Depth profiling of Pu, ^{241}Am and ^{137}Cs in soils from southern Belarus measured by ICP-MS and α and γ spectrometry

Sergei F. Boulyga,*[a] Myroslav Zoriy,[b] Michael E. Ketterer[c] and J. Sabine Becker[b]

[a]*Institute of Inorganic and Analytical Chemistry, University of Mainz, Duesbergweg 10-14, 55099 Mainz, Germany. E-mail: boulyga@uni-mainz.de*
[b]*Central Department of Analytical Chemistry, Research Center Jülich, 52425 Jülich, Germany*
[c]*Northern Arizona University, Department of Chemistry, Flagstaff, AZ 86011-5698, USA*

Received 31st March 2003, Accepted 11th June 2003
First published as an Advance Article on the web 24th June 2003

The depth distribution of plutonium, americium, and ^{137}Cs originating from the 1986 accident at the Chernobyl Nuclear Power Plant (NPP) was investigated in several soil profiles in the vicinity from Belarus. The vertical migration of transuranic elements in soils typical of the 30 km relocation area around Chernobyl NPP was studied using inductively coupled plasma mass spectrometry (ICP-MS), alpha spectrometry, and gamma spectrometry. Transuranic concentrations in upper soil layers ranged from 6×10^{-12} g g^{-1} to 6×10^{-10} g g^{-1} for plutonium and from 1.8×10^{-13} g g^{-1} to 1.6×10^{-11} g g^{-1} for americium. These concentrations correspond to specific activities of $^{239+240}$Pu of 24–2400 Bq kg^{-1} and specific activity of ^{241}Am of 23–2000 Bq kg^{-1}, respectively. Transuranics in turf-podzol soil migrate slowly to the deeper soil layers, thus, 80–95% of radionuclide inventories were present in the 0–3 cm intervals of turf-podzol soils collected in 1994. In peat-marsh soil migration processes occur more rapidly than in turf-podzol and the maximum concentrations are found beneath the soil surface (down to 3–6 cm). The depth distributions of Pu and Am are essentially identical for a given soil profile. $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios vary by up to a factor of 5 at some sites while smaller variations in these ratios were observed at a site close to Chernobyl, suggesting that ^{137}Cs is dominantly particle associated close to Chernobyl but volatile species of ^{137}Cs are of relatively greater importance at the distant sites.

Introduction

The 1986 failure of the Chernobyl nuclear power plant (NPP) Reactor 4 produced widespread regional contamination of the environment with transuranic elements. A previous study[1] demonstrated that irradiated uranium concentrations in relocation areas of Belarus varied from 5×10^{-12} g g^{-1} to 2×10^{-6} g g^{-1} in the top 10 cm of the soil profile; surface concentrations were also inversely related to distance from the reactor. According to calculations of isotope production in irradiated uranium of the Chernobyl reactor,[2] the abundance of total transuranic elements (Np, Pu, Am, Cm) is about 0.4% of the core U inventory. The released radionuclides can possibly enter the biosphere, which represents a serious perturbation of the environment's indigenous content of radionuclides. The initial surface inventory of Chernobyl transuranics has now been redistributed to various soil- and radionuclide-specific depth profiles.[3] The mechanisms of migration of radionuclides in soils and groundwater will define the long-term dynamics of radioactive contamination. The migration processes depend upon the initial physical and chemical state of radioactive fallout, the soil humidity and acidity, and the effect of vegetation and soil fauna. In the initial deposition from the Chernobyl NPP, the majority of transuranic elements were contained in individual "hot" fuel particles.[4] Therefore, the migration of radionuclides in the environment was mainly determined by physical properties of fuel particles, but it only slightly depended on the chemical properties of individual actinides. However, under the effect of chemically "active" soil components, as a result of nuclear decay and oxidation of tetravalent uranium UO_2 (*i.e.* matrix of fuel particles), the particles are destroyed, and the fine particle fraction increases[1,4] which raises the leaching rate of transuranics and fission products from the fuel particles. The soluble fraction of radionuclides increases with time and the transuranics become more accessible for plants, animals and humans. Under these circumstances, biogeochemical processes at work in different soils play an increasingly important role in defining radionuclide migration. Studies of the distribution of transuranium elements in different soils in comparison to ^{236}U[1] are therefore relevant for forecasting the behavior of radioactive fallout in contaminated regions.

Along with α-spectrometry and γ-spectrometry, which are traditionally used for determination of Pu and Am, inductively coupled plasma mass spectrometry (ICP-MS) is a method of choice for the sensitive and accurate determination of long-lived transuranics in environmental samples at ultratrace (pg g^{-1}) levels. ICP-MS is attractive for these studies on account of high sensitivity, good accuracy of isotopic measurements, and relatively simple sample preparation procedures.[5–7] The objective of the present work has been to apply ICP-MS for an experimental study of the depth profiling of Pu and Am in Belarus soils typical of the 30 km relocation area around the Chernobyl NPP and to compare ICP-MS data with earlier results obtained by radioanalytical techniques on the same soil samples.

