Similar to the soil data, probability distributions of P_{Sr}, P_{Mg}, P_{Ca}, and P_{Ba} in brown rice samples are shown in Fig. 1b, and they were a log-normal type. Compared to the number of samples for brown rice, we had smaller numbers of samples of other crops, but we assumed that the other crops also showed a log-normal type distribution. Thus, GM values for each crop were calculated and the results are shown in Table 3.

To supplement the plant concentration data, we also used the data reported by Takeda et al. (2004). They collected wild plant leaves throughout Japan and measured elemental concentrations using neutron activation analysis. We selected plant species with more than three individual plant data and calculated their GMs which are shown in Table 4. These data included 35 trees and two herbaceous plants (perennial) so that they had different growing periods from those of the agricultural crops. The GM values were also used for comparisons among P_{Sr}, P_{Mg}, P_{Ca}, and P_{Ba}.

Correlations for P_{Sr}, P_{Mg}, P_{Ca}, and P_{Ba} are plotted in Fig. 2 using all the dry weight basis concentration data for crops (edible part) and wild plant leaves. Correlation factor (R) was calculated based on the logarithm of the raw concentration data. When $\log(P_{Sr})$ values were compared with $\log(P_{Mg})$, $\log(P_{Ca})$ and $\log(P_{Ba})$, all these results were found to be significantly correlated by t-test ($p < 0.001$), and R values decreased in the following order: Ca ($R = 0.943$) > Ba ($R = 0.888$) > Mg ($R = 0.532$). Correlations among Mg, Ca, and Ba were not as good as we found between P_{Sr}, $-P_{Ca}$ and P_{Sr}, $-P_{Ba}$.

Interestingly, P_{Sr}, P_{Mg}, P_{Ca}, and P_{Ba} in edible part of crops and wild plant leaves (mostly trees) were not separated in the Fig. 2 plots. Especially from the results of $P_{Sr} - \mathrm{P}_{Ca}$, we may roughly estimate P_{Sr} in a wild tree leaf from P_{Ca} using correlations between $P_{Sr} - P_{Ca}$ observed by agricultural crop data as we reported previously (Tagami and Uchida 2010).

Comparisons of Soil-to-Crop Transfer Factors of Alkaline Earth Elements

Transfer factors were calculated for Sr, Mg, Ca, and Ba (TF_{Sr}, TF_{Mg}, TF_{Ca}, and TF_{Ba}, respectively) on dry weight basis for agricultural crop samples using the following equation.

$$TF_M = P_M\left(\text{mg kg}^{-1}\text{-dry}\right) / S_M\left(\text{mg kg}^{-1}\text{-dry}\right), \tag{1}$$

where $_M$ is Sr, Mg, Ca, or Ba.

Table 3 Concentrations of Mg, Ca, Sr and Ba in edible part of crops and their soil-to-crop transfer factors calculated on dry weight basis

Sample name	*N*		Mg	Ca	Sr	Ba	Mg	Ca	Sr	Ba
			mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	TF	TF	TF	TF
Brown rice	68	**GM**	**1380**	**84**	**0.3**	**0.5**	**2.4E-1**	**8.9E-3**	**3.2E-3**	**1.7E-3**
		Min	850	45	0.1	0.1	7.4E-2	1.4E-3	6.6E-4	1.8E-4
		Max	1890	130	0.5	1.5	2.4E+0	5.2E-2	1.6E-2	7.8E-3
Wheat	7	**GM**	**1320**	**200**	**1.3**	**2.2**	**1.7E-1**	**1.3E-2**	**1.4E-2**	**1.1E-2**
		Min	1040	106	0.8	0.3	6.3E-2	5.1E-3	5.6E-3	2.7E-3
		Max	1570	335	1.9	5.6	1.9E+0	3.0E-2	2.7E-2	4.3E-2
Soybean	6	**GM**	**2570**	**1690**	**4.7**	**7.3**	**7.4E-1**	**2.4E-1**	**7.7E-2**	**3.0E-2**
		Min	2200	1390	2.3	4.2	3.0E-1	1.1E-1	3.6E-2	1.3E-2
		Max	2910	2290	7.5	20.4	3.0E+0	6.1E-1	1.9E-1	1.0E-1
Cabbage	11	**GM**	**1610**	**6870**	**15.4**	**4.7**	**2.4E-1**	**5.8E-1**	**1.9E-1**	**2.0E-2**
		Min	1090	4070	7.6	0.8	1.2E-1	2.4E-1	7.3E-2	1.6E-3
		Max	2190	12,050	87.7	13.3	4.6E-1	2.1E+0	1.0E+0	7.3E-2
Chinese cabbage	9	**GM**	**1620**	**7550**	**23.7**	**12.5**	**1.7E-1**	**6.7E-1**	**3.1E-1**	**5.5E-2**
		Min	950	4600	11.1	8.3	5.1E-2	2.7E-1	1.1E-1	2.5E-2
		Max	1960	11,630	54.1	19.3	5.0E-1	2.2E+0	8.1E-1	9.2E-2
Spinach	6	**GM**	**8810**	**7440**	**16.5**	**7.0**	**9.4E-1**	**5.1E-1**	**2.3E-1**	**3.5E-2**
		Min	6180	5540	11.9	1.8	4.4E-1	3.6E-1	9.2E-2	1.0E-2
		Max	11760	11,620	40.3	50.6	2.1E+0	9.1E-1	4.4E-1	1.1E-1
Lettuce	3	**GM**	**1650**	**5080**	**16.1**	**4.1**	**3.0E-1**	**4.1E-1**	**2.4E-1**	**1.8E-2**
		Min	1500	4430	10.3	2.4	2.4E-1	3.1E-1	1.2E-1	4.8E-3
		Max	1740	6140	29.6	7.4	3.6E-1	6.2E-1	6.0E-1	5.0E-2
Welsh onion	8	**GM**	**1920**	**6240**	**12.9**	**3.7**	**1.9E-1**	**4.0E-1**	**1.3E-1**	**1.8E-2**
		Min	1020	2350	3.0	0.4	9.2E-2	9.4E-2	4.5E-2	1.2E-3
		Max	3490	20,120	72.5	18.1	6.2E-1	1.4E+0	7.5E-1	1.2E-1
Other leafy vegetables	11	**GM**	**2110**	**9000**	**24.6**	**8.8**	**2.2E-1**	**5.6E-1**	**2.4E-1**	**3.6E-2**
		Min	740	2120	4.8	1.4	4.2E-2	8.2E-2	6.2E-2	1.2E-2
		Max	3900	34,400	131	41.1	1.4E+0	5.5E+0	3.0E+0	1.3E-1
Cucumber	5	**GM**	**3130**	**4440**	**13.6**	**7.3**	**3.5E-1**	**2.3E-1**	**1.2E-1**	**2.8E-2**
		Min	2700	3270	6.1	2.3	2.4E-1	1.6E-1	6.4E-2	1.7E-2
		Max	3430	6690	20.6	18.0	4.5E-1	4.4E-1	1.8E-1	5.5E-2
Tomato	7	**GM**	**1580**	**1370**	**2.8**	**0.7**	**4.1E-1**	**1.4E-1**	**4.7E-2**	**4.5E-3**
		Min	1430	1030	1.5	0.3	2.1E-1	7.2E-2	1.1E-2	5.4E-4
		Max	1630	1770	10.8	1.9	1.3E+0	2.8E-1	1.4E-1	1.8E-2
Eggplant	5	**GM**	**2350**	**2380**	**3.2**	**1.4**	**3.1E-1**	**2.1E-1**	**4.6E-2**	**6.1E-3**
		Min	2110	1810	2.6	0.6	1.4E-1	8.5E-2	1.6E-2	2.4E-3
		Max	2500	2800	4.3	3.1	1.0E+0	3.3E-1	1.2E-1	1.2E-2
Other fruit vegetables	3	*GM*	**2090**	**1490**	**2.4**	**0.5**	**1.6E-1**	**6.7E-2**	**2.3E-2**	**2.5E-3**
		Min	1530	720	0.8	0.2	1.0E-1	3.6E-2	6.0E-3	9.8E-4
		Max	3430	4410	22.1	1.0	2.1E-1	9.3E-2	1.2E-1	5.4E-3

(continued)

Table 3 (continued)

Sample name	N		Mg	Ca	Sr	Ba	Mg	Ca	Sr	Ba
			mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	mg kg^{-1}	TF	TF	TF	TF
Sweet potato	3	**GM**	**570**	**940**	**4.4**	**1.9**	**1.3E-1**	**9.0E-2**	**4.8E-2**	**6.1E-3**
		Min	480	740	3.0	0.6	1.1E-1	6.5E-2	1.5E-2	1.1E-3
		Max	700	1270	6.5	6.6	2.0E-1	1.7E-1	1.4E-1	2.2E-2
Potato	7	**GM**	**980**	**150**	**1.2**	**1.7**	**2.9E-1**	**2.2E-2**	**1.9E-2**	**5.2E-3**
		Min	700	8	0.6	0.3	7.5E-2	7.8E-4	4.8E-3	1.4E-3
		Max	1220	810	3.7	5.5	1.9E+0	1.9E-1	7.2E-2	1.3E-2
Japanese radish	8	**GM**	**1180**	**6670**	**18.5**	**7.6**	**2.0E-1**	**4.8E-1**	**2.5E-1**	**3.5E-2**
		Min	940	3230	5.1	1.3	4.2E-2	8.4E-2	2.2E-2	5.4E-3
		Max	1590	150,400	305	236	9.2E-1	8.8E+0	4.5E+0	1.4E+0
Onion	4	**GM**	**770**	**2060**	**6.5**	**1.1**	**3.5E-1**	**2.8E-1**	**8.3E-2**	**3.2E-3**
		Min	600	1250	4.8	0.1	1.7E-1	1.1E-1	6.0E-2	7.6E-4
		Max	991	3930	9.5	4.6	6.2E-1	6.4E-1	1.8E-1	9.8E-3
Carrot	4	**GM**	**1100**	**2300**	**17.5**	**22.0**	**1.2E-1**	**1.2E-1**	**1.1E-1**	**7.3E-2**
		Min	1060	1710	7.2	12.8	5.9E-2	6.6E-2	3.0E-2	4.5E-2
		Max	1170	2730	44.6	53.0	3.8E-1	1.7E-1	2.2E-1	1.1E-1

Additional $\mathrm{TF}_{\mathrm{Sr}}$, $\mathrm{TF}_{\mathrm{Ca}}$, and $\mathrm{TF}_{\mathrm{Ba}}$ values were added from the data reported by Tsukada and Nakamura (1998); because these TFs were reported in wet weight crop basis, the data were changed into dry weight basis using dry–wet ratios. All TF results are summarized in Table 3. Unfortunately, S_{Sr}, S_{Mg}, S_{Ca}, and S_{Ba} were not available for wild plants so that we could not include wild plant TF data.

Figure 3 shows correlations among the TF values of alkaline earth elements. When we compared $\mathrm{TF}_{\mathrm{Sr}}$ with $\mathrm{TF}_{\mathrm{Mg}}$, $\mathrm{TF}_{\mathrm{Ca}}$, and $\mathrm{TF}_{\mathrm{Ba}}$ in the logarithm values, R values decreased in the following order: Ca ($R = 0.929$) > Ba ($R = 0.816$) > Mg ($R = 0.275$). Because of this high correlation between $\mathrm{TF}_{\mathrm{Sr}}$ and $\mathrm{TF}_{\mathrm{Ca}}$, it was implied that crops having high $\mathrm{TF}_{\mathrm{Ca}}$ would show high $\mathrm{TF}_{\mathrm{Sr}}$. $\mathrm{TF}_{\mathrm{Sr}}$ values were high in leafy vegetables and root crops, for example, Chinese cabbage (GM = 0.31) and Japanese radish (GM = 0.25); while those in cereals were one to two orders of magnitude lower. Similar values were found for $\mathrm{TF}_{\mathrm{Ca}}$. Nisbet and Shaw (1994) also reported a similar tendency in their radiotracer experiment using ^{90}Sr: $\mathrm{TF}_{90\mathrm{Sr}}$ values for cabbage and carrot were higher than for barley.

It is also interesting to point out that $\mathrm{TF}_{\mathrm{Sr}}$ and $\mathrm{TF}_{\mathrm{Ca}}$ data points were almost on the 1:1 line; thus, $\mathrm{TF}_{\mathrm{Sr}}$ can be estimated using the following equation.

$$\log\left(\mathrm{TF}_{\mathrm{Sr}}\right) = -0.472 + 0.927\ \log\left(\mathrm{TF}_{\mathrm{Ca}}\right). \qquad (2)$$

From this equation, it was clear that $\mathrm{TF}_{\mathrm{Sr}}$ tended to be lower than $\mathrm{TF}_{\mathrm{Ca}}$. Another $\mathrm{TF}_{\mathrm{Sr}}$ estimation method was used with S_{Sr} and P_{Ca} (Ishikawa et al. 2009), and the equation is as follows.

Table 4 Calculated geometric mean concentrations of Mg, Ca, Sr and Ba in leaves of wild plants collected throughout Japan on dry weight basis

		Mg		Ca		Sr		Ba	
Scientific name	Plant type	*N*	mg kg^{-1}	*N*	mg kg^{-1}	*N*	mg kg^{-1}	*N*	mg kg^{-1}
Bischofia javanica	Tree	12	4950	13	17,600	11	112	3	22
Pinus densiflora	Tree	14	1030	16	4350	14	77	10	18
Mallotus japonicus	Tree	7	3140	7	18,000	6	180	4	29
Hydrangea macrophylla	Tree	9	4370	9	19,700	8	135	8	45
Distylium racemosum	Tree	5	1790	5	20,000	5	65	3	34
Ginkgo biloba	Tree	5	3610	5	14,400	5	126	5	30
Ilex crenata	Tree	7	3370	9	11,300	8	776	9	113
Deutzia crenata	Tree	6	3550	6	19,800	6	266	6	63
Prunus speciosa (Koidz.) Nakai 1915	Tree	8	3570	8	15,200	8	179	8	86
Macaranga tanarius (L.) Muell. Arg.	Tree	5	2980	5	12,900	4	110	5	34
Dendropanax trifidus	Tree	5	3780	5	17,800	4	230	3	44
Lindera umbellata	Tree	5	2570	5	11,200	5	135	5	28
Cleyera japonica	Tree	8	3720	8	12,800	8	118	8	82
Smilax china	Tree	11	1810	10	11,600	10	127	5	20
Sapium japonicum	Tree	11	2550	15	15,800	13	232	8	42
Cryptomeria japonica	Tree	16	1840	16	13,700	15	141	12	27
Cycas revoluta	Tree	5	2380	5	14,400	4	54	–	–
Cerasus × yedoensis (Matsum.) A.V.Vassil. ‘Somei-yoshino’	Tree	8	3830	7	15,400	8	151	8	43
Camellia sinensis	Tree	5	2170	12	8170	7	64	11	49
Camellia japonica	Tree	14	3920	19	12,600	20	113	17	57
Weigela coraeensis	Tree	5	3400	5	22,300	5	204	5	85
Eurya japonica	Tree	9	5250	17	13,900	17	257	17	145
Chamaecyparis obtusa	Tree	7	1840	11	10,700	9	149	10	38

(continued)

Table 4 (continued)

		Mg		Ca		Sr		Ba	
Scientific name	Plant type	*N*	mg kg^{-1}	*N*	mg kg^{-1}	*N*	mg kg^{-1}	*N*	mg kg^{-1}
Metasequoia glyptostroboides	Tree	13	2500	14	13,100	14	177	11	54
Prunus jamasakura Sieb. ex Koidz. 1911	Tree	5	3610	5	18,300	5	171	4	75
Artemisia indica var. maximowiczii	Herbaceous plant	7	2260	6	10,300	7	82	6	26
Clethra barbinervis	Tree	4	3730	6	13,600	4	409	6	111
Mallotus paniculatus	Tree	3	3420	3	14,600	3	256	3	69
Diospyros kaki	Tree	4	3240	4	7290	2	48	2	12
Glochidion zeylanicum	Tree	3	4310	3	12,200	3	281	3	41
Bambuseae Kunth ex Dumort.	Tree	3	1550	3	2690	3	71	3	37
Aralia elata	Tree	3	2450	3	12,300	3	221	2	121
Triadica sebifera	Tree	3	1230	3	10,800	3	58	3	9
Cirsium japonicum	Herbaceous plant	3	2420	3	18,000	3	194	3	32
Aucuba japonica var. *borealis*	Tree	3	3330	3	9770	3	150	3	27
Hydrangea serrata	Tree	3	3890	3	11,900	3	182	3	63
Pinus luchuensis	Tree	4	1040	6	5160	4	159	2	59

Data source: Takada et al. (1994)

$$\log\left(\mathrm{TF}_{\mathrm{Sr}}\right) = -0.88\ \log\left(S_{\mathrm{Sr}}\right) + 0.93\ \log\left(P_{\mathrm{Ca}}\right) - 2.53. \tag{3}$$

Although P_{Ca} data were replaced with Ca concentration in crops from a food composition database compiled by the Ministry of Education, Culture, Sports, Science and Technology (MEXT 2017), the estimated $\mathrm{TF}_{\mathrm{Sr}}$ values still agreed well with the measured $\mathrm{TF}_{\mathrm{Sr}}$ (Ishikawa et al. 2009). Thus, even if we measured only S_{Sr}, $\mathrm{TF}_{\mathrm{Sr}}$ could be easily provided with equation (3) using officially available P_{Ca} in the crop, e.g., from the food composition database in Japan (MEXT 2017).

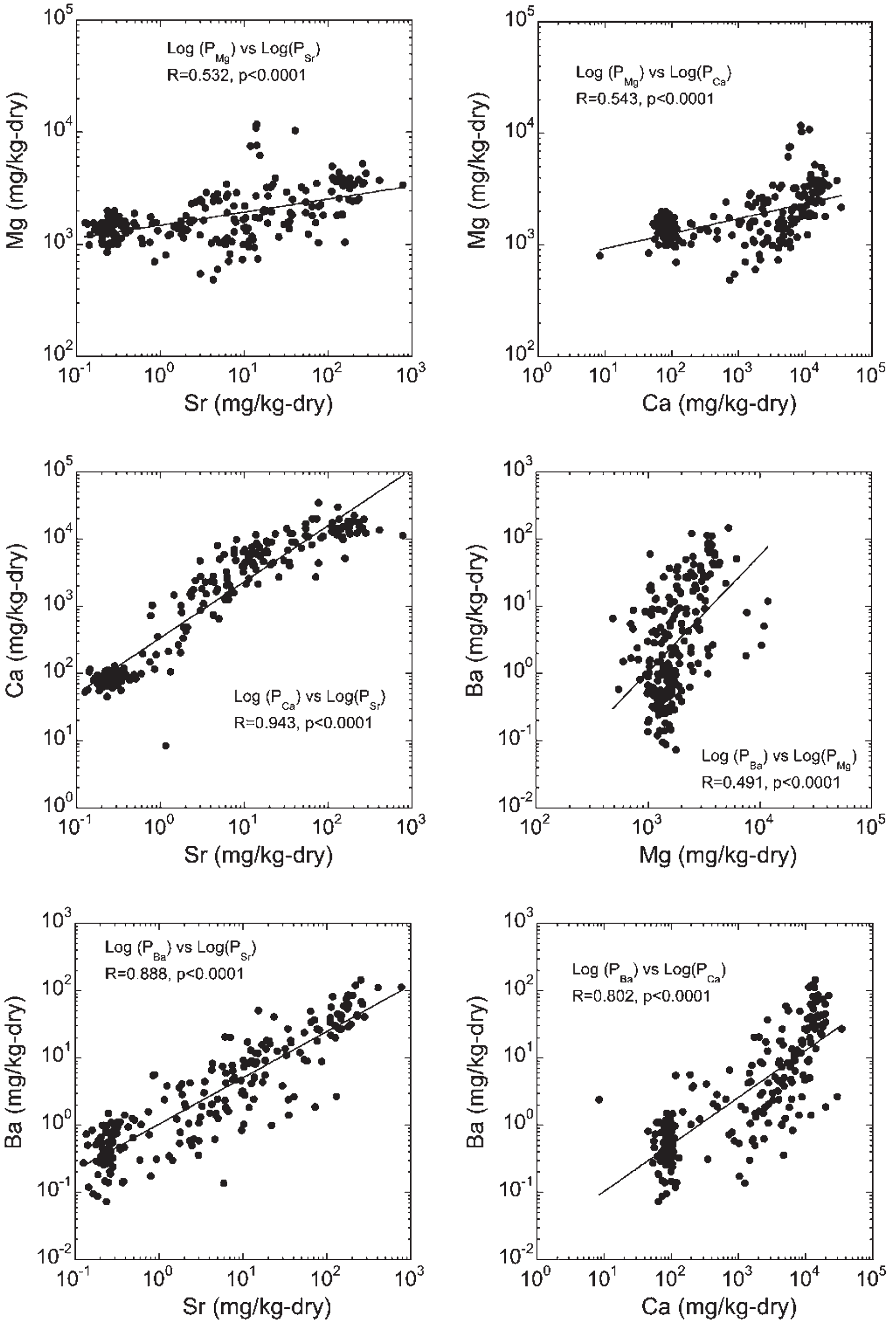

Fig. 2 Correlations between concentrations of Mg, Ca, Sr and Ba in plant samples (P_{Mg}, P_{Ca}, P_{Sr} and P_{Ba}) calculated on dry weight basis

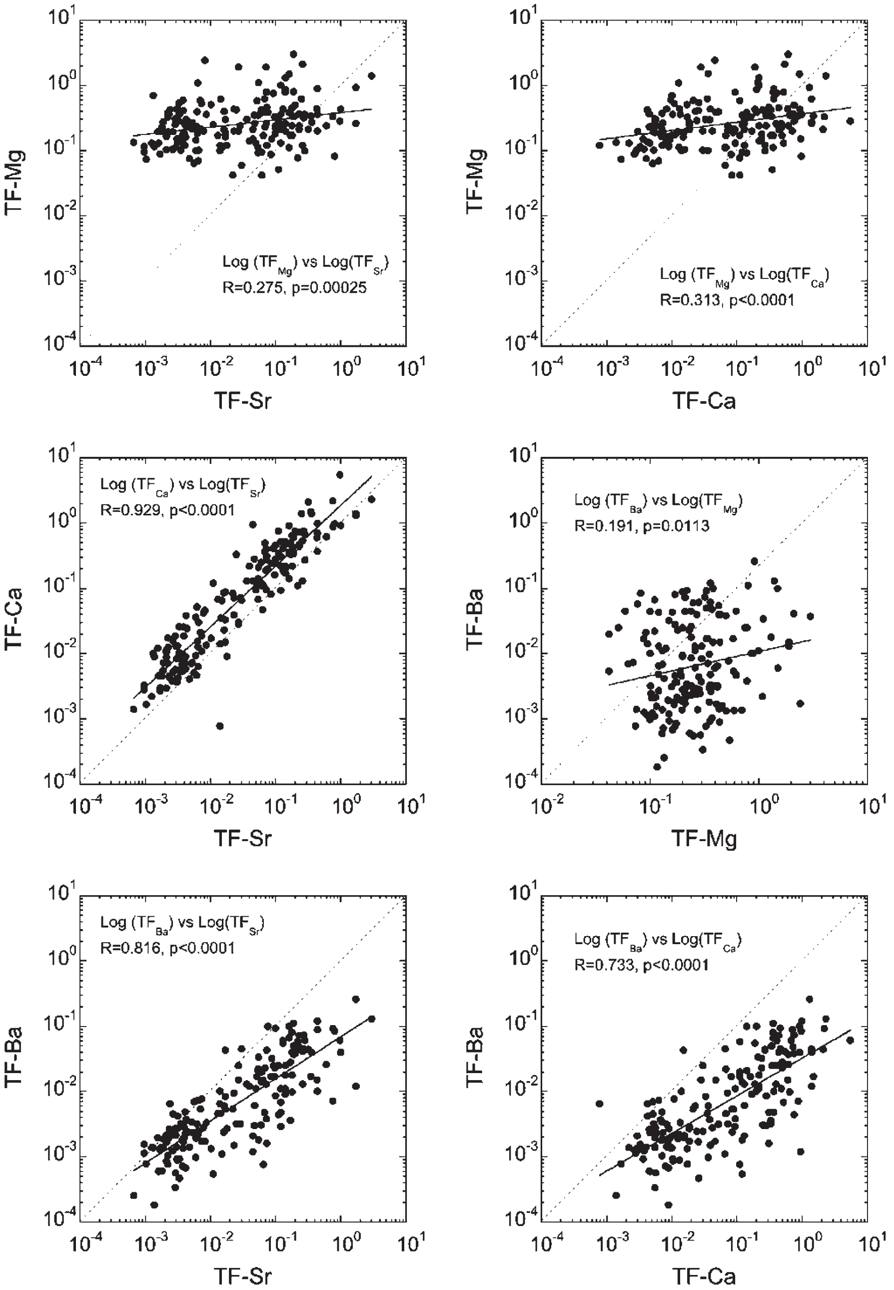

Fig. 3 Correlations between transfer factors of Mg, Ca, Sr and Ba in plant samples (TF_{Mg}, TF_{Ca}, TF_{Sr} and TF_{Ba}) calculated on dry weight basis

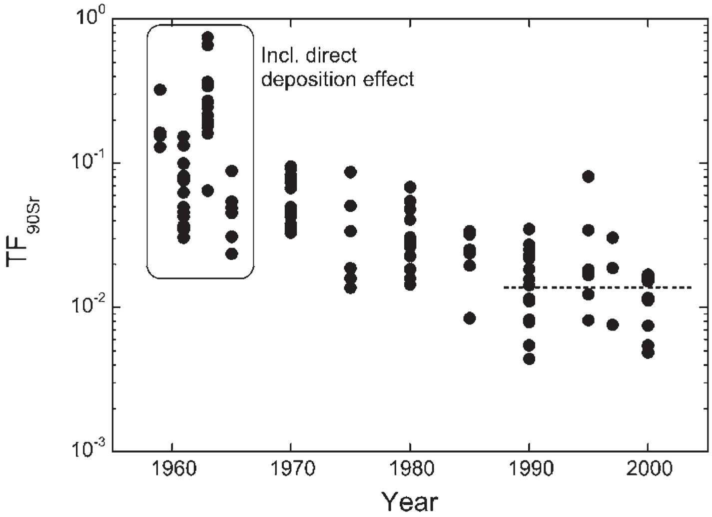

Fig. 4 Time dependence of transfer factors of global fallout ^{90}Sr (TF$_{90Sr}$) in brown rice samples collected in Japan. The dotted line shows the geometric mean value of TF$_{90Sr}$ in 1990–2000

Comparisons of Transfer Factors of Stable Sr and ^{90}Sr

TFs of Stable Sr and ^{90}Sr

It is well known that mobility of a freshly added radionuclide in soil is higher initially and decreases later due to an aging effect. For ^{90}Sr, Nisbet and Shaw (1994) found TF decreased with time for cabbage grown in loam and sand. In Japan, Komamura and her group measured global fallout ^{90}Sr in soil and brown rice from 1959 to 2000 (Komamura et al. 2005). Using their data, we previously calculated TF values (TF$_{90Sr}$) (Uchida et al. 2009). We have plotted these TF$_{90Sr}$ values in Fig. 4. Because of the direct deposition effect until the 1960s, the TF$_{90Sr}$ values were high, and indeed, Komamura et al. (2006) estimated that more than half of the ^{90}Sr content in rice was from the direct deposition effect in the 1960s. After that period, however, a decreasing tendency until 1980s was found possibly due to the aging effect. Wang et al. (1998) reported brown rice TF$_{90Sr}$ data observed in Taiwan for samples collected in 1982 and the value (GM = 0.07) was slightly higher than that observed in Japan in 1980 (GM = 0.03, calculated by using Komamura et al. (2006)). TF$_{90Sr}$ data for brown rice in 1990, 1995 and 2000 became almost constant as seen from Fig. 4. Tsukada et al. (2005) also reported ^{90}Sr concentrations in white rice, bran, and soil from three sites collected in 1997. Using white rice and bran data, we reconstructed ^{90}Sr concentrations in brown rice and then TF in brown rice was calculated and the data are also plotted in Fig. 4. All these data were similar in 1990–2000 and therefore we calculated the GM value.

