

Chapter 3

Mobility and Bioavailability of the Chernobyl-Derived Radionuclides in Soil–Water Environment: Review



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Abstract The chapter reviews the studies of the behavior of Chernobyl-derived radionuclides in the soil–water environment that have been carried out over more than 30 years after the accident at the Chernobyl nuclear power plant on 26 April 1986, the worst nuclear accident ever. As a result of post-Chernobyl investigations, the role of released fuel particles in the environmental behavior of radionuclides has been revealed. A conceptual model accounting for transformation of radionuclide chemical forms in soils and sediments is outlined and data on key kinetic and equilibrium parameters for this model are presented. Newly introduced after Chernobyl accident, parameters of radiocesium selective sorption by soils and sediments (capacity of frayed-edge sites [FES] and radiocesium interception potential [RIP]) and methodologies for their determination and application are discussed. Another important addressed issue is establishing the exchangeable distribution coefficient K_d^{ex} , which can be estimated on the basis of ion-exchange equilibrium from commonly accepted characteristics of soils and sediments. Development of methods to parameterize radionuclide bioavailability through soil and soil solution characteristics is also examined. The reviewed advances in post-Chernobyl studies help to reduce significantly the uncertainty associated with predicting radionuclide fate and transport in the environment.

Keywords Chernobyl · Radiocesium · Radiostrontium · Speciation · Fuel particles · Transformation · Sorption · Fixation

3.1 Introduction

In terrestrial and aquatic environment, behavior of accidentally released radionuclide is governed by the ratio of its chemical forms in fallout and site-specific environmental characteristics determining the rates of leaching and fixation–remobilization,

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as well as sorption–desorption of radionuclide (its solid–liquid distribution) (Konoplev et al. 1992; Beresford et al. 2016).

In terms of environmental conditions, the major part of the close-in areas contaminated due to the Chernobyl accident occurs in Pripyat Polesye. By terrain, this is outwash lowland plain. The vegetation is primarily broad leaved and pine woods located on drained terraces of alluvial origin. The climate is moderate continental, with the mean annual precipitation of 500–600 mm. The water table is 8 m at maximum, the groundwater outcropping in Pripyat floodplain depressions, forming lakes. In the Chernobyl region 45% of the territory is occupied by forest, bogs, and water bodies, while 54% are arable lands. The soil cover is represented by soddy weakly podzolic and medium podzolic soils (53% of arable lands), whereas soddy and meadow soils account for 30%, boggy soil—10%, and weakly sodded sands—7% (Konoplev 1998).

3.2 Speciation of Chernobyl-Derived Radionuclides in the Environment and Their Transformation

3.2.1 Fuel Particles: Major Distinguishing Feature of the Chernobyl Release

The speciation of radionuclides released to air from the damaged reactor varied with time after the accident (Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a, b). In the process of atmospheric transport, particles depositing on the surface were differentiated by size and density. Therefore, the fallout was not uniform and was a function of direction and distance from the ChNPP. All hot particles deposited on the surface after the accident could be attributed to two main types: fuel particles and condensation particles (Loschilov et al. 1991; Kashparov et al. 1996). Fuel particles were mainly particles of released uranium dioxide nuclear fuel containing its decay products at ratio close to irradiated fuel makeup, with some depletion of volatile fissile products of ^{131}I , $^{134,137}\text{Cs}$, ^{106}Ru , etc. (Bogatov et al. 1990; Konoplev and Bobovnikova 1991; Victorova and Garger 1991; Sandalls et al. 1993; Salbu et al. 1994, 2018; Kashparov et al. 1996; Salbu 2001; Beresford et al. 2016).

Due to high temperature in the damaged reactor (about 2000 °C), a large amount of volatile fission products (iodine, cesium, ruthenium, etc.) were released to the atmosphere, part of which were precipitated on dust particles—inert carriers in air. The resulting condensation particles were similar by their properties to those formed in the last stage of a nuclear explosion, characterized by surface contamination and relatively low activity concentration as compared to fuel particles (Kashparov et al. 1996). The condensation particles formed as a result of the Chernobyl accident contained 1–2 radionuclides and resembled global fallout following the nuclear weapons tests. For this reason, predicting behavior of these particles could be

based on abundant data about the global fallout (Pavlotskaya 1974; Wicker and Schultz 1982). As regards fuel particles, prior to the Chernobyl accident, such particles were released to the environment only during the accidental release in Windscale (UK) in October 1957 and destruction of the satellite Kosmos 954 in 1978 (Sandalls et al. 1993; Salbu et al. 1994). These releases, however, were small scale and the environmental behavior of particles was not well understood. Because of this, the behavior of fuel particles determining the features of the Chernobyl footprint and occurring primarily within 60 km around the ChNPP was a major scientific challenge.

The occurrence of fuel particles in the Chernobyl depositions led to considerable nonuniformity of area radioactive contamination. Nonuniformity of soil contamination was seen over distances from several meters to macroscale (Izrael et al. 1990; Salbu et al. 1994; Ivanov 1997; Kashparov et al. 2018). Particle size and makeup were a function of direction and distance to the reactor, and therefore the behavior of radionuclides in soil and their characteristics determining transfer to plants and aquatic systems varied (Beresford et al. 2016).

Formation of particles at elevated temperatures in airflow was discovered in 1960 (Megaw et al. 1960). When heated to 600–1000 °C, particles with a median diameter of 100–600 µm were formed. Higher temperatures, in the authors' opinion, were conducive to formation of larger particles. This, however, has not been supported by the studies (Kashparov et al. 1996) in which oxidation of uranium fuel at 400 and 900 °C led to formation of particles with lognormal size distribution. The median size of particles was not dependent on temperature, decreased from 10 to 3 µm with increase in the annealing time from 3 to 21 h. The researchers (Kashparov et al. 1996) concluded that the main process of fuel particle formation during the accident was high-temperature oxidation of nuclear fuel.

The diameter of fuel particles in the first years after the accident varied from hundreds to fractions of micrometers (Loschilov et al. 1991; Sandalls et al. 1993; Kashparov et al. 1996). The particle density was found to be as high as 10^5 particles/m² within the 30-km zone of ChNPP (Victorova and Garger 1991). With distance from the reactor, the fraction of fuel component declined, and particle size distribution changed (Sandalls et al. 1993).

The presence of fuel particles in the Chernobyl release led to a lower mobility of radionuclides, as compared to the global fallout (Konoplev et al. 1992; Beresford et al. 2016), since fuel particles are insoluble in water. From environmental standpoint, this played a positive role in the early phase after the accident by reducing secondary contamination of water bodies and root transfer of radionuclides from soil to plants (IAEA 2006). With time, however, radionuclides tend to leach from fuel particles. Leaching of radionuclides from fuel particles is a crucial process because it influences long-term changes in mobility and bioavailability of radionuclides in terrestrial and aquatic ecosystems (Bobovnikova et al. 1991a; Avdeev et al. 1990a, b; Smith et al. 2009).

3.2.2 *Basic Chemical Forms of Radionuclides and Their Transformation in the Environment*

Physicochemical state of radionuclides controls their mobility and bioavailability in the environment. Ratio of different chemical forms is a function of the initial state of radionuclide in depositions and its changes due to transformation processes occurring in the environmental compartments (Konoplev and Bobovnikova 1991; Konoplev et al. 1992; Beresford et al. 2016).

Radionuclides can occur in the environment in the dissolved state and within the solid phase. When in solution, radionuclide can occur as a cation, as a complex compound with organic and inorganic ligands and colloid particles. In the solid phase, radionuclide can occur in the exchangeably sorbed state, i.e., at adsorption-desorption equilibrium with solution; be embedded in insoluble compound or insoluble hot particles (such as fuel particles), or fixed by clay mineral particles (nonexchangeably sorbed) (Konoplev et al. 1988, 1992; Bobovnikova et al. 1991a).

The long-lived radionuclides of radiocesium and radiostrontium, which were of most environmental significance after the Chernobyl accident, occur in solution mainly as cations and virtually do not form stable complex compounds. Conducted laboratory and field experiments (Konoplev et al. 1996; Matsunaga et al. 2004) have shown that the colloid form of these radionuclides, as a rule, does not play a major role in most of environmental conditions.

Therefore, with all the diversity of radiocesium and radiostrontium speciation in soil and water bodies, in terms of radionuclide transport, exchange between environmental compartments, and biological availability, it seems appropriate to distinguish the water-soluble, exchangeable, and nonexchangeable forms.

The ratio of key chemical forms of individual radionuclides is governing their mobility in the environmental compartments. This ratio, in turn, depends on the initial state of radionuclides in depositions and its change due to physicochemical processes occurring in soil or bottom sediments. For separation of different chemical forms, the method of sequential extraction by different chemical agents is usually used. In many works (Hilton et al. 1992; Oughton et al. 1992; Salbu et al. 1994; Askbrant et al. 1996; Tsukada et al. 2008), the technique proposed by Tessier et al. (1979), or its modification, was used to separate chemical forms of heavy metals, including the following extraction stages:

1. Deionized water for separation of water-soluble compounds (solid phase: liquid phase ratio 1:10).
2. 1 N ammonium acetate solution to separate exchangeable form of radionuclides (sorbed by ion-exchange mechanism), phase ratio 1:10.
3. Acetate buffer NaOAc + HOAc at pH = 5 for extracting carbonates.
4. 0.04 M solution of hydroxylamine $\text{NH}_2\text{OH}\cdot\text{HCl}$ in 25% acetic acid to extract easily reducible compounds (solid to liquid phase ratio 1:10).

5. Mixture of 30% hydrogen peroxide solution reduced to pH = 2 by nitric acid with 3.2 M ammonium acetate solution in 20% HNO₃ at ratio 3:1 (phase ratio 1:10) to extract radionuclides bound to organics.
6. 7 M nitric acid solution to extract acid soluble fraction of radionuclide (phase ratio 1:10).

Other authors (Pavlotskaya 1974; Konoplev et al. 1988, 2002; Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a) used the methodology for separating chemical forms of radionuclides in the solid phase of the environmental compartments (atmospheric aerosols, soil, bottom sediments suspension etc.) consisting of four stages:

- Extraction by distilled water at solid phase:water ratio 1:5 for 24 h.
- Extraction of 1 N NH₄Ac solution at phase ratio 1:8 for 24 h.
- Extraction by solution 6 N HCl with boiling, phase ratio 1:5.
- Extraction by mixture of nitric and hydrofluoric acid until total decomposition.