Experimental

Instrumentation

A double-focusing sector-field ICP-MS (ICP-SFMS ELEMENT, Finnigan MAT, Bremen, Germany) was used for measurements of Pu and Am performed at Research Center Jülich. The ICP torch was shielded with a grounded platinum electrode (Guard Electrode™, Finnigan MAT). A microconcentric low-flow nebulizer with membrane desolvation (Aridus, CETAC Technologies Inc., Omaha, Nebraska, USA) was used for

solution introduction in ICP-SFMS. Aqueous solution was introduced into the Aridus in the continuous flow mode *via* a peristaltic pump (Perimax 12, Spetec GmbH, Erding, Germany). Optimization of experimental parameters of ICP-SFMS was performed to maximize $^{238}U^{+}$ ion intensity while minimizing $^{238}U^{1}H^{+}$ formation; a 1 μg l^{-1} natural uranium solution was used for optimization. The measured isotopic ratio was corrected using a mass discrimination factor (assuming a linear correlation[8]) determined experimentally from measurements of a CCLU-500 laboratory standard solution; further corrections for UH^{+} at *m*/*z* 239 and detector dead time were also performed as described previously.[9] The combined uncertainty of isotopic ratio measurements was calculated by combining contributions from standard deviations of the measured ratio and background (including instrument background and interfering hydride ions), the uncertainty of mass discrimination factors, and uncertainties associated with activities of the internal standards ^{242}Pu and ^{243}Am.[10] The ICP-MS experimental parameters are summarized in Table 1. Further details of the ICP-SFMS and the measurement procedure used can be found elsewhere.[11]

Comparative analysis of Pu concentrations in three soil samples measured previously using alpha spectrometry[12] were performed at Northern Arizona University with respect to a possible influence of sample inhomogeneity on measurement accuracy. A VG Axiom MC inductively coupled plasma mass spectrometer (Thermo Elemental, Winsford, Cheshire, UK) was equipped with an ultrasonic nebulizer (CETAC U-5000AT) for high efficiency sample introduction. Sample solutions were introduced at 400 μl min^{-1}; signals were collected in an "E-Scanning" mode for $^{239}Pu^{+}$, $^{240}Pu^{+}$, and $^{242}Pu^{+}$. A dwell time of 10 ms was used to collect data at 10 narrowly spaced points (0.25 peak widths) at the summit of each peak; 50 such sweeps were recorded for each integration. Each integration required about 45 s. Integrations were collected in blocks of three. Preliminary measurements indicated that signals for $^{238}U^{+}$ were about 10,000 cps indicating satisfactory removal of U; since $^{238}U^{1}H^{+}/^{238}U^{+}$ was found to be 0.00002–0.00003, the contribution of $^{238}U^{1}H^{+}$ to the signal measured at *m*/*z* 239 was negligible. The raw ratio data ($^{239}Pu/^{242}Pu$ and $^{240}Pu/^{242}Pu$) were corrected for mass discrimination based upon a mass bias factor of 1.007 determined for $^{238}U/^{235}U$ in a natural U solution (true value = 137.88). The concentrations of ^{239}Pu and ^{240}Pu radionuclides were determined by isotope dilution technique; these were converted into the conventionally reported $^{239+240}Pu$ activity using the specific activities of each isotope.

Alpha spectrometric determinations of $^{239+240}Pu$ activity were performed using a 450 mm^2 solid state alpha detector (Canberra Industries, Inc., 800 Research Parkway, Meridien, CT 06450, USA) having a 28% counting efficiency, a background of $<10^{-5}$ s^{-1} and a resolution (FWHM) of 25 keV (^{238}U). Activities of ^{241}Am in samples (ashed soil samples, precipitation of iron hydroxide from alkali-treated samples of soils) were performed using a spectrometer suitable for X-ray and soft gamma-radiation (X-γ spectrometer). This spectrometer was equipped with a planar semi-conductor detector (PSD, detector surface of 2000 mm^2, thickness of a sensitive layer of 7 mm, an entrance window made from beryllium with a thickness of 300 μm) fabricated from pure germanium (EGP 2000-07R, Euromeasures). Germanium detector represents a semiconductor diode having a *p*-i-n structure which is sensitive to ionizing radiation, particularly X rays and gamma rays. The $E_{\gamma} = 59.6$ keV ^{241}Am emission line was used as described elsewhere.[13] Determinations of ^{137}Cs and ^{134}Cs were performed *via* gamma-lines with energies of 661.66 keV and 604.7 keV, respectively. The ^{137}Cs–^{134}Cs spectrometer was equipped with a DGDK-80B-3 semiconductor detector (Aspect Ltd., Dubna, Russia). The overall efficiency of the spectrometer-detector combination was performed using known ^{137}Cs standards of 10.3, 5.3, 1.01, and 0.49 kBq.