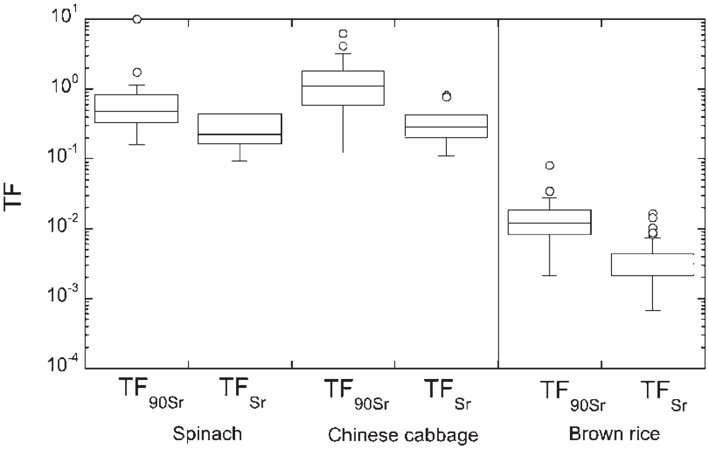

Fig. 5 Comparisons between transfer factors of global fallout ^{90}Sr (TF_{90Sr}) and stable Sr (TF_{Sr}) in spinach, Chinese cabbage and brown rice

For other crops, we assumed that ^{90}Sr data from 1990 would be appropriate to obtain TF values under equilibrium conditions according to the brown rice results mentioned above. To have TF_{90Sr} for other crops, we surveyed the environmental radiation database (Nuclear Regulation Authority 2017) and found ^{90}Sr data in Chinese cabbage and spinach together with those in associated soils. Finally, 37 values of TF_{90Sr} for Chinese cabbage and 24 values of TF_{90Sr} spinach were calculated. These data together with brown rice data were compared with TFs of stable Sr (TF_{Sr} values in Table 3) and the results are shown in Fig. 5. For all crop types, TF_{Sr} values were significantly lower than TF_{90Sr} values by *t*-test ($p < 0.01$). For example, GM of TF_{Sr} for brown rice was 0.0032 and that of TF_{90Sr} was 0.014; GM of TF_{Sr} for Chinese cabbage was 0.31 and that of TF_{90Sr} was 0.98; GM of TF_{Sr} for spinach was 0.23 and that of TF_{90Sr} was 0.56. Previously, Ban-nai et al. (1995) measured TF using ^{85}Sr as a tracer and, for Chinese cabbage, they found a TF value of 0.93 (calculated using dry/wet ratio) similar to the TF_{90Sr} value. The difference between TF_{Sr} and TF_{90Sr} would be due to the different physicochemical form in soil of stable Sr and ^{90}Sr, because stable Sr would be fixed in the soil particles.

Effect of Differences of Stable Sr and ^{90}Sr Distributions in Soil and Their Uptakes

For a relatively short-time experiment, Forsberg et al. (2001) reported ^{90}Sr distribution in soil contaminated for 3–4 years using the sequential extraction technique with H_2O (soil water), 1 M NH_4Ac (exchangeable), 0.04 M $NH_2OH \cdot HCl$ (Fe-Mn oxides), 30% H_2O_2 (organic matter), 9 M HNO_3 (strongly bound) and residual. The results showed that ^{90}Sr decreased in the following order: NH_4Ac (44%) > $NH_2OH \cdot HCl$

(27%) > HNO_3 (15%) > H_2O_2 (8%) > H_2O (6%) and there was no ^{90}Sr in the residual fraction. Puhakainen et al. (2001) measured ^{90}Sr (from global fallout and the Chernobyl accident) and stable Sr in Finish organic soil collected in 1997 and found different distributions. From their results, we calculated average ^{90}Sr distributions, and the values decreased in the following order: NH_4Ac (42%) > H_2O_2 (36%) > $NH_2OH{\cdot}HCl$ (21%) > HNO_3 (1%), while those of stable Sr were as follows: $NH_2OH{\cdot}HCl$ (42%) > NH_4Ac (25%) = H_2O_2 (25%) > HNO_3 (8%). For ^{90}Sr that originated from global fallout and the Chernobyl accident, similarly high extractability in NH_4Ac was reported by Rigol et al. (1999) using soils from the Mediterranean area and Chernobyl area. It should be noted that ^{90}Sr mobility in soil would not be the same as that of global fallout because fuel particles including ^{90}Sr were discharged to the environment. Probably due to fuel fragment particles in the soil, NH_4Ac extractable ^{90}Sr changed with time (Krouglov et al. 1998). In mineral soil, stable Sr was distributed so that 30% was in NH_4Ac, ca. 10% was in 6 M HCl and ca. 60% was distributed in the residual fraction (Sánchez et al. 2005). From these results, it was reasonable to attribute the higher TF_{90Sr} to the different physicochemical forms of ^{90}Sr and stable Sr as mentioned above. Yokoo and Nakano (2001) showed that only the exchangeable fraction of Sr in soil is available to plants; thus, if the exchangeable fraction was used for stable Sr, then TF_{90Sr} would agree well with the results.

Thus, we carried out a further analysis using P_{Sr} and S_{Sr} for Chinese cabbage as an example, since in Table 2, we reported exchangeable Ca and total Ca concentrations in soil (hereafter [ex. Ca] and [Ca], respectively). We assumed that exchangeable Sr fraction in a soil could be estimated using the [ex. Ca]/[Ca] ratio in the soil. Based on the Sr exchangeable fraction in soil, $TF_{exSr,}$ was calculated using the following equation.

$$\mathrm{TF}_{\mathrm{exSr}} = P_{\mathrm{Sr}} / \left(S_{\mathrm{Sr}} \cdot [\mathrm{ex.Ca}] / [\mathrm{Ca}] \right)$$

The calculated TF_{exSr} values ranged 0.39–0.98 with GM of 0.55. When these values were compared with TF_{90Sr}, no significant difference was observed by *t*-test. Therefore, as reported by Yokoo and Nakano (2001), only exchangeable Sr fraction in soil would be bioavailable. However, the bioavailable Sr fraction in soil has not been clarified yet, thus further studies are necessary to provide more reliable transfer factors of Sr in soil-to-crops systems under equilibrium conditions.

Consideration on the Crop ^{90}Sr Concentrations from the Fukushima Daiichi Nuclear Accident

We would like to discuss the ^{90}Sr transfer to crops after the Fukushima Daiichi Nuclear Power accident. As we showed above, Sr is more mobile compared to cesium (Cs) in soil (e.g., Nisbet and Shaw 1994; Krouglov et al. 1998; Rigol et al.

1999; Forsberg et al. 2001; Puhakainen et al. 2001), and scientists suspected that internal radiation from ^{90}Sr released by the accident would increase. Merz et al. (2015) reported that because they found a maximum ^{90}Sr/^{137}Cs activity ratio in foods of 0.1, the current Japanese food policy underestimated the ^{90}Sr content of Japanese foods. However, this assumption would lead to an overestimation because they did not consider global fallout ^{90}Sr and ^{137}Cs contributions and long-term ^{90}Sr/^{137}Cs activity ratio. The effect of global fallout can be considered as follows. Igarashi et al. (2005) found ^{90}Sr/^{137}Cs was ca. 0.15 in soil in the 1990s. According to the brown rice data by Komamura et al. (2005), ^{90}Sr/^{137}Cs data in the 1990s ranged from 0.01 to 4.7 with GM of 0.50. Thus, compared to the ratio in soil (Igarashi et al. 2005), we could conclude that TF of ^{90}Sr was indeed 3.3 times higher than TF of ^{137}Cs based on GM values. From these results, if ^{90}Sr/^{137}Cs activity ratio in soil was more than 0.03 then the ^{90}Sr/^{137}Cs activity ratio in foods of more than 0.1 could be achieved. But in such places, ^{137}Cs concentrations should be lower than 1000 Bq kg^{-1} when we calculated them using soil data of Fukushima Prefecture (data from Nuclear Regulation Authority 2017). For example, in areas contaminated with Fukushima ^{137}Cs fallout of more than 1000 Bq kg^{-1}, we calculated the GM of the ^{90}Sr/^{137}Cs activity ratio was 0.0008 (details of the calculation are not given here); thus much less than 0.03. So, if we had soil with ^{137}Cs and ^{90}Sr at concentrations of 1000 and 30 Bq kg^{-1}, respectively, according to the ^{90}Sr/^{137}Cs of 0.03 in soil, and using the TF values we obtained in this study, ^{90}Sr concentration in brown rice was estimated to be 0.42 Bq kg^{-1}-dry (GM of TF_{90Sr} = 0.014) and that in Chinese cabbage was 30 Bq kg^{-1}-dry (GM of TF_{90Sr} = 0.98). On wet weight basis, these concentrations should be 0.36 Bq kg^{-1}-wet for brown rice (water content = 15%) and 1.5 Bq kg^{-1}-wet for Chinese cabbage (water content = 95%). Thus, only a very small amount of ^{90}Sr concentration was expected in crops grown in such soil. Moreover, there is no evidence of ^{90}Sr increases for food in Japan after the Fukushima accident (Ministry of Health, Labour and Welfare 2016). ^{90}Sr concentration in crops in Japan after the Fukushima accident has been within the range we used of global fallout ^{90}Sr, and the concentration will decrease with time in the future.

Conclusions

Soil-to-crop transfer factors (TFs) of Sr were compared with TFs of other alkaline earth elements, i.e., Mg, Ca, Ba in Japanese crops to provide more reliable TF data for Japan and other Asian countries. Among TFs of these elements, Sr and Ca showed a high correlation and it was implied that TF_{Sr} could be simply estimated by TF_{Ca} or concentrations of Sr in soil together with Ca concentration data (measured or openly available data). We made a further comparison with stable Sr and ^{90}Sr TFs; because of their different forms in soils, GMs of TF for Chinese cabbage, spinach and brown rice were 2–4 times different. It is likely that the exchangeable Sr fraction in soil would be bioavailable to crops; however, further studies are necessary to clarify the bioavailable fraction of Sr in soil.AcknowledgmentThis work was partially supported by the Agency for Natural Resources and Energy, the Ministry of Economy, Trade and Industry (METI), Japan.

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^{90}Sr Distribution in System "Soil-Scots Pine" (*Pinus sylvestris* L.)

Ingrida Pliopaitė Bataitienė

Introduction

^{90}Sr is a radioisotope that has artificial origin; it is long-lived beta emitter with a half-life of 28.8 year. ^{90}Sr is formed by decay of ^{235}U and released in the environment during atomic explosions and accidents at nuclear power plants. This radionuclide integrates into metabolism together with biological and chemical analogue (Ca) in the environment (Ladygienė 2006).

Radionuclides can enter into the trees:

- By the tree foliage, through epidermis together with atmospheric fallout, i.e., atmospheric radioactive fallout is absorbed by overground wood cell surfaces. For example, 20–30% of all radioactive contamination was deposited on the forests, and the major part (40–90%) was withhold by canopy after the Chernobyl nuclear power plant (CAE) accident (Avila et al. 2001; Ciuffo et al. 2003; Butkus and Konstantinova 2004; Nedveckaitė 2004; Cybulka et al. 2004; Ščeglov and Cvetnova 2001, 2004). In this way, release of radionuclides into the wood depends on the following factors: tree age, roughness and type of tree foliage, moisture content in foliage, radionuclide properties, precipitation, time of the year, the structure of tree surface elements, and density. The released radioactive fallout can be washed or absorbed from the tree folliage. Radionuclides absorption takes place during radionuclides penetration through the epidermis of the folliage element, i.e., during ion and molecular diffusion; or gaseous radionuclides can be absorbed in cells stoma, and later radionuclides can be absorbed in plasmalemma. The absorption of radionuclides through the tree folliage is a quick process (Gerzabek et al. 1998).

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- Radionuclides can be dropped from the surface of tree leaves during the resuspension (because of wind, soil tillage, and other actions). Such radionuclides can deposit on other plant in other location. Resuspension is the most effective immediately after the deposition, as recently deposited aerosol deposition is easier to lift than the older fallout;
- From the soil through the root system with nutrients, namely, the secondary entering of radionuclides into the wood that is available because of flush and leaf litter contamination. Eighty percent of radioactive contamination deposited in canopy falls on the forest floor after 1–2 months, and full folliage purification can take up to 3 years (Avila et al. 2001; Ciuffo et al. 2003; Butkus and Konstantinova 2004; Nedveckaitė 2004; Cybulka et al. 2004; Ščeglov and Cvetnova 2001, 2004). Radionuclides migrate deep down through the soil toward the tree roots, and enter the tree together with nutrients (Nedveckaitė 2004; Butkus and Pliopaitė Bataitienė 2006; Seidel 2010). The trees absorb the maximum quantity of radionuclides (up to 70%) through the tree root system.
- Radionuclides redistribute in wood elements during the transpiration and assimilation (Gerzabek et al. 1998).

Artificial radionuclides deposited on the soil from the atmosphere (e.g., ^{90}Sr) and exposed to natural biogenic factors are involved in the migration process. Radionuclides migrate in both vertical and horizontal directions. Ionic diffusion, convective transfer together with the soil solution, and migration by plant root systems are the basic radionuclide migration processes in soil. Horizontal migration of radionuclides, occurring with soil erosion, is also possible (Ciuffo et al. 2003; Butkus and Konstantinova 2004; Nedveckaitė 2004; Cybulka et al. 2004; Ščeglov and Cvetnova 2004). Radionuclide migration in the environment is determined by physical and chemical properties of corresponding element, weather conditions, environmental terrain, soil type, hydrological regime of the territory, vegetation type, agrochemical features of agriculture, etc. (Vasilenko and Vasilenko 2002).

Geochemical migration of radionuclides and accumulation in the soil depend not only on the quantity of organic material, and inorganic soil matrix dispersity grade (the more dispersed the rock, the greater its relative active surface), but especially on manganese and iron colloidal compounds in the soil (Mažeika et al. 1998). Accumulation of radionuclides in the wood depends not only on the instantaneous contamination, but also on the secondary contamination, its amount especially depends on soil contamination of the plant habitat.

Radionuclides are found in various forms in soil, such as: soluble in water, absorbed by ion-exchange mechanism, included in mineral crystal lattice, or forming complex with insoluble soil organic materials, insoluble in water, having the form of large dispersion aerosol of nuclear fuel particle. The diverse interaction between the soil and radionuclides is going on in the soil leading to variations in the forms of radionuclides, which are determined by soil type, water treatment, and radionuclide properties (Butkus et al. 2014).

^{90}Sr of global fallout, in most cases, is soluble in water and has a mobile form in the soil. ^{90}Sr takes part in plant metabolism as Ca analogue after having entered into the environment. ^{90}Sr enters the plants through contaminated overground parts with atmospheric fallout deposition on them. The same process is also going within the scope of soil and the roots; the soil type, its pH environment, humidity, Ca and the amount of organic substances have a significant impact on ^{90}Sr migration. ^{90}Sr fixation in a plant depends on the dispersion forms of this radionuclide and specific characteristics of the plant. Biological structure of the tree is one of the factors, influencing the acces of radionuclides into the trees. For example, based on the data presented in the paper, the (Sazykina 2005) conifers are the most sensitive to the effects of ionizing radiation.

^{90}Sr comparing to ^{137}Cs is not characterized by radial mobility in softwood (Rochon et al. 1998; Soukhova et al. 2003). The rate of fixation in plants ^{90}Sr from global depositions can reach 25%. ^{90}Sr is absorbed after deposition on the plant because of active biological processes. ^{90}Sr penetration into the plant depends on water solubility of particles and species of the plant. The ability of the plant to absorb ^{90}Sr from the soil depends on its bioavailability; ^{90}Sr solubility in water; agrochemical soil properties; metabolic Ca content in the soil (Kanapickas and Raupelienė 2002; Vasilenko and Vasilenko 2002; Nuclear contamination …2004). Having entered the soil, artificial ^{90}Sr is withheld on the top layer of minerals. The water-soluble part of the radionuclide fallout moves to metabolic state, and the insoluble part remain unchanged and do not take part in water exchange processes (Butkus et al. 2014).

Artificial radionuclides (e.g., ^{90}Sr) entered Lithuanian soil in nuclear fuel particle form, and radionuclides can disolve and become available to the trees or can be bound with soil components during the decay. The rate of ^{90}Sr fixation in soil is less than disolving from the particles (Beinaravičius 2005; Pliopaitė Bataitienė 2011). The soil in Lithuania was contaminated by artificial radionuclides because of nuclear weapon test in atmosphere (mid of 1940s–mid of 1960s), ChNPP accident in April 26, 1986; as well, although a small influence for other artificial radionuclides (Iodine-131, Caesium isotopes) was felt because of the Fukushima nuclear power plant accident in March 11, 2011 (Lujanienė et al. 2012). According to some articles, ^{90}Sr of "Fukushima genesis" cannot be found in the terrestrial areas around the Fukushima Daichi NPP. The structure of spotty radionuclides on soil surface was determined; hot spots were detected there (Nedveckaitė 2004). Figure 1 presents Lithuanian soil contamination by ^{90}Sr after the ChNPP accident (Butkus et al. 1999, 2001).

The soil contamination by ^{90}Sr has increased on average by 1.1 times, and only in certain areas the amount of this radionuclide has increased by 1.5 times or more after the ChNPP accident, because of meteorological conditions of radioactive contamination of air transport that has passed through Lithuania. Radioactive strontium and plutonium isotopes were rejected out of the emergency reactor into the environment later, when the temperature of hot reactor was thousands degrees, and the air mass was moving in the northeast and later in south directions, so Lithuanian soil contamination by these isotopes changed a little (Butkus 1999; Butkus et al. 2001).

Atlantic Ocean and the short distance from the sea have the main impact on air temperature and its distribution in Lithuania. The increase of soil water level

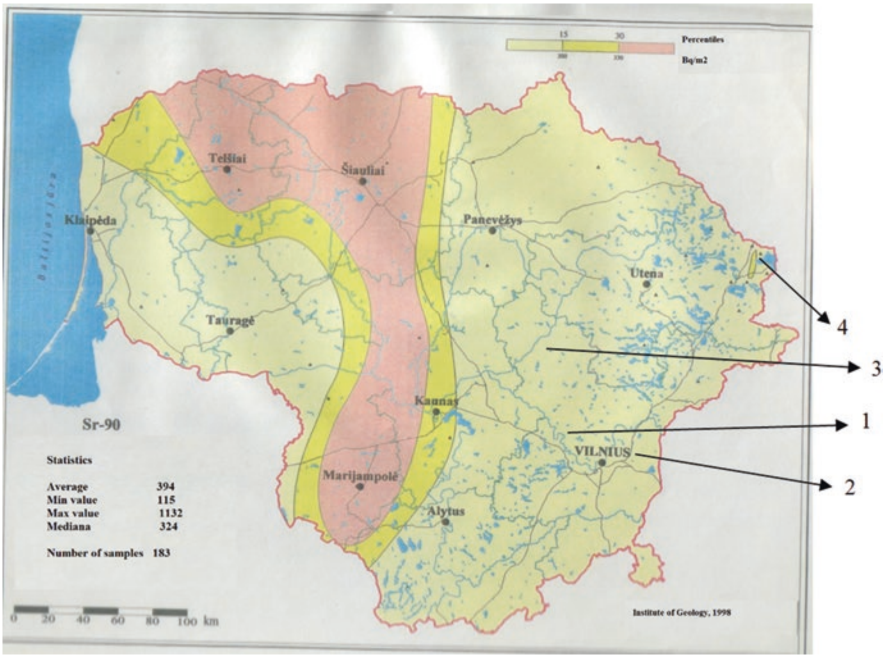

Fig. 1 Radioactive contamination of Lithuanian soils by ^{90}Sr after the Chernobyl NPP accident (Adopted from, Butkus 1999; Butkus et al. 2001): (1) Neris Regional Park, (2) Rūkainiai, (3) Ukmergė district, (4) district of the Ignalina nuclear power plant

promotes the growth of woody biomass in organic soils, and vice versa in minerals (Stravinskienė and Erlickytė 2003). Plant growth and transpiration (when water evaporation is greater) are promoted in the years, when average annual temperature increases are higher than the average (Ruseckas 2002; Beinaravičius 2005). Beinaravičius (2005) stated that the access of radionuclides into the plant indirectly depends on the temperature (ambient air temperature decreases with increasing precipitation, and rainfall decreases at higher ambient temperatures, and it is more difficult for plants to get supply of water). Three-month drought has the greatest negative impact on the growth of trees. Intensive nutrient metabolism occurs at a time of intensive growthof the plant. Accumulation of radionuclides in the tree depends on its growth, development, and intensity of maturation, i.e., on the intensity of the absorption of nutrients that depends on the age of the tree. Biomass growth rate determines this process (Ozolinčius and Sujetovienė 2002; Butkus and Pliopaitė Bataitienė 2006).

The alternation of the accumulation in radionuclides in wood can be associated not only with the radioactive contamination (only in the case of artificial radionuclides), but also with a year with the most favorable conditions for the tree growth. The microclimatic conditions of the habitat (temperature, humidity mode) form (non)favorable conditions for tree growth (Stravinskienė 2002).

Radionuclides accumulated in the plant emit ionizing radiation, which can cause ecotoxicological effects, such as changes of cells, tissues, organs, functional and morphological changes of the whole body, radiation effects (biological, synergistic, and antagonistic effects, genomic instability, and the phenomenon of adaptation). Radionuclide ionizing radiation exposure for the plants of terrestrial ecosystems can be of two types: external and internal. Exposure effects on the tree depend on the type of radionuclide forming the exposure and its amount. In (Sazykina 2005) her work, the author mentions that the threshold dose rate for the deterministic effects is approximately 0.5–1 mSv day^{-1} in case of the long-term influence of ionizing radiation, as well there is the threshold value, where ionizing radiation impact effects associated with the growth of conifers trees is evident and some vertebrate animals have experienced and survived 10 mSv day^{-1}. Conifers are killed at permanent ionizing radiation dose greater than 1 mSv day^{-1}, a danger to deciduous trees also exists (Sazykina 2005).

The aim of this chapter is to determine the distribution of ^{90}Sr in pines (*Pinus sylvestris* L.) during their vegetation periods.

Materials and Methods

Conifer forests occupy the largest forest area (5602%) in Lithuania with the predominant species of conifers, and deciduous cover 43.9% (39.7% of soft hardwood and 4.2% of hardwood) of all forests (Lithuania 2012).

In this study, three locations have been selected for the research: the Neris Regional Park (soil has been contaminated with radionuclides by nuclear tests and accident at Chernobyl NPP), a field in the vicinity of the Ignalina nuclear power plant (was polluted during the Chernobyl NPP accident; later at the Ignalina NPP, the possible source of pollution was localized), and the field in the village in Ukmergė district.

The samples for the determination of radioactive contamination of the soil by ^{90}Sr were collected at the intersection points of concentric circles and the world country directions. They were taken by metal ring (Fig. 2a) and taking into account the prevailing winds (Fig. 2b).

Metal ring is driven into the soil, its bottom is covered by the trowel, and the soil sample is raised for taking soil samples at the intersection points of concentric circles and the world country directions. Each sample is placed in a plastic bag and marked. Concentric circles are drawn every 1 m. Their number is determined according to the diameter of the test tree folliage. The folliage tangent should be the last concentric circle. Soil samples are taken from 30 cm depth in 2–5 cm distance from each other. The samples are collecteded according to the prevailing winds. The depth of sampling is determined by the peculiarities of the layout of tree root systems. When collecting the soil samples based on winds prevailing in the growth place of the tree and having dug 30 cm deep sap, up tp 30 cm deep samples of the soil are taken. The depth of sampling is determined by the peculiarities of the layout

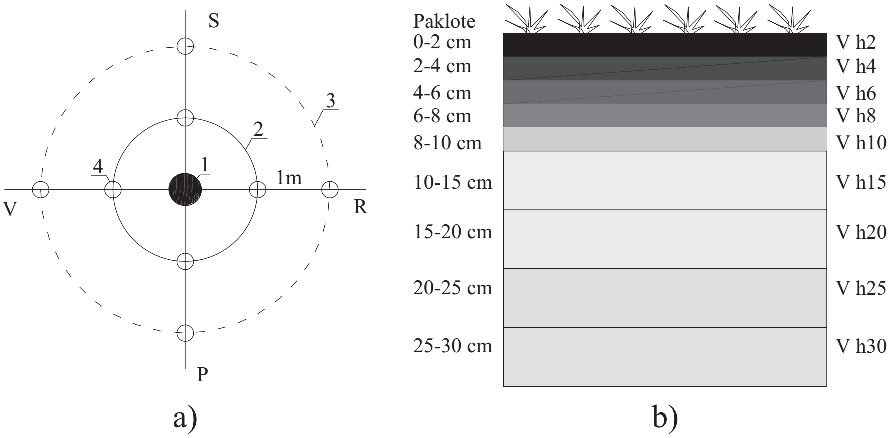

Fig. 2 Soil samples collection: (**a**) at intersection points of the concentric circles and world country directions (1: tree trunk; 2: concentric circle at 1 m from the tree trunk; 3: crown boundary; 4: soil sample); (**b**) taking into account the prevailing winds [V: the prevailing winds in tree habitat; *h*: depth of the soil layer (Adopted from, Pliopaitė Bataitienė 2011)]

of the tree root system. The collected soil samples are weighed dried in natural conditions.