In the process of sequential extractions, cations desorbed by ion-exchange mechanism and soluble complex compounds of radionuclides (neutral and anionic forms) pass into the aqueous extract. Concentration of cations in the aqueous extract solution is matching the position of ion-exchange equilibrium governed by soil cation-exchange capacity and concentration of exchangeable ions in solution. The key exchangeable ions in soil are known to be Ca²⁺, Mg²⁺, Na⁺, K⁺, NH₄⁺, and H⁺. Also extracted by water are colloid forms of radionuclides, i.e., particles of the size smaller than filter pores (ranging from 1 to 100 nm). These particles are normally transported by water flow, yet their biological availability is much lower than that of truly dissolved radionuclides.

Radionuclides extracted by ammonium acetate solution are those adsorbed in soil by ion-exchange mechanism, while nonexchangeable radionuclides, i.e., the forms, which do not normally pass to water in the environment, are extracted by acid. The nonexchangeable form, in turn, includes radionuclides incorporated in poorly soluble compounds, fuel particles, and the so-called irreversibly sorbed states (inclusions in the lattice of minerals, radionuclide-organic compounds with insoluble organic matter of soil, etc.) (Konoplev et al. 1988).

The initial ratio of radionuclide chemical forms in the environmental compartments is determined by speciation in atmospheric deposition. In the global fallout after nuclear tests, the water-soluble and exchangeable forms of ¹³⁷Cs and ⁹⁰Sr were predominant, as shown by Pavlotskaya (1974).

During the Kyshtym accident caused by explosion in a tank with high-level radioactive waste, radioactive materials were released in the form of liquid pulp to the height of 1–2 km and formed a plume of liquid and solid aerosols (Alexakhin et al. 2001; Il'in and Gubanov 2001). Data about the speciation of ⁹⁰Sr and other radionuclides in Kyshtym fallout do not exist. It is known, however, that radionuclides in the released aerosols were present primarily in the form of highly soluble compounds such as nitrates and acetates (Martyushov et al. 1995).

Table 3.1 Comparison of initial ^{90}Sr and ^{137}Cs mobility in fallout of different origin

Type of fallout	Part of mobile forms, %		Reference
	^{90}Sr	^{137}Cs	
Kyshtym	~80–90	~80–90	Pavlotskaya (1974)
Global fallout	>90	>50	Pavlotskaya (1974)
Chernobyl: 30-km zone	11.2	26.8	Konoplev and Bobovnikova (1991)
Western Europe	–	85	Hilton et al. (1992)

The radionuclides of ^{137}Cs and ^{90}Sr deposited on soil surface after the Chernobyl accident occurred mainly in nonexchangeable form, as follows from the analysis of fallout samples collected in April–May 1986 at Chernobyl meteorological station (Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a, b). This is also suggested by data on radionuclides speciation in soil at the same location in the first year after the accident (Bobovnikova et al. 1991a). Since ^{137}Cs tends to be easily fixed in soil, i.e., transferring to nonexchangeable form, the ratio of exchangeable and nonexchangeable forms in soil does not change much with time as a result of ^{137}Cs leaching from fuel particles.

Since the compounds of Cs isotopes are more volatile than those of Sr, essential part of deposited Cs occurred in mobile form embedded in condensation particles (see Table 3.1), and hence radiocesium was transported over longer distances than strontium. Radiostrontium, on the other hand, largely deposited within the 30-km zone and only a small part of this radionuclide was transported to southern areas of Belarus and Bryansk region (Izrael et al. 1990).

Hilton et al. (1992) analyzed ^{134}Cs and ^{137}Cs chemical forms in fallout in UK at the time of the Chernobyl accident. More than 85% of these isotopes were found to be in water-soluble and exchangeable forms, suggesting condensation origin of depositions on remote parts of the Chernobyl footprint. Unfortunately, there are no data available about radionuclide speciation in Chernobyl fallout at medium range, such as Bryansk region of the Russian Federation or central areas of Belarus. It can be assumed that in these regions the situation was “half-way” between the near zone (Chernobyl) and remote footprint (UK). The proportion of the mobile ^{137}Cs in depositions in Bryansk region can be expected to be 40–60% (Konoplev et al. 1996), which was actually supported by the analysis of chemical forms in soils in the first years after the accident (Konoplev et al. 1988; Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a). The initial forms of ^{90}Sr and ^{137}Cs in depositions of different types are compared in Table 3.1.

Therefore, when analyzing radionuclide speciation in soils of the Chernobyl-contaminated areas, two factors should be considered:

1. Nonuniformity of radionuclide speciation in the fallout as a function of distance from ChNPP.
2. Variety of soil sorption and fixation ability on different locations.

In the first years after the Chernobyl accident, ^{90}Sr speciation in the 30-km zone soils was notable for high proportion of nonexchangeable forms as compared to global fallout and East Ural Radioactive Trail (Konoplev et al. 1988; Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a).

A relatively high content of water-soluble ^{90}Sr forms was typical of sandy and boggy soils, which seems to be associated with a weak sorption capacity of sandy soils and high mineralization of aqueous extract in boggy soil (in particular, high concentration of Ca and possible formation of colloids: strontium-organic compounds) (Konoplev et al. 1988, 1992).

For ^{137}Cs the fraction of nonexchangeable form is even higher, being as high as 99% in some cases (Konoplev et al. 1988; Bobovnikova et al. 1991a). A higher proportion of ^{137}Cs nonexchangeable form, as compared to ^{90}Sr , was seen in the global fallout too (Pavlotskaya 1974), which is explained by fixation processes typical of radiocesium, i.e., transfer of exchangeable form to nonexchangeable due to incorporation into the interlayer space of clay mineral crystal lattice (Tamura and Jacobs 1960; Jacobs 1962; Sawhney 1972; Von Reichenbach 1968). Even though the part of ^{137}Cs mobile forms in the fallout was much higher than that of ^{90}Sr , as a result of ^{137}Cs fixation with time the proportion of ^{137}Cs mobile forms becomes much lower than that of ^{90}Sr . What is more, while leaching of ^{90}Sr from fuel particles leads to increase in its mobility in soil, leached ^{137}Cs immediately passes to nonexchangeable form, i.e., time scale of its fixation (several weeks or months) is much shorter than time scale of leaching from fuel particles (Konoplev et al. 1992; Beresford et al. 2016).

Subsequent to fallout on the surface of soil or water bodies, radionuclides become involved in physicochemical and biological processes, which lead to changes in their speciation. The dissolved fraction of radionuclide is subjected to sorption by solid particles, the main mechanism of sorption being ion exchange (Sawhney 1972; Konoplev et al. 1992). Exchangeable form undergoes fixation and transfer to colloid forms or complex compounds with organic and inorganic ligands. Fuel particles disintegrate, the effective surface of particle contact with water increases, and additional amount of radionuclide passes to solution. The main processes of radionuclide chemical forms transformation are shown in Fig. 3.1. In terms of speciation, fixation of radionuclides is transfer of their exchangeable form to nonexchangeable one.

Apart from the differences in radionuclide speciation between Chernobyl fallout and the global fallout, the radionuclides also differ by the pattern of their transformation in soil. The mobility of radionuclides from nuclear weapons testing decreased with time because of fixation by soil components, while in the Chernobyl zone in the first years after the accident the predominant process was leaching of radionuclides from fuel particles, resulting in increased mobility, especially for ^{90}Sr (Konoplev et al. 1992; Kashparov et al. 1999, 2004; Konoplev and Bulgakov 1999).

The main chemical forms of radionuclides shown in Fig. 3.1 differ by mechanisms and rates of migration in environmental compartments. Therefore, when predicting radionuclide transport in the environment and food chains, it is essential to measure, or predict, the ratio of different chemical forms for each radionuclide of

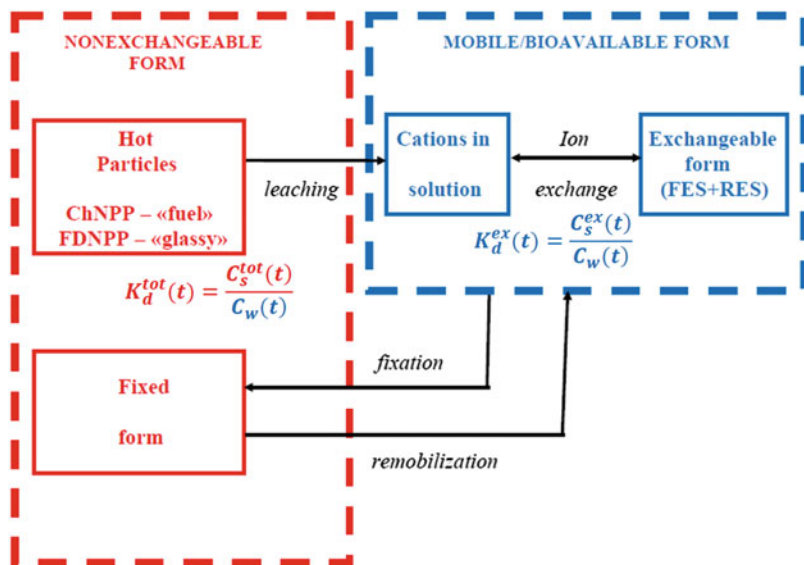


Fig. 3.1 Conceptual model of transformation processes for radiocesium speciation in soils/sediments (modified and updated after Konoplev et al. 1992, 1993; Konoplev and Bulgakov 2000b)

interest. In turn, the key parameters of radionuclide behavior used for migration prediction such as the distribution coefficient (K_d), wash-off coefficients, soil–plant transfer factor, and bioconcentration factors are governed by the speciation ratio to a great degree. Therefore, determination and parameterization of rates of transformation processes shown in Fig. 3.1, specifically leaching from fuel particles, sorption–desorption, fixation and remobilization, was an important task after the Chernobyl accident (Konoplev et al. 1992; Salbu et al. 2004).

3.2.2.1 Kinetics of Radionuclide Leaching from Fuel Particles

In the first years after the accident, radionuclide leaching from fuel particles was the key process with respect to radionuclide chemical form transformation in the environment. Radionuclides are released from fuel particles by three mechanisms: (1) due to diffusion from particle to the surface and passing into solution; (2) by mechanical disintegration of particle and correspondent increase in surface of interphase interaction, and (3) with dissolution of fuel matrix (Konoplev and Bulgakov 1999). Pure uranium dioxide is known to dissolve slowly even in concentrated acids (Vdovenko 1960). In natural conditions, however, four-valent uranium U(IV) is easily oxidized by air oxygen to six-valent U(VI) state, three-oxide of which is better dissolving in aqueous media. Crystals of UO_2 , when in water, are quickly covered by oxide film of U_{2+x} , where $0 < x < 1$ and the rate of further oxidation is determined by diffusion of oxygen and surface area of crystal. In

addition to oxygen, UO_2 can be oxidized by other oxidizers such as three-valent iron Fe (III) or hydrogen peroxide, molecules of which can be formed in the vicinity of hot particle as a result of water radiolysis. Dissolution of oxidized uranium is facilitated by the presence of inorganic carbonates and sulfates, as well as organic ligands (Vdovenko 1960).