Table 1 Experimental parameters for the ICP-MS (Element, Finnigan MAT, situated at Research Center Jülich)

RF power/W	1200
Cooling gas flow rate/l min^{-1}	18
Auxiliary gas flow rate/l min^{-1}	1.36
Nebulizer gas flow rate/l min^{-1}	1.01
Sweep gas flow rate/l min^{-1}	4.82
Solution uptake rate/ml min^{-1}	0.08
Spray chamber temperature/°C	70
Membrane temperature/°C	160
Total acquisition time per replicate/min	0.2
Number of replicates	10
Number of measurements per sample	6
Mass resolution/m Δm^{-1}	300
Measured radionuclides	^{239}Pu, ^{240}Pu, ^{241}Am

Standards and reagents

The ^{242}Pu and ^{243}Am tracer solutions were supplied by Amersham International Ltd. (UK). The isotopic purity of the tracer solutions was monitored by ICP-MS in order to demonstrate the absence of impurities (^{239}Pu, ^{240}Pu, ^{241}Am). An isotopic standard solution of uranium (CCLU-500 laboratory standard, Nuclear Research Centre, Prague, Czech Republic) with $^{235}U/^{238}U$ isotope ratio of 0.9999 was used to determine mass discrimination for the isotope ratio measurement by ICP-MS. The solutions were diluted to the necessary concentrations for determining the isotopic ratio of uranium by ICP-MS with deionized Milli-Q water (18 MΩ) obtained from a Millipore Milli-Q-Plus water purifier and acidified to 1% subboiled HNO_3.

Samples

Soil samples were collected in May, 1994 from the relocation zone of Belarus (8–45 km to the north and north-west of Chernobyl NPP) in areas that have been undisturbed by technogenic and anthropogenic activities since the 1986 accident. The general features of these sites and the soil characteristics are representative of southern Belarus; the terrain consists of low-lying bogs, elevated terraces, and rolling plains. The soil types investigated ranged from turf-podzols to alluvial peat-marsh soils. The peat deposits analysed were moderately or strongly minerotrophic. Descriptions of sampling areas, with indications of the nearest settlement, distances to the Chernobyl NPP, relief, and soil type are given in the figure captions. The soil was collected using a coring device that was specially designed to cut 1 cm thick soil layers down to a depth of 20 cm.

Sample preparation

After homogenization, 250 g of the air-dried soil samples were ground, sieved through a 1.0 mm screen for the removal of stones and fragments of plant roots, and carefully mixed; then the samples were dried to constant weight at 105 °C for 24 h and ashed at 600 ± 50 °C for 1 h. Plant roots and vegetation were incinerated separately at 550 ± 50 °C for 2 h and then the ashed plant material was mixed with the ashed soil. For plutonium isotope analysis 2 g of the soil sample was spiked with a known quantity of ^{242}Pu yield tracer (from 1 pg g^{-1} to 100 pg g^{-1} depending on expected ^{239}Pu concentration); the mixture was milled and stirred with 20 ml HNO_3 and then with 20 ml HCl. The residue was then dissolved with warm HF, the solution was evaporated, the residue was boiled with concentrated $HClO_4$, evaporated again, and the residue was dissolved in 100 ml 7.5 M HNO_3 and filtered. After that 2 ml of NH_2OH HCl and 5 ml of 8 M $NaNO_2$ were added consecutively. After heating at 90 °C the solution was passed

through a column of Microthene supporting tri-n-octylamine (TNOA) for the plutonium retention. The column was washed with 100 ml 7.5 M HNO_3 and 100 ml of 6 M HCl. Plutonium was eluted with oxalic acid in nitric acid. The eluted Pu fractions were divided into two portions for alpha spectrometry and ICP-MS, respectively. For alpha spectrometry, Pu was electroplated on a stainless steel disk at pH 1.5–2.0 for 90 min at 550 mA cm^{-2} current density. Precipitation of iron hydroxide from alkali-treated solution of soils for ^{241}Am measurement *via* gamma-spectrometry was performed as described elsewhere.[13]

In addition, a digestion of soil samples with potassium pyrosulfate preparation procedure was performed for ICP-MS determinations of $^{239+240}Pu$ activities at Northern Arizona University. Approximately 0.30–0.35 g of ashed soil were mixed with 1.0–1.1 g of potassium pyrosulfate in a 40 ml borosilicate glass vial. A ~0.008 Bq solution aliquot of NIST 4334 g ^{242}Pu tracer was added, and the mixture was dried at 110 °C. The sample–spike–pyrosulfate mixtures were sintered at 600 °C for 2.25 h. This treatment recovers refractory non-silicate forms of Pu. After cooling, 5 ml of 16 M HNO_3 and 5 ml water were added, and the mixtures were heated at 75–80 °C for 16 h. The mixtures were filtered with a glass wool-packed pipet tip and were then diluted to 20 ml. 0.4 g of $NaNO_2$ (s) and 30 mg TEVA resin beads (EIChrom, Darien IL, USA; TE-B25-A, 100–150 μm) were added to retain Pu. The resin was collected in a pipet tip, and was rinsed using two 5 ml portions of 2 M HNO_3 followed by 1.5 ml of 8 M HCl. Pu was eluted using 1 ml of 0.05 M ammonium oxalate. The Pu fractions were diluted to 5 ml prior to analysis.