The 3 cm height trunk rollers were taken in order to form pine wood samples. These samples are split by chisels of different size tangentially at every few rings. Branches and root specimens were collected taking into account the thickness of pine segments. Formed samples were dried to a constant weigh and burned to ash at 450 °C temperatures. The chemical concentration of samples conducted in order to determine ^{90}Sr contamination in investigated samples with the separation of ^{90}Y, and ^{90}Sr specific activity was measured by beta radiometer based on this value (LAND 2005). ^{90}Sr specific activity in the sample is calculated according to the equation (Pliopaitė Bataitienė 2011):

$$A_{\text{Sr-90}} = A \div (\mathrm{Y}(^{90}\mathrm{Y}) \cdot m), \tag{1}$$

where $A_{\text{Sr - 90}}$ is ^{90}Sr specific activity in the sample, Bq kg^{-1}; A is the activity in the sample, Bq; Y(^{90}Y) is ^{90}Y chemical yield, %; m is the mass of the sample, kg.

The transfer factor *PF* is used to assess radionuclides insertion through the root system. The system soil–plant transfer factor is the ratio of radionuclide specific activity in the plant A_{a} and radionuclide specific activity in the soil A_{d} (Ban-nai and Muramatsu 2002; Butkus et al. 2003; Nedveckaitė 2004; Marčiulionienė et al. 2008; IAEA 2010):

$$\mathrm{PF} = \frac{A_{\mathrm{a}}}{A_{\mathrm{d}}}, \tag{2}$$

Radionuclides transfer rate from the soil to the root surface is determined by the diffusion and conventional radionuclide transport through the soil layer burrowing to the root system. The concentration of radionuclide activity in the soil is decreased because of radionuclide absorption within the tree (Ban-nai and Muramatsu 2002; Butkus et al. 2003; Nedveckaitė 2004; Marčiulionienė et al. 2008; IAEA 2010). The plants, including trees, absorb certain ions from the soil through the root system depending on physiological characteristics of each type of the plant. Ions absorbed by the tree can compete as competition is going on (Solecki and Chibowski 2002; Sanches et al. 2008). Plants absorb other bivalent ions, such as strontium, in case of lack of calcium. This phenomenon is called the influence of strontium discrimination on calcium transfer. Discrimination factor can be assessed knowing the factor of radionuclide transfer that is equal to the ratio of the rate of radionuclide activity in the tree and soil values on condition that the speed of the radionuclides entering from the soil to the plant is equal to the speed of radionuclide entering from a plant to the soil. Competitiveness factors can be evaluated by discrimination factors that are determined by Ca and ^{90}Sr concentrations in the soil and plants, according with the following equation (Solecki and Chibowski 2002; Sanches et al. 2008):

$$K_{\mathrm{d}} = \frac{\mathrm{PF}_{^{90}\mathrm{Sr}}}{\mathrm{PF}_{\mathrm{Ca}}}, \tag{3}$$

where K_{d} is discrimination factor; $\mathrm{PF}_{^{90}\mathrm{Sr}}$, $\mathrm{PF}_{\mathrm{Ca}}$is ^{90}Sr and Ca transfer factors from soil to the plant.

The conclusion from (Solecki and Chibowski 2002) research work was that calcium ions can determine the absorption of strontium. The discrimination factor expression from (Solecki and Chibowski 2002) can be applied to ^{90}Sr. Ca is chemical analogue of ^{90}Sr radionuclide. The authors argue that the impact of Ca discrimination influence on ^{90}Sr absorption depends on the species of the plant. Accumulated ^{90}Sr in the plant also depends on the presence of adequate calcium forms in the soil solution near the plant root area. Calcium accumulates in microorganisms, plants and animals, and performs essential physiological functions (White and Broadley 2003). Ca is important macroelement to the plants that enters them from the soil solution to sprouts (White 2003). Ca and ^{90}Sr are competing elements during the transfer from the soil to the plant.

Results and Discussion

Figure 3 provides specific activities of ^{90}Sr in soils of investigated areas. Distribution of ^{90}Sr artificial radionuclide in the soil depth is uneven. The nature of this distribution was determined not only by the fundamental processes of radionuclide migration, radionuclide and soil characteristics, but also by human activities. Redistribution

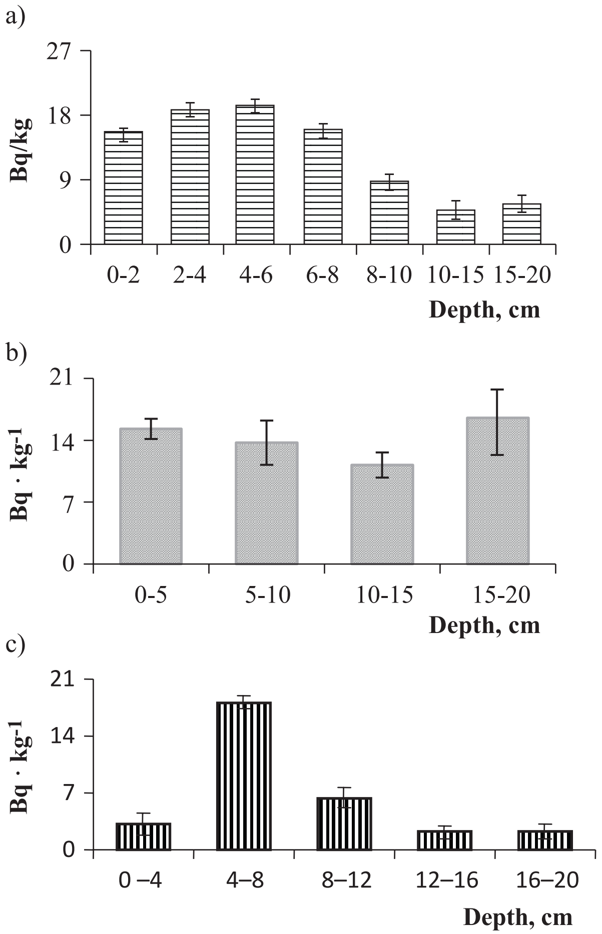

Fig. 3 The vertical profile of ^{90}Sr specific activity concentration in soils of different habitats: (**a**) the Neris Regional Park; (**b**) Ukmergė district; (**c**) district of the Ignalina nuclear power plant (Adopted from, Pliopaitė Bataitienė 2011; Alienčik 2011; Butkus et al. 2010)

of radionuclides will be of different types in forest area and the area with intensive agricultural activity.

As it was mentioned above, soil contamination by ^{90}Sr after ChNPP was not significant in Lithuania (increased by 1.1 times), so it can be suggested that the existing pollution by this radionuclide has happened during the tests of nuclear weapon. ^{90}Sr maximum specific activity concentrations determined are up to 8 cm in depth, and the decline is seen from 8 cm in the habitats with minimal human activities, i.e., in the forest (the Neris Regional Park), meadow (surroundings of the Ignalina nuclear power plant). ^{90}Sr specific activity is approximately constant in the soil up to the depth of 10–15 cm, but much lower ^{90}Sr specific activity concentration than the average in this area is determined in accordance with the results of the analysis at a depth of 5–10 cm in Ukmerge region with intensive horticulture and oreliculture activities. The results of the research reflect the processes of migration of radionuclides deeper in the soil in the areas, where human activity has not disturbed the nature processes or has changed them minimally; the same cannot be said about man-affected areas. Comparison of ^{90}Sr vertical distribution in the soil of the Neris Regional Park Paaliosės area and Ignalina NPP Stabatiškės shows that this radionuclide migrates vertically, and the highest specific activity concentration is set at soil depth of 4–8 cm.

The scientific literature mentions that main ways of the acces of radionuclides into the tree are through atmospheric falls, root system together with nutrients. However, in addition to the abovementioned ways of the acces of radionuclides, there are other mechanisms of redistribution of radionuclides in the tree. Radial mobility of radionuclides between the rings of the tree is one of them (i.e., radionuclide migrates from outer rings towards the central ring (the core). ^{90}Sr is less mobile in conifers than ^{137}Cs, but some sources consider it to be stable in conifers. Katayama et al. (2006) mention that Japanese scientists conducted an experiment, related to mobility of ^{90}Sr between annual rings of cedar during which strontium chloride solution was injected into a living tree. The profile of strontium distribution in the trunk was studied after 8 months. Strontium moves radially in cedrum sapwood, but it is almost completely stopped in wood. In cases, when the concentration of specific activity of radionuclides in tree rings where the yer of formation are earlier than the beginning of nuclear testing (1945 year) can be substantiated by the characteristic of radionuclide to migrate towards the core. The results of the research of radionuclides accumulation in trees (*Pinus sylvestris* L.) in Lithuania reflect the stages of radioactive environment contamination. Figure 4 shows few examples reflecting radioactive contamination from several Lithuanian habitats, such as the Neris Regional Park and Rukainiai. Both habitats are in Vilnius county; Scots pines and the predominant sandy loams were analyzed there.

Two periods of the change in specific ^{90}Sr activity concentration in the wood, when ^{90}Sr specific activity concentration is close to the average value of tree growth throughout the period or more (1949–1964, 1977–1996) can be distinguished. The concentration levels of higher specific activities in the first period are associated with nuclear testing. More pollution (secondary redistribution of radionuclide through the root system after nuclear tests, the redistribution of contamination caused by ChNPP accident in the tree through the root system) factors can be distinguished during the second period. ^{90}Sr specific activity concentrations in Scots pine tree rings in Rukainiai habitat are lower than in the Neris Regional Park. However, these results show similar change character also, i.e., the concentration increases of specific activity of this radionuclide can be attributed to the effects of nuclear testing; and at the tree ring formed during the year of Chernobyl NPP accident, though the higher than the average ^{90}Sr specific activity levels (0.6 ± 0.1) Bq kg^{-1} were determined, they are not the highest during the pine growth periods. The analysis of perrenial ^{90}Sr change in the trees leads to the statement that it is of cumulative character. ^{90}Sr is accumulated in the wood to a certain extent, and later the stabilization of the quantity of this radionuclide takes place, resulting in the

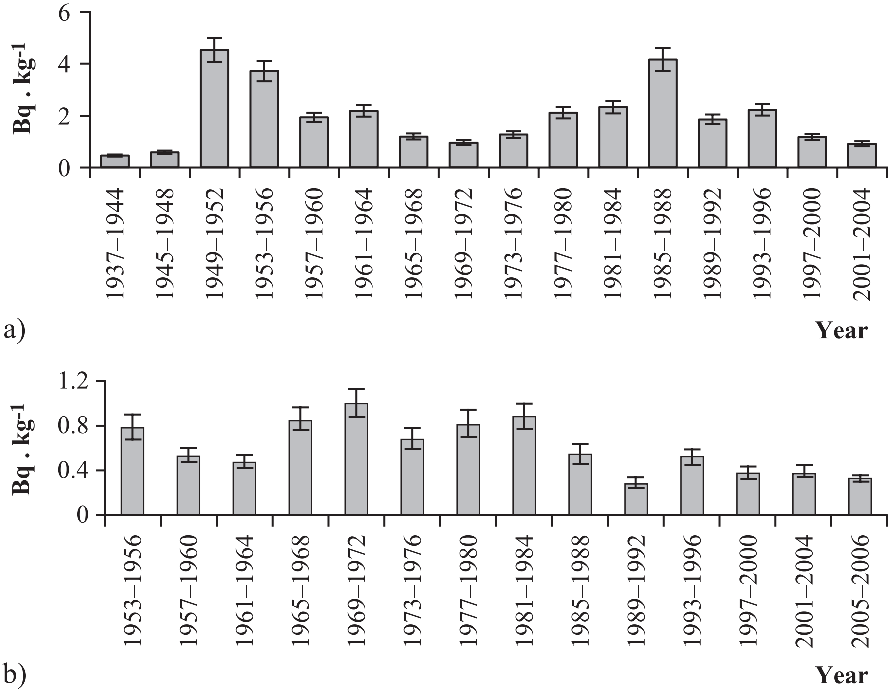

Fig. 4 ^{90}Sr specific activity concentration in the wood (**a**) the Neris Regional Park; (**b**) Rūkainiai habitats (Adopted from Pliopaitė Bataitienė 2011)

reduction of levels because of radioactive division and irreversible fixation in the soil. The minimum level of this radionuclide is seen in the first year after fallout during the perrenial change in ^{90}Sr levels in the wood. ^{90}Sr level is increasing next year. Such change of ^{90}Sr level is determined by the property of ^{90}Sr to accumulate in aging tissues; in addition, the mobility of this radionuclide is low in plants. The annual wood increase provides the perrenial dynamics of ^{90}Sr level with cumulative nature.

The accumulated ^{90}Sr amount in annual tree ring can provide the information about the radioactive environment contamination and its change. Forest plants could be one of the best monitoring research objects during the monitoring of radioactive environmental contamination. Richvanov (2005) suggests evaluating the contamination of retrospective radioactive fallout using fragment radiography in dendrochronological sequence of the tree. The former radioactive contamination can be determined according to accumulated ^{90}Sr amount in trees. For example, the conclusion that a change of environmental contamination can be recorded after the investigation of ^{90}Sr specific activities of cedar (Cryptomeria) in annual tree rings in Japan (Tsuki and Takao areas) can be done.

^{90}Sr in gymnosperm trees is of cumulative nature. The analyses have shown, however, that ^{90}Sr has radial mobility between the annual rings, but it is negligible.

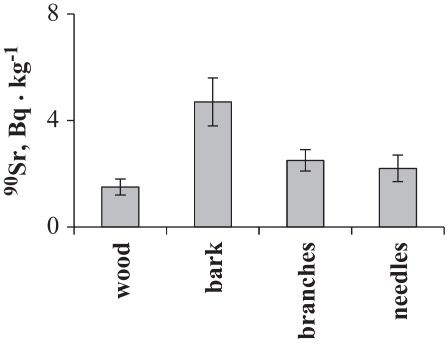

Fig. 5 ^{90}Sr distribution in tree components "the Neris Regional Park" (Adopted from Pliopaitė Bataitienė 2011)

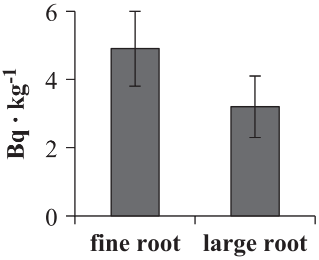

Fig. 6 Specific activity concentration of ^{90}Sr in fine and large Scots pine roots (Paaliosės habitat)

The conclusion that the research on the accumulation of this radionuclide in annual tree rings of gymnospermous plant is a great opportunity for reconstructive research of radioactive contamination. It was also argued that cumulative amount of ^{90}Sr in the annual tree rings show both local and global ^{90}Sr fallout. It is important to note that ^{90}Sr specific activity concentration in the wood highly depends on its amount in the soil (Kagawa et al. 2002; Malek et al. 2002).

Researchs on ^{90}Sr absorption in pine wood shows the influence of climate condition in habitats varies during different maturity periods of the plants. Therefore, it is likely that one factor cannot determine forest condition; it must be looked in complex manner, and the contamination must be evaluated by radionuclides, habitat climate conditions, and biological peculiarities of the plant.

Concentration of radionuclide specific activity (Bq kg^{-1}) in wood according to the literature data is slightly lower than in the needles, leaves, bark, or roots. Figure 5 illustrates trends of radionuclide distribution in tree components.

The lowest concentration of ^{90}Sr specific activity was determined in wood 1.5 ± 0.2 Bq kg^{-1}, needles 2.2 ± 0.5 Bq kg^{-1}, and branches 2.5 ± 0.4 Bq kg^{-1}. Different rate of the access of radionuclides into individual components of the tree, and different rate of natural purification processes of the tree in the literature is referred to as the basic reasons for the accumulation of radionuclids in different parts of the tree.

The biomass of tree root system on average contains 10% of ground wood biomass. However, the access of radionuclides into the tree through the root system is stronger than through aboveground parts of the tree, as the main stream of

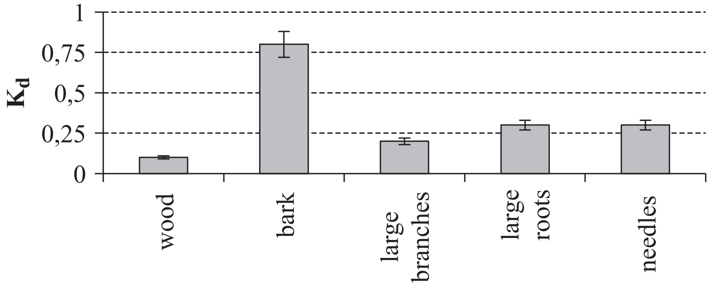

Fig. 7 ^{90}Sr discrimination factors in Common Scots pine components (Adopted from Pliopaitė Bataitienė 2011)

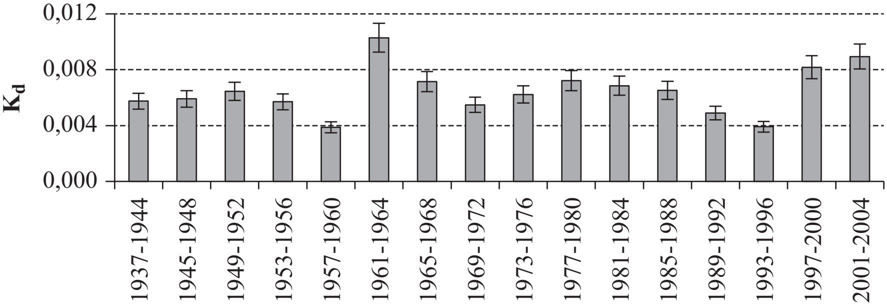

Fig. 8 ^{90}Sr multi-annual change of discrimination factor in common Scots pine wood

radionuclides moves together with nutrients. According to the data of scientific literature (Shcheglov and Tsvetnova 2004), the concentration of specific activity of radionuclides in wood is slightly lower than in the elements of root system. Figure 6 shows ^{90}Sr specific activity concentrations in fine (diameter to 4 mm) and large (diameter greater than 20 mm) roots of Common Scots pine tree.

Fine roots of Scots pine accumulate more radionuclides of artificial origin than larger roots, but the small mass in comparison with the large makes on average 6–10% of the root mass. Therefore, the greater importance for larger roots is provided when assessing the radioactive contamination of the tree, entering within the root system.

Figure 7 presents discrimination factors of ^{90}Sr**,** describing the peculiarities of access of artificial radionuclide (^{90}Sr) and its chemical analogues from the soil into the Common Scots pine (*Pinus sylvestris* L.) components (Pliopaitė Bataitienė and Butkus 2011).

Discrimination factor describes the access of radionuclide and its chemical analogue from the soil into the plant. The highest values of ^{90}Sr discrimination factors are established for Scots pine bark, needles, and roots, i.e., for those components of the tree, where more intense metabolism is taking part. For some components, bark and needles, with the exception of the access of radionuclides from the soil with nutrients, other way of access of radionuclides—from the atmosphere—is quite important, and therefore, perhaps, higher values of ^{90}Sr discrimination factor were obtained. Figure 8 provides ^{90}Sr multi-annual change of discrimination factor in Scots pine tree rings.

The change of ^{90}Sr discrimination factors is uniform, i.e., there are no significant increases in ^{90}Sr transfer from the soil to the wood, though higher than average ^{90}Sr discrimination factor values were determined in the periods of 1961–1968, 1973–1988, and 1997–2004. This can be explained by the fact that the contamination of nuclear explosions (in the periods of 1961–1968, 1973–1988) has reached the soil depth, where the large part of pine tree rootball biomass is located. The contamination caused by Chernobyl NPP accident in 1997–2004 was insignificant in Lithuanian territory and was higher only by 1.1 times than before the Chernobyl NPP accident has reached a depth of soil, where a large part of pine rootball biomass lies.

Comparing discrimination factors presented in (Butkus et al. 2014) ^{137}Cs with ^{90}Sr discrimination factors presented in Fig. 8, it can be seen that ^{137}Cs discrimination factors on average are by two orders higher than ^{90}Sr discrimination factors. The distribution of discrimination factors in annual tree rings located closer to the core is also different. ^{137}Cs discrimination factors increase towards the core and are by 1.2–1.9 times higher than average, and ^{90}Sr remains close to the average ($K_{\mathrm{d\,,\,vid}}$ = 0.006). Such distribution of discrimination factors in tree rings located closer to the core supports the idea that ^{90}Sr mobility in gymnosperm (including conifers) is so low that it can be considered stable in conifers.

Conclusion

The soil in Lithuania was contaminated by artificial radionuclides because of nuclear weapon test, ChNPP accident a small influence was felt because of the Fukushima nuclear power plant accident. The structure of spotty radionuclides on soil surface was determined; hot spots were detected there. Ionic diffusion, convective transfer together with the soil solution, and migration by plant root systems are the basic radionuclide migration processes in soil. The trees absorb the maximum quantity of radionuclides (up to 70%) through the tree root system. ^{90}Sr in most cases of global fallout is soluble in water and mobile form. ^{90}Sr takes part in plant metabolism as Ca analog when entered into the environment. Lithuanian forestland covers 33.3% of the country, the predominant species are conifers (scots pine occupies the largest forest area (56.2%)). Then this species of trees is more sensible for ionising radiation. Distribution of ^{90}Sr artificial radionuclide in the soil depth is uneven. The nature of this distribution was determined not only by the fundamental processes of radionuclide migration, radionuclide and soil characteristics, but also by human activities. By the investigation results are two periods of specific ^{90}Sr activity concentration change in the wood when ^{90}Sr specific activity concentration is close to the average tree growth throughout the period value or more can be distinguished. The concentration levels of higher specific activities in the first period are associated with nuclear testing. More pollution (secondary radionuclide redistribution through the root system after the nuclear tests, the contamination redistribution in the tree through the root system caused by ChNPP accident) factors can be excluded during the second period. The minimum quantity of this radionuclide in trees is seen in the first year after fallout during the perrenial ^{90}Sr amount change in the wood. ^{90}Sr amount is increasing in the next year. Such ^{90}Sr change of amount is determined by the property of ^{90}Sr to accumulate in aging tissues; in addition, the mobility of this radionuclide is low in plants. The distribution of discrimination factor in tree rings, located closer to the core, supports the idea that ^{90}Sr mobility in gymnosperm (including conifers) is so small that it can be considered stable in conifers.

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Contamination of Firewood Taken from the Exclusion Zone of the Chernobyl NPP by ^{90}Sr According to Data from 2005–2016

Viachaslau Zabrotski

Introduction

The Polessye State Radiation–Ecological Reserve (PSRER) began operating in the territory of the Belarusian sector of the exclusion zone of the Chernobyl Nuclear Power Plant (ChNPP) in September 1988. The primary goals of the reserve include performing radiobiological research and executing radiation–ecological monitoring of the exclusion zone. The PSRER is located in the south-east of Belarus and stretches from east to west for 70 km and from north to south for 48 km. At present, the area of the reserve is 2162 km^2.

The maps of contamination of the PSRER territory by ^{137}Cs, ^{90}Sr, ^{238}Pu, ^{239,240}Pu, and ^{241}Am were created only in 2009 and were due to the work of several Belarusian organizations. The levels of contamination of the reserve by ^{90}Sr are presented in Fig. 1. The purpose of the current work was to analyze the analytical data accumulated during radiation examination of the forest in the territory of the Belarusian part of the Chernobyl exclusion zone in the period from 2003 to 2016, with a discussion and generalization.

Object and Methods of Research

One of the main tasks of the laboratory of spectrometry and radiochemistry at the PSRER is radiation examination of the forests. The examination is carried out in accordance with the relevant normative document (TKP 239-2010).

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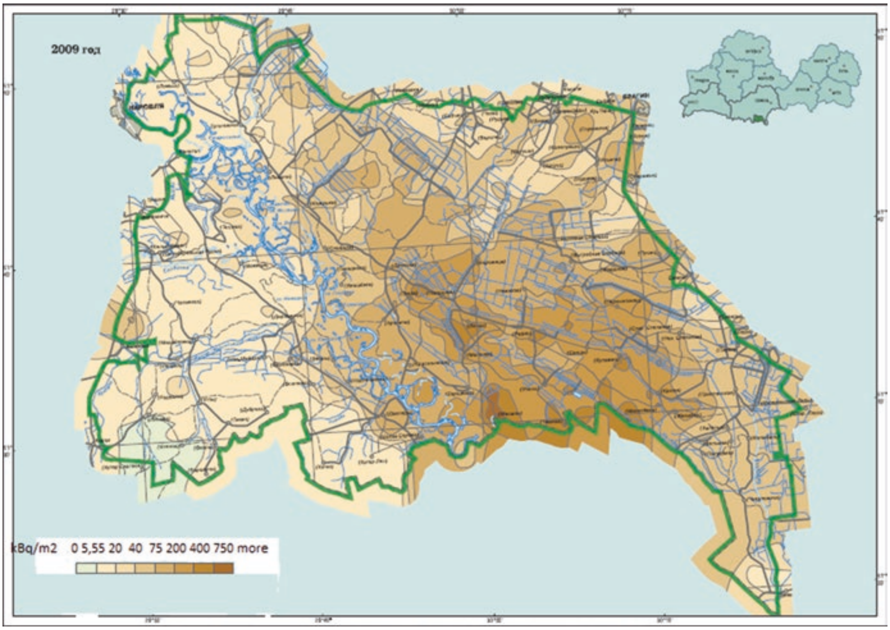

Fig. 1 Contamination (expressed in kBq m^{-2}) of the PSRER territory by ^{90}Sr in 2009 (Adapted from Izrael and Bogdevich 2009)

The following equipment is used in radiation examination:

- A dosimeter with a lower measurement limit of 0.1 μSv h^{-1} and less
- A soil sampler with an internal core diameter of 4 cm and depth of 20 cm
- A portable chainsaw for wood sampling

Verifying the Homogeneity of the Test Sites on Some Factors and Test Sites Selection

To be sure the density of the radioactive contamination of the forest site is quite uniform, the dose rate is measured at several control points. The test site is regarded as homogeneous if the difference between the maximal (minimal) and mean dose rate values is not more than 50%. If it is concluded that the site is inhomogeneous, it is divided into the minimal number of smaller homogeneous sites and the following sampling of the wood and soil is performed in each of them.

Soil Sampling

A rectangular area measuring approximately (30–50) × (30–50) m is selected for soil sampling in the part of the site characterized by the maximal dose rate. In each corner of that quadrangle, soil (including litter) to a depth of 20 cm is taken. The dose rate is registered in the same places at a height of 1 m from the soil surface. According to the data received, the values of dose rate changed from 0.1 and 11.1 μSv h^{-1} are, correspondingly, the minimal and maximal value of dose rate measured in examination of about 4000 test sites in the period 2003–2016.