Dissolution of fuel matrix has been confirmed experimentally for the first time by the presence of uranium in the aqueous extract from a soil (chernozem from the vicinity of the ChNPP) that was in contact with hot particles for 290 days (Avdeev et al. 1990a). In that case, $(1-2) \times 10^{-2}\%$ of cesium and ruthenium isotopes, $(3-4) \times 10^{-3}\%$ of ^{95}Zr and ^{144}Ce , $(4-7) \times 10^{-3}\%$ of total α -activity, and $(1-4) \times 10^{-3}\%$ of uranium were leached from hot particles. Better passage to solution was seen for radionuclides from graphite particles: 1.7–4.0% for cesium isotopes; 0.6–1.5% for ^{106}Ru , 0.9–1.7% for ^{95}Zr , and 1.2–1.8% for ^{144}Ce . It has also been shown that ^{144}Ce , ^{106}Ru , and to a lesser degree cesium isotopes are leached from high-density fuel particles more slowly as compared to particles with loose structure.

Bogatov et al. (1990) stated that the main mechanism of radionuclide release from fuel particles is dissolution of uranium matrix. In their study, the dissolution rate was estimated to be $(0.57-16) \times 10^{-5} \text{ g/cm}^2 \text{ day}$ at contact of particles of different size with solutions modeling aqueous media.

The diffusion release rate for radionuclides was determined by these researchers using the method of layer dissolution of particles (Bogatov et al. 1990). Based on depletion of surface layers the diffusion coefficients were estimated to be close and equal to $(2-4) \times 10^{-17} \text{ cm}^2/\text{s}$ for ^{137}Cs , ^{90}Sr , $^{238,239,240}\text{Pu}$, ^{241}Am . This value seems to be a slight underestimate, because besides diffusion the most depleted layers are subjected to dissolution in soil and bottom sediments and accordingly the radionuclide concentration gradient in particles is increasing.

The ability of fuel particles, being aggregates of fine crystals of uranium oxides, to break down easily at physical impact was indicated by many authors. Breakdown of particles was due to exposure of soil sample to ultrasound and cooling-heating cycles (Petryaev et al. 1993). The ability of fungi, widespread in the 10-km zone around the Ch NPP, to grow in immediate vicinity of hot particles and cause their breakdown was reported by Borisyuk et al. (1990). Breakdown of fuel particles is facilitated by a large number of radiation-thermal defects in their structure. As a result of disintegration, the mean size of hot particles decreases and the part of particles with high activity declines. Consequently, the rate of radionuclide release from fuel matrix by both mechanisms (diffusion and dissolution) should be growing.

Chernobyl-derived fuel particles did not dissolve in water or neutral solution of salts and therefore the proportion of mobile forms of radionuclides (exchangeable and water-soluble) in the soils of Chernobyl close-in areas in the first years after the accident was much lower than in similar soils after the global fallout or Kyshtym accident (Konoplev et al. 1988; Petryaev et al. 1993). This difference could be used for quantifying the part of radionuclides embedded in fuel particles (Konoplev and Bobovnikova 1991; Bobovnikova et al. 1991a; Konoplev et al. 1992; Kashparov et al. 1999, 2000, 2004; Konoplev and Bulgakov 1999). Data on ^{90}Sr speciation are

preferable to be used because this radionuclide has sufficiently long half-life (28.8 years) enabling to follow changes in its exchangeability in soil over extended time period. Sorption equilibrium of radiostrontium in soils is achieved in several days (Frere and Champion 1967) and therefore its speciation, except for embedded in fuel particles, can be assumed to be at equilibrium. Furthermore, the steady-state fraction of exchangeable radiostrontium is 80–100% in most of soils and weakly dependent on soil characteristics (Pavlotskaya 1974).

The fraction of ^{90}Sr embedded in fuel particles in different points of the Chernobyl-contaminated zone can be calculated following the equation (Konoplev and Bulgakov 1999)

$$F_t = 1 - \frac{\alpha_{\text{ex}}^t}{\alpha_{\text{ex}}^\infty}, \quad (3.1)$$

where F_t is the fraction of ^{90}Sr present in soil and embedded in fuel particles at the time moment t ; α_{ex}^t and $\alpha_{\text{ex}}^\infty$ are the fractions of exchangeable ^{90}Sr at the time moment t and at equilibrium, e.g., after fuel particles decomposition in soil, respectively.

The exchangeable ^{90}Sr fraction at a given time moment (α_{ex}^t) was determined by extraction of 1 M ammonium acetate solution using the technique discussed above at phase ratio 1:8 and extraction time 24 h.

The equilibrium fraction of ^{90}Sr exchangeable form ($\alpha_{\text{ex}}^\infty$) can be determined by three different methods. The most accurate way is spiking of a tracer such as ^{90}Sr in the form of solution and determining the species ratio after equilibrium is achieved (Kashparov et al. 1999, 2004). Another method of $\alpha_{\text{ex}}^\infty$ determination consists in approximation with the stationary fraction of exchangeable ^{90}Sr in the lower soil layers not containing fuel particles having low mobility (Konoplev et al. 1992; Konoplev and Bulgakov 1999).

Modeling of the processes resulting in radionuclide release from hot particles is complicated by the diversity of their sizes, shapes, and chemical characteristics. For this reason, it would be appropriate to use integral parameters accounting for the rate of radionuclide leaching from fuel particles in different parts of the contaminated zone. Such parameters could be the first-order rate constants k_1 (year^{-1}) or fuel matrix dissolution rate v ($\text{g}/\text{cm}^2 \text{ year}$) (Konoplev and Bulgakov 1999).

In the process of breakdown or dissolution of fuel particles, their size tends to decrease, and relative content of radionuclides embedded in most stable particles increases. The first process leads to an increase in the k_1 value, whereas the second one—to its decrease. The assumption of mutual compensation of these two processes is the basis for using the first-order kinetics for describing radionuclide-leaching rate from fuel particles. In this case, a decrease in the fraction of radionuclide embedded in particles as a function of time follows the equations (Konoplev and Bulgakov 1999)

$$\frac{dF_t}{dt} = -k_1 F_t, \quad (3.2)$$

and hence

$$F_t = F_0 e^{-k_1 t}, \quad (3.3)$$

where F_t and F_0 are the fractions of radionuclides in fuel particles at time moment t and initial depositions, respectively; t is the time since the accident.

If particles have approximately the same size and dissolution resistance, the leaching rate will increase as a function of time due to increase in the ratio of surface area of particles and their weight. In this case, it is better to use dissolution rate normalized over particles surface area v (g/cm^2 year). By definition (Konoplev and Bulgakov 1999),

$$v = \frac{1}{S} \cdot \frac{dP_t}{dt}, \quad (3.4)$$

where P_t is the particle weight at time moment t ; S is the particle surface area.

Assuming that the radionuclide is distributed uniformly in a particle volume and considering that

$$\frac{dP_t}{dt} = \rho S \frac{dR_t}{dt}, \quad (3.5)$$

and

$$\left(\frac{F_t}{F_0}\right)^{1/3} = \frac{R_t}{R_0}, \quad (3.6)$$

where R_t and R_0 are the current and initial radius of the particle, respectively, ρ is the fuel matrix density, F_0 is the initial fraction of the radionuclide embedded in fuel particles:

$$\left(\frac{F_t}{F_0}\right) = 1 - \left(\frac{v}{\rho R_0}\right)t. \quad (3.7)$$

Thus, with particles characterized by a single effective size and assuming the uranium matrix dissolution rate for unit surface to be constant under given conditions, the time dependence of $(F_t/F_0)^{1/3}$ should be linear. The analogue of the rate constant in this case is normalized dissolution rate $w = v/\rho R_0$.

On the other hand, when radionuclide leaching is described by the first-order kinetics, the time dependence of $\ln(F_t)$ should be linear in accordance with the following equation:

$$\ln(F_t) = \ln(F_0) - kt. \quad (3.8)$$

Applicability of these models for predicting radionuclide leaching from Chernobyl-derived fuel particles was validated by comparison of experimental and theoretical curves representing the changes in the concentration of ^{90}Sr embedded in fuel particles in soil samples collected in 1987–1991 near the settlement of Benevka and on the experimental plot near Korogod (Konoplev and Bulgakov 1999). Results of regressive analysis of the time changes in F_t/F_0 by Eqs. (3.7) and (3.8) showed a good agreement between experimental and theoretical dependencies (Konoplev and Bulgakov 1999). In this regard, Eq. (3.7) showed a slightly better agreement of calculation results and experimental data. The values of correlation coefficients, for the most part, are higher for Eq. (3.7).

What is also in favor of Eq. (3.7) is a good fit of the values of dissolution rate constants calculated by ^{90}Sr speciation changes in soil to those determined in laboratory experiment in which fuel particles separated from Chernobyl close-in area soils were exposed to solutions modeling natural aqueous media (Bogatov et al. 1990).

A great amount of data on radiostromtium speciation in soils of contaminated areas were used to obtain site- and soil-specific rate constants of radionuclide leaching from fuel particles k_1 , which ranged from 10^{-3} to 10^{-4} day^{-1} (Konoplev and Bulgakov 1999) or 0.05 – 0.3 year^{-1} (Kashparov et al. 2004) depending on location and soil type.

Konoplev et al. (1992) used data on dynamics of ^{137}Cs speciation in soils of the 30-km zone to reveal the rate constant of its leaching with consideration of the rate of its subsequent fixation by clay minerals. The rate constants k_1 for ^{137}Cs and ^{90}Sr were close, which indicated that the chemical nature of radionuclide has a minor effect on the rate at which it breaks loose from particles during their disintegration.