Results and discussion

Fig. 1–7 present experimentally measured distributions of radionuclides in soil profiles of various types. In general, the

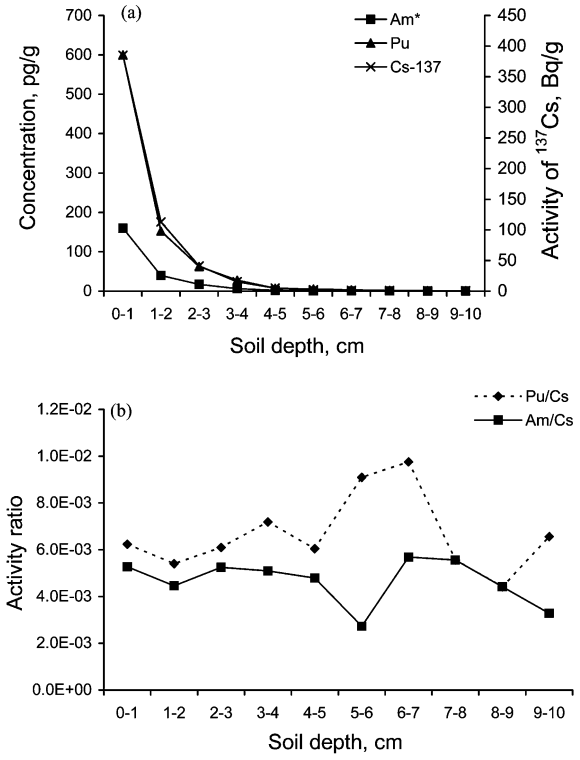

Fig. 1 Distributions of plutonium ($^{239}Pu+^{240}Pu$) and americium (^{241}Am) concentrations in comparison to distribution of ^{137}Cs specific activity (a) and $^{239+240}Pu/^{137}Cs$ and $^{241}Am/^{137}Cs$ activity ratios (b) in soil profile collected in Masany. Distance from Chernobyl NPP: 8 km; relief: withered pine forest; soil type: turf-podzol, sand. *Note, that Am concentration is multiplied by 10 in all figures with index (a) for better presentation.

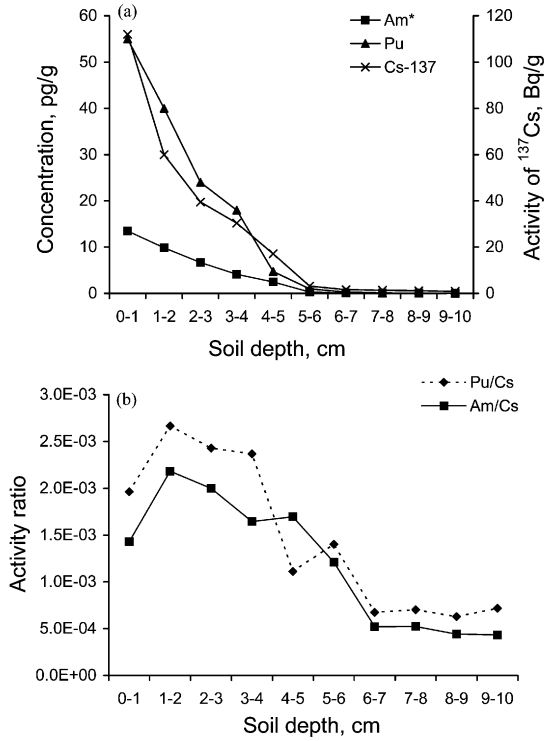

Fig. 2 Distributions of radionuclides (a) and activity ratios (b) in soil profile Krasnosel'e. 18 km from Chernobyl NPP, dune base, turf-podzol, sand.

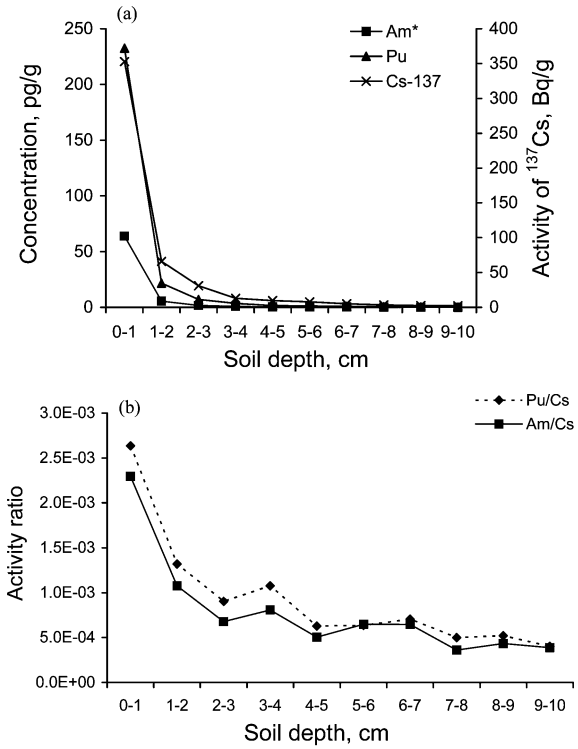

Fig. 3 Distributions of radionuclides (a) and activity ratios (b) in soil profile Lesok. 19 km from Chernobyl NPP, plain meadow, light turf-podzol, sand-clay.