Wood Sampling

It is usual to distinguish two types of firewood in Belarus, depending on its origin:

1. Fuelwood that is the residue of merchantable wood (the spires of trees). We can identify this as "fuelwood from timber."
2. Fuelwood that is obtained by cutting trees that are inapplicable for production of merchantable wood. We can identify this as "fuelwood from cull."

Three test trees from each species are cut on the test site for the wood sampling. The wood sampling from the timber trees is fulfilled near the stem base (butt) and at the upper point of the log. In the case of firewood obtained from the top of a timber tree, the wood is sampled from the middle of the spire (crown).

A saw cut through the whole diameter of the trunk is made using a portable chainsaw. The resulting sawdust is collected on polyethylene film. The samples taken from the three test trees are combined separately for the timber and firewood parts of the tree.

In total, for the period of 2003–2016, about 4000 test sites with a total area of more than 140 km^2 were examined. The area of each site varied in a range from several thousand to several hundred thousand square meters.

According to the relevant normative documents adopted in Belarus (RDU/LH-2001), the content of only one radionuclide, ^{137}Cs, should be controlled in the wood used in the country. Instrumental determination of ^{137}Cs was carried out with the help of an AT1315 gamma–beta radiation spectrometer (Atomtex, Belarus). In that device, a detection block based on an NaI(Tl) scintillation crystal with a height and diameter equal to 63 mm is used to register gamma radiation. An organic scintillator on a base of polystyrene with a height and diameter equal to 9 mm and 128 mm, respectively, is used to register beta radiation. In spite of the high limit of determination of ^{90}Sr (20 Bq kg^{-1}), this device is suitable for use and is widely applied in the laboratory in the analysis of environmental samples contaminated by Chernobyl fallout. Beginning in 2005, apart from the content of ^{137}Cs in the samples that were analyzed, the content of ^{90}Sr also was registered. By the end of 2016, the total number of analyzed samples of firewood amounted to about 8000. The main species were Scotch pine (*Pinus sylvestris*)—about 60% of all samples, birch

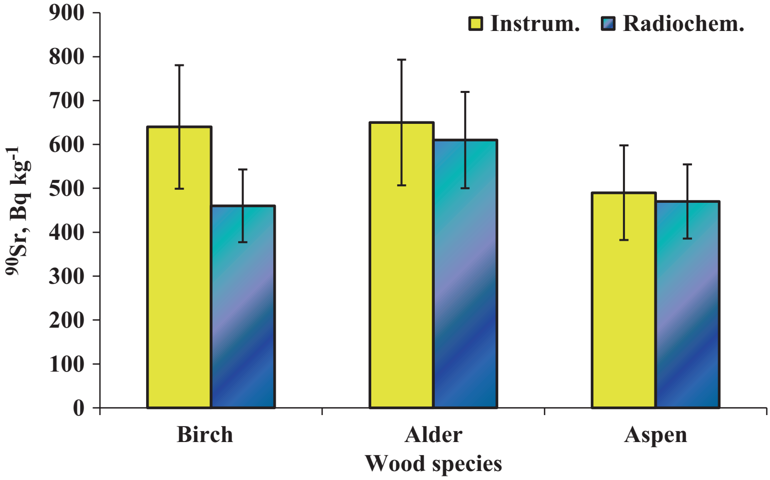

Fig. 2 Comparison of the results of instrumental and radiochemical determinations of ^{90}Sr in wood (Adapted from Zabrotski 2013)

(*Betula*)—22%, and alder (*Alnus glutinosa*)—8%, as well as aspen *(Populus tremula)*, oak (*Quercus*), etc. It should be emphasized that we did not control for any factors (conditions of forest growth, contamination density, soil type, etc.) influencing the transfer of the radionuclides from soil to trees. Nevertheless, taking into account certain peculiarities of the obtained data, such as the long-term character and absence of the influence of man in natural processes since 1986, consideration of the data received could provide new fundamental knowledge.

Special experiments were conducted to ensure the reliability of the instrumental radioanalytical data on the content of ^{90}Sr in the wood. Directly after instrumental determination of the activity concentration of ^{90}Sr in the wood, these samples were burned and the resulting ash was analyzed by a radiochemical method. At the final stage of that procedure, the activity of the precipitate of ^{90}Y oxalate was measured by an LB 4100 gas flow proportional counter (Canberra). The chemical recovery of the strontium was determined by an atomic absorption spectrometer (Solaar S2), and the chemical recovery of the yttrium was determined gravimetrically. The data obtained are presented in Fig. 2. The fact that the differences between the instrumental and radiochemical data are lesser than the experimental error supports the reliability of the results obtained.

Contamination of the Firewood in the Exclusion Zone of the Chernobyl NPP by ^{90}Sr

The distributions of the samples of firewood with ^{137}Cs and ^{90}Sr activity concentration values are presented in Fig. 3a. According to those data, the activity concentration of ^{90}Sr in the wood varies by two orders of magnitude. The density contamination

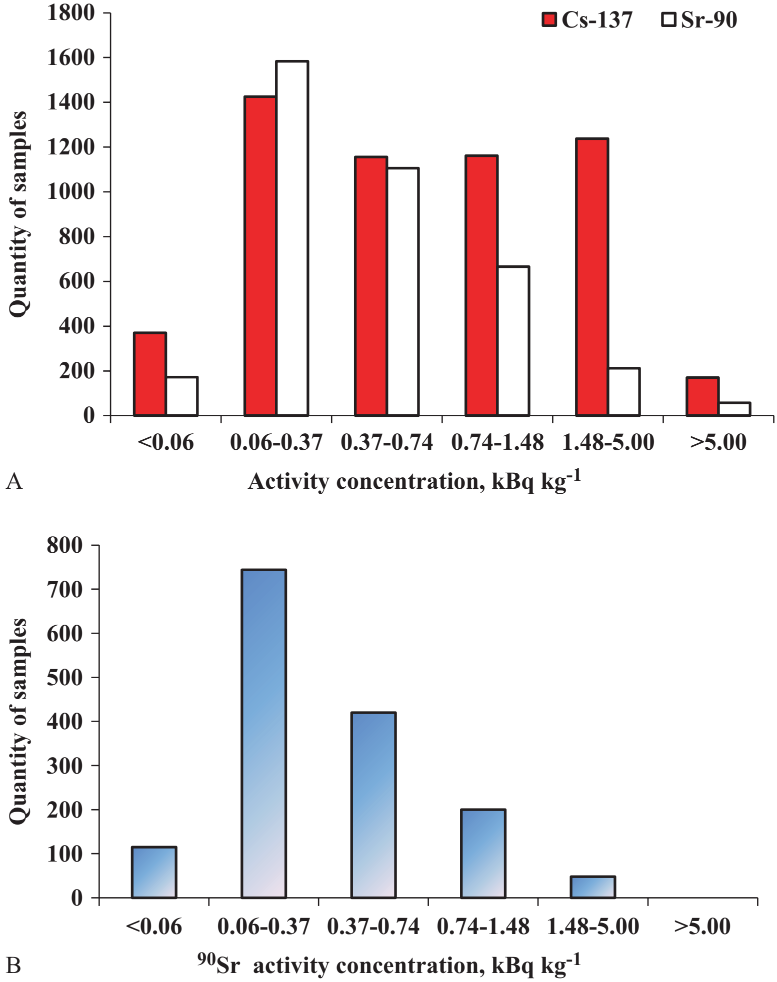

Fig. 3 Distribution of samples of firewood (pine, birch, alder, aspen, oak, etc.) with activity concentrations of ^{137}Cs and ^{90}Sr: **a** without limitations in the ^{137}Cs activity concentration, and **b** in samples with a ^{137}Cs activity concentration of less than 740 Bq kg^{-1}

by ^{90}Sr in the sampling territory varies by the same limits (i.e., by the same number of times: from 10 to 1000 kBq m^{-2}) (Fig. 4).

According to Fig. 3a, the distribution of samples of firewood with activity concentrations of ^{137}Cs do not differ very much from the distribution of ^{90}Sr. However, in the case of some specific samples, the high (low) levels of ^{137}Cs content do not always coincide with the high (low) levels of ^{90}Sr content. The data in Fig. 3b, which presents the content of ^{90}Sr in firewood with a ^{137}Cs activity concentration of less than 740 Bq kg^{-1}, as well as the variation in the ^{137}Cs/^{90}Sr ratio in the same wood samples (Fig. 5), support this suggestion. In analysis of the data in Fig. 3b, we

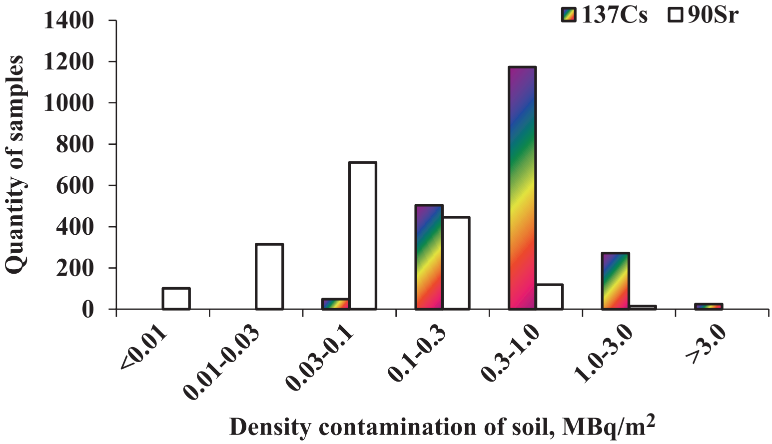

Fig. 4 Distribution of sampling sites with density contamination by ^{137}Cs (2003–2015) and ^{90}Sr (2005–2016)

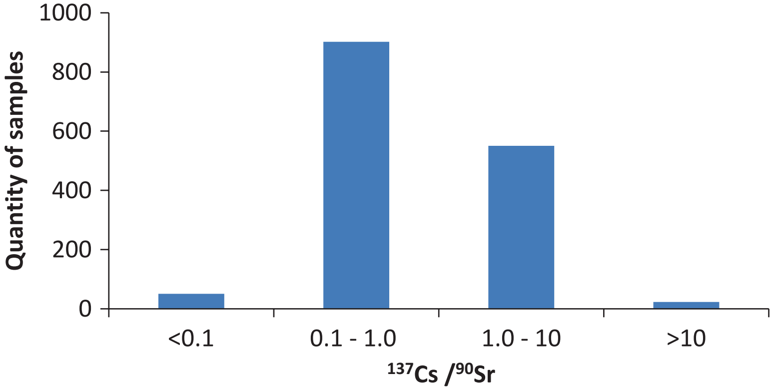

Fig. 5 ^{137}Cs/^{90}Sr ratio in firewood samples from all species in 2005–2016

should remember that 740 Bq kg^{-1} is the permissible level of ^{137}Cs in firewood, while the content of ^{90}Sr in wood is not controlled in Belarus. In other words, all of the wood—the results of analysis of which are presented in Fig. 3b—could be used as firewood in accordance with the normative document (RDU/LH-2001). The probability of finding ^{90}Sr in firewood in a concentration higher than, for example, 740 Bq kg^{-1} could be evaluated using the percentage of such samples in the total number of samples analyzed, i.e., 16%. Luckily, the probability of finding firewood with a higher concentration of ^{90}Sr is low—for 1480 Bq kg^{-1}, it amount to little more than 3%.

Uptake of ^{137}Cs and ^{90}Sr by the wood could be regarded as two independent processes that do not correlate with each other. Such a conclusion could be drawn on the basis of the data presented in Fig. 3a, b. It can be seen that the form of the curve describing the distribution of the firewood samples with activity concentrations of ^{90}Sr does not depend on the activity concentrations of the other radionuclide, ^{137}Cs. The form of the curve is the same both for the wide interval of ^{137}Cs content from 60 to 5000 Bq kg^{-1} (Fig. 3a) and for the substantially more restricted interval of less than 740 Bq kg^{-1} (Fig. 3b). It is worth noting that more than half of the samples analyzed fall within this more restricted interval of ^{137}Cs content.

After the Chernobyl accident, forests were contaminated primarily as a result of both dry and wet (i.e., rain) deposition. Tree canopies are efficient filters of atmospheric pollutants of all kinds. The primary mechanism of tree contamination after the Chernobyl accident was direct interception of radionuclides by the tree canopy, which effectively intercepted the major part of the initial deposition (Report of the Chernobyl Forum 2006). At that stage, absorption of radioactivity by leaf surfaces also occurred, although this was difficult to measure directly. Within roughly 1 year, the bulk of the radionuclides were transferred from the tree canopy to the underlying soil, which became the major repository of radioactivity within the forests. Subsequently, the trees became contaminated because of root uptake, which has continued as radionuclides have migrated into the soil profile.

So, there were two ways of radionuclide uptake by trees—through the tree canopy and through the roots. Another way was found in recent investigations of the forest contaminated after the Fukushima nuclear accident. The authors of that work (Mahara et al. 2014) showed that radionuclides absorbed through tree bark could be directly transported into sapwood. As the result, the radionuclide concentration in tree rings depends on the directional abundance of radionuclides deposited on the bark surface.

Different approaches to quantitative description of the transfer of radionuclides to biota are presented in the literature (IAEA-TECDOC-1616 2009, TRS 472 2010, TRS 479 2014). According to the International Atomic Energy Agency (IAEA) (IAEA-TECDOC-1616 2009), the aggregated transfer factor (T_{ag})—defined as the ratio of the radionuclide activity concentration in a plant (Bq kg^{-1}) divided by the total deposition on the soil (Bq m^{-2})—has the following advantage. There may be less variation in the case of transfer from the soil because the effects of differences in the soil bulk density between organic and mineral soils may be overcome. The T_{ag} concept has been adopted as a reasonable empirical measure to normalize radionuclide accumulation in forests and wood regardless of variations in the vertical radionuclide distribution and availability in the soil profile.

On the other hand, the authors of a more recent document (TRS 479 2014) prefer to use the concentration ratio (CR) to describe the transfer from media to wildlife organisms. This approach is justified because of:

1. Its simplicity, transparency, and user friendliness
2. The relatively large amount of relevant information available for organisms, elements, and ecosystems in comparison with other methods for quantifying the transfer

3. The common use of this parameter in the existing environmental exposure assessment models
4. Its compatibility with tools used for human and non-human biota exposure assessments

T_{ag} values were used in our work; when it was necessary, we transformed them into CR values, using the data on the activity concentration of radionuclides in the 20 cm soil layer.

The basic features of the seasonal and long-term dynamics of ^{137}Cs and ^{90}Sr in wood were examined by Shcheglov and Tsvetnova (Shcheglov and Tsvetnova 2004), who concluded that the content of ^{137}Cs in the canopy of forest ecosystems is decreased from spring to autumn. Such a character of the dynamics is accounted for by the dependence of the ^{137}Cs concentration—as well as the concentration of its chemical analog potassium—on the dynamics of the intensity of growth processes in plants. It is known that maximal quantities of potassium (and ^{137}Cs) are present in young growing tissues, characterized by a high level of metabolism—meristem, cambium, etc. This promotes an increase in the water-retaining ability and permeability of the cellular protoplasm, being highly mobile and easily recovered (Polevoy 1989). For that reason, the maximal content of potassium and ^{137}Cs in the plants occurs in spring and the minimal content occurs in autumn, when growth processes fade out and the water-retaining ability of the plants is decreased.

In contrast to ^{137}Cs, the seasonal dynamics of ^{90}Sr concentration in the assimilating organs of wood species is characterized by an increase from spring to autumn. This is because ^{90}Sr and its non-isotopic analog calcium are accumulated in aging organs and tissues. In the aging of cells and the decrease in their physiological activity, these elements are accumulated as insoluble salts of organic acids, making transport of calcium and ^{90}Sr and their reuse difficult. In addition, because of their concentrations in aging tissues and organs, calcium and ^{90}Sr regulate water content in vegetation and result in it decreasing at the end of that period (spring to autumn) (Polevoy 1989).

The most interesting data for the public are the long-term variations in the accumulation of radionuclides by wood, because it is the main forestry product. The most integrated approach existing between research studies concerns the long-term dynamics of ^{90}Sr. It has been shown both by model experiments and by the main observations made after the Kyshtym and Chernobyl accidents that the long-term dynamics of the content of ^{90}Sr in wood are characterized as cumulative (until a certain value), followed by some relative stabilization of the content of that radionuclide, with further lowering because of radioactive decay and non-exchangeable fixation by the soil (Alexakhin and Naryshkin 1977; Tihomirov 1976). According to Shcheglov and Tsvetnova (2004), the minimal content of ^{90}Sr in wood is registered in the first year after the radioactive deposition, with subsequent growth in that indicator. In other words, a cumulative effect of ^{90}Sr takes place from the first year onward after the depositions and is clearly pronounced. As was stated earlier, this is due to accumulation of the radionuclide by aging tissues and organs, as well as its low mobility in plants and minimal reuse.

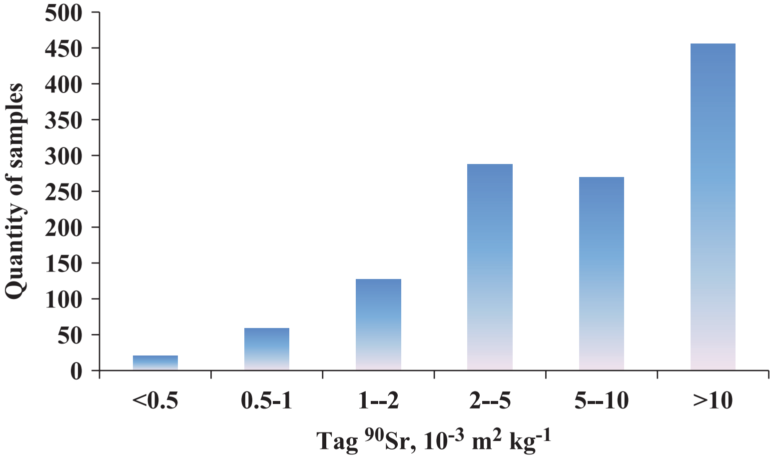

Fig. 6 Distribution of T_{ag} values for ^{90}Sr in firewood samples from all species (2005–2016)

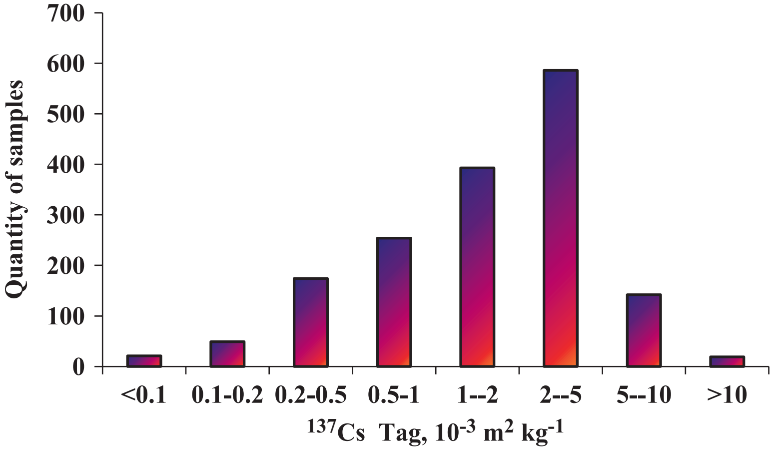

Fig. 7 Distribution of T_{ag} values for ^{137}Cs in pine firewood samples (2003–2015)

The data presented in Fig. 6 support the suggestion about the cumulative character of uptake of ^{90}Sr by the wood. Comparison of that figure with Fig. 7 emphasizes the difference between the mechanisms of accumulation of ^{137}Cs and ^{90}Sr by the wood. The fact that the largest number of samples of firewood belong to the rank with the highest content of ^{90}Sr could be regarded as being due to both a continuation of the process of uptake of that radionuclide and the cumulative character of that absorption, i.e., absence of elimination of ^{90}Sr from the plant (Shcheglov and Tsvetnova 2004).

There is no such unanimous position among different authors regarding the long-term dynamics of ^{137}Cs in wood. Some of them suggest that the long-term dynamics of ^{137}Cs are close to those of ^{90}Sr (Avila et al. 1999). Others suppose that the long-term dynamics of ^{137}Cs differ from those of ^{90}Sr and have a two-stage character. According to Shcheglov and Tsvetnova (Shcheglov and Tsvetnova 2004), the long-term dynamics of ^{137}Cs in wood vary from those of ^{90}Sr and are closely connected with the landscape features in the place of plant growth.

For treatment of our experimental data, methods of statistical analyses (Ellison et al. 2009) were used. They allowed us to decide whether the two independent sets of data are really different. The following data sets were compared:

1. Set of T_{ag} ^{90}Sr for pine firewood and set of T_{ag} ^{90}Sr for firewood from other wood species

Mean 1	Mean 2	s1	s2	n1	n2	t_{crit}	t_{calc}
0.0169	0.0075	0.0352	0.0116	1034	186	1.96	3.63

The calculated value (3.63) exceeds the critical value (1.96), so the null hypothesis is rejected, and a conclusion could be drawn about the significant difference between the means of the two sets of data.

2. Set of T_{ag} ^{90}Sr for pine fuelwood from cull and set of T_{ag} ^{90}Sr for pine firewood from timber (in the first case, we have the total tree, but in the second case, we have only its spire)

Mean 1	Mean 2	s1	s2	n1	n2	t_{crit}	t_{calc}
0.0169	0.0155	0.0352	0.047	1034	812	1.96	0.75

The calculated value (0.75) is less than the critical value (1.96), so the null hypothesis is accepted, and the two sets belong to the same general data collection.

3. Set of T_{ag} ^{90}Sr for pine fuelwood from cull and set of T_{ag} ^{137}Cs for pine fuelwood from cull (every one of the 1034 samples was analyzed for ^{90}Sr and ^{137}Cs content)

Mean 1	Mean 2	s1	s2	n1	n2	t_{crit}	t_{calc}
0.0169	0.0021	0.0352	0.0023	1034	1034	1.96	13.49

The calculated value (13.49) exceeds the two-tailed critical value of 1.96, so the null hypothesis is rejected, and a conclusion could be drawn about a significant difference between the mean T_{ag} ^{90}Sr and T_{ag} ^{137}Cs values for pine fuelwood.

The results for control of ^{90}Sr content in firewood produced in the reserve in the period of 2005–2016 are presented in Fig. 8. The number of samples analyzed annually varied from 30 to 200. To evaluate the spread of the aggregated transfer factor values for ^{90}Sr relative to the mean year transfer factor, coefficients of variation (relative standard deviations) were calculated. In most cases, their values exceeded 100%, pointing out the substantial spread of values inside the annual set. Nevertheless, the statement about increasing transfer factors for ^{90}Sr up to 2012 (or

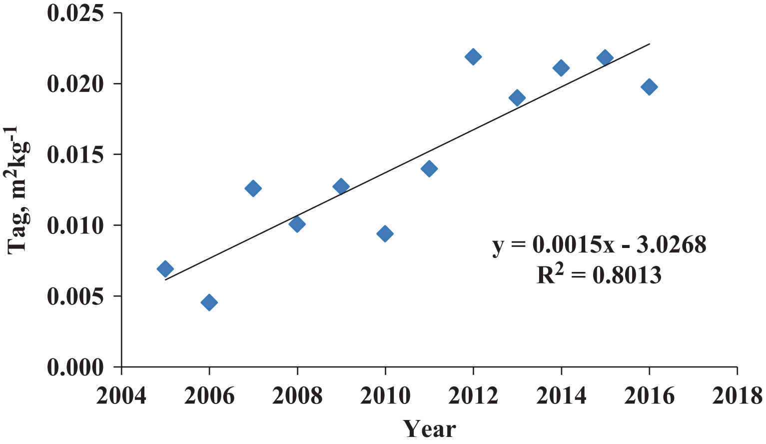

Fig. 8 Dynamics of ^{90}Sr transfer into pine firewood in the exclusion zone of the ChNPP

Table 1 Comparison of our data on ^{90}Sr uptake by wood taken from the forest in the exclusion zone with previously published data

T_{ag}, m^2 kg^{-1}		Concentration ratio	
IAEA (TRS 472 2010)	Our data	IAEA (TRS 479 2014)	Our data
4.7×10^{-4} to 1.0×10^{-2}	2×10^{-4} to 2.6×10^{-1}	1.2×10^{-3} to 5.3×10^{0}	4.4×10^{-2} to 45.1

at least attainment of a plateau since 2012) seems to be well grounded. The dependence presented in Fig. 8 corresponds to the rate of weathering of fuel particles of Chernobyl origin. It is known that no less than 90% of the ^{90}Sr laid down in the territory of the exclusion zone was included in the matrix of fuel particles (Report of the Chernobyl Forum 2006). Just the process of destruction of fuel particles explains the accumulation of ^{90}Sr by vegetation in the exclusion zone of the ChNPP (Kashparov et al. 2004). As a result of the weathering of fuel particles, ^{90}Sr goes from a fixed form in the matrix of fuel particles to a mobile form available to be consumed by plants. There are numerous experimental pieces of evidence that the content of ^{90}Sr in wood in the exclusion zone exceeds that of ^{137}Cs by ten times (Kashparov et al. 2004; Zabrotski 2013).

According to the source (TRS 472 2010), the T_{ag} values for ^{90}Sr in individual species vary practically by two orders of magnitude (Table 1). Possibly because of this large variation, there are no data on specific species in the newer IAEA document (TRS 479 2014), which contains only broader information concerning two groups of trees—broadleaf and coniferous.

According to the data of Table 1 the transfer of ^{90}Sr to the wood of trees growing in exclusion zone is characterized by higher values of T_{ag} and CR than in case of earlier published data. That could be due to accumulation of ^{90}Sr in the wood during the long period since the Chernobyl accident. According to the results obtained in

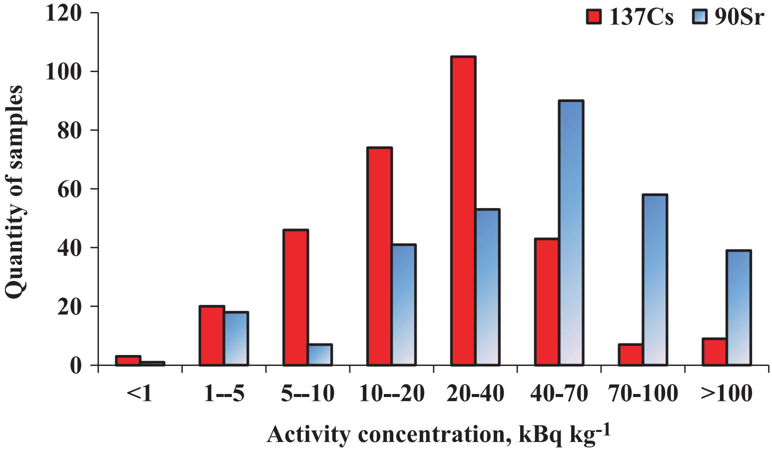

Fig. 9 Distribution of activity concentrations of ^{137}Cs and ^{90}Sr in stove ash samples. The samples were taken in the territory of the reserve in the period from 2006 to 2016

research studies connected with the Kyshtym accident (IAEA-TECDOC-1616 2009), T_{ag} values achieve a plateau 10–15 years after the accident. In the territories contaminated after the Chernobyl accident, that time interval would be substantially increased because additional time is needed for preliminary destruction of fuel particles containing the major part of ^{90}Sr.