Rate of Radionuclide Leaching from Fuel Particles as a Function of Direction and Distance to ChNPP

Radionuclide-leaching rate from fuel particles is a function of the medium in which they are present, as well as characteristics of particles themselves. For example, in laboratory experiment on determining dissolution rate of fuel particles collected in the Chernobyl close-in area, when exposed to solutions modeling natural aqueous media, the fuel particles showed dissolution rates varying by two orders of magnitude from 5.7×10^{-6} to 1.6×10^{-4} $\text{g U}/(\text{cm}^2 \text{ day})$ (Bogatov et al. 1990). It is common knowledge that in the initial phase of the Chernobyl accident radioactive materials released from the reactor were first transported north and northwest and then west, south, east, and northeast (Izrael et al. 1990; IAEA 2006; Talerko et al. 2020). Since the reactor state was changing, characteristics of deposited particles were most likely to be different in different directions. It is also clear that the particle mean size and density are decreasing with distance from the source. This being the

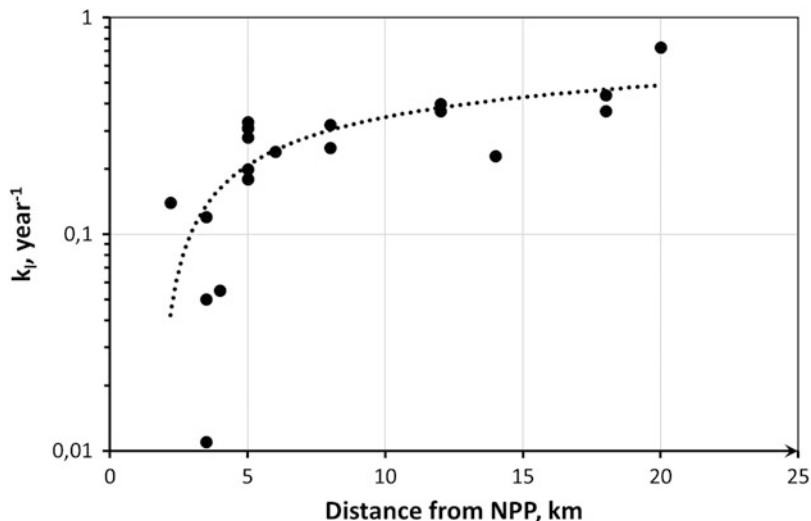


Fig. 3.2 Rate constants for ^{90}Sr leaching from fuel particles as a function of distance to the reactor for the south sector of the footprint (Konoplev 1998)

case, the impact of particles characteristics on leaching rate of radionuclides from these particles can be studied by comparison of rate constants determined at different locations in the contaminated zone. By the classification proposed by Izrael et al. (1990), the region contaminated after the Chernobyl accident can be divided into three sectors: west (northwest, west, and southwest), south (south, southeast, east), and north (north and northeast).

The leaching rate constants were ranging from $1 \times 10^{-2} \text{ year}^{-1}$ to $4 \times 10^{-2} \text{ year}^{-1}$, the mean being $2.4 \times 10^{-2} \text{ year}^{-1}$ in the west and southwest directions, and from $4 \times 10^{-2} \text{ year}^{-1}$ to $2.9 \times 10^{-1} \text{ year}^{-1}$, the mean being $1.1 \times 10^{-1} \text{ year}^{-1}$ in the northwest direction. In these directions, no dependence of leaching constants on distance from the reactor has been revealed (Konoplev and Bulgakov 1999; Kashparov et al. 2004) contrary to the south and southeast directions in which a clear trend for an increase in leaching rate constant with distance from the NPP was reported (Konoplev and Bulgakov 1999) (Fig. 3.2). At distances less than 5 km the mean constant value is estimated to be $7.7 \times 10^{-2} \text{ year}^{-1}$, while at 5 km and more— $3.3 \times 10^{-1} \text{ year}^{-1}$. The dissolution rate constants have demonstrated similar behavior.

Since the soil cover in the 30-km zone is quite homogeneous, it may be reasonable to conclude that the particles more stable in terms of decomposition were deposited in the north, northwest, west, and southwest of the reactor, as well as south and southeastward in the immediate vicinity of the NPP, where the leaching rate constants varied from 4×10^{-3} to $4 \times 10^{-2} \text{ year}^{-1}$. With distance from the NPP in the southern direction, the rate constants increase and at the distance of about 20 km are as high as $4 \times 10^{-1} \text{ year}^{-1}$.

From this, it follows that the characteristics of fuel particles in the south direction changed with distance to ChNPP more drastically than in the west direction. Outside the 30-km zone, the leaching rate constants should be even higher due to smaller size of particles. In soils of Bryansk region of Russia, for example, speciation was determined for the first time in 1987. At that time, ^{90}Sr species were found to be practically at equilibrium, which leads us to the conclusion that the leaching rate constant was at least $1\text{--}2\text{ year}^{-1}$. Similar conclusion can be drawn for the regions of Belarus occurring at the distance 200–250 km, where ^{90}Sr species in soils in 1987 were also close to equilibrium (Petryaev et al. 1993).

Therefore, depending on characteristics of fuel particles, rate constants of radionuclide leaching can vary by several orders of magnitude.

Rate of Radionuclide Leaching from Fuel Particles as a Function of Soil Properties

Since rate of radionuclide leaching from fuel particles largely depends on characteristics of particles themselves, impact of soil properties on their breakdown rate can be studied only using experimental plots on which soils of different types are in close proximity. In this case, differences in the nature of hot particles responsible for contamination in each point will be minimized. Comparison of the leaching rate constants determined at different points showed that there is no clear dependence on soil type, humus content, and cation-exchange capacity (Konoplev and Bulgakov 1999). This can be associated with the fact that the role of soil properties is impossible to discern against the influence of nature of particles and nonuniformity of their area distribution on leaching rate, as well as a truly weak dependence of leaching rate on characteristics of the environment in which they occur. The latter is possible when radionuclide leaching from particles is determined by processes of their oxidation by air oxygen or processes within particles resulting, for example, from radioactive decay of isotopes in them.

On the basis of large amount of statistically reliable data, Kashparov et al. (1999) established the dependence of k_l on the soil pH as follows (Kashparov et al. 2000, 2004):

For the Western sector:

$$\begin{aligned} k_l &= 0.6 \times 10^{-0.15 \cdot \text{pH}} \text{ at } \text{pH} < 7.0, \\ k_l &= 0.05 \text{ at } 7.0 < \text{pH} < 7.5. \end{aligned} \quad (3.9)$$

For the Southern and Northern sectors:

$$\begin{aligned} k_l &= 40 \times 10^{-0.45 \cdot \text{pH}} \text{ at } \text{pH} < 6.5, \\ k_l &= 0.05 \text{ at } 6.5 < \text{pH} < 7.5. \end{aligned} \quad (3.10)$$

During the years immediately after the accident, fuel particles primarily occurred in the upper centimeters of soil, both close to the reactor and at distances to 250 km (Konoplev 2001). Most of the particles were concentrated in the 0–1 cm soil layer and their proportion decreased markedly with depth. The vertical profile of radioactive particles in soil was found to be practically independent of the distance from the ChNPP and is mainly governed by soil type. The lack of dependence of the vertical distribution of particles on their size and chemical nature results from the fact that the primary mechanism of radionuclide migration in the upper soil layer is mixing by soil flora and fauna (bioturbation) (Bulgakov et al. 1991; Konoplev and Golubenkov 1991; Konoplev et al. 1992).

At present, fuel particles have almost completely disintegrated (Beresford et al. 2016) in terrestrial soils. Opposite situation is observed in the cooling pond (CP) of the Chernobyl NPP, where the vast majority of long-lived radionuclides deposited embedded in fuel particles. It is remarkable that in the cooling pond sediments the majority of ^{90}Sr activity still occurs in the form of fuel particles (Bulgakov et al. 2009; Kanivets et al. 2020). Due to low dissolved oxygen concentration and high pH, dissolution of fuel particles in the CP sediments is significantly slower than in soils. However, at the present time, these conditions are changing. After the cessation of water pumping from the Pripyat River to the pond in 2014, a significant part of the sediments is being drained and exposed to the air, which significantly enhances the dissolution rate of fuel particles in exposed sediments, and hence mobility and bioavailability of radionuclides will increase with time (Bulgakov et al. 2009). The rate constant was expected to increase almost instantaneously to the typical value for the aerated layer of neutral and slightly alkaline soils in the Chernobyl exclusion zone. After that, the dissolution rate may increase slowly due to potential acidification of the newly formed soils as a result of vegetation and microbiological activity. The dynamics of this acidification was estimated from the data on the decrease in soil pH as a function of time after soil liming ceased. Along with empirical equations relating the dissolution rate to soil pH, this allowed the prediction of fuel particle dissolution and ^{90}Sr mobility dynamics for different remediation scenarios. Model calculations have shown that in newly exposed sediments, fuel particles will be almost completely dissolved in 15–25 years, while in parts of the pond that remain flooded, fuel particle dissolution will take about 100 years (Bulgakov et al. 2009). Recent data on dynamics of ^{90}Sr and ^{137}Cs in the cooling pond confirm the predictions (Kanivets et al. 2020): dissolved ^{90}Sr activity concentration in the cooling pond increased from about 1 Bq/L in 2014 before the cessation of water pumping to 3–5 Bq/L in spring 2017. Also, as was predicted, dissolved ^{137}Cs in cooling pond water does not show any noticeable increase because of its fast fixation on clay minerals after release from fuel particles.

3.2.2.2 Radionuclide Sorption–Desorption by Soils and Sediments

One of the major processes governing migration of radionuclides in environmental compartments is their sorption by the solid phase of soil and bottom sediments

(Fig. 3.1). In a general form, sorption–desorption of a radionuclide from solution by sorption sites of the solid phase can be written by the equation



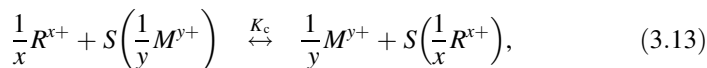
where R is the radionuclide in solution; S is the sorption site on the solid-phase surface; (RS) is the radionuclide sorbed form.

The position of sorption–desorption equilibrium is characterized by the equilibrium constant K (Orlov et al. 2005):

$$K = \frac{[RS]}{[R][S]}, \quad (3.12)$$

where $[R]$, $[RS]$, and $[S]$ are the concentration of a radionuclide in solution, the radionuclide sorbed form concentration, and the capacity of sorption sites, respectively.