concentration of Pu and Am correlated inversely with distance from Chernobyl NPP. Thus, average plutonium concentration in 10 cm soil layer decreased from 86 pg g^{-1} at 8 km (Masany) to 6 pg g^{-1} at 45 km from Chernobyl NPP (Lomachi). Isotope ratios of $^{240}Pu/^{239}Pu$ varied from 0.36 to 0.42 in upper layers of seven soil profiles analyzed. No correlation of plutonium

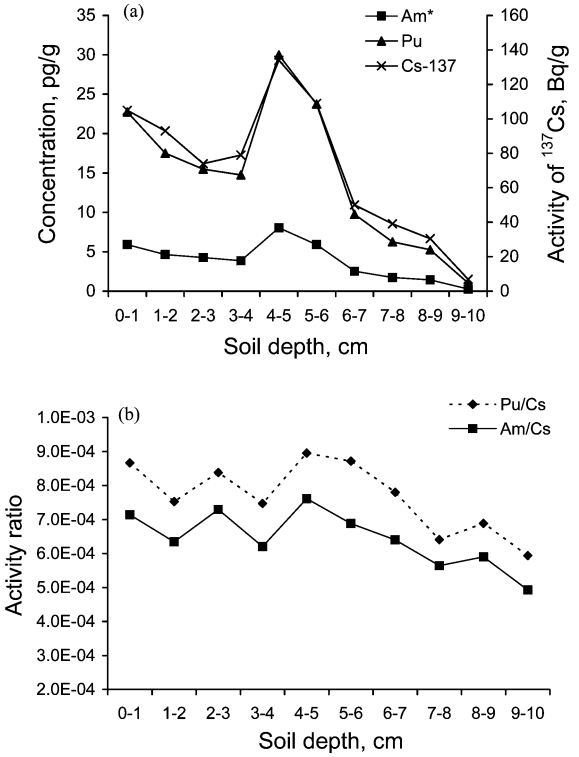

Fig. 4 Distributions of radionuclides (a) and activity ratios (b) in soil profile Kulazhin. 20 km from Chernobyl NPP, degenerating hayfield, peat-gley.

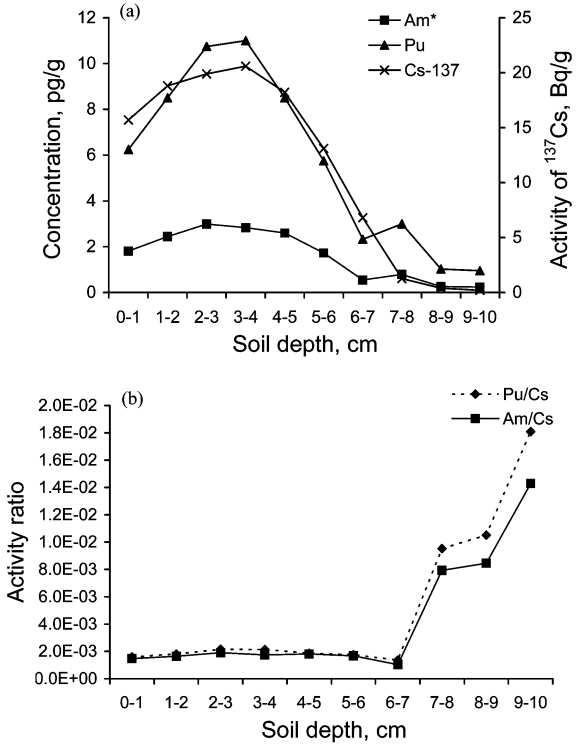

Fig. 6 Distributions of radionuclides (a) and activity ratios (b) in soil profile Dernovichi. 33 km from Chernobyl NPP, bushed meadow in lowered flood land, peat-marsh.