The Danger of Probable Damage to Health Through Use of Firewood Contaminated by ^{90}Sr

A substantial part of the living space and utility rooms both on the periphery of the reserve and near its borders are heated by the burning of firewood. The high content of radionuclides in the wood leads to the following problems with treatment of the ash formed by its burning.

The importance of the problem of normalization of the radionuclide content in wood ash is emphasized by the results of an analysis of furnace ash conducted in the territory of the reserve (Fig. 9). According to the data in that figure, about 80% of the ash samples have an activity concentration of ^{137}Cs higher than its exemption limit (10^4 Bq kg^{-1}). For ^{90}Sr, the corresponding value is 10^5 Bq kg^{-1}. Therefore, the values seen in 80% of the samples analyzed refer to radioactive waste in terms of their ^{137}Cs concentrations, and the values in 13% refer to their ^{90}Sr concentrations.

In 54 of more than 300 samples, the results of analysis of which are presented in Fig. 9, ^{241}Am was registered in concentrations from 2 to 170 Bq kg^{-1}. Taking into account the substantially lower transfer coefficients of ^{241}Am into plants in comparison with ^{137}Cs and especially ^{90}Sr (TRS 472 2010), it seems unlikely that ^{241}Am gets

Table 2 Enrichment of stove ash with ^{90}Sr

Ratio ^{137}Cs/^{90}Sr			
In nuclear fuel from the ChNPP (Kuriny et al. 1993; Kashparov et al. 2003)	In soil in the territory of the reserve (20 cm)	Wood	Stove ash
1.17	1.2–108	0.01–1 (40% of samples) 1–119 (60% of samples)	0.05–1 (85% of samples) 1–7.6 (15% of samples)

into ash through the "soil–tree–ash" chain. A more probable reason is contamination of the external surface of the trunk by soil in timber-harvesting operations.

The data pointing out alterations in the ^{137}Cs/^{90}Sr isotopic ratio in movement through the "reactor–soil–firewood–stove ash" chain are presented in Table 2.

About 33% of the ^{137}Cs contained in the nuclear reactor of the ChNPP and only 1.5% of the ^{90}Sr were thrown out into the environment in the Chernobyl accident. The inhomogeneous character of the radioactive fallout explains the variability in the ^{137}Cs/^{90}Sr isotopic ratio (from 1.2 to 108) in the territory of the exclusion zone. Because of selective uptake of ^{137}Cs by soil and higher absorption of ^{90}Sr by trees, up to 40% of wood samples have a ^{137}Cs/^{90}Sr ratio of less than 1. The greater change in the ratio between ^{137}Cs and ^{90}Sr observable in stove ash could be explained by the higher volatility of cesium in burning of firewood.

Handling of the Ash

During the heating season (October–April), the firewood produced in the reserve is used for the heating of utility rooms and living space in the territory of the exclusion zone. The formation of stove ash presents a problem for its handling. One of the possible scenarios that has been discussed to handle the ash is use of it as an additive to concrete in construction work. According to the Finnish Radiation and Nuclear Safety Authority (STUK 2003), to be sure that the additional exposure dose is not higher than 0.1 mSv per year, the activity concentration of the ^{137}Cs in the ash should be less than 1000 Bq g^{-1} and the portion of ash in the concrete should be 120 kg m^{-3}. If the portion of ash is lesser, its activity concentration may be greater.

One possible way of treating the ash is mounding it. In that case, to safeguard against inhalation of radioactive substances, the ash mounds should always be covered by a material layer that prevents dust from rising.

Another possible way of handling the ash is to return it to the forests contaminated by the Chernobyl radionuclides. In that case, apart from solving the problem of handling the radioactive ash, additional fertilization of the forest would be achieved.

The countries contaminated after the Chernobyl accident restrict the use of firewood in contaminated territories (Report of the Chernobyl Forum 2006), but their approaches are different. In particular, Russia and Ukraine suggest it is necessary to

Table 3 Permissible activity concentrations of ^{137}Cs and ^{90}Sr in firewood in different countries contaminated after the Chernobyl accident

	^{137}Cs, kg^{-1}	^{90}Sr, Bq kg^{-1}
Russia	1400	370
Ukraine	600	60
Belarus	740	No control

control radioactive contamination of firewood by both ^{137}Cs and ^{90}Sr, but there is no control of ^{90}Sr in firewood in Belarus (Table 3).

To evaluate the permissible level of ^{90}Sr in firewood, we used the approach stated in the relevant IAEA document (IAEA-TECDOC-1376 2003). It was suggested that wood ash is traditionally used as a fertilizer in vegetable gardens. In that case, part of the radionuclides can transfer from the ash to the growing plants. Consumption of those plants as food can increase the dose of internal exposure of the person. Knowledge of the dose limit for internal exposure to ^{90}Sr and use of the calculation methodology recommended by the IAEA (IAEA-TECDOC-1376 2003) allows us to obtain the value of the permissible level of ^{90}Sr content in firewood.

The following formula is used for calculation of the annual effective dose for internal exposure (E_{ing}, mSv) in consumption of vegetables contaminated by ^{90}Sr.

$$E_{\text{ing}} = C_{\text{w}} \times \text{CF}_{\text{ash}} \times (M_{\text{ash}} / M_{\text{s}}) \times \text{TF} \times M_{\text{v}} \times \text{DG},$$

where:

C_{w} is the content of ^{90}Sr in used firewood, expressed in Bq kg^{-1}.

CF_{ash} is the coefficient of the concentration of radionuclides in the burning of firewood (dimensionless); according to IAEA recommendations, $\text{CF}_{\text{ash}} = 50$.

M_{ash} is the amount of ash used annually for fertilization of 1 m^2 of kitchen garden; $M_{\text{ash}} = 1$ kg by default (IAEA-TECDOC-1376 2003).

M_{s} is the amount of arable soil in 1 m^2 with a layer thickness of 15 cm and soil density of 1500 kg m^{-3}; $M_{\text{s}} = 225$ kg (IAEA-TECDOC-1376 2003).

TF is the soil-to-plant transfer factor for ^{90}Sr (TF = 0.2); it varies from 0.02 to 3.0 for peas, root crops, and green vegetables (IAEA-TECDOC-1376 2003).

M_{v} is the annual consumption of vegetables taken from a kitchen garden, varying from 20 to 300 kg; we presume $M_{\text{v}} = 100$ kg.

DG is the internal dose coefficient for ingestion; DG = 8×10^{-5} mSv Bq^{-1} (IAEA-TECDOC-1376 2003).

Suggesting that uptake of ^{90}Sr through vegetables from a kitchen garden is not the only way ^{90}Sr enters an organism, we have decreased that value by two times. After that, our task would be to determine the value of C_{w} (denoted as $\text{PL}_{\text{Sr-90}}$) at which E_{ing} would be equal to 0.04 mSv. In other words, the following equation should be resolved:

$$0.04 \text{ mSv} = \text{PL}_{\text{Sr}-90} \times 50 \times (1 \text{ kg} / 225 \text{ kg}) \times 0.2 \times 100 \text{ kg} \times 8 \times 10^{-5} \text{ mSv Bq}^{-1}.$$

The result is that $PL_{Sr\text{-}90}=113$ Bq kg^{-1}.

Taking into account the wide interval of possible TF values (0.02–3.0), it could be supposed that the spread of possible $PL_{Sr\text{-}90}$ values varies from 1/10 × 113 to 10 × 113. Mention could be made that the calculated $PL_{Sr\text{-}90}$ value is intermediate between the Ukrainian (60 Bq kg^{-1}) and Russian (370 Bq kg^{-1}) permissible levels of ^{90}Sr content in firewood.

Conclusions

The current use of firewood taken in the exclusion zone amounts to formation of a large amount of solid radioactive waste, which demands special handling. It is still endurable in the territory of the exclusion zone itself, but beyond the reserve, the economic profit from use of contaminated firewood is not clear. It seems reasonable to institute some temporary norms—without any financial costs—to limit the permissible level of ^{90}Sr in firewood in the territory of the reserve. Significant differences between ^{137}Cs transfer factors and ^{90}Sr transfer factors in pine wood have been shown by use of statistical analysis. Similar results were achieved in comparison of ^{90}Sr transfer factors in pine wood and ^{90}Sr transfer factors in the wood of other species. The difference between ^{90}Sr uptake by wood and that of ^{137}Cs was demonstrated using analysis of a large number of wood samples (more than 2000). The absorbance of ^{90}Sr has a cumulative character, which has been experimentally supported by dominance of the fraction of samples with a maximal activity concentration of ^{90}Sr.

Comparison of the transfer factors for ^{90}Sr in wood obtained by us with already published IAEA data shows that our values are higher. The time necessary to achieve a plateau in transfer factor values is also greater. This could be due to certain peculiarities of the Chernobyl accident after which the exclusion zone was contaminated by fuel particles. For that reason, additional time is needed for destruction of fuel particles containing the major part of ^{90}Sr and their transformation into mobile forms.

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The Behavior of ^{90}Sr in Macrophytes Inhibiting Water Reservoirs in the Belarussian Sector of the Chernobyl NPP Exclusion Zone

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Introduction

As a result of the accident at the Chernobyl nuclear power plant, thousands of water bodies of various types were affected by the disaster. The ecosystems of the basins of the Dnieper, Sozh, and Pripyat rivers became most polluted in Belarus (Konoplya and Rolevich 1998).

At present, the water bodies in the Belarusian sector of the exclusion zone of Chernobyl accident contain the highest concentrations of radioisotopes. Closed and weakly flowing reservoirs are the most contaminated ones. The components of the river ecosystems are characterized by a lower activity of radioisotopes owing to the processes of natural self-purification of bottom sediments. Since several years have passed after the accident, the bottom sediments of these reservoirs no more serve as the secondary source of contamination to the water masses and biota. In this context, floods and periods of spring floods have an important place. As of today, the main flow of radioisotopes into river systems comes from the flushing of catchment areas and the inflow from more polluted water bodies.

Unlike river systems, the accumulation of radioisotopes by hydrobionts in closed water bodies is affected by the increasing activity of radioisotopes; these radioisotopes are retained in the ecosystem and deposited in the bottom sediments, leading to cyclicity and secondary contamination of water masses and an increase in the radioisotopes specific activity in the aquatic ecosystem components. Radioactive isotopes can accumulate in varying degrees in different aquatic ecosystem

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components, creating different radiation conditions for each ecological group of the aquatic biota. Autorehabilitation of closed reservoirs in the exclusion zone is extremely slow; therefore, the ecosystems of most of the lakes, oxbows, and backwaters are still characterized by a high level of radioactive contamination (Pavlutin and Makarevich 1998; Gudkov et al. 2009; Kazakov and Utkin 2009; Vakulovskij et al. 2009; Volkova et al. 2009).

Radioactive contamination of water ecosystems in and around the territories affected by the Chernobyl accident depends on the several factors such as the type of water body, pollution density of the water and catchment areas, features of landscape elements, physical and chemical properties of radioisotopes, and biological features of investigated objects (Worner and Harrison 1999; Levina 2008; Tryapitsyna 2011). The activity concentration of some radioisotopes in hydrobionts can be thousand times higher in water due to their ability of selectively accumulating in some components of aquatic ecosystem (Kulikov and Chebotina 1988). Often, macrophytes are the main components of the water ecosystem in terms of biomass and can actively accumulate radioisotopes, playing an important role in the redistribution of radioactive isotopes between the elements of the reservoir (Levina et al. 2006).

In other words, water macrophytes, which produce large amounts of organic matter, are great accumulators of radioisotopes. Simultaneously, the parameters of Chernobyl-origin radioisotopes migration and the magnitude of their accumulation in macrophytes may extensively vary. It is related to certain factors such as the degree of bottom sediments contamination, the absorptive capacity of the species, season, and growth conditions. In this regard, the ecological group of macrophyte is one of the integrating factors. Due to the diversity of occupied ecological niches and significant differences in the nutritional conditions, hydrophytes possess high species specificity and selectivity in the accumulation of radioisotopes, even within one ecological group (Kalinichenko 2010).

Chemical characteristics of water, such as ionic composition, ionic strength, the presence of organic ligands, oxidation–reduction potential (Eh), and acidity (pH), can play a significant role in the migration of radioisotopes and their accumulation by higher aquatic plants. This is particularly noticeable in closed water bodies, where the ecosystem is subject to the so-called "aging" and gradual eutrophication processes due to an increase in the amount of humic and fulvic acids, causing a decrease in the pH of the medium, increase in the desorption of radioisotopes, and their transition into a dissolved state; this can be related to varying ^{90}Sr concentrations (Rovinskij 1976; Kondo et al. 2003; Shyrokaya et al. 2005; Tryapitsyna 2011).

In the last decade, many researchers have noted a significant increase in ^{90}Sr concentration in macrophytes as compared to ^{137}Cs. In the early post-accident period, ^{90}Sr was relatively less absorbed by higher hydrophytes because unlike ^{137}Cs, its deposition was predominantly fixed in poorly soluble particles with a silicate matrix. At present, there is a decrease in the availability of ^{137}Cs, which is gradually fixed in bottom sediments in non-exchange forms. Therefore, the transfer of ^{137}Cs into macrophytes has reduced. The ability of ^{90}Sr for the non-exchange fixation by the soil is much weaker than ^{137}Cs. In addition, the release of ^{90}Sr from the sparingly soluble matrices of primary depositions accumulated by bottom sediments has led to the increasing ^{90}Sr specific activity in macrophytes with the passing years (Rovinskij 1976; Shyrokaya et al. 2008; Gudkov et al. 2006).

The above aspects have a significant impact on the accumulation of radioisotopes by aquatic plants. Currently, radioecological studies on water ecosystems in the Belarusian sector territories of the exclusion zone are extremely few and unsystematic. An attempt is made to eliminate this gap and analyze the processes of accumulation of ^{90}Sr by macrophytes in three types of reservoirs that were most characteristic for the given territory.

Methods, Objects, and Conditions of the Research

Aquatic Ecosystems

The investigations were conducted in the Belarusian sector of the Chernobyl Exclusion Zone, which comes in the territory of the Polesie State Radiation Ecological Reserve. The floodplain Lake Prestok, the oxbow of River Pripyat and the Pogonyansky Canal close to the former settlement Borshchevka were chosen as the model reservoirs (Fig. 1).

All these ecosystems are in the southern part of the reserve near the Belarusian–Ukrainian border at a 10–13 km from the Chernobyl nuclear power plant. Lake Perstok is an oxbow type reservoir extending from the north to the south for 1.42 km; it has a width of 0.062 km and an average depth of 1.84 m. According to the Hutchinson system, this completely enclosed lake belongs to the water-accumulative genetic type of lakes (Hatchinson 1969). It is in the floodplain of the River Pripyat and is fueled mainly by the floods, groundwater, and atmospheric precipitation. The volume of the water is approximately 0.162 million m^3. The area of the lake is 0.0883 km^2, and the length of the coastline is 2.98 km. Its shores are low (except the eastern part), sandy, and overgrown with shrubs and wetland vegetation, and the catchment area is small (approximately 1.1 km^2).

The oxbow of River Pripyat (Masany Oxbow) is located on a near-river part of the river floodplain and has direct access to the riverbed. The Oxbow branches out into two arms in the north. The maximum depth of the reservoir is 2.2 m, the flow velocity at the mouth is 0.02 m/s, and the thickness of bottom sediments ranges from 5 to 15 cm. The coasts are flat and meadow grass grows on the sand soil.

The Pogonyansky Canal has a maximum depth of 1.5–2.0 m. After the closure of the canal in 1992–1993, a large area was flooded, resulting in a marshy shallow water reservoir with a maximum width of 2.1 km; this reservoir stretched over 16.7 km from the northwest to the southeast. The flooding area depends on seasonal fluctuations in water level. Its shores are unformed, partly peaty and overgrown with willow, black alder, birch, and pine (Fig. 2).

Fig. 1 Location of waterbodies studied within the exclusion zone; (1) Lake Perstok; (2) Pogonyansky Canal; (3) oxbow of River Pripyat

Species and Ecological Groups of Macrophytes

This study focused on macrophytes belonging to four different ecological groups.

1. The plants of the first ecological group (floating) do not root on the ground and move with the water masses throughout the reservoir; hence, they do not form stable habitats (e.g., *Hidrocharis morsus-ranae* L.*, Spirodela polyrhiza* (L.) *Schleid*, and *Salvinia natans* L.).
2. The plants of the second group (with leaves floating on the surface) grow at the depths of 2–3 m, form intermittent groups along the reed strip and reach the greatest width in bays with a muddy bottom (e.g., *Nymphaea alba* L., *Nuphar lutea* (L.) *Smith*, *Trapa natans* L., and *Potamogeton natans L*).
3. The plants of the third group (completely submerged) usually grow at the depths of up to 3 m and can take root in the ground or drift along the entire body of water basins (e.g., *Myriophyllum spicatum* L. and *Stratiotes aloides* L.).
4. The plants of the fourth ecological group (semi-submerged) form a peculiar "second shore" of reservoirs from the littoral to a depth of 1.0–1.5 m (e.g., *Phragmites australis Cav*., *Typha angustifolia* L., and *Carex* sp.). Some species, such as *Acorus calamus* L., form almost pure associations.

Thus, ten species (three species from I and IV groups, respectively, and two from II and III groups, respectively) of the dominant hydrophytes growing on Lake

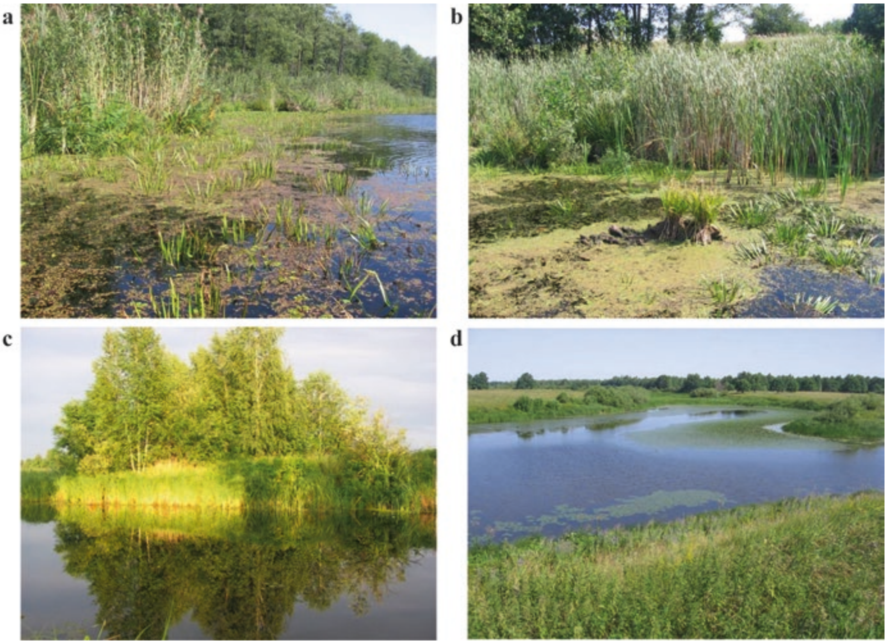

Fig. 2 The investigated reservoirs in the exclusion zone of Chernobyl nuclear power plant; (**a**) Lake Perstok (north direction); (**b**) Lake Perstok (west direction); (**c**) Pogonyansky Canal (the area of the former settlement Borshchevka); (**d**) oxbow of River Pripyat

Perstok were analyzed. In addition, five species (two species are from IV group and three species are from group II) growing on the oxbow of River Pripyat and eight species (one species from I group, three species from IV group, and two II and IV groups, respectively) growing on the Pogonyansky Canal were analyzed.

The phenology of macrophytes in the investigated reservoirs was practically the same as the established norms for the south of the Belarus (Gigevich et al. 2001; Parfenov et al. 2002) (Table 1).

Sampling of Plants and Other Components of Water Ecosystems

Investigations and sampling of water and plants were replicated thrice for each species every month from May to September annually and the wet weight was determined. Either the whole plant (roots and shoots) or only shoots were sampled depending on the species. The samples were rinsed to remove the remains of bottom sediments and silt. The plant samples, except *S. polyrhiza* (L.) *Schleid*, *H. morsus-ranae* L., *S. natans* L., *T. natans* L., and *M. spicatum* L., were ground into 1–2-cm pieces and placed in plastic bags.

Table 1 Phenology of macrophytes in the investigated reservoirs of the exclusion zone

		Vegetation period, month				
Plant species—ecological group		5	6	7	8	9
Lake Perstok						
Spirodela polyrhiza (L.) Schleid	I	X	X	X	X	X
Hidrocharis morsus-ranae L.	I		X	X	X	X
Salvinia natans L.	I			X	X	X
Nuphar lutea (L.) Smith	II	X	X	X	X	X
Trapa natans L.	II		X	X	X	X
Myriophyllum spicatum L.	III	X	X	X	X	X
Stratiotes aloides L.	III	X	X	X	X	X
Phragmites australis Cav.	IV	X	X	X	X	X
Acorus calamus L.	IV	X	X	X	X	X
Typha angustifolia L.	IV	X	X	X	X	X
Oxbow of River Pripyat						
Nuphar lutea (L.) Smith	II	X	X	X	X	X
Phragmites australis Cav.	IV	X	X	X	X	X
Carex acuta L.	IV	X	X	X	X	X
Channel Pogonyansky						
Hidrocharis morsus-ranae L.	I		X	X	X	X
Nymphaea alba L.	II	X	X	X	X	X
Potamogeton natans L.	II		X	X	X	X
Myriophyllum spicatum L.	III	X	X	X	X	X
Stratiotes aloides L.	III	X	X	X	X	X
Phragmites australis Cav.	IV	X	X	X	X	X
Carex riparia Curt.	IV	X	X	X	X	X
Typha angustifolia L.	IV	X	X	X	X	X

The sampling of the bottom sediments and soil was carried out by a cylindrical sampler with a diameter of 7 cm. These samples along with water samples were placed in plastic bottles.

Measuring Activity Concentration of Radionuclides in the Samples

The plant samples, bottom sediments, and soil were air-dried at 20–25 °C in laboratory conditions and were placed in the measuring vessels to determine the specific activity (*SA*) ^{90}Sr. The activity concentrations of the radioisotopes in plants were present in Bq kg^{-1} dry weight; the measurement error was within the 20% range. The content of ^{90}Sr and pH was analyzed in water.

The ^{90}Sr activity concentration was estimated using the γ-β spectrometer "MKS-AT 1315" (Atomtex, Belarus) with a detector unit based on a scintillation NaI (Tl) crystal.

The concentration factor (CF) was calculated as the ratio of ^{90}Sr specific activity in the plant to the radioisotope-specific activity in the bottom sediments in the analysis of specific features of accumulation of radioisotopes by macrophytes. The ratio of ^{90}Sr/^{137}Cs was used for assessing the species specificity for radioisotope intake. The ^{90}Sr/^{137}Cs ratio was used to calculate the ratio CF ^{90}Sr to CF ^{137}Cs in the plant species. Statistical processing of the results was carried out by conventional methods (Lakin 1980).

Results and Discussion

The increasing percentage of ^{90}Sr mobile forms ensures its additional flow with surface runoff into the hydrological network, its localization in closed drainage water systems, and its rapid incorporation into the biotic cycles. Consequently, the contaminated areas of the exclusion zone can be considered as open sources that supply radioactive substances to water bodies, where chronic ionizing radiation affects the biota in aquatic ecosystems.

The multifactorial process occurring in the freshwater ecosystems is reflected in the intensity of radioisotope accumulation by different taxonomic groups of aquatic organisms and creates conditions of permanent radiation exposure, thus leading to radiobiological and genetic effects. Observing the dynamics of the accumulation processes will allow us to further assess the extent of such influences and consequences for each group of freshwater organisms.

Accumulation of ^{90}Sr by Different Macrophyte Species

The intensity of ^{90}Sr intake in macrophytes is much higher than that in terrestrial plants because it has a higher bioavailability in the water ecosystems. The specific activity of the radioisotope in aquatic plants depends on the degree of contamination of the reservoir (primarily bottom sediments), the absorptive capacity of the species, season, and growth conditions. According to long-term observations and analysis of the content of long-lived radioisotopes in various types of water macrophytes in Lake Perstok, there was a significant difference in the accumulation of ^{90}Sr in the plants' phytomass. The species concentrators and antagonists were selected based on it.

The highest absolute values of ^{90}Sr activity during the entire observation period on Lake Perstok was recorded for *H. morsus-ranae* L. (36.9 Bq kg^{-1} × 10^{3}) from I group and *S. aloides* L. (30.9 Bq kg^{-1} × 10^{3}) from III group. Species of IV group had the lowest absolute values of ^{90}Sr activity (*Ph. australis Cav.*: 0.62 Bq kg^{-1} × 10^{3}). At different periods, plants of III group were immersed in the water (*M. spicatum* L. and *T. aloides* L.), and *T. natans* L. from II group showed a high degree of radioisotope accumulation. Plants from the Oxbow have the lowest absolute values of radioisotope content; it is associated with lower levels of bottom sediment pollution and washing character of the water area (Table 2).

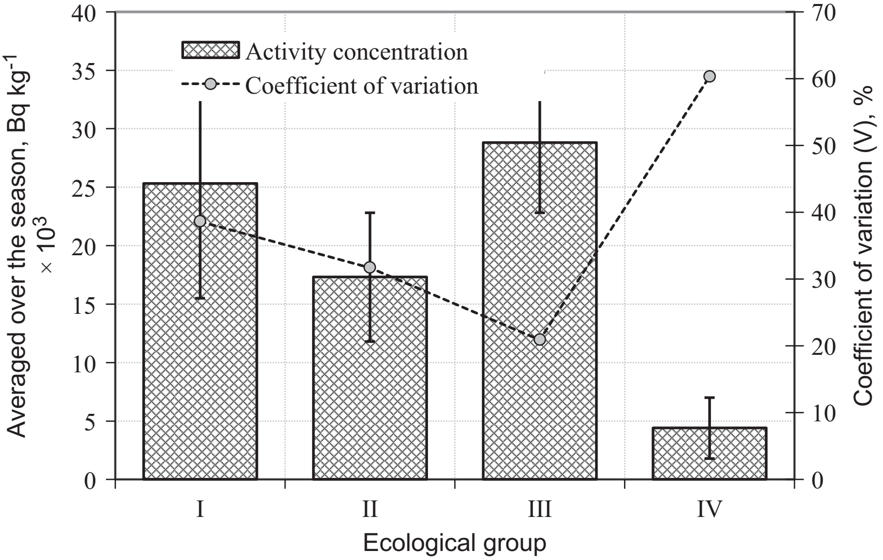

Fig. 3 Activity concentration of ^{90}Sr in various ecological groups of the macrophytes in Lake Perstok

The average values do not always correctly represent the samples, as seen from the presented data. This is due to a significant intraspecific variability in the accumulation of radioisotopes by hydrophytes. In addition, the maximum content of ^{90}Sr varied during the vegetation periods of different years depending on the ecological group. This is confirmed by the high standard deviation values and the variation coefficient (Fig. 3).