Many radionuclides are present in aqueous solution in the form of ions and then the main mechanism for their sorption–desorption is ion exchange (Timofeev-Resovsky and Titlyanova 1966; Cremers and Maes 1986; Konoplev et al. 1992). As a rule, the surface of naturally occurring solid particles in aqueous solution is characterized by excessive negative charge and hence cations are better sorbed by them than anions. The radionuclides of radiocesium and radiostrontium under consideration occur in solution primarily as cations. The cation-exchange equation for radionuclide cation R^{x+} with charge $x+$, in the general case, takes the form (Kokotov and Wilken 1969):



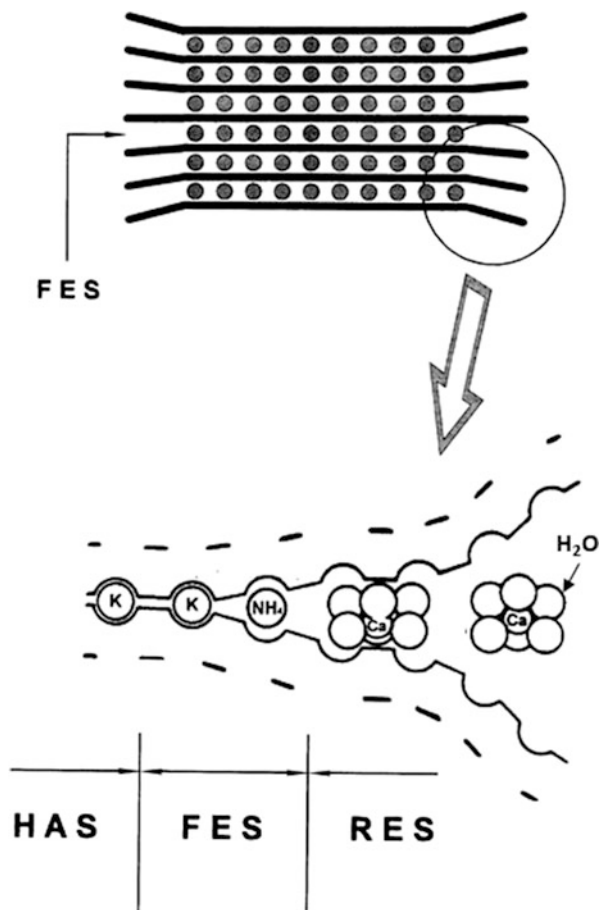
where M^{y+} is the competing ion present in the medium. In environmental compartments, the key exchange ions are Ca^{2+} , Mg^{2+} , K^+ , NH_4^+ , and Na^+ (in acid medium also H^+ and Al^{3+}).

The equilibrium constant of reversible process (3.13) is referred to as the selectivity coefficient. It accounts for relative affinity of the cation R^{x+} to sorption sites with respect to exchange ion M^{y+} (Kokotov and Wilken 1969; Orlov et al. 2005):

$$K_c(R^{x+}/M^{y+}) = \frac{[S(\frac{1}{x}R^{x+})]^{\frac{1}{x}}[M^{y+}]^{\frac{1}{y}}}{[S(\frac{1}{y}R^{x+})]^{\frac{1}{y}}[R^{x+}]^{\frac{1}{x}}}. \quad (3.14)$$

It is now commonly believed that high ability of soils and sediments to sorb selectively radiocesium is determined by the presence of clay minerals with crystal lattice of group 2:1 (Sawhney 1972; Cremers et al. 1988; Comans et al. 1991). In work by Brouwer (Brouwer et al. 1983), based on the study of Cs sorption on illite, the multitude of ion-exchange sorption sites have been divided, at least, into three

Fig. 3.3 Schematic structure of 2:1 micaceous clay mineral and frayed edges of interlayer (modified from Konoplev 1998)



main types with different selectivity with respect to cesium. The proportion of these sites was 0.25%, 2.5%, and 97.25% of total ion-exchange capacity in case of illite saturated with potassium; and 0.55%, 3.35, and 96.15% in case of illite saturated with calcium. The sorption sites of maximum capacity and minimum selectivity with respect to cesium were attributed to regular surface ion-exchange sites (RES). Two other types of sorption sites with smaller capacity, the first of which has extremely high (the highest) selectivity with respect to cesium, were attributed to sorption sites occurring in the area of frayed edges of neighbor layers of illite crystal lattice. Schematically, the structure of layer mineral of group 2:1 and its frayed edges are shown in Fig. 3.3. Specific location of frayed-edge sites (FES) is responsible for their high selectivity with respect to ions with low values of cation radius and low hydration energy in solution.

Since most of cations in aqueous solution occur in hydrated form, i.e., surrounded by water molecules, they are too large to approach FES. Yet, ions with low hydration

energy, such as K^+ , Rb^+ , NH_4^+ , and especially Cs^+ , can easily lose a hydration water shell, penetrate to the frayed edges of mineral's interlayers, and be adsorbed on selective sorption sites FES. On the other hand, the ions Ca^{2+} , Mg^{2+} , and Sr^{2+} , surrounded by a large hydration shell, are not capable of approaching FES and can be adsorbed only on regular exchange sites RES located on the surface of mineral particles. Quantitatively, the selectivity of sorption sites with respect to cesium is characterized by the selectivity coefficient. Radiocesium is adsorbed on surface sorption sites RES nonselectively, i.e., the selectivity coefficient of its sorption with respect to one-charge ions K^+ , Na^+ , NH_4^+ , and others is close to 1. With that, the selectivity coefficient of Cs sorption on FES is equal to 1000 for K^+ and 200 for NH_4^+ (De Preter 1990; Wauters et al. 1996a). The selective sorption sites FES constitute a relatively small portion of ion-exchange sorption sites (1–5%) for most of soils and bottom sediments (De Preter 1990). Nevertheless, as follows from the published data (Brouwer et al. 1983; Comans et al. 1991; Konoplev et al. 2002), FES are nonuniform in terms of selectivity with respect to radiocesium and can be divided, at least, into two types: regular FES and sites of extremely high selectivity HAS (High-Affinity Sites).

Due to high selectivity of FES with respect to cesium and because radiocesium, and even stable cesium, occurs at trace concentrations in the environment, practically the entire radiocesium in most of soils and bottom sediments is adsorbed on FES.

The ability of soils and sediments to sorb radiocesium selectively is characterized by the capacity of selective sorption sites [FES] or by the so-called radiocesium interception potential (RIP), which is the product of [FES] and the selectivity coefficient of radiocesium in relation to the corresponding competitive ion (Sweeck et al. 1990).

Cremers and coworkers (Cremers et al. 1988; Sweeck et al. 1990; De Preter 1990; Valcke and Cremers 1994; Wauters et al. 1994) developed a method for quantitative determination of [FES] and RIP. The method is based on using silver thiourea $Ag(TU)^+$ as a masking agent for regular exchange sites, RES. RES selectively binds this complex, while $Ag(TU)^+$ does not interact with FES because of the molecule's large size. Thus, the masking blocks RES and allows the study of cesium sorption-desorption on FES. Later, the original method was simplified using blocking RES by sufficient concentration of hydrated Ca^{2+} cations (Wauters et al. 1996a, b).

It was found, however, that even during a relatively short laboratory procedure of [FES] and RIP determination (both original and modified) Cs was partly fixed by the solid phase and the method overestimated the amount of reversibly sorbed Cs (Konoplev and Konopleva, 1999a; Konoplev et al. 2002). Given highly organic soils and bottom sediments, collapse of the interlayers of micaceous clay prevents determination of [FES] when using this method. To avoid this drawback, modification of the procedure was proposed (Konoplev and Konopleva, 1999a; Konoplev et al. 2002), which included:

- An additional step of ammonium acetate extraction after equilibration time is used. This step allows avoiding the influence of fast fixation on [FES] or RIP determination.
- The [FES] is calculated using the linearized form of Langmuir isotherm and its value is given by the intercept on the ordinate of the plot of reversibly sorbed cesium inverse concentration versus inverse equilibrium concentration of cesium in solution.
- Taking the initial range of the isotherm at low cesium concentrations, the capacity of high-affinity sites [HAS] located between layers of micaceous clay particles can be determined. HAS have higher selectivity for cesium as compared to average FES and are therefore occupied by cesium in the first stage. Notice that HAS represent 1–10% of FES.

3.2.2.3 Radiocesium Fixation by Soils and Sediments

In terms of chemical forms, fixation of radionuclides is a transfer from their mobile to fixed form. Fixation represents long-term replacement of interlattice K^+ by Cs^+ ions due to collapse of expanded edges of mineral's crystal lattice interlayers. Models have been proposed in which radiocesium fixation is treated as an irreversible process (Comans and Hockley 1992). The available data suggest that this assumption is warranted at the initial stage of fixation and for processes of relatively short timescale. On the other hand, data about long-term transformation of radionuclides chemical forms in soil after nuclear weapon testing (Pavlotskaya 1974) and Kyshtym accident (Konoplev and Bobovnikova 1991) and 30-year studies after the Chernobyl accident are indicative of the existence of remobilization process that is the reverse of fixation (Konoplev et al. 1992; Smith and Comans 1996; Konoplev and Bulgakov 2000a). After deposition of radiocesium onto the soil, the fraction of its exchangeable form α_{ex} does not decrease to zero, as expected for irreversible fixation, but only decreases to a certain steady-state level, and then does not change significantly (Konoplev and Bobovnikova 1991; Konoplev et al. 1996).

Rate constant of radiocesium fixation k_f (see Fig. 3.1) for typical soils of the Chernobyl 30-km zone was found to be $(1-4) \times 10^{-2} \text{ day}^{-1}$ (Konoplev et al. 1992, 1996). Remobilization was about an order of magnitude slower: $k_r = (2-6) \times 10^{-3} \text{ day}^{-1}$, which is comparable with rate constant of radionuclide leaching from fuel particles (Fig. 3.1).

3.3 Radionuclide Distribution Between Solid and Liquid Phases in the Soil–Water Environment

Up to now, modeling and predicting of radionuclide behavior in the environment, and dose assessment is based on using, instead of specific rate and equilibrium constants, an integral parameter—distribution coefficient K_d (L/kg) (Zheleznyak et al. 1992, 2016; Borzilov et al. 1993; Konoplev et al. 1996a; Nair et al. 1996; Smith et al. 2005a, b; Monte et al. 2009), which is equal to the ratio of radionuclide equilibrium concentrations of particulate radionuclide $[R]_p$ (Bq/kg) and dissolved radionuclide $[R]_w$ (Bq/L):

$$K_d = \frac{[R]_p}{[R]_w}. \quad (3.15)$$

In case of radiocesium, for example, $[R]_p$ includes radiocesium embedded in fuel particles, exchangeably sorbed radiocesium and fixed or nonexchangeable radiocesium in solid phase.