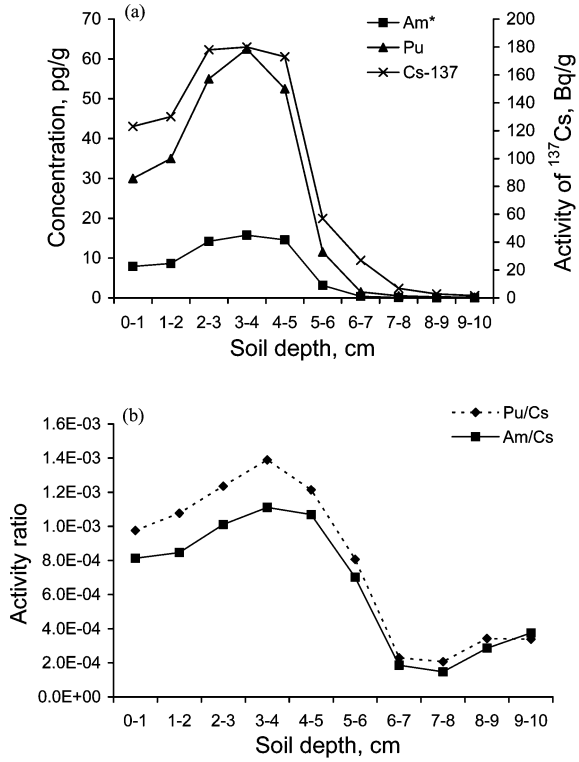

Fig. 5 Distributions of radionuclides (a) and activity ratios (b) in soil profile Radin. 21 km from Chernobyl NPP, plain meliorated peatbog, peat-gley.

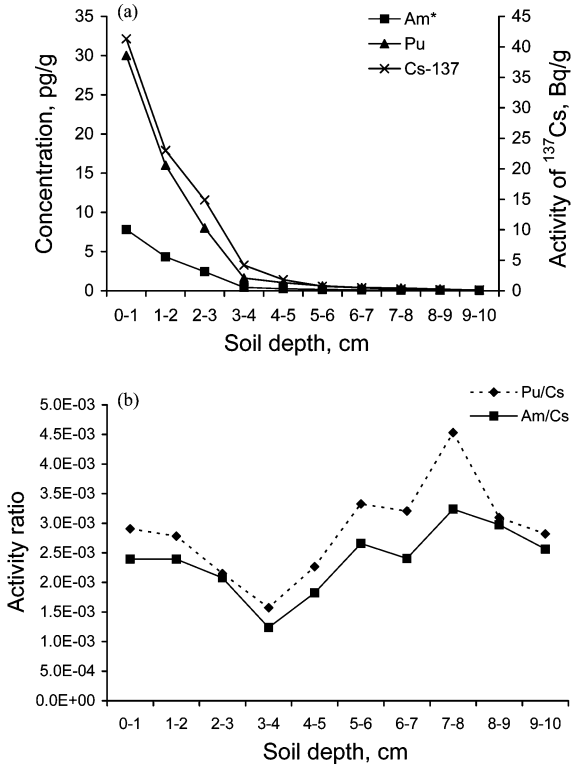

Fig. 7 Distributions of radionuclides (a) and activity ratios (b) in soil profile Lomachi. 45 km from Chernobyl NPP, meliorated massif in a low terrace above water meadow, peat-marsh on a light sedge peat.

isotope ratios with the distance from Chernobyl NPP was observed, possibly due to the concentrations of plutonium originating from Chernobyl exceeded plutonium concentration from global fallout by 1 to 4 orders of magnitude. On the other hand, precision of $^{240}Pu/^{239}Pu$ isotope ratios measured in deep soil layers *via* routine ICP-MS procedure was 5 to 30% because of low plutonium concentration (in pg g^{-1} and sub pg g^{-1}

range). A sharp decrease of radionuclide concentrations (Am, Pu and ^{137}Cs) with the depth of turf-podzol soil was observed both in areas close to Chernobyl NPP (Massany, Krasnosel'e, Lesok) and in more distant locations (*e.g.* Lomachy). The depth distribution is well-described by an exponential function. Below a depth of 3–4 cm, a less pronounced decrease of activity is observed. Thus, the top soil layer (0–1 cm), including vegetation, contained from 50 to 90% of the radionuclide inventory, and only minor amounts have penetrated to soil layers below 5 cm. These results correspond to the results of a previous study[1] where a similar decrease of Chernobyl NPP-derived U concentration with soil depth was also observed for turf-podzol soils, based upon using ^{236}U as an indicator of Chernobyl uranium.

According to a model of vertical migration of radionuclides,[14] there are two main mechanisms of migration in soil: (1) rapid migration of radionuclides added in a water-soluble form; (2) slow migration of a radioactive substance, which is fixed in hardly soluble soil complexes or "hot" particles. The results obtained for turf-podzol soils show, that the portion of actinides migrating slowly is about 80–95%. Hence one can conclude that the main part of nuclear fuel fallout is contained in low-soluble matrix and the mass transfer of radioactive substances is very slow. $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios in turf-podzol soil profiles collected in Masany (at 8 km from Chernobyl NPP) were almost constant within experimental errors down to a 10 cm depth, except for a slight peak of $^{239+240}$Pu/^{137}Cs ratio observed at the 5–7 cm depth (Fig. 1b), whilst in more distant locations Krasnosel'e and Lesok (about 20 km from Chernobyl NPP) the $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios decreased with soil depth by 3 to 5 folds (Fig. 2b and 3b). In the close Chernobyl vicinity (Masany) the radioactive fallout consists mainly of fine-dispersive particles of destroyed nuclear fuel (UO_2), aggregates of fuel particles with reactor graphite and carbon-bitumen particles *etc.*, where both actinides (Pu and Am) and fission products are "encapsulated". Therefore, the measured activity ratios in Masany soil coincided with the calculated activity ratios in the core of the Chernobyl reactor[2] which accounts for the decay of ^{137}Cs and production of ^{241}Am. In this case the radionuclides migrate as a result of mechanic migration of fuel particles and due to particle destruction, oxidation and leaching processes. On the contrary, the fallout in Krasnosel'e and Lesok represented a superposition of the fuel component (UO_2 particles) and condensation component (volatile fission products, among them caesium, captured by atmospheric aerosols), which resulted in generally lower $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios in these soils. In addition, the solubility and mobility of the condensation component is significantly higher than the mobility of the fuel component, therefore, the ratio of activities of Am and Pu to the ^{137}Cs activity decreased further with the soil depth in Fig. 2b and 3b.