Dependences of ^{90}Sr Accumulation in Macrophytes from Type of Water Bodies and Season

An analysis of the activity concentration of ^{90}Sr and ^{137}Cs in the bottom sediments of three different reservoirs near the Chernobyl nuclear power plant showed significantly higher values of specific activity of cesium ($p < 0.001$). However, the average activity concentration of ^{137}Cs in the dominant species of macrophytes from Lake Perstok was two times lower than that of ^{90}Sr (Fig. 4); the lowest pH values were also noted here. This dependence is naturally related to the factors that affect the redistribution of radioisotopes within the aquatic ecosystem.

As mentioned above, the illumination intensity and the pH of the environment can significantly impact the behavior of man-made radioisotopes in enclosed water bodies subject to the so-called “aging” (bogging). Strontium is an active element; it can form several water-soluble compounds including complex compounds.

Table 2 Activity concentration of ^{90}Sr in the dominant macrophyte species, Bq kg^{-1} × 10^{3}

Plant species	The sample size (n)	$\frac{\bar{X} \pm S_{\bar{X}}}{x_{\text{min.}} - x_{\text{max.}}}$	Conf. interval $\frac{(P < 0.001)}{\text{SS},\%}$
Lake Perstok			
Spirodela polyrhiza L. Schleid	32	$\frac{18.88 \pm 0.89}{13.20 - 25.47}$	$\frac{15.95 - 21.81}{4.72}$
Hidrocharis morsus-ranae L.	31	$\frac{24.58 \pm 1.05}{17.20 - 36.85}$	$\frac{21.11 - 28.05}{4.29}$
Salvinia natans L.	16	$\frac{18.06 \pm 0.88}{13.42 - 21.57}$	$\frac{15.18 - 20.95}{4.85}$
Nuphar lutea (L.) Smith	39	$\frac{15.79 \pm 0.30}{12.23 - 17.86}$	$\frac{14.80 - 16.77}{1.90}$
Trapa natans L.	21	$\frac{16.53 \pm 1.22}{10.25 - 21.51}$	$\frac{12.50 - 20.55}{7.41}$
Myriophyllum spicatum L.	40	$\frac{19.46 \pm 0.63}{12.34 - 25.43}$	$\frac{17.39 - 21.53}{3.24}$
Stratiotes aloides L.	41	$\frac{25.85 \pm 0.58}{18.33 - 30.87}$	$\frac{23.93 - 27.78}{2.26}$
Phragmites australis Cav.	46	$\frac{1.47 \pm 0.12}{0.62 - 2.81}$	$\frac{1.08 - 1.85}{8.02}$
Acorus calamus L.	48	$\frac{5.09 \pm 0.15}{3.56 - 6.91}$	$\frac{4.58 - 5.60}{3.04}$
Typha angustifolia L.	55	$\frac{4.52 \pm 0.19}{2.79 - 6.62}$	$\frac{3.91 - 5.13}{4.11}$
Oxbow of River Pripyat			
Nuphar lutea (L.) Smith	27	$\frac{0.28 \pm 0.01}{0.21 - 0.33}$	$\frac{0.24 - 0.31}{3.35}$
Trapa natans L.	3	$\frac{0.19 \pm 0.07}{0.08 - 0.30}$	$\frac{0 - 0.40}{35.14}$

(continued)

Table 2 (continued)

Plant species	The sample size (n)	$\frac{\bar{X} \pm S_{\bar{X}}}{x_{\text{min.}} - x_{\text{max.}}}$	Conf. interval $\frac{(P < 0.001)}{\text{SS},\%}$
Nymphaea alba L.	2	$\frac{0.38 \pm 0.02}{0.36 - 0.39}$	$\frac{0.33 - 0.42}{4.0}$
Phragmites australis Cav.	23	$\frac{0.45 \pm 0.02}{0.25 - 0.61}$	$\frac{0.37 - 0.52}{5.08}$
Carex acuta L.	22	$\frac{0.10 \pm 0.004}{0.09 - 0.12}$	$\frac{0.09 - 0.11}{3.69}$
Pogonyansky Canal			
Hidrocharis morsus-ranae L.	3	$\frac{3.83 \pm 0.06}{3.76 - 3.90}$	$\frac{3.64 - 4.02}{1.49}$
Nymphaea alba L.	10	$\frac{0.35 \pm 0.02}{0.29 - 0.45}$	$\frac{0.29 - 0.41}{5.40}$
Potamogeton natas L.	2	$\frac{1.47 \pm 0.09}{1.36 - 1.58}$	$\frac{1.11 - 1.83}{7.48}$
Myriophyllum spicatum L.	2	$\frac{2.21 \pm 0.08}{2.11 - 2.30}$	$\frac{1.89 - 2.52}{4.31}$
Stratiotes aloides L.	2	$\frac{3.22 \pm 0.07}{3.13 - 3.31}$	$\frac{2.92 - 3.52}{2.80}$
Phragmites australis Cav.	13	$\frac{0.87 \pm 0.10}{0.62 - 1.66}$	$\frac{0.55 - 1.18}{11.05}$
Carex riparia Curt.	9	$\frac{0.94 \pm 0.41}{0.20 - 2.36}$	$\frac{0 - 2.29}{43.88}$
Typha angustifolia L.	13	$\frac{2.86 \pm 0.20}{2.35 - 3.37}$	$\frac{2.20 - 3.52}{6.99}$

SS statistical significance

According to Rovinsky (1976), the ratio of mobility ^{90}Sr to ^{137}Cs for non-flow water bodies is 1:0.12. After analyzing the data obtained for several years, we can infer that the ^{137}Cs and ^{90}Sr activities in water increase with the decreasing pH.

In most cases, the seasonal dynamics of changes in the specific activity of ^{137}Cs and ^{90}Sr in macrophytes shows that their activity concentration increases towards

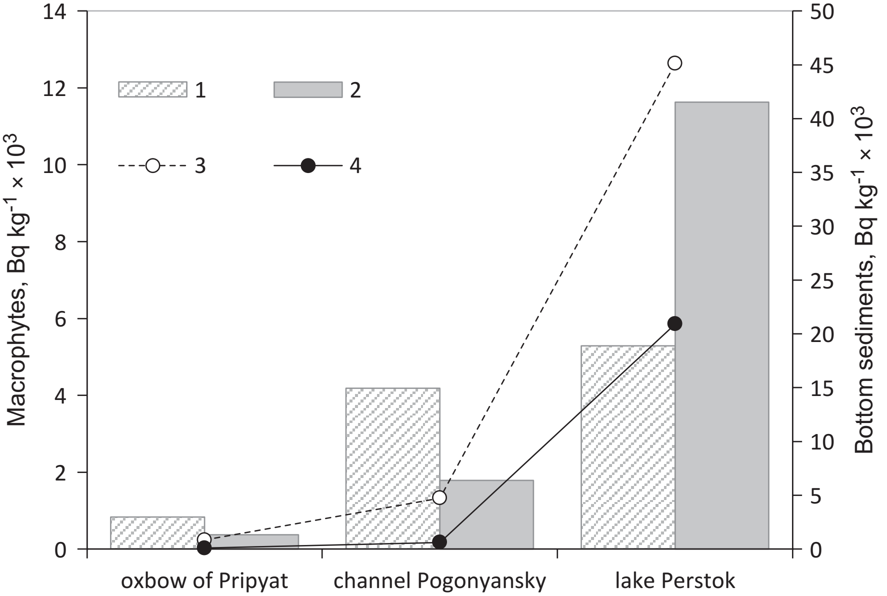

Fig. 4 Comparative content of ^{90}Sr and ^{137}Cs in macrophytes and bottom sediments of various reservoirs of the exclusion zone; (1) ^{137}Cs in macrophytes; (2) ^{90}Sr in macrophytes; (3) ^{137}Cs in bottom sediments; (4) ^{90}Sr in bottom sediments

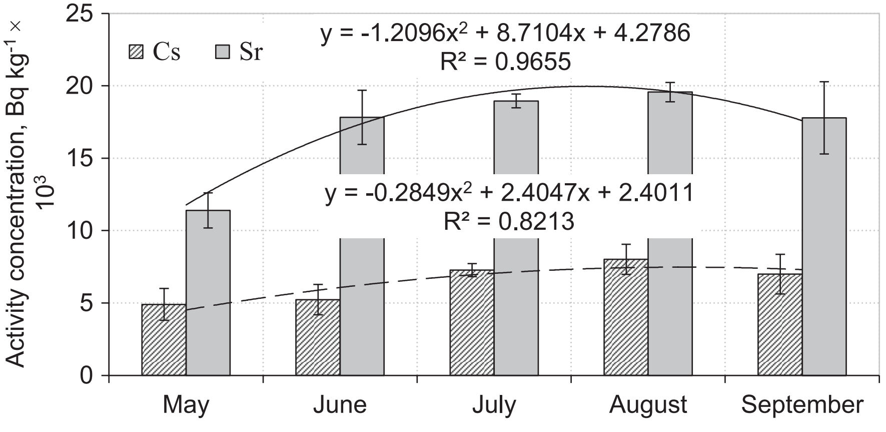

Fig. 5 Seasonal dynamics of ^{90}Sr and ^{137}Cs activity concentration in the macrophytes in Lake Perstok

the end of the growing season and decreases towards the beginning of the wilting period (September). The rate of ^{90}Sr intake by macrophytes is generally higher than ^{137}Cs intake during the period of growth onset. The tendency to increase the activity concentration of the radioisotopes in the macrophytes during the growing season is associated with the high meristematic activity of plant tissues in the summer. When

Table 3 Concentration factors of ^{90}Sr, ^{137}Cs and ^{90}Sr/^{137}Cs-ratio for the dominant species in the investigated water reservoirs

Plant species	Ecological group	CF		^{90}Sr/^{137}Cs—ratio
		^{137}Cs	^{90}Sr	
Lake Perstok				
Spirodela polyrhiza L. Schleid	I	0.20	1.89	3.9
Hidrocharis morsus-ranae L.	I	0.48	2.46	2.4
Salvinia natans L.	I	0.24	1.81	2.9
Nuphar lutea (L.) Smith	II	0.23	1.58	2.7
Trapa natans L.	II	0.32	1.65	2.1
Myriophyllum spicatum L.	III	0.31	1.94	1.9
Stratiotes aloides L.	III	0.22	2.58	6.6
Phragmites australis Cav.	IV	0.09	0.15	0.6
Acorus calamus L.	IV	0.03	0.51	9.5
Typha angustifolia L.	IV	0.04	0.45	12.6
Oxbow of River Pripyat				
Nuphar lutea (L.) Smith	II	1.02	2.85	1.8
Trapa natans L.	II	1.10	3.07	0.3
Nymphaea alba L.	II	2.89	4.03	0.2
Phragmites australis Cav.	IV	0.14	4.65	3.2
Carex acuta L.	IV	0.91	1.02	7.1
Pogonyansky Canal				
Hidrocharis morsus-ranae L.	I	1.63	3.94	0.3
Nymphaea alba L.	II	0.73	0.30	0.1
Potamogeton natas L.	II	0.28	1.59	0.8
Myriophyllum spicatum L.	III	1.07	2.32	0.3
Stratiotes aloides L.	III	0.20	3.34	2.3
Phragmites australis Cav.	IV	0.16	0.88	1.1
Carex riparia Curt.	IV	0.30	1.32	0.1
Typha angustifolia L.	IV	0.01	3.41	40.2

physiological processes slow down, the osmotic current of mineral substances, including their radioactive analogs, in the tissues of plants also decreases (Fig. 5).

The behavior of radioisotopes in various aquatic ecosystems can substantially differ. This is due to a number of biotic and abiotic factors. Biotic factors are determined by the individual physiological and ecological characteristics of the species inhabiting the reservoir. Abiotic factors include different hydrodynamic and chemical processes occurring in various water bodies; these factors are directly related to weather and climatic and topographical conditions. All these factors need to be considered while evaluating the processes occurring during the transformation of radioisotopes in aquatic ecosystem components.

The CF for ^{137}Cs, ^{90}Sr and ^{90}Sr/^{137}Cs ratio for the dominant species of the macrophytes in the model water bodies were established based on empirical research (Table 3). A comparative analysis of CF indicates that it is inversely dependent on the level of bottom sediment contamination. The ^{90}Sr/^{137}Cs ratio reflects the propensity of one or another plant to accumulate a certain radioisotope. According to the obtained data, the ^{90}Sr/^{137}Cs ratio for different types of macrophytes in water bodies near the zone of radioactive contamination can differ by ten times. Overall, the ^{90}Sr/^{137}Cs ratio in the Chernobyl exclusion zone was 4.5. The macrophytes from the oxbow of Pripyat River have the least value of ^{90}Sr/^{137}Cs ratio (2.8), while plants from the Pogonyansky Canal have the largest value (5.6). In the early post-accident period, the ^{90}Sr/^{137}Cs ratio did not exceed 0.17. The selectivity of ^{90}Sr accumulation by *T. angustifolia* L. was confirmed in various reservoirs. The maximum difference between the intake of ^{137}Cs and ^{90}Sr was 40.2 times, which is typical for plants from the Pogonyansky Canal. The differences in the values of ^{90}Sr/^{137}Cs ratio were not associated with an ecological group of plant species. The ratio mainly depends on species features.

The analysis of the fluctuations of the ^{137}Cs and ^{90}Sr activity concentration in the macrophytes belonging to the various ecological groups in Lake Perstok shows a significant ^{90}Sr accumulation (up to 30 times) by dominant macrophytes compared to ^{137}Cs accumulation (Fig. 6).

Such a significant predominance in ^{90}Sr accumulation can be attributed to the following reasons: the initial amount of radioisotope in the environment (in bottom sediments in this case) and eight times higher ^{90}Sr mobility in non-flow reservoirs than ^{137}Cs mobility (Rovinskij 1976). The other types of reservoirs (the Oxbow and the channel of the former meliorative canal) do not have such clear dependence. Only *P. australis* (*Cav.*) does not show such a trend; it accumulates ^{137}Cs to a greater extent (up to two times).

The high accumulation of ^{90}Sr in the aquatic plants is due to an increase in its mobile forms in soils of the catchment areas of the exclusion zone, which also enhances the amount of biologically available radioisotope in the reservoirs. The processes of remobilization intensively take place in waterlogged ecosystems contaminated with radioisotopes, which become more active due to ongoing bogging processes. The intensity of ^{90}Sr intake into hydrobionts (such as macrophytes, mollusks, and fish) and its contribution to exposure dose of biota are increasing in such ecosystems. While analyzing the radioisotope accumulation by plants, species specificity and their ecological niches need to be considered. These topics have been discussed in detail in our previous papers (Kalinichenko and Nenashev 2012; Kalinichenko 2013).

Long-Term Trends of ^{90}Sr and Their Behavior in Aquatic Ecosystems

Over the years, the activity concentrations of ^{137}Cs and ^{90}Sr have significant ranges of intraspecific fluctuations and do not follow the general trend towards a decrease in the accumulation of radioisotopes in macrophytes. The content of radioisotopes in *P. australis Cav* has greatly varied in this 8-year study, with the maximum and minimum values ranging from five times for ^{137}Cs to 4.5 times for ^{90}Sr. The range for *A. calamus* L. is 2.2 and 1.9 times, respectively, that for *T. angustifolia* L. is 16.6 and 2.4 times, respectively (Fig. 7).

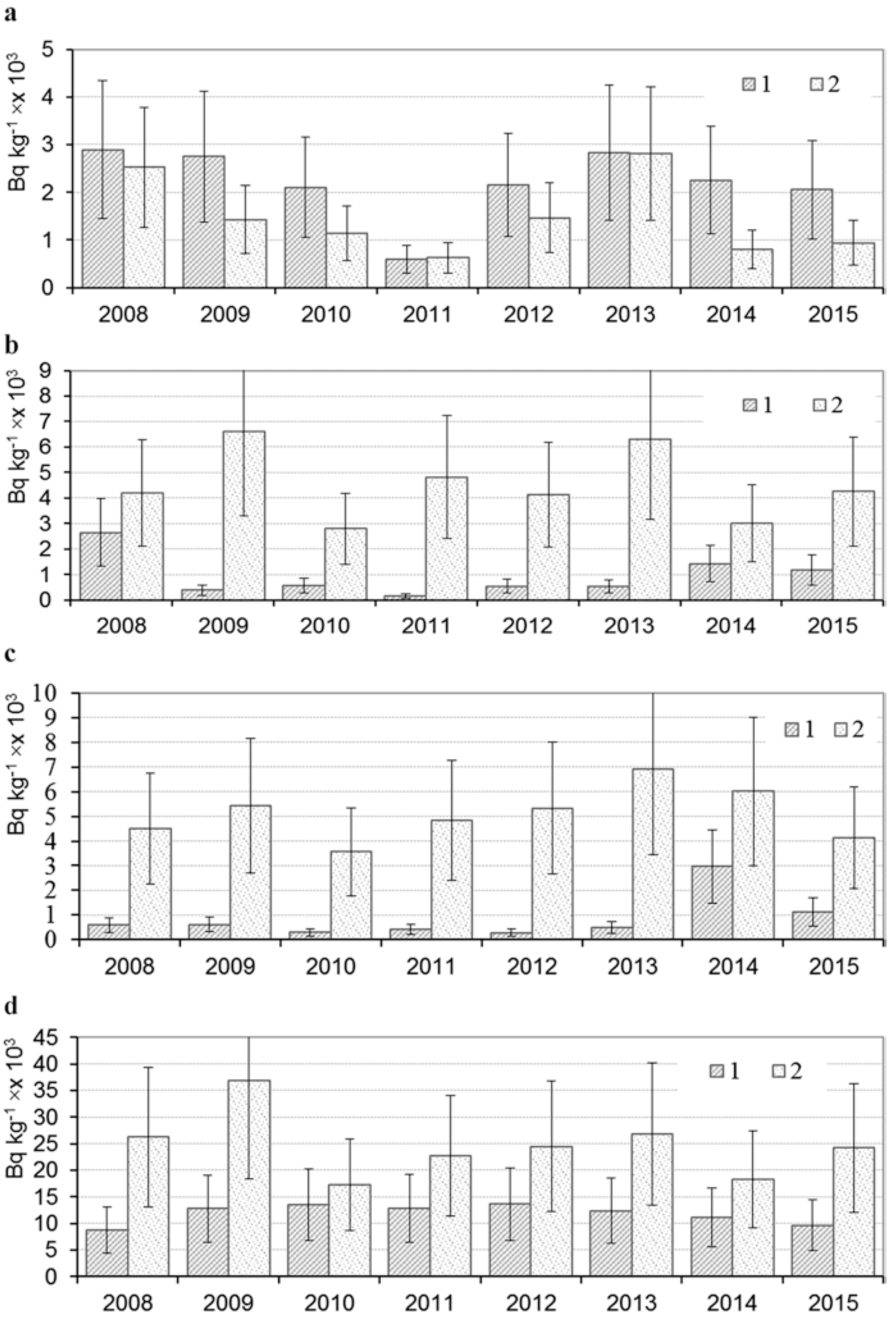

Fig. 6 Long-term dynamics of the content of ^{137}Cs and ^{90}Sr in the macrophytes in the Lake Perstok averaged over the season; (**a**) *Ph. australis Cav.*; (**b**) *T. angustifolia* L.; (**c**) *A. calamus* L.; (**d**) *H. morsus-ranae* L.; (1) ^{137}Cs; (2) ^{90}Sr

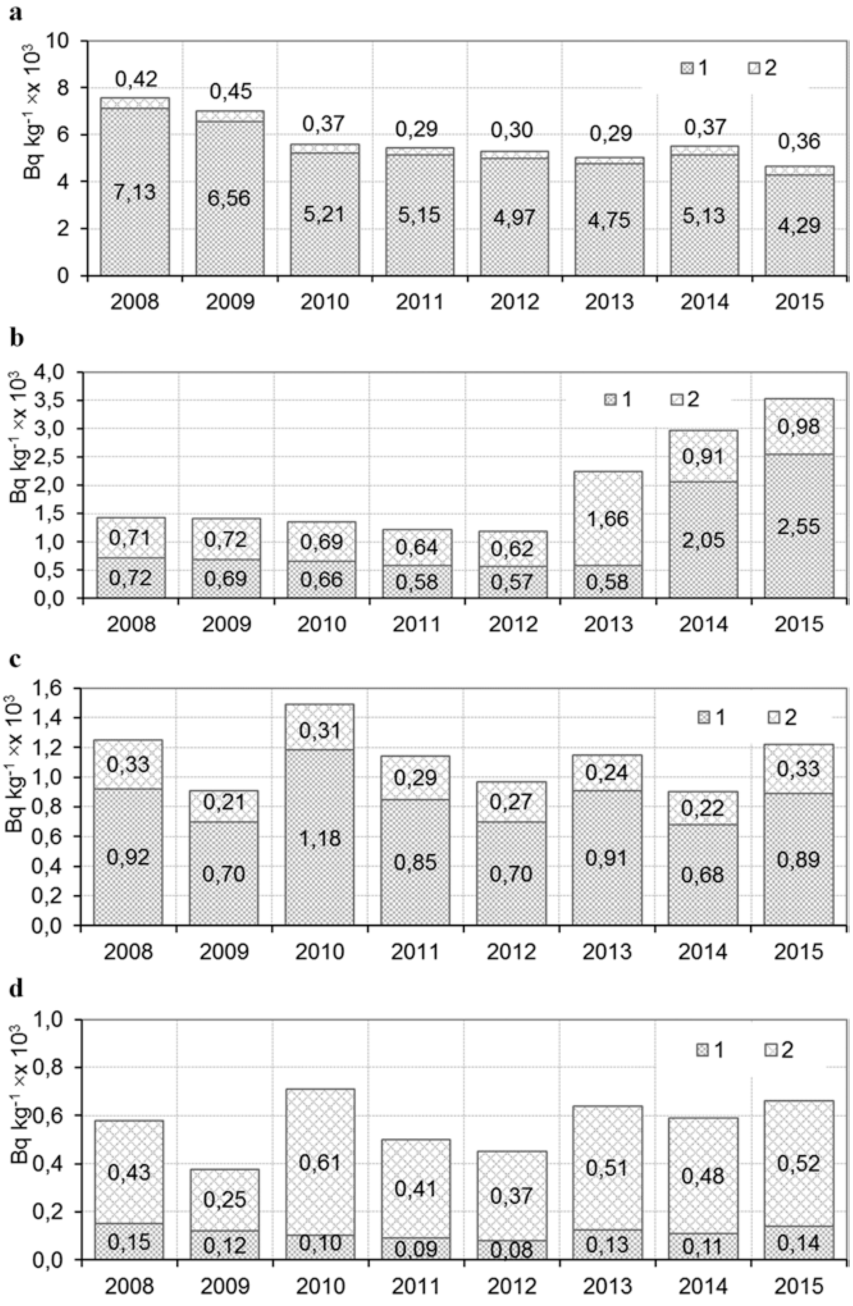

Fig. 7 Dynamics of ^{137}Cs and ^{90}Sr activity concentration in some types of macrophytes in the Pogonyansky Canal and the oxbow of River Pripyat averaged over the season; (**a**) *N. lutea* (L.) *Smith* in channel Pogonyansky; (**b**) *P. australis Cav.* in Pogonyansky Canal; **c:** *N. lutea* (L.) *Smith* in oxbow of river Pripyat; (**d**) *P. australis Cav.* in oxbow of river Pripyat; (1) ^{137}Cs; (2) ^{90}Sr

All the groups of biotic and abiotic factors continually impact the aquatic ecosystem, changing the physiological parameters of aquatic organisms and the state of their environment; hence, the probability of such fluctuations that are influenced by the abovementioned factors is minimal. Furthermore, the conditions for the development of macrophytes, such as microclimatic factors and the hydrological regime, lead to a redistribution of favorable conditions from one species to another. This is reflected in the variation of the levels of radioisotopes accumulation in plants in different years and the redistribution of radioactive isotopes between the macrophyte species and ecological groups. Moreover, it is important to consider the physicochemical transformation of the radioisotopes in this ecosystem. The reactions of leaching radioisotopes from fuel particles and their transition into the exchange forms play a significant role. The rate of this process can vary depending on the geochemical parameters of the environment during aging of the reservoir.

Species specificity in the accumulation of radioisotopes is most observed in the oxbow in the channel of the former meliorative canal. Macrophytes species such as *T. angustifolia* L. and *S. aloides* L. have an affinity for ^{90}Sr accumulation, irrespective to the ecosystem. These species belong to different ecological groups and differ in their biological and physiological characteristics. *Cyperaceae* species predominantly accumulate ^{137}Cs. For example, *C. acuta* L. growing in the oxbow of River Pripyat accumulate ^{137}Cs 2.4 times more than ^{90}Sr, whereas *C.* riparia *Curt*. from the Pogoniansky Canal accumulate ^{137}Cs approximately 16 times more than ^{90}Sr. The third groups of plants also have the advantage of accumulating a particular radioisotope, which is manifested in each ecosystem in different ways. *Nuphar lutea* (L.) *Smith* shows a greater tendency to accumulate ^{137}Cs in the Pogonyansky Canal and oxbow of River Pripyat.

The histograms reveal that the activity concentration of ^{90}Sr in the tissues is same, but the ratio between the radioisotopes varies due to a decrease in ^{137}Cs accumulation in the oxbow where its levels are lower. In this case, the processes of absorption of ^{137}Cs and ^{90}Sr by plant tissues proceed identically, but there is a difference in the quantity of biologically accessible compounds of these isotopes in the environment. Similar processes of radioisotopes accumulation are observed for *P. australis* (*Cav.*). However, its affinity for ^{137}Cs absorption by the root system decreases with the increase of the flow ability of the reservoir. Compared to plants from other groups, this plant species, similarly as all macrophytes from group IV, absorbs the lowest concentration of radioisotopes from the environment.