3.3.1 Applicability of K_d in Nonequilibrium Conditions

The distribution coefficient K_d is by definition an equilibrium parameter and therefore should be measured at equilibrium or in close to equilibrium conditions. At the same time, using the distribution coefficient is appropriate only to predict processes for which time scale is much longer than the time of establishing equilibrium. In addition, nonequilibrium distribution of radionuclides can be caused by their deposition on the ground as water-insoluble particles, such as fuel particles following the Chernobyl accident. Part of radionuclides occurring in the solid phase will not take part in establishing equilibrium concentration in solution. The contribution of this part to total concentration of radionuclides in soil or depositions decreases with time because of disintegration of particles. As a result, the apparent distribution coefficient estimated as a ratio of the radionuclide concentration in the solid phase to its concentration in the liquid phase is a function of time even for radionuclides with a relatively fast sorption–desorption, such as ^{90}Sr (Konoplev et al. 1992; Beresford et al. 2016).

It has been proven that high retention of radiocesium in soil and bottom sediments is determined by two different processes: reversible selective sorption by micaceous clay minerals (Sawhney 1972; Cremers and Maes 1986; Cremers et al. 1988) and fixation/remobilization (Konoplev et al. 1992, 1996, 2002). Kinetics of the processes of leaching of radiocesium from fuel particles and its fixation/ remobilization determines dynamics of the integral parameter K_d .

3.3.2 Prediction of Solid–Liquid Distribution of Radionuclide Exchangeable Form: Exchangeable Distribution Coefficient

The existing methods for estimating the distribution coefficient have a common drawback, which is the absence of a clear division of radionuclide speciation based on their ability to exchange with the liquid phase. The problem can be resolved by using the concept of exchangeable distribution coefficient (K_d^{ex}). This concept provides that the total amount of the radionuclide in the solid phase of the soil–water system can be divided into two components: the exchangeable part, which occurs at instantaneous equilibrium with the liquid phase; and the nonexchangeable part, which does not contribute to the radionuclide concentration in solution (Fig. 3.1). In the immediate term, only the exchangeable form of radionuclide contributes to the solid–liquid interphase exchange. Therefore, the notion of the exchangeable distribution coefficient K_d^{ex} was proposed as a ratio of the concentration of the radionuclide in exchangeable form in the solid phase $[R]_{\text{ex}}$ to its concentration in solution at equilibrium (Konoplev et al. 1992, 2002; Madruga et al. 1996; Konoplev and Bulgakov 2000a, b):

$$K_d^{\text{ex}} = \frac{[R]_{\text{ex}}}{[R]_{\text{w}}} = \alpha_{\text{ex}} K_d^{\text{tot}}, \quad (3.16)$$

where α_{ex} is the fraction of exchangeable radionuclide, and K_d^{tot} is the apparent or total distribution coefficient.

The advantage of K_d^{ex} is that it is a function of environmental characteristics (sorption properties of the solid phase and cation composition in solution) and can be assessed/predicted on their basis (Konoplev and Bulgakov 2000a, b). On the other hand, the value of total K_d^{tot} is strongly dependent on the fraction of nonexchangeable radionuclide. Using the equation of ion-exchange equilibrium for Cs sorption on FES in the presence of M^+ ion, an equation can be written to relate the exchangeable distribution coefficient of radiocesium and the solid-phase FES capacity and M^+ concentration in solution:

$$K_d^{\text{ex}}(\text{Cs}) = \frac{K_c(\text{Cs}/M)[\text{FES}]}{[M^+]}, \quad (3.17)$$

where $K_c(\text{Cs}/M)$ is the selectivity coefficient for ion exchange of Cs^+ on FES with respect to cation M^+ . Or

$$K_d^{\text{ex}}[M^+] = K_c(\text{Cs}/M)[\text{FES}] = \text{RIP}^{\text{ex}}(M). \quad (3.18)$$

Equation (3.18) accounts for the exchangeable radiocesium interception potential $\text{RIP}^{\text{ex}}(M)$ (Konoplev and Konopleva, 1999a; Konoplev et al. 2002). The

exchangeable radiocesium interception potential is a constant value for a given sorbent and characterizes its ability to sorb cesium selectively and exchangeably. The significance of the radiocesium interception potential is explained by the fact that it can be used to determine the exchangeable distribution coefficient of radiocesium.

In a general case, the value of ^{137}Cs distribution coefficient can be predicted based on separate consideration of sorption on FES and RES (Wauters et al. 1996c). Then

$$K_d^{\text{ex}} = K_d^{\text{ex}}(\text{FES}) + K_d^{\text{ex}}(\text{RES})$$

$$= \frac{K_c^{\text{FES}}(\text{Cs}/\text{K})[\text{FES}]Z_K}{[\text{K}]_w} + \frac{K_c^{\text{RES}}(\text{Cs}/\text{K})[\text{K}]_{\text{RES}}}{[\text{K}]_w}, \quad (3.19)$$

where K_c^{FES} is the selectivity coefficient for exchange of cesium and potassium cations on FES; [FES] is the capacity of selective sorption sites in the solid phase; Z_K is the FES fraction occupied by potassium cations; $[\text{K}]_{\text{RES}}$ is the concentration of potassium bound with nonselective adsorption sites.

For nonselective sorption sites, $K_c^{\text{RES}}(\text{Cs}/\text{K})$ is close to 1 and besides that the second term in the right part of Eq. (3.19) is usually negligible for most of soils and bottom sediments as compared to the first term. With this in mind, when potassium is a predominant ion competing with ^{137}Cs (Wauters et al. 1994, 1996a, b, c; Konoplev et al. 2002),

$$K_d^{\text{ex}}(\text{Cs}) = \frac{\text{RIP}^{\text{ex}}}{[\text{K}]_w}. \quad (3.20)$$

In those cases when ammonium cation is present in the system in noticeable amounts (Wauters et al. 1994, 1996c; Konoplev et al. 2002),

$$K_d^{\text{ex}}(\text{Cs}) = \frac{\text{RIP}^{\text{ex}}(\text{NH}_4)}{[\text{NH}_4]_w}. \quad (3.21)$$

In mixed cases, when the contributions of potassium and ammonium to competition with ^{137}Cs are comparable, the following ratios are applicable (Wauters et al. 1994, 1996a, b, c, d):

$$K_d^{\text{ex}}(\text{Cs}) = \frac{\text{RIP}^{\text{ex}}(\text{K})}{[\text{K}]_w + K_c^{\text{FES}}(\text{NH}_4/\text{K})[\text{NH}_4]}, \quad (3.22)$$

where $K_c^{\text{FES}}(\text{NH}_4/\text{K})$ is the selectivity coefficient of ammonium in relation to potassium for FES sites. Post-Chernobyl studies showed that for most soils and sediments $K_c^{\text{FES}}(\text{NH}_4/\text{K}) \approx 5$, and simplified Eq. (3.22) can be written as follows (Wauters et al. 1994, 1996a; Konoplev et al. 2002):

$$K_d^{\text{ex}}(\text{Cs}) = \frac{\text{RIP}^{\text{ex}}(\text{K})}{[\text{K}]_w + 5[\text{NH}_4]} \quad (3.23)$$

Knowing K_d^{ex} one can calculate the apparent K_d^{tot} on the basis of Eq. (3.16); α_{ex} can be easily measured using 1 M ammonium acetate extraction or estimated on the basis of rate constants of radionuclide leaching from hot particles, fixation, and remobilization (Fig. 3.1) (Konoplev et al. 1992; Konoplev and Bulgakov 2000a, b).

Using this method enables reducing uncertainty involved in assessing the exchangeable distribution coefficient and the apparent K_d^{tot} . The downside of this method is its complexity and unavailability of data for many territories. For all that, measurement of RIP^{ex} can be recommended for critical regions, in order to improve reliability of predictions in emergency and remediation strategy development.

The methodology has been used for a number of water bodies contaminated as a result of the Chernobyl accident: Kiev reservoir, Glubokoe Lake (Ukraine), Prip'yat and Dnieper rivers (Ukraine and Belarus), Svyatoe and Kozhanovskoe lakes (Russia), Devoke Water (UK), Lakes Constance and Vorse (Germany), Lake Lugano (Switzerland), etc. (Konoplev et al. 1996, 2002; Konoplev and Konopleva, 1999a), and demonstrated its applicability to estimate and predict K_d^{ex} and K_d^{tot} for radiocesium in various water bodies.

3.4 Radionuclide Bioavailability in Soil and Its Parameterization Through Soil Characteristics

Uptake of radionuclides from soil to plant is a significant pathway for transfer to humans and animals (IAEA 2006). For characterization of soil–plant transfer, the concentration ratio (CR) is normally used, which is the ratio of radionuclide concentration in plant and its concentration in the upper soil layer. Values of CR determined experimentally in different conditions for a particular plant can differ by hundreds or even thousands of times (IAEA 2010). That is why, using a plant-average CR leads to a considerable uncertainty in prediction. Numerous attempts were therefore made to develop methods for estimating site-specific values of CR. Empirical dependencies of radionuclides accumulation in plant on soil properties were derived (IAEA 2010), but the problem is that purely empirical characteristics are not all-purpose. In order to be applied for soils different from those for which they were obtained, they should be substantiated based on information about mechanisms of sorption–desorption of radionuclides in soil and their soil–plant transfer.

3.4.1 Conceptual Model of Radionuclide Soil–Plant Transfer

It is generally agreed that a key parameter governing soil–plant transfer of radionuclide is its exchangeability in soil (Oughton et al. 1992; Konoplev et al. 1993). Yet it is not a single factor; account should also be taken of radionuclide and major cations concentrations in soil solution (Shaw 1993; Konoplev et al. 1996; Smolders et al. 1997; Absalom et al. 1999). In this respect, two hypotheses were proposed to account for the influence of soil solution composition on radionuclide transfer to plant. By the first hypothesis, radiocesium concentration in plant is proportional to the ratio of its concentration in soil solution and potassium concentration. Comparison with experimental data, however, has shown that there is no meaningful correlation between the concentration ratio and $^{137}\text{Cs}/\text{K}$ ratio in soil solution (ECP-2 1996). The other hypothesis, which seems more reasonable, is that the concentration ratio is proportional to the radionuclide fraction in the root-exchange complex as a function of composition of soil solution (Smolders et al. 1997). Using this hypothesis, a model of radiocesium bioavailability in soil was proposed (Konoplev et al. 1999; Konoplev and Konopleva 1999a).