Different types of radionuclide distribution in soil profiles were observed for peat-marsh soils (Fig. 4–7). In peat (Kulaszhin, Radin), maximum specific activities of actinide radionuclides were found in layers of 4–5 and 3–4 cm, respectively (Fig. 4 and 5). In these two profiles the $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios were lower than the ratios in the upper layers of the turf-podzol soils (Fig. 1b–3b), however, the averaged over profile ratios of $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs coincided within experimental errors in four soil profiles collected in Krasnosel'e, Lesok, Kulaszhin and Radin. That points to the fact, that the ratio of fuel and condensation components was similar in the four profiles collected at 18 km to 21 km from Chernobyl NPP, although the contamination level with radionuclides was different (average Pu concentration was 16 pg g^{-1}, 27 pg g^{-1}, 15 pg g^{-1}, 24 pg g^{-1} in Krasnosel'e, Lesok, Kulaszhin and Radin, respectively). The sub-surface maxima in radionuclide concentrations observed in Fig. 4a–6a can be explained by physical and chemical properties of peat profiles. The upper layer consists of partially decomposed vegetation rests, which do not bind radionuclides. In addition, the structure of this layer is loose and porous, and radioactive particles can penetrate it. The deeper layers are more solid and consist of humus complexes which have relatively high binding capacity for metals. This can explain the migration of radionuclides to deeper peat layers and fixing them at a depth of 3 to 6 cm, depending on the structure and properties of a particular soil.

A very similar distribution of Pu, Am and Cs radionuclides down to 7 cm was observed in soil profiles collected in Dernovichi (Fig. 6). However, activity ratios $^{239+240}$Pu/^{137}Cs and ^{241}Am/^{137}Cs increased significantly in the profile below 7 cm. Obviously, this increase cannot be explained by plutonium from global fallout, because measured Pu concentration in deeper soil collected in Dernovichi exceeds the global fallout concentration by more than one order of magnitude (compare, for instance, with 7–10 cm layer collected in Lomachi, Fig. 7a). Contaminations of deep soil layer in Dernovichi with Pu and Am are similar and might be due to mechanical transfer of contaminated upper soil shortly after the Chernobyl accident. It should be mentioned, that the relocation zone included initially only areas within 30 km around Chernobyl NPP.

Table 2 presents results of plutonium measurement in three soil samples collected in Masany, Krasnosel'e and Radin using alpha spectrometry (average results obtained during a previous interlaboratory comparison program in Minsk, Belarus[12]) and ICP-MS. Agreement is rather poor between these $^{239+240}$Pu activities and the previous alpha spectrometric results for samples collected in Masany and Krasnosel'e. The potassium pyrosulfate preparations were performed using very small (*ca.* 0.3 g) aliquots of solid sample and therefore the apparent disparities may largely arise from inhomogeneity of samples and the presence of varying numbers of high-activity hot particles in individual subsamples. Note that the $^{239+240}$Pu specific activity agreement is relatively good for NIST 4357, in which Pu is homogeneously distributed. Our ongoing work with soil samples from Belarus in the vicinity of the Chernobyl reactor shows that the soils are very heterogeneous with respect to specific activity (Bq kg^{-1}) and isotopic compositions. The isotopic compositions of samples from Krasnosel'e, Masany, and Radin will be the subject of a future paper.

Conclusions

The depth distributions of Pu, ^{241}Am and ^{137}Cs were investigated in soils from seven locations in Belarus from the Chernobyl vicinity. The specific activity *vs.* depth profiles indicate that migration in the soil profiles is occurring relatively slowly, and 80–95% of radionuclide inventories are present in

Table 2 Results of plutonium specific activity measurement in three soil samples collected in Masany, Krasnosel'e and Radin (1–10 cm soil layer). Uncertainties of ICP-MS are given as one standard deviation of a block of three repetitive measurements of the same sample preparation