The distribution and migration of radioisotopes in aquatic ecosystems are affected by complex and interrelated processes such as the following: physical (dilution, dispersion, mixing, and sedimentation), physicochemical (sorption–desorption), chemical (isotopic exchange, hydrolysis, polymerization, and transformation), and biological (accumulation and excretion) processes. The significance of these processes in the behavior of the radioisotopes is far from equal, and it is determined not only by the chemical nature of the radioactive isotopes but also by the specific features of the particular reservoir, such as the characteristics of sediment, water properties, species composition, and biomass of hydrobionts (Kuzmenko 1990). Biogeochemical processes taking place in bottom sediments and water play

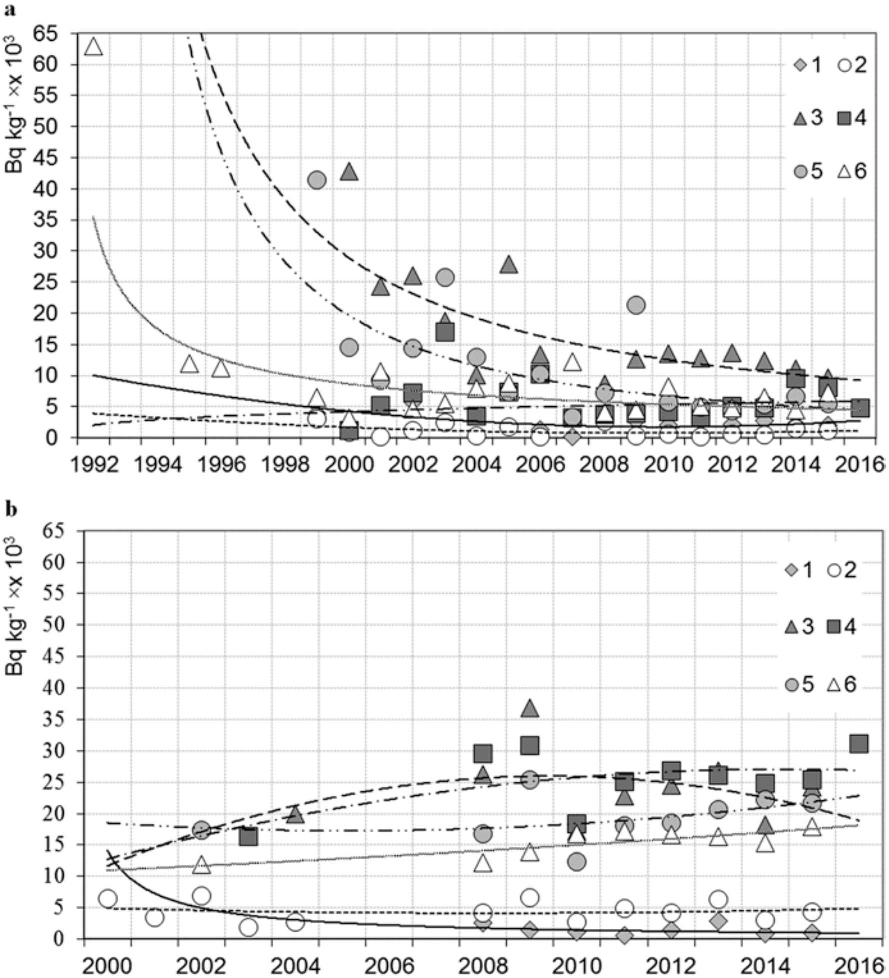

Fig. 8 Trends in the content of radioisotopes over the time in the macrophytes from Lake Perstok; a: ^{137}Cs; b: ^{90}Sr ((1) *P. australis Cav.*; (2) *T. angustifolia* L.; (3) *H. morsus-ranae* L.; (4) *S. aloides* L.; (5) *M. spicatum* L.; (6) *N. lutea* (L.) *Smith*)

an important role because the bulk activity of radioisotopes that enter the reservoir are concentrated in bottom sediments due to sedimentation.

In the long-term period after the accident, a significant source of radioisotope stock replenishment in the bottom sediments is the flow from nearby areas due to rains and flood waters (such as streams, rivulets, and ditches). We cannot confidently comment on a dynamic balance in the radioisotope accumulation because a substantial uncertainty can be made by biotic factors that can significantly change the state of the environment by accumulating biogenic elements. Furthermore, the soils of freshwater reservoirs are highly diverse. Therefore, their ability to accumulate radioisotopes varies depending on the type of bottom sediments and the conditions of the aquatic environment.

Over the time, the analysis long-term dynamics of radioisotopes accumulation in macrophytes revealed a different behavior for ^{137}Cs and ^{90}Sr after the Chernobyl fallout (Fig. 8).

The presented functional dependencies reveal a clear tendency to decrease the activity concentration of ^{137}Cs in hydrophytes. In most cases, the trend is opposite for ^{90}Sr. The analysis showed a steady decline only for *P. australis Cav*. In other cases, the activity concentration of ^{90}Sr in plants has increased by a little in recent years. However, this analysis does not consider the radioisotope content in the initial post-accident years, which could significantly affect the overall picture of the dynamic processes. For predicting the behavior of Chernobyl-origin radioisotopes in higher hydrophytes, many characteristics need to be considered for a more accurate description; however, long-term monitoring is required under full-scale conditions. In this case, the usual representations of dynamic processes using the exponential curve do not reflect the real picture. The presence of radioisotopes in the exclusion zone in various forms, taking into account the light dispersed fraction and the fuel component, whose behavior has not yet been adequately studied, leaves its imprint.

It should be noted that the species composition of macrophytes in the studied water bodies has not changed in the last 8 years, allowing us to judge the changes in environmental conditions, fertility, and chemical composition of the water environment. Water macrophytes could serve as an excellent object for bio indicator of water bodies for the determination of chemical peculiarities of water and direction of ecological processes. However, further investigations are warranted to study macrophytes as indicator organisms.

Conclusions

- Macrophytes, an important component of water ecosystems, remain as the accumulators of radioactive substances in the exclusion zone of Chernobyl nuclear power plant. The investigation of radioisotopes accumulation peculiarities in macrophytes is an important link for an understanding of the state and development of ecosystems under conditions of enormous radioisotope contamination. The study results open up new aspects of strontium behavior in aquatic ecosystems.
- The concentration factors of ^{137}Cs and ^{90}Sr in macrophytes indicate significant differences in the accumulation of the radioisotopes by aquatic plants from different ecological groups. A higher mobility of ^{90}Sr in comparison with ^{137}Cs in the components of aquatic ecosystems is established, which is confirmed by higher CF and ^{90}Sr/^{137}Cs values. The activity concentration of ^{90}Sr in macrophytes during the season has a high rate of increasing (up to four times more against ^{137}Cs) and decreasing (up to 1.6 times). Furthermore, the analysis of CF indicates a tendency to reduce the accumulation of radioisotopes by hydrophytes as a function of the content of ^{137}Cs and ^{90}Sr in bottom sediments.

- Dynamic processes of accumulation of radioisotopes in macrophytes are very complex, and they are caused by many factors such as biological peculiarities and environment properties. These include the following: physicochemical properties and density of radioactive fallout (which includes a large number of fuel particles in the exclusion zone), chemical characteristics of water, an ecological group of plant, type of reservoir, and seasonal fluctuations. The accumulation of radioisotopes by aquatic plants can depend on the concentration elements with similar chemical properties (calcium and potassium) in water, pH of the water, illumination level, and temperature regime.

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Strontium Isotopes in Biological Material: A Key Tool for the Geographic Traceability of Foods and Humans Beings

Simone Tommasini, Sara Marchionni, Ines Tescione, Martina Casalini, Eleonora Braschi, Riccardo Avanzinelli, and Sandro Conticelli

Introduction

In this contribution we will discuss the variation of the $^{87}Sr/^{86}Sr$ isotope ratio in the solid Earth and its link to that of biological material. The great potential of $^{87}Sr/^{86}Sr$ is that it represents a ratio between two isotopes of the same element, hence with identical geochemical behaviour. However, while ^{86}Sr is stable, ^{87}Sr is radiogenic being the product of the radioactive decay of ^{87}Rb, an isotope of a different element, hence with different geochemical behaviour.

Long lived isotope ratios of heavy elements of geological interest, such as $^{87}Sr/^{86}Sr$, $^{143}Nd/^{144}Nd$, $^{206}Pb/^{204}Pb$, $^{207}Pb/^{204}Pb$ and $^{208}Pb/^{204}Pb$ have gained importance in the last decades for assessing the issue of traceability in food, archaeological, environmental, medical and forensic sciences (Kelly et al. 2005; Podio et al. 2013;

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Voerkelius et al. 2010). These new lines of application stem from the extensive and successful use of such radiogenic isotopic ratios in several 'classic' field of the Geoscience, as precious tools for Geochronology (in both terrestrial and meteoritic samples) and for tracking petrological, geological and environmental processes (Horn et al. 1993; Capo et al. 1998; Tommasini et al. 2000).

This chapter is focused on strontium isotopes and presents a review aimed at describing the variety of pathways and different source materials that characterize the cycle of strontium in the solid Earth and in the biological systems. First, we describe how strontium isotopes are used in geochronology and how $^{87}Sr/^{86}Sr$ differs in the major Earth's reservoirs and rock types, then how strontium isotopes are transferred to weathered materials (i.e., soils), plants, and animals, and eventually we highlight a number of examples pointing to the added value of strontium isotopes in the abovementioned studies.

Solid Earth and Strontium (and Rubidium) Distribution

Strontium (Sr) belongs to the II group of chemical elements of the Periodic Table (alkaline earth metals) along with Beryllium, Magnesium, Calcium, Barium, and Radium, and has an atomic number $Z = 38$. Sr has four naturally occurring isotopes (i.e., ^{84}Sr, ^{86}Sr, ^{87}Sr, and ^{88}Sr) one of which produced by radioactive decay of ^{87}Rb (i.e., the parent). Rubidium (Rb) belongs to the alkaline metals, the I group of chemical elements of the Periodic Table, along with Lithium, Sodium, Potassium, Caesium and Francium, and has an atomic number $Z = 37$.

The distribution of Sr and of Rb within Earth depends upon the mineralogy of its interior and the deep cycle of these elements is governed by the Earth's geodynamics, and thus by the geological processes that determine enrichment vs. depletion of such elements within the different geochemical Earth's reservoirs or shells (e.g., mantle vs. crust).

The Earth's interior is subdivided into three major shells: the crust (ca. 1 vol.%) that can be either oceanic or continental, the mantle (ca. 84 vol.%), and the core (ca. 15 vol.%). The crust and the mantle make up the silicate fraction of our planet, whilst the core consists of a Fe-Ni alloy. Seven chemical elements [Oxygen (50.7 wt.%), Magnesium (15.3 wt.%), Iron (15.2 wt.%), Silicon (14.4 wt.%), Sulphur (3.0 wt.%), Aluminium (1.4 wt.%), and Calcium (1.0 wt.%)] account for 97% of the Earth's mass, but all the other elements of the Periodic Table can be found in our planet in minor amounts and they can be preferentially partitioned in a given Earth's layer or rock material (e.g., Sun and McDonough 1989).

Following the classification scheme of Goldschmidt (1937), both Sr and Rb are lithophile (*stone-loving*) elements that are concentrated, albeit at ppm level (i.e., $mg\ kg^{-1}$), in the outermost Earth's layer (continental and oceanic crust). Given their low content in Earth's rocks, Sr and Rb rarely form their own minerals, such as Strontianite ($SrCO_3$), Celestine ($SrSO_4$), Rubicline [$(Rb_{0.75}K_{0.25})AlSi_3O_8$] and Ramanite [$RbB_5O_6OH_4 \cdot 2(H_2O)$]. Strontium is ubiquitous, however, in common rock-forming minerals as bivalent Sr with an ionic radius (1.18 Å) similar to that of bivalent Calcium (1.00 Å). The similar charge to ionic radius ratio of Sr and Ca allows Sr to enter readily in the crystal lattice of the most common Ca-bearing minerals such as tectosilicates (plagioclase), carbonates (calcite), sulphates (gypsum), and phosphates (apatite). Conversely, monovalent Rb has a larger ionic radius (1.52 Å) similar to that of monovalent Potassium (1.38 Å), hence favouring Rb substitution for K in the crystal lattice of the most common K-bearing minerals such as phyllosilicates (muscovite, biotite, illite) and tectosilicates (K-feldspar).

The different geochemical characteristics of Sr and Rb (i.e., bivalent vs. monovalent cations, preferential enrichment in Ca-bearing vs. K-bearing minerals) determine a significant fractionation of Rb from Sr during the major rock-forming processes ($0 < \mathrm{Rb/Sr} < 300$, e.g., Table 1) such as (1) partial melting of rocks and magma differentiation (igneous processes), (2) mineral reaction at high pressure and temperature (metamorphic processes), and (3) mineral weathering and authigenic mineral formation (sedimentary processes). The latter processes occur at the Earth surface and are responsible also for the transfer of Sr and Rb from the solid Earth to the Biological cycle. Sr and Rb are variably released during weathering and up-taken as non-nutrient elements in the different trophic levels (plants, herbivores, carnivores) of life on Earth (Fig. 1).

The Rubidium-Strontium Isotope System

The Sr isotope ratio (i.e. $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$) represents a powerful geochemical tool to establish the link, if any, between solid Earth materials and Biosphere because it records the time elapsed since the last geochemical equilibrium achieved within rocks.

Rubidium has two naturally occurring isotopes ($^{85}\mathrm{Rb}$ and $^{87}\mathrm{Rb}$). The present day atomic ratio of $^{85}\mathrm{Rb}/^{87}\mathrm{Rb}$ is 2.593 and is constant throughout the Earth. $^{87}\mathrm{Rb}$ is radioactive and decays to the stable isotope $^{87}\mathrm{Sr}$ by emission of a β^- and antineutrino (ν) particles (eq. 1), which share, as kinetic energy, the decay energy (Q) of 275 keV:

$$^{87}\mathrm{Rb} \rightarrow {}^{87}\mathrm{Sr} + \beta^- + \nu + Q \tag{1}$$

The decay constant λ of $^{87}\mathrm{Rb}$ is 1.42×10^{-11} year^{-1}, corresponding to a half-life $t_{1/2}$ of 48.8×10^{-9} year (e.g. Faure 1986; Dickin 2003). This means that after 48.8 billion years, half of the amount of $^{87}\mathrm{Rb}$ initially present at the time of Earth formation will have decayed to $^{87}\mathrm{Sr}$. Strontium has four stable isotopes characterized by different atomic abundances (Fig. 2): $^{84}\mathrm{Sr}$, $^{86}\mathrm{Sr}$, and $^{88}\mathrm{Sr}$ are not radiogenic, whilst $^{87}\mathrm{Sr}$ is radiogenic.

Consequently, the atomic abundance ratios among not radiogenic Sr isotopes have remained constant since the formation of the Solar System ($^{88}\mathrm{Sr}/^{86}\mathrm{Sr} = 8.3752$; $^{84}\mathrm{Sr}/^{86}\mathrm{Sr} = 0.05658$), whilst $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ is strongly variable and depends upon both the initial content of Rb and Sr in any given rock or mineral and the time elapsed since its formation (Fig. 3; e.g., Faure 1986; Dickin 2003). Accordingly, $^{87}\mathrm{Sr}/^{86}\mathrm{Sr}$ is often referred as representative of the *time integrated* $^{87}\mathrm{Rb}/^{86}\mathrm{Sr}$.

Table 1 The values reported in the Table are meant to provide a guideline and to represent examples of the wide variability encountered within natural material on Earth

Material	Age	Sr (ppm)	Rb (ppm)	Rb/Sr	$^{87}Sr/^{86}Sr$	References
Geologic materials						
Average						
Bulk Crust		320	49	0.15	–	Rudnick and Gao (2003)
Upper Crust		320	83	0.26	0.710–0716	Rudnick and Gao (2003), McLennan (2001), Goldstein and Jacobsen (1988)
Global Subducted Sediments (GLOSS)		327	57	0.17	0.717	Plank and Langmuir (1998)
Mid Oceanic Ridge Basalt (MORB)		90	0.56	0.01	0.703	Sun and McDonough (1989), Salters and Stracke (2004)
Example values for single rock types						
Granite	Permian	9.3	182	19.6	0.923	*Georoc database*
Granite	Cretaceous	9.4	182	19.4	0.721	*Georoc database*
Ryolite	Quaternary	6	184	30.7	0.704	*Georoc database*
Plateau basalt	Cretaceous	4.5	200	44.4	0.704	*Georoc database*
Basalt	Pleistocene/Holocene	697	15	0.02	0.704	*Georoc database*
Pelagic clay	Jurassic	163	121.7	0.75	0.713	Plank and Langmuir (1998)
Shale	Cretaceous	132	33	0.25	0.714	Casalini (2016)
Ultamafic rock	Jurassic	12	2	0.17	0.707	Casalini (2016)
Carbonate	Quaternary	1504	7.7	0.01	0.709	Plank and Langmuir (1998)

Material	Age	Sr (ppm)	Rb (ppm)	Rb/Sr	$^{87}Sr/^{86}Sr$	References
Carbonate	Jurassic	850	–	–	0.707	Faure et al. (1978)
Gneiss	Paleozoic	142	144.5	1.02	0.722	Conticelli (1989), Conticelli (1998), Conticelli et al. (2015)
Phyllite	Paleozoic	63	70.2	1.11	0.729	Conticelli (1989), Conticelli et al. (2015)
Water						
Seawater		7.6	0.11		0.7069–0.7092	Holland (1984), Taylor and McLennan (1985)
Rivers		0.006–0.8	0.0013–0.0031		variable (0.704–0.922 da Capo et al. 1998)	Holland (1984), Wadleigh et al. (1985), Goldstein and Jacobsen (1987), Yang et al. (1996)
Rain		0.0001–0.0002	0.0002		variable (depending of areosol composition)	Reimann and Caritat (1998)
Snow		0.00001–0.001				Andersson et al. (1990), Baisden et al. (1995)
Biological material						
Edible		1–100			variable	Burton et al. (1999)
Mammal (incl. Human) bone		100–1000			variable	Burton et al. (1999), Elias et al. (1982)
Mammal (incl. Human) enamel		50–500			variable	Kohn et al. (1999)

As discussed in the text the resulting $^{87}Sr/^{86}Sr$ of any rock is the result of its time-integrated Rb/Sr ratios: a clear example of it is provided by the different value of the two granites, which have similar Rb/Sr but different $^{87}Sr/^{86}Sr$ depending on their different age

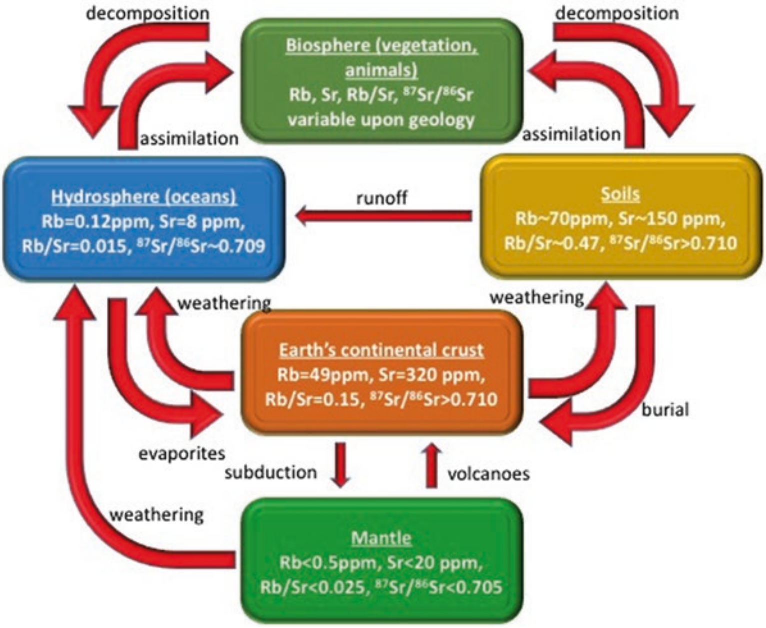

Fig. 1 Sr, Rb, Rb/Sr and ^{87}Sr/^{86}Sr cycle in the different Earth's layers with average composition reported

This decay process sets the basis for the Rb-Sr absolute radiometric dating of igneous and metamorphic rocks. According to the laws of radioactive decay, the number of ^{87}Sr atoms occurring in a rock or mineral at any time *t* is given by:

$$^{87}\mathrm{Sr} = {}^{87}\mathrm{Sr}_i + {}^{87}\mathrm{Rb} \cdot \left(e^{\lambda t} - 1\right) \tag{2}$$

where the second term of the right part of eq. (2) represents the number of ^{87}Sr atoms produced by the radioactive decay of ^{87}Rb since *t* years ago, and it is summed up to $^{87}\mathrm{Sr}_i$, which is the number of ^{87}Sr atoms present initially in the system.

Equation (2) can be rearranged dividing all terms by ^{86}Sr (which is not radiogenic and therefore remains constant with time) to yield isotopes ratios that allows for more precise measurements than absolute atomic abundance:

$$\left({}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr}\right)_m = {}^{87}\mathrm{Sr}/{}^{86}\mathrm{Sr}_i + {}^{87}\mathrm{Rb}/{}^{86}\mathrm{Sr} \cdot \left(e^{\lambda t} - 1\right) \tag{3}$$

The present-day $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_m$ can be measured by various types of mass spectrometers. The best results (up to a precision of $2\sigma < 20$ ppm) are obtained by Multi-Collector Thermal Ionisation Mass spectrometers (i.e., TIMS; e.g., Thirlwall 1991; Avanzinelli et al. 2005). Recently similar, yet slightly lower, precisions ($2\sigma \sim 30$ ppm) have been achieved through double focusing Multi-Collector Inductively Coupled Plasma Mass Spectrometers (i.e., MC-ICPMS, Waight et al. 2002). Few studies,

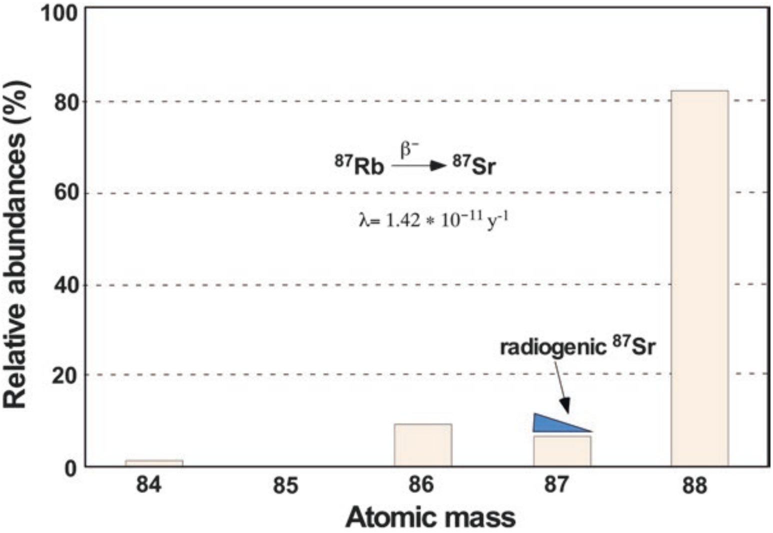

Fig. 2 Relative natural abundances of strontium isotopes

however, determine Sr isotope composition through significantly less precise methods, such as High Resolution Sector Field ICP-MS (i.e., HR-ICPMS) or Quadrupole ICPMS, both of which are without a multi-collector system. The former yields lower precision by more than an order of magnitude with respect to TIMS (2σ = 200–1000 ppm: Latkoczy et al. 1998, 2001), whilst the latter provide even larger errors (2σ = 4000–6000 ppm: Almeida and Vasconcelos 2001, 2004). Such large uncertainties are inadequate not only for geochronological purposes, but also for establishing a link between a plant and its substratum, since, as we will see later, an uncertainty of 6000 ppm covers a large portion of the Sr isotope variability occurring on Earth material (e.g. almost the whole range of volcanic rocks from MORBs to the most enriched subduction-related rock).

The atomic ^{87}Rb/^{86}Sr can be calculated from the weight ratio of Rb/Sr, measured by isotope dilution mass spectrometry with a precision <1%, according to eq. (4):

$$^{87}\mathrm{Rb}/^{86}\mathrm{Sr} = \mathrm{Rb}/\mathrm{Sr}\cdot\left[2.69295 + 0.28304\cdot\left(^{87}\mathrm{Sr}/^{86}\mathrm{Sr}\right)_m\right] \tag{4}$$

provided that the (^{87}Sr/^{86}Sr)$_i$ is known, or can be estimated, then the time t elapsed from the formation of a rock or mineral can be determined from eq. (3):

$$t = 1/\lambda\cdot\ln\left\{1+\left[\left(^{87}\mathrm{Sr}/^{86}\mathrm{Sr}\right)_m - \left(^{87}\mathrm{Sr}/^{86}\mathrm{Sr}\right)_i\right]/^{87}\mathrm{Rb}/^{86}\mathrm{Sr}\right\} \tag{5}$$

Due to the different geochemical affinities of elemental Sr and Rb (see above), the Rb/Sr of either the rock-forming minerals of an igneous rock or a suite of cogenetic [i.e., starting with the same initial (^{87}Sr/^{86}Sr)$_i$ igneous rocks is expected to vary significantly (Fig. 4a, b, respectively).