Figure 3.4 shows the conceptual scheme of the equilibrium model of radiocesium soil–plant transfer. This model is based on the following assumptions:

1. Radiocesium is taken up by plant from soil solution and its concentration in plant is a linear function of radionuclide loading in the root-exchange complex, which is determined by cation composition of soil solution. The root-exchange complex is not selective, i.e., selectivity coefficient of Cs^+ in relation to K^+ and NH_4^+ is equal to unity (Konoplev et al. 1996b; Smolders et al. 1997);
2. Only the exchangeable fraction of ^{137}Cs inventory in soil is involved in immediate exchange with soil solution (Konoplev et al. 1992, 1993);
3. Exchangeable radiocesium interception potential (RIP^{ex}) and cation composition of soil solution control ^{137}Cs concentration in soil solution in line with ion-exchange equilibrium (Konoplev and Konopleva, 1999a).

The model includes ion-exchange sorption–desorption process in soil, ion exchange in the root-exchange complex, and uptake from root surface to plant. The fixed part of radionuclide is not involved in the immediate transfer to plant.

Prior to plant uptake, the radionuclide must pass through the cell wall free space characterized by specific cation-exchange capacity (Haynes 1980). This speculation differs from the approaches usually applied to radionuclide soil–plant transfer based on linearity of the radionuclide concentration in plant with its concentration in soil solution. The distinguishing feature of the proposed model is that Cs loading in the root-exchange complex is influenced by cationic composition of soil solution. The root-exchange complex is associated with carboxylic groups, and hence not selective, as opposed to soil exchange complex. This approach was used for modeling of radiocesium transfer in the soil solution–plant system (Smolders et al. 1997; Absalom et al. 1999). As the root-exchange complex is nonselective, the selectivity coefficients of both K/NH_4 and the Cs/K are equal to unity. The same can be

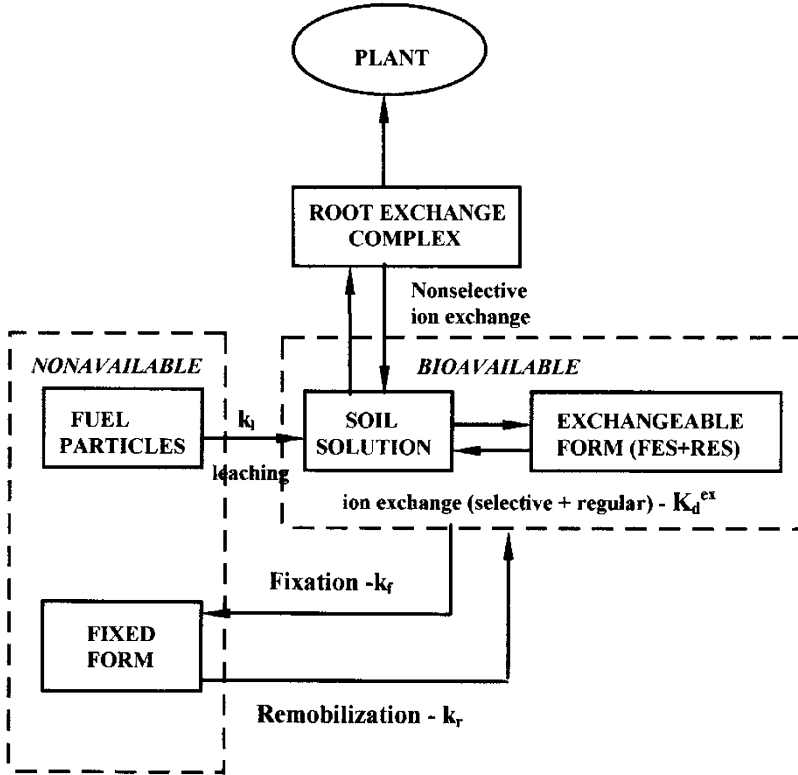


Fig. 3.4 Conceptual model of radiocesium soil-plant transfer (Konoplev et al. 1999)

assumed for the Ca/Mg pair. Accordingly, K^+ and NH_4^+ can be treated as one ion M^+ , whereas Ca^{2+} and Mg^{2+} as one ion M^{2+} . On the basis of these assumptions (Smolders et al. 1997),

$$[^{137}Cs]_{\text{plant}} = k \frac{Z_{M^+}}{[M^+]_w} [^{137}Cs]_w, \tag{3.24}$$

where Z_{M^+} is the fraction of M^+ in the root cation-exchange complex (potassium window); $[M^+]_w$ is the concentration of M^+ in soil solution; $[^{137}Cs]_w$ is the ^{137}Cs concentration in soil solution; $[^{137}Cs]_{\text{plant}}$ is the ^{137}Cs concentration in plant; k is the proportionality constant, which reflects efficacy of transport across plasmalemma. At the same time, the concentration of ^{137}Cs in soil solution is determined by the sorption-desorption equilibrium of the radionuclide in the “soil-soil solution” system. This means that radiocesium occurs in soil solution at simultaneous equilibrium with two cation exchangers: the selective exchanger of soil and the nonselective one of root surface. The ^{137}Cs concentration in soil solution at

ion-exchange equilibrium of radiocesium on the FES can be presented as follows (Konoplev et al. 1996b, 1999, 2000):

$$[^{137}\text{Cs}]_w = \frac{\alpha_{\text{ex}} [^{137}\text{Cs}]_{\text{tot}}}{\text{RIP}^{\text{ex}}(\text{K})} ([\text{K}]_w + K_c^{\text{FES}}(\text{NH}_4/\text{K})[\text{NH}_4]), \quad (3.25)$$

where α_{ex} is the portion of exchangeable ^{137}Cs in soil; $[^{137}\text{Cs}]_{\text{tot}}$ is the total concentration of ^{137}Cs in soil; $\text{RIP}^{\text{ex}}(\text{K})$ is the radiocesium interception potential of soil in relation to potassium; $[\text{K}]_w$ and $[\text{NH}_4]_w$ are the concentrations of correspondent cations in soil solution; $K_c^{\text{FES}}(\text{NH}_4/\text{K})$ is the selectivity coefficient of ammonium in relation to potassium at FES.

3.4.2 Parameterization of Radiocesium Bioavailability Through Soil Characteristics

The “potassium window” Z_{M^+} in the nonselective root-exchange complex can be expressed in terms of cation composition of soil solution. For most soils, $Z_{\text{M}^{2+}} \gg Z_{\text{M}^+}$, and Z_{M^+} is proportional to the following parameter $\frac{[\text{K}^+]_w + [\text{NH}_4^+]_w}{\sqrt{[\text{Ca}^{2+}]_w + [\text{Mg}^{2+}]_w}}$ of soil solution. Substituting this and (3.25) in (3.24) and taking into account that $\text{CR} = [^{137}\text{Cs}]_{\text{plant}}/[^{137}\text{Cs}]_{\text{tot}}$, at equilibrium conditions, CR should be a linear function of A (availability factor) (Konoplev et al. 1996b, 1999, 2000):

$$\text{CR} = B \times A, \quad (3.26)$$

where B is the parameter dependent on plant characteristics (first of all, capacity of the root-exchange complex). The availability factor A is taken to be proportional to the fraction of radiocesium in the root-exchange complex and is parameterized through the composition of soil solution and sorption properties of soils as follows (Konoplev et al. 1999, 2009; Konoplev and Bulgakov 2000b):

$$A = \frac{\alpha_{\text{ex}} \text{SPAR}}{\text{RIP}^{\text{ex}}}, \quad (3.27)$$

where SPAR is the sorption potassium–ammonium ratio (Konoplev and Bulgakov 2000b):

$$\text{SPAR} = \frac{[\text{K}]_w + K_c^{\text{FES}}(\text{NH}_4/\text{K})[\text{NH}_4]}{\sqrt{[\text{Ca}]_w + [\text{Mg}]_w}}, \quad (3.28)$$

where $[K]_w$, $[NH_4]_w$, $[Ca]_w$, $[Mg]_w$ are the concentrations of corresponding cations in soil solution, $K_c^{FES}(NH_4/K)$ is the selectivity coefficient of potassium exchange for ammonium at selective adsorption sites of soil FES.

Equation (3.27) can actually serve as a basis for prediction of CR using soil and soil solution characteristics. In fact, availability factor A is a combination of three key soil parameters: fraction of exchangeable form of ^{137}Cs (α_{ex}) accounting for fixation ability of soil; exchangeable radiocesium interception potential (RIP^{ex}) representing ability of soil to adsorb radiocesium selectively and reversibly, and parameter of soil solution cation composition SPAR. A similar approach to model soil–plant transfer through soil and soil solution parameters was applied by Absalom et al. (1999, 2001).

The above method can be used for improving the accuracy of soil-specific estimates of CR. However, this requires knowing soil characteristics such as the content of exchangeable ammonium and exchangeable radiocesium interception potential (RIP^{ex}). Values of these parameters are known for a limited number of soils, methods for their evaluation have not been developed yet, and experimental determination of RIP^{ex} is rather complicated. In view of this, a simpler method was proposed for parameterization of the radiocesium availability factor using only those soil characteristics, which can either be taken from reference literature or estimated using known correlation ratios (Konoplev et al. 1999, 2000).

A simplified parameterization of the availability factor for “potassium scenario” can be obtained with the equation calculating radiocesium concentration in soil solution, as proposed in (Konoplev and Bulgakov 2000b):

$$[^{137}Cs]_w = \frac{[^{137}Cs]_{ex}[K]_w}{K_c^{eff}[K]_{ex}}, \quad (3.29)$$

where $[^{137}Cs]_{ex}$, $[K]_{ex}$ is the concentration of exchangeable ^{137}Cs (Bq/kg) and potassium (meq/kg) in soil, respectively; K_c^{eff} is the effective selectivity coefficient of the potassium cation exchange for caesium cation in the soil exchange complex.

In this case, the availability factor A^* can be written as

$$A^* = \frac{\alpha_{ex} PAR}{K_c^{eff}(Cs/K)[K]_{ex}}, \quad (3.30)$$

where PAR is the potassium adsorption ratio ($mM^{1/2}$),

$$PAR = \frac{[K]_w}{\sqrt{[Ca]_w + [Mg]_w}}, \quad (3.31)$$

Basically, Eq. (3.30) is similar to Eq. (3.27), the only difference is that Eq. (3.30) includes the effective selectivity coefficient, which allows estimation of the availability factor using the classification of soils by values of $K_c^{eff}(Cs/K)$ proposed in

(Konoplev and Bulgakov 2000b). A better accuracy can be achieved if the value of $K_c^{\text{eff}}(\text{Cs}/\text{K})$ is measured experimentally, which is much easier than to measure RIP^{ex} .