	Alpha spectrometry[a]		ICP-MS
	Specific activity/Bq kg^{-1}		Specific activity/Bq kg^{-1}
Sample	$^{239+240}$Pu	^{238}Pu[a]	$^{239+240}$Pu
Krasnosel'e	27 ± 5	10 ± 2	15.7 ± 0.5
Masany	153 ± 23	56 ± 9	109 ± 1
Radin	15 ± 3	5 ± 1	18.4 ± 0.3
NIST 4357	10.4 ± 0.2[b]		9.7 ± 0.1, 10.9 ± 0.2

[a]Reference date 01.12.1998. [b]Certified $^{239+240}$Pu specific activity, NIST 4357 = Ocean Sediment.

the 0–3 cm intervals of turf-podzol soils. In addition, the depth distributions of actinide radionuclides (^{239}Pu, ^{240}Pu, ^{241}Am) in a specific soil profile exhibited generally similar trends. This suggests that the depth distributions of the radionuclides depend to a limited extent upon the chemical properties of the specific elements, but rather are controlled by soil type and its effect over time on the deposited particles, *e.g.* formed at high temperatures during the burning of reactor core. Upon decomposition of the deposited fuel particles, chemical fractionation of individual elements is expected whereby specific elements will be more mobile in the soil profile. $^{239+240}$ Pu/^{137}Cs and ^{241}Am/^{137}Cs activity ratios vary by up to a factor of 5 at some distant sites while smaller variations in these ratios were observed at site Masany, which is close to Chernobyl. This can be explained by the fact that ^{137}Cs is dominantly associated with fuel particles in the close Chernobyl vicinity (giving similar distributions to the actinides). In general, at more distant sites a "condensation component" representing volatile species including ^{137}Cs is of greater importance. In peat-marsh soils, the vertical transport processes are more pronounced, and maximum activities are found at 3–5 cm beneath the surface.

Acknowledgements

The authors are grateful to Prof. O. I. Yaroshevich, Dr. I. V. Zhuk and Dr. V. P. Kudrjashov for valuable discussions as well as preparation of selected samples and gamma spectrometric measurements of ^{241}Am. M. E. K. acknowledges support from the US National Science Foundation (CHE-0116804) for purchase of the Axiom ICPMS.

References

1 V. P. Mironov, J. L. Matusevich, V. P. Kudrjashov, S. F. Boulyga and J. S. Becker, *J. Environ. Monit.*, 2002, **4**, 997–1002.
2 S. N. Begichev, A. A. Borovoj, E. B. Burlakov, A. J. Gagarinsky, V. F. Demin, A. A. Khrulev and I. L. Khodakovsky, in *Fission Product Transport Processes in Reactor Accidents*, ed. J. T. Rogers, Hemisphere, New York, USA, 1990, pp. 717–734.
3 S. F. Boulyga, E. M. Lomonosova, I. V. Zhuk, O. I. Jaroshevich, V. P. Kudjashov and V. P. Mironov, *Radiat. Meas.*, 1999, **30**, 703.
4 V. Mironov, J. Matusevich and V. Kudrjashov, Destruction of fuel particles from the Chernobyl fallout on the territory of Belarus, in *Radionuclides and Heavy Metals in Environment*, ed. M. V. Frontasyeva, Kluwer Academic Publishers, The Netherlands, 2001, pp. 55–60.
5 J. S. Becker and H.-J. Dietze, Mass spectrometry of long-lived radionuclides, in *Encyclopedia of Analytical Chemistry*, ed. R. A. Meyers, John Wiley, Chichester, 2000, pp. 12947–12961.
6 J. S. Becker, *J. Anal. Atom. Spectrom.*, 2002, **17**, 1172.
7 M. E. Ketterer, B. R. Watson, G. Matisoff and C. G. Wilson, *Environ. Sci. Technol.*, 2002, **36**, 1307–1311.
8 K. G. Heumann, S. M. Gallus, G. Raedlinger and J. Vogt, *J. Anal. At. Spectrom.*, 1998, **13**, 1001.
9 W. Kerl, J. S. Becker and H. J. Dietze, *Fresenius J. Anal. Chem.*, 1997, **359**, 407.
10 *EURACHEM/CITAC Guide, Quantifying Uncertainty in Analytical Measurement*, ed. S. L. R. Ellison, M. Rosslein, A. Williams, London, 2nd edn., 2000.
11 S. F. Boulyga, C. Testa, D. Desideri and J. S. Becker, *J. Anal. At. Spectrom.*, 2001, **16**, 1283.
12 I. V. Zhuk, private communication.
13 A. V. Bushuev, V. N. Zubarev, E. V. Petrova, V. I. Popov, I. M. Proshin, O. I. Yaroshevich, I. V. Zhuk, E. M. Lomonosova, M. K. Kievets, N. N. Glubokii, Y. I. Bondar, V. P. Mironov, V. P. Kudryashov, L. E. Grusheich, S. F. Bulyga, V. S. Shkolnik, O. G. Tyupkina and M. A. Akhmetov, *Atomic Energy*, 1997, **82**, 116–123.
14 B. M. Prokhorov, *Migration of Radioactive Contamination in Soils*, Energoizdat, Moscow, 1981, p. 183.