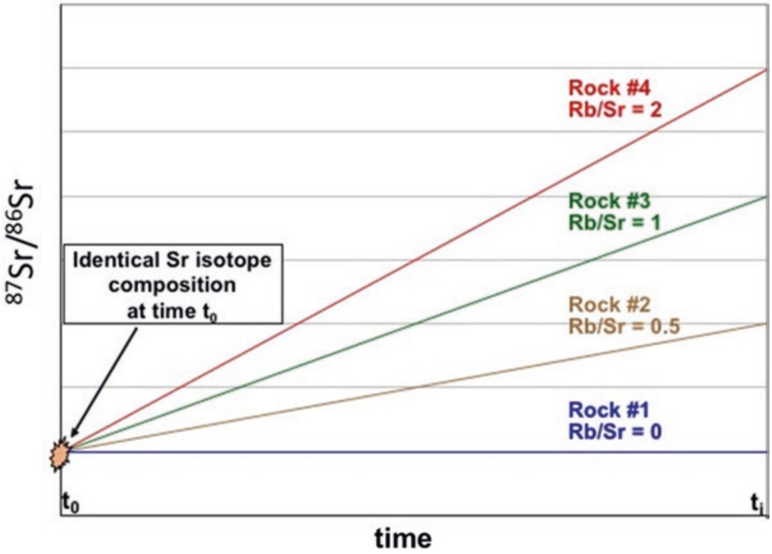

Fig. 3 Qualitative diagram highlighting the development of different Sr isotope compositions depending upon aging (from t_0 to t_1), and parent/daughter (Rb/Sr) ratios

This difference in Rb/Sr results in different time-dependent $^{87}Sr/^{86}Sr$ data points distributed along a straight line (eq. 3: i.e., an *isochron*) on a plot $^{87}Sr/^{86}Sr$ vs. $^{87}Rb/^{86}Sr$, and allows to calculate a robust regression line (York 1968; Ludwig 2003) yielding the intercept c with the x-axis and the angular coefficient m of the regression line, corresponding to $(^{87}Sr/^{86}Sr)_i$ and $(e^{\lambda t} - 1)$ parameters, respectively. Hence the time t elapsed since the crystallisation of the rock (Fig. 4a), or the differentiation of the suite of cogenetic igneous rocks (Fig. 4b), is given by:

$$t = 1/\lambda \cdot \ln\left(m+1\right) \tag{6}$$

Sr Isotopes in Earth's Materials

In addition to its use as an absolute radiometric clock, the Rb-Sr isotope system has proven as a robust tracer for the wide variety of geologic materials that had different parent-daughter ratios (^{87}Rb-^{87}Sr) and formed at different times during the 4.6 billion years of Earth's history (e.g., Faure 1998, 2001). Because of the different geochemical properties of Rb and Sr, their ratio in rocks can vary by 2–3 orders of magnitude (Table 1), resulting in significant differences in $^{87}Sr/^{86}Sr$. As a schematic example, the Earth's mantle has a relatively uniform and low $^{87}Sr/^{86}Sr$, and magmas formed by partial melting of the mantle, such as Mid-Ocean Ridge Basalts (MORB) are characterised by unradiogenic $^{87}Sr/^{86}Sr$ (0.702–0.7035) owing to the extremely

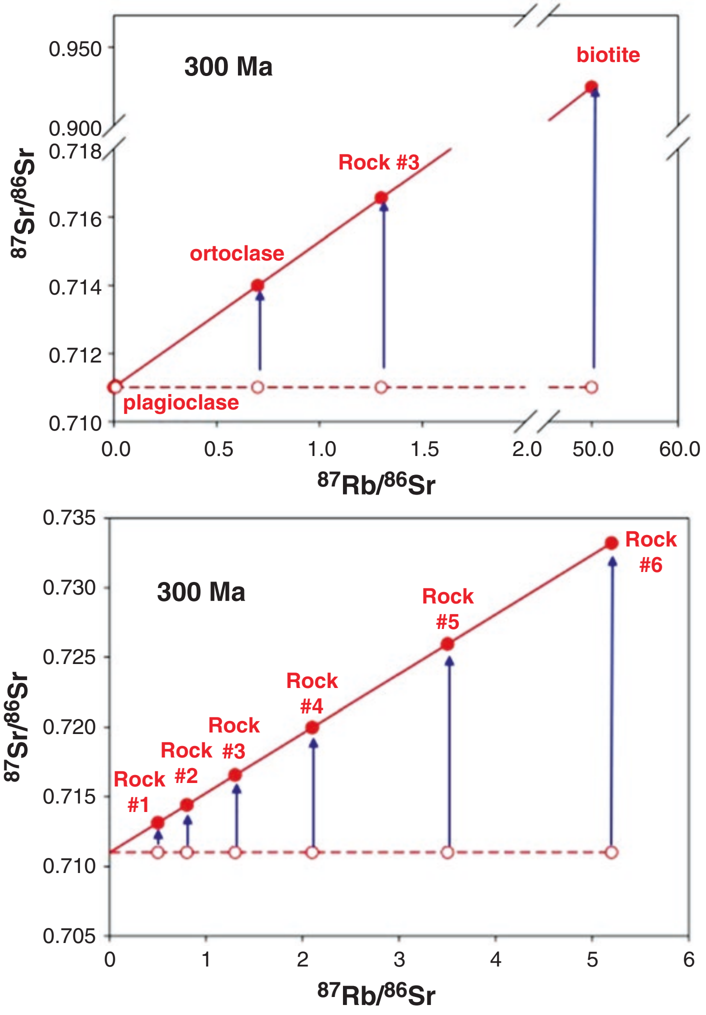

Fig. 4 Classic "isochron" plot for (**a**) a series of minerals crystallised in equilibrium at the same time (t_0) and evolved as a closed system for $t = 300$ Ma, and (**b**) a group of co-genetic magmatic rock with variable degree of differentiation and thus Rb/Sr (e.g. from basalt at low Rb/Sr to ryolite at high Rb/Sr). The slope of the regression line of the points at eh time t is proportional to the crystallisation age, whilst the intercept on the y axis represent the initial isotope ratio of the magmatic system [i.e., $(^{87}\mathrm{Sr}/^{86}\mathrm{Sr})_i$]. The variation of ^{87}Rb/^{86}Sr with time appears small (almost constant) with respect to that of ^{87}Sr/^{86}Sr: this is only due to the relatively small time (300 million years) considered, with respect to the half-life of ^{87}Rb (48.8 By), and to the difference in the scale of the two axes. After $t = 48.8$ billion years (geologically meaningless given the 4.6 Ga age of the Earth) each point would show ^{87}Rb/^{86}Sr halved with respect to its initial value

low Rb/Sr of this portion of the mantle (e.g., Fig. 5). In contrast, the continental crust (and the anatectic magmas formed from it) is characterised by a high (i.e., radiogenic) ^{87}Sr/^{86}Sr signature (>0.710) due to its high Rb/Sr and generally old age (e.g., Faure 1986; Dickin 2003; Fig. 5).

As far as sedimentary rocks are concerned, they also have extremely variable and heterogeneous Sr isotope composition (Table 1). The ^{87}Sr/^{86}Sr of clastic sedimentary rocks depends upon the Sr isotope composition of the source material outcropping in the feeding basin of the sediments. Conversely, the ^{87}Sr/^{86}Sr of Phanerozoic marine limestone (biochemical and chemical) are directly inherited from the sea water ^{87}Sr/^{86}Sr, which has varied throughout the Earth history depending on the variable contribution of different inputs (e.g. hydrothermal fluids at Mid Oceanic Ridges, Sediments inputs); therefore ^{87}Sr/^{86}Sr of carbonates may also provide qualitative information on their age of formation (Fig. 6; e.g., Banner 2004 and reference therein).

Overall, the ^{87}Sr/^{86}Sr in rocks of the continental crust vary generally between 0.703 and 0.750, from younger basalts with low ^{87}Sr/^{86}Sr ratios (around 0.703–0.704, Table 1) to granites with ^{87}Sr/^{86}Sr typically above 0.710, but that may reach extreme values (even >1.0) in particularly old granite with high Rb/Sr (e.g. Table 1). It is worth noticing that these variations are large and significant relative to the instrumental error of modern TIMS and MC-ICPMS measurements (typically ±40 ppm or better), but less so when compared to other less precise methods (e.g. Quadrupole ICPMS: see above). In addition, in old poly-mineralic rocks (e.g., granites), the ^{87}Sr/^{86}Sr of the single mineral phases making up the rocks might be significantly different, depending on their Rb/Sr (e.g., more radiogenic Sr in biotite than in feldspar: Fig. 4a).

This becomes important when considering the Sr content and Sr isotope composition of soils, which depends on the nature of the bedrock and reflects the complex processes occurring during weathering (e.g. hydrolysis of silicates, dissolution of carbonates, oxidation reactions, etc.).

As a general rule, each pedogenetic substrate has a distinct Rb/Sr ratio, and therefore a characteristic ^{87}Sr/^{86}Sr. For example, a soil derived from a 300 Ma granitic bedrock will have ^{87}Sr/^{86}Sr > 0.710 because of the high Rb/Sr of granites, resulting in a high production of radiogenic ^{87}Sr. In contrast, soils overlying a basaltic bedrock or a sedimentary carbonate one, which have low Rb/Sr ratios, will have ^{87}Sr/^{86}Sr < 0.710 owing to a low production rate of radiogenic ^{87}Sr.

The Sr isotope composition of a given soil, however, will not be necessarily identical to that of its bedrock, in particular when soils originate from rocks consisting of minerals with different Rb/Sr that experienced long (>10 Ma) ^{87}Sr/^{86}Sr evolution (i.e., different production rate of radiogenic ^{87}Sr, Fig. 3) and have different weathering potential (e.g., Borg and Banner 1996; Capo et al. 1998). As a result, the ^{87}Sr/^{86}Sr tends to vary considerably from soil to soil, and can be considered a robust parameter to fingerprint a given terrain.

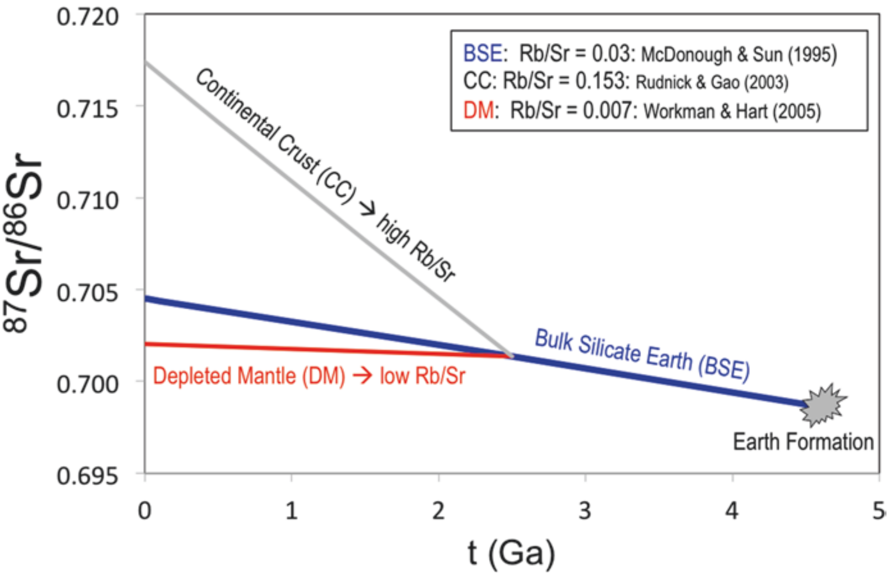

Fig. 5 Simplified Sr isotopic evolution model of the Bulk Silicate Earth (BSE; McDonough and Sun 1995) showing the effect of the hypothetical separation at 2.5 Ga of a continental crust with high Rb/Sr (0.44, Rudnick and Gao 2003) and a corresponding depleted mantle with low Rb/Sr (0.02, Workman and Hart 2005), which are let evolve till the present developing different $^{87}Sr/^{86}Sr$

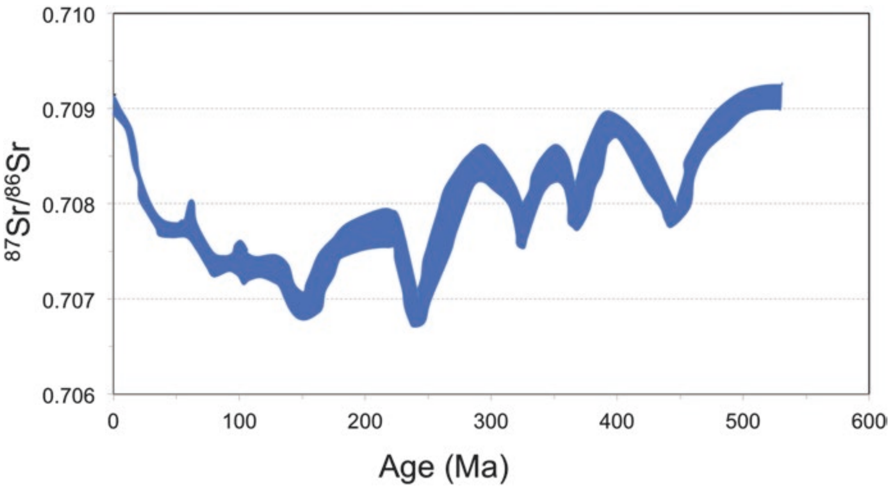

Fig. 6 Secular curve of seawater Sr isotope variation during the Phanerozoic based on analyses of marine carbonate and evaporate rocks and fossil (adopted from Burke et al. 1982). *PC* pre-Cambrian

Sr and Its Isotopes in the Biological Environment

In biological systems Strontium is neither a toxic nor an essential element, and bioavailable Sr is transferred from soils to plants, herbivores, and carnivores as a trace constituent that mimics the metabolic pathway of Ca due to their similar atomic properties (i.e. ionic radius and valence). Combined studies of the behaviour of Ca, Sr, and Ba in terrestrial trophic chains (Balter et al. 2002; Balter 2004, and references therein) have demonstrated that Sr/Ca and Ba/Ca ratios concomitantly decrease during bio-purification of Ca in the metabolic processes of organisms.

This has important implications to study complex predator/prey relationships within mammals, in addition to $\delta^{13}C$ and $\delta^{15}N$ stable isotope ratios, and highlights the potential role of not essential inorganic trace elements, such as Sr, to assess the relative contribution of different food sources to an animal's diet. This had been already perceived by Jean Anthelme Brillat-Savarin (1755–1826) in one of his famous aphorisms "*Dis-moi ce que tu manges, je te dirai ce que tu es*" from his work Physiologie du goût (The Physiology of Taste), published in 1825.

The study of the Sr isotope composition of a given trophic chain (soils, plants, herbivores, and carnivores) permits a step forward in assessing and tracing ecosystem processes. This is because Sr is a relatively high-mass element implying that kinetic and equilibrium fractionation of $^{87}Sr/^{86}Sr$ is negligible at the low temperatures of biological environments (unlike lighter elements such as H, C, N and O). Sr isotopes are, therefore, transferred from bedrocks and soils into biologically-available solutions to plants and animals without any measurable fractionation (i.e. retaining the same $^{87}Sr/^{86}Sr$; Graustein 1989; Graustein and Armstrong 1983; Hurst and Davis 1981; Kawasaki et al. 2002; Flockhart et al. 2015). Moreover, any possible mass-dependent fractionation in $^{87}Sr/^{86}Sr$ is corrected for during mass spectrometric measurement by routine normalization to the constant $^{88}Sr/^{86}Sr = 8.375209$ (e.g., Thirlwall 1991; Dickin 2003). This is to say that the measured $^{87}Sr/^{86}Sr$ reflects only variations in the amount of radiogenic ^{87}Sr present in a sample, which depends on both the age and Rb/Sr of its source. The net result is that the geochemical signature of Sr isotopes is paramount to fingerprint a geologic area because $^{87}Sr/^{86}Sr$ is conveyed unchanged by local temperature variations or internal biologic processes, from eroding geologic materials to soils and successive trophic levels.

This has set the basis to use Sr isotopes as a powerful tracer not only for geological studies but also, especially in the last decades, for a number of applications to archaeological, environmental, medical, forensic, and fishery sciences (e.g., Horn et al. 1993, 1998; Banner et al. 1994; Aberg et al. 1998; Capo et al. 1998; Beard and Johnson 2000; Tommasini et al. 2000; Almeida and Vasconcelos 2003, 2004; Evans et al. 2004; Hölzl et al. 2004; Kelly et al. 2005; Bentley 2006; Degryse et al. 2006; Kutschera and Müller 2006; Rummel et al. 2010; Voerkelius et al. 2010; Kennedy et al. 2011; Marchionni et al. 2013, 2016; Durante et al. 2015; Tescione et al. 2015; Petrini et al. 2016). In the next paragraphs we summarize some key studies that have applied Sr isotopes in these fields of science.

Food Sciences

Food authentication is an important and growing field within modern society. Health and safety of merchandise along with a reliable and certificate food provenance have gained increasing consideration in present-day European government legislation. This is important since some foods sourced from established areas tend to be more valued than those from other, less renowned, areas. There have been many studies characterizing foods using a Sr isotope. Plants uptake nutrients through their roots from the bioavailable fraction of the soil. Along with nutrients, inorganic not essential trace elements, such as Sr, are also adsorbed, making the Sr isotope composition of plants dependent upon that of the soils they grow. This constitutes a powerful fingerprint for traceability and food provenance studies (e.g., Kelly et al. 2005).

The most valuable and pioneering application of Sr isotopes to food sciences is related to the certification of authenticity and geographic provenance of high-quality wines, the commodity with the highest commercial value and whose market has strict regulations concerning the "*appellation contrôleé*"(e.g., Horn et al. 1993; Martin et al. 1999; Barbaste et al. 2002). Almeida and Vasconcelos (2004), Durante et al. (2013) and Marchionni et al. (2013, 2016) examined high-quality Portuguese and Italian wines, grapes and soils and showed that the Sr isotope composition of wine mirrors that of the soil, and provides a robust geochemical fingerprint of geographic provenance. Also, these authors established that during the wine making process the Sr isotope composition of wine remained unchanged.

In addition to wine authenticity, Sr isotopes have been applied to the traceability of an increasing number of food products. Olive oil is perhaps the other food commodity whose sensorial and nutritional properties (i.e., commercial value), akin high-quality wines, depend upon the geographical *terroir* (climate, bedrock, morphology of the terrain, etc.) and the specific cultivar. Unfortunately, the application of Sr isotopes to olive oil traceability was not extensively employed as that of high-quality wines, and only recently Medini et al. (2015) published a preliminary study on the traceability of French and Moroccan olive oil. Song et al. (2014), Bong et al. (2012) and Kawasaki et al. (2002) used Sr isotopes to trace the origin of hot peppers, cabbage and rice, whilst Fortunato et al. (2004) used Sr isotopes for tracking the food provenance of cheese from different European source regions. A comprehensive study integrating stable isotopes of light elements (C, N, O, S) and Sr isotope investigated the origin of butter from a variety of different worldwide sources (Roßmann 2001). Voerkelius et al. (2010) examined the Sr isotope composition of honey and wheat from specific sample regions and found a correlation with the Sr isotope composition of bioavailable Sr that is subsequently transferred into the food chain through plant uptake.

Anthropological and Bio-archaeological Sciences

As illustrated above, the Sr isotope composition of human and animal hard tissues is a function of their dietary habits (e.g., Capo et al. 1998), and, along with other stable and radiogenic isotope systematics, has been successfully applied to track migrational pathways, to assess the region of origin of archaeological population, and to study their hunting and trading areas, along with dietary habits (e.g., Price et al. 1994, 2000; Hoppe et al. 1999; Beard and Johnson 2000; Budda et al. 2000; Burton et al. 2003; Gregoricka 2013; McManus et al. 2013; Price et al. 2014; Slater et al. 2014; Waterman et al. 2014).

The rationale of these Sr isotope studies arises from the similar chemical properties of Sr and Ca, which allow Sr substitution for Ca in bioapatite [empirical formula $Ca_5(PO_4,CO_3)_3(OH, F)$], the mineral constituent of bones and teeth. Moreover, unlike bones and dentine, dental enamel forms during childhood (Hillson 1996), and remains unaltered throughout the years. Thus, Sr isotopes of dental enamel can be used as tracer of the trophic chain down to the soil (i.e., a specific geologic terrain) during the formation period of the teeth (Price et al. 2014; Slater et al. 2014; Waterman et al. 2014), being a fingerprint of the geological substrate in which the food ingested was grown. This possibility is also due to the fact that dental enamel is unaffected by post mortem diagenesis unlike dentine and bones (Budda et al. 2000; Sponheimer and Lee-Thorp 2006).

English et al. (2001) examined Sr isotope composition of roof timbers at the Native American site of Chaco Canyon, New Mexico, to determine the source of the felled trees. The Sr isotopes provided enough resolution to show that 2/3 of the logs originated in the Chuska Mountains and 1/3 from the San Mateo mountains, adding new insights into the sociological behaviour of the native Indians.

In addition to anthropological studies, Sr isotopes along with stable isotopes (C, N, S and H; Hobson 1999) are also valuable techniques to tackle the origins and migration of wildlife. Organisms moving between distinctly different food webs will acquire the isotopic composition of the different geological substrata. This technique can be applied to a range of different animals from insects to birds and large mammals. For example, Hoppe et al. (1999) determined the migration behaviour of mammoths and mastodons using Sr isotopes. Vogel et al. (1990) used a combination of N, C, Sr and Pb isotopes in elephant ivory and bones to determine the origin of elephant history and for examining different elephant populations.

Environmental Sciences

Aeolian mineral dust is an important component of the Earth's environmental systems, playing roles in the planetary radiation balance, as a source of fertilizer for biota in both terrestrial and marine realms and as an archive for understanding atmospheric circulation and paleoclimate in the geologic past. Crucial to understanding all of these roles of dust is the identification of dust sources. Present-day identification of dust sources can be readily identified by means of Earth-orbiting Satellites and back-trajectory analyses (wind fields modeling and air masses movement over periods of several days).

Identification of dust sources from the past requires novel approaches that are part of the geologic toolbox of provenance studies, namely mineralogical, geochemical, and radiogenic isotope analyses of dust deposits (Olivarez et al. 1991; Muhs et al. 2014). Physical properties include systematic spatial changes in dust deposit thickness and particle size away from a source. Mineralogy and geochemistry can pinpoint dust sources by clay mineral ratios and Sc-Th-La abundances, respectively. However, radiogenic isotopes (Sr, Nd, Pb) are among the most common and powerful tools to tackle the origin of dust archives in deep-sea cores, ice cores, and loess (e.g., Pettke et al. 2002; Grousset and Biscaye 2005; Chen et al. 2007; Meyer et al. 2011; Scheuvens et al. 2013, and references therein). All these methods have shown that dust sources have changed over time, with far more abundant dust supplies existing during glacial periods. Greater dust supplies in glacial periods are likely due to greater production of glaciogenic dust particles from expanded ice sheets and mountain glaciers, but could also include dust inputs from exposed continental and insular shelves now submerged.

The combination of the Sr, Nd and Pb isotope systems was used by Stille et al. (2012) to test the reliability of spruce (*Picea abies*) growth rings as environmental archives through time (from 1916 to 1983) in a forest ecosystem affected by acid atmospheric deposition. The coupled Sr-Nd isotope composition of tree rings of spruce permitted to reconstruct the chemical evolution of the uppermost soil compartments that are accessible for root uptake. The innermost rings have Nd and Sr isotope compositions closer to those of soils whilst the outermost rings have significantly lower Sr and Nd isotopic compositions and approach values similar to those of industrial or preindustrial "natural" aerosols like Saharan dust. Thus, both the Sr and Nd isotopic compositions of spruce tree rings record the chemical and isotopic changes in the soil and, therefore, allow identifying the principal sources of Sr and Nd in the wood.

Forensic Sciences

The application of Sr isotopes, along with other radiogenic isotope techniques, in forensic sciences is based upon employment of analytical methods for traceability studies well established in environmental, anthropological, bio-archaeological, and food sciences.

In forensic sciences it is of foremost importance to determine the source of a material or characterize its transportation history. Examination of fragments of material may help to determine these sources or history by a variety of techniques such as looking at colour, grain sizes, optical properties, and chemical composition. The last one has been widely used to determine the source of materials by fingerprinting the chemical composition of the material to be identified and comparing it to the chemical composition of potential sources. This approach was used extensively for major elements, trace elements, and stable isotopes (Ulrich et al. 2004).

The use of isotopes in forensic applications is relatively new. Forensic isotope geochemistry relies on the difference in isotopic composition of elements to characterize a particular material. These different isotopic abundances give rise to a unique isotopic composition that can identify a material having sourced from a particular region and help to trace its origin. Radiogenic isotope ratios have a major advantage over elemental ratios in characterizing a material since isotope ratios are significantly more sensitive tracers than elemental ratios or even elemental concentrations. These materials can vary from soils and rocks to bones, hair, feathers, wines, chocolate, pottery, metal ores and teeth among other substances (e.g., Pye and Croft 2004; Montgomery et al. 2006; Aggarwal et al. 2008; West et al. 2009).

The Sr isotope composition measured in skeletal elements (e.g., bone, teeth, or antlers) can be used to infer the geographic region that an animal or human inhabited, because different regions tend to have distinct Sr isotope compositions, and natural variations in the relative abundance of Sr isotopes are not changed as Sr is processed through the food chain (e.g., Beard and Johnson 2000). Therefore, an organism that ingests Sr from one region can have a Sr isotope composition that is different than that of an organism that ingests Sr from another region. The Sr isotope composition of skeletal elements is a reflection of the concentration-weighted average of dietary Sr that was ingested while that skeletal element was produced. Because different skeletal elements grow and exchange Sr at different stages during the life times of organisms, Sr isotope analysis of different skeletal elements can be used to infer changes in geographic location at different stages in an organism's life. The Sr isotope composition measured in human teeth enamel will reflect the average Sr isotope composition that was ingested as a child, due to the immobile nature of Sr and Ca in teeth enamel after formation, whereas the Sr isotope composition of bone will reflect the average isotopic composition over the last 10 years of life, due to continuous biological processing of Sr and Ca in bone (e.g., Grupe et al. 1997; Aberg et al. 1998; Price et al. 2000; Prohaska et al. 2002; Bentley 2006).

However, with modern populations, the Sr isotope technique can be less successful due to the introduction of packaged foods in the diet that shifts the Sr isotope signature away from the local geology and inhibits the usefulness of a soil or faunal sample baseline. Nonetheless, a positive result can be acquired if the Sr isotope composition is oriented to measure within population variation (control groups) to determine a statistically acceptable baseline of a given socio-cultural and geographic area. This could help to account for the affinity of a selected population towards certain local and packaged foods. This is not to suggest that boundaries assigned through socio-cultural and geographical means will maintain unique and individual geochemical signatures but rather, from a socio-cultural standpoint, this categorization of people can provide a baseline for the analysis.

As an example of successful radiogenic and stable isotope application in forensic sciences, Font et al. (2015) carried out a multi-isotope investigation (Sr and Pb isotopes and $\delta^{18}O$, $\delta^{13}C$ and $\delta^{15}N$) to bones and teeth from an unidentified male found drowned in the "*IJ*", Amsterdam, The Netherlands in March of 1999. The individual remained unidentified until mid 2013, after the isotope study was completed. Coupled $\delta^{13}C$ and $\delta^{15}N$ values in bone collagen recovered from rib and femur were consistent with an omnivore living in a region where C3-type diet dominates (i.e., Europe). Integrated Sr and Pb isotopes and $\delta^{18}O$ values in canine, third molar teeth and femur and rib bone data excluded extended residence in north-west Europe and particularly The Netherlands. Characteristic Pb isotope ratios coupled with inferred $\delta^{18}O$ values of drinking water argued for a most probable place of origin for the unidentified individual in west and south Poland, south-east Slovakia and the region of Ukraine–Romania–Bulgaria, specifically the region associated with the Carpathian Mountains. Independent investigations of the Amsterdam Police achieved a positive identification of the individual, successively confirmed by DNA analyses. The individual originated from south-west Poland, where he spent most of his life. The region lies within the area proposed by the study of Font et al. (2015) and consequently provides strong validation of the potential power of the combined multi-isotope methodology to forensic sciences.

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