Using expression of PAR through EPR (exchangeable potassium ratio) equal to the ratio of the exchangeable potassium and the sum of exchangeable calcium and magnesium in soil (Richards 1954), A^* can be parameterized through the sum of exchangeable Ca/Mg and exchangeable K:

$$A^* = \frac{9.5\alpha_{\text{ex}}}{K_c^{\text{eff}}(\text{Cs}/\text{K})} \left\{ \frac{1}{[\text{Ca}]_{\text{ex}} + [\text{Mg}]_{\text{ex}}} - \frac{0.036}{[\text{K}]_{\text{ex}}} \right\}. \quad (3.32)$$

Equation (3.32) enables parameterization of bioavailability via commonly used soil characteristics such as exchangeable cations Cd, L, and Mg that are available in conventional soil maps.

3.4.3 Radionuclide Bioavailability in Agricultural and Forest Soils

It is worth dwelling on limitations of the proposed method for parameterization of radiocesium soil–plant transfer. First, the presented model is based on the assumption that only exchangeable form of radionuclide is potentially involved in transfer to plant. At the same time, the exchangeable radionuclide fraction determined by the sequential extractions characterizes the state of dynamic equilibrium between radiocesium fixation and remobilization (see Fig. 3.1). In the long term, the total radionuclide inventory in soil can be potentially bioavailable as a result of remobilization. Therefore, the steady state of exchangeable radiocesium does not always correspond to the biologically available fraction of radionuclide. A more appropriate technique would be extraction from soil during time close to time scale of soil–plant transfer. Secondly, when estimating radionuclide uptake from soil, CR should be compared for the same vegetation species. Thirdly, an appropriate parameter of radionuclide soil–plant transfer needs to be selected. For example, for agricultural soils uniform distribution of radionuclides in the upper 20-cm horizon is normally taken, while in forest soils the upper soil layer is stratified, and the properties of layers L, Of, Oh, and Ah differ from each other. Therefore, for analysis of radionuclide bioavailability in forest soils, it is important to know in what layer the roots of plant under study occur. It is reasonable to compare CR_r , calculated specifically for root zone.

To test applicability of the proposed model, the data on radiocesium transfer from soil to understory in forests of Chernobyl exclusion zone (Ukraine), Baden Wuerttemberg (Germany), Bryansk region (Russia), Uppsala district (Sweden), and Ticino (Switzerland) were used (Konoplev et al. 1999, 2000; Victorova et al. 2000). As a reference plant for analysis, fern (*Pteridium aquilinum* (L.) Kuhn and *Dryopteris filix-mas* (L.) Schot) was chosen. For some sites, data on bilberry

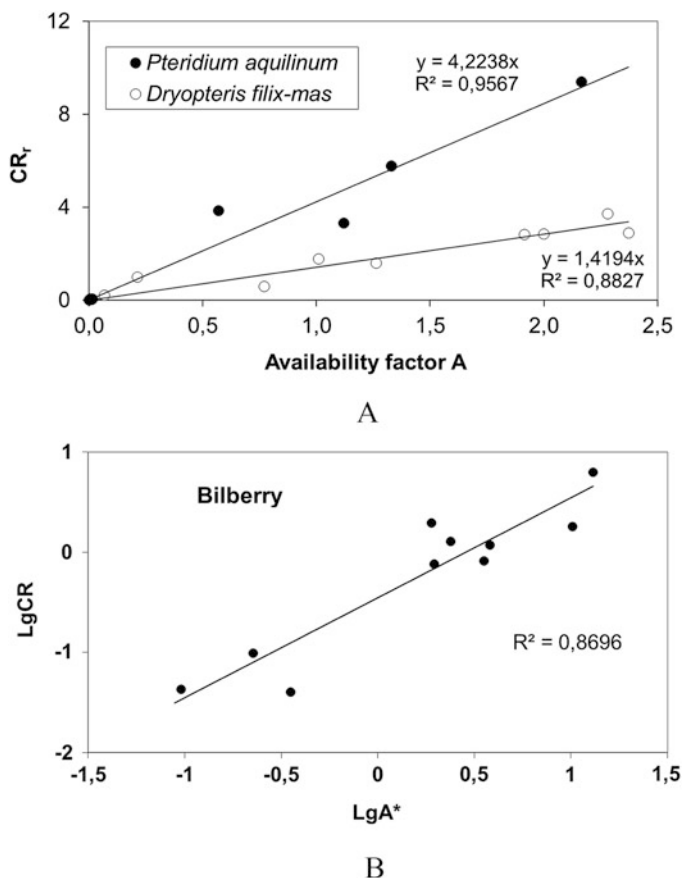


Fig. 3.5 Dependencies of CR_r for two species of fern on the availability factor A in different forest soils (a). Dependence of $LgCR_r$ for bilberry on LgA^* (b) in case of simplified parameterization of bioavailability factor (Konoplev et al. 1999)

(*Vaccinium myrtillus*) and blackberry (*Rubus fruticosus*) were obtained and applied. Roots of these plants occur in the humus layer of soil (O_h or A_h). Since most of radiocesium was found in the same layers, soil characteristics involved in equations (Eqs. 3.27 and 3.30) were measured for these layers.

Figure 3.5 presents dependencies of CR_r on the availability factor A for two species of fern in forest soils (Konoplev 1998). As can be seen, they are very close to a linear function and the agreement of the theoretical dependence with experimental data can be considered as satisfactory.

Extended data sets on ^{137}Cs soil-plant transfer for maize and oat on arable soils have been used to validate the simplified model (Bulgakov and Shkuta 2004). For these plant species, satisfactory agreement between experimental values of CR and model predictions has been found. The model by Absalom et al. (1999, 2001) was validated against data sets for agricultural crops and grass (Smolders et al. 1997a;

Sanchez et al. 1999; Absalom et al. 2001) and also demonstrated ability to predict radiocesium dynamics across a wide range of arable and meadow soils.

The agreement in the theoretical and experimental dependencies (Konoplev et al. 1999, 2000, 2009; Konoplev and Konopleva, 1999b; Absalom et al. 2001; Bulgakov and Shkuta 2004) suggests that the described method can be used for estimation of site-specific concentration ratios of radiocesium in plants. Part of the parameters required for calculating the availability factor can be obtained by expert judgment or measured using a rather simple procedure (α_{ex} , K_c^{eff}), while others are important agrochemical indicators and can be taken from reference literature for many regions ($[\text{Ca}]_{\text{ex}}$, $[\text{Mg}]_{\text{ex}}$, $[\text{K}]_{\text{ex}}$). All this is made possible using the proposed method for mapping radiocesium phytoavailability. Incorporation of such maps into GIS can lead to essential reduction in uncertainty associated with dose and risk assessment.

Similar line of approach can be used for radiostrontium soil–plant transfer, but the situation is simpler since radiostrontium is not sorbed by soil selectively. Assuming that Ca is the main competitive cation for Sr in solution, the following equation was proposed by Sysoeva et al. (2005):

$$CR(^{90}\text{Sr}) \sim A(^{90}\text{Sr}) = \frac{\alpha_{\text{ex}}(^{90}\text{Sr})}{K_c(\text{Sr}/\text{Ca})[\text{Ca}]_{\text{ex}}}, \quad (3.33)$$

where $\alpha_{\text{ex}}(^{90}\text{Sr})$ is the exchangeable fraction of ^{90}Sr in soil, $[\text{Ca}]_{\text{ex}}$ is the concentration of exchangeable Ca in soil. The authors tested the applicability of Eq. (3.33) for barley and lupine on a wide range of soils and showed reliability of such predictions (Sysoeva et al. 2005).

3.5 Conclusions

The studies of the behavior of Chernobyl-derived radionuclides became a basis for establishing a scientific framework for methods to estimate and predict mobility and bioavailability of accidentally released radiocesium and radiostrontium, which can be used for developing decision-support systems in case of nuclear emergency, environmental impact assessment for facilities of radiation hazard, and evaluating efficacy of countermeasures to reduce risk of living in contaminated areas.

Post-Chernobyl studies have allowed to reveal the role of hot particles in environmental behavior of radionuclides. In the case of Chernobyl, most of long-lived radionuclides were incorporated in nuclear fuel particles. Because of occurrence of fuel particles, initial mobility and availability of radionuclides in the near zone were lower than in similar conditions in the case of global fallout and Kyshtym accident. Moreover, deposition of fuel particles mainly in the near zone resulted in localized contamination by refractory radionuclides and strong dependence of initial mobility of more volatile radiocesium on distance to the accidental reactor. Disintegration of fuel particle resulted in leaching of radionuclides and hence changes in mobility and

bioavailability. Studies of changes in radiostrontium exchangeability have made it possible to determine rates of decomposition and leaching of radionuclides from fuel particles in the soils of the Chernobyl 30-km zone. The time scale of radionuclide leaching from fuel particles in soil was determined to vary from 1 to 10 years and more.

Radionuclides leached from fuel particles become involved in the processes of transformation of radionuclide chemical forms in the soil–water environment. To account for major processes of radiocesium and radiostrontium transformation, a conceptual model was developed, including radionuclide leaching from fuel particles, sorption–desorption by ion-exchange mechanism, fixation, and remobilization. It was found that high retention of radiocesium in soils and sediments is associated with two distinct processes: high selective exchangeable sorption by micaceous clay minerals and fixation. In addition to radionuclide fixation, a reverse process of remobilization was shown to play a certain role and should be taken into account.

A major advancement in studies of radiocesium behavior following the Chernobyl accident consisted in introducing and using the concepts of frayed-edge sites (FES) capacity and radiocesium interception potential (RIP) for soils and sediments, which allowed parameterization of radiocesium behavior in the soil–water environment and prediction of its mobility and bioavailability.

Data have been generated regarding quantitative characteristics of radiocesium fixation kinetics on different soils for both laboratory and field conditions. The evidence suggests that the total, or apparent, distribution coefficient of radionuclide K_d can vary in a wide range, depending on deposition characteristics and environmental conditions. Its value is a function of contact time and depends on transformation rates of radionuclide chemical forms. Another important step forward was establishing the exchangeable distribution coefficient instead of the total K_d^{tot} . K_d^{ex} can be estimated on the basis of ion-exchange equilibrium from characteristics of soil and sediments such as cation-exchange capacity, RIP^{ex} , and cation composition of the solution. This allows reducing significantly the uncertainty involved in prediction of radiocesium behavior in the soil–water environment.

In terms of bioavailability, the achievement was the parameterization of radionuclide soil-to-plant concentration ratio CR through soil and soil solution characteristics. As a result, the uncertainty associated with estimates of soil–plant transfer in agricultural and seminatural environment can be reduced to a significant degree. This has been verified against numerous experimental data on radiocesium and radiostrontium soil-to-plant transfer.

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