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Radioisotope contaminations from releases of the Tomsk–Seversk nuclear facility (Siberia, Russia)

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Abstract

Soils have been sampled in the vicinity of the Tomsk–Seversk facility (Siberia, Russia) that allows us to measure radioactive contaminations due to atmospheric and aquatic releases. Indeed soils exhibit large inventories of man-made fission products including ^{137}Cs (ranging from 33,000 to 68,500 Bq m^{-2}) and actinides such as plutonium (i.e. $^{239+240}Pu$ from 420 to 5900 Bq m^{-2}) or ^{241}Am (160–1220 Bq m^{-2}). Among all sampling sites, the bank of the Romashka channel exhibits the highest radioisotope concentrations. At this site, some short half-life gamma emitters were detected as well indicating recent aquatic discharge in the channel. In comparison, soils that underwent atmospheric depositions like peat and forest soils exhibit lower activities of actinides and ^{137}Cs.

Soil activities are too high to be related solely to global fallout and thus the source of plutonium must be discharges from the Siberian Chemical Combine (SCC) plant. This is confirmed by plutonium isotopic ratios measured by ICP-MS; the low $^{241}Pu/^{239}Pu$ and $^{240}Pu/^{239}Pu$ atomic ratios with respect to global fallout ratio or civil nuclear fuel are consistent with weapons grade signatures.

Up to now, the influence of Tomsk–Seversk plutonium discharges was speculated in the Ob River and its estuary. Isotopic data from the present study show that plutonium measured in SCC probably constitutes a significant source of plutonium in the aquatic environment, together with plutonium from global fallout and other contaminated sites including Tomsk, Mayak (Russia) and Semipalatinsk (Republic of Kazakhstan). It is estimated that the proportion of plutonium from SCC source can reach 45% for ^{239}Pu and 60% for ^{241}Pu in the sediments.

Keywords: Tomsk–Seversk; Radioisotope; Soil contamination; Plutonium isotopes; Ob–Irtysh catchment

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1. Introduction

The region of Tomsk-7 (or Seversk) is known for it Siberian Chemical Combine (SCC), which consists of five nuclear reactors originally designed for military production: a chemical separation plant, a reprocessing facility for uranium and plutonium, an uranium enrichment plant and storage facilities for radioactive waste. Together with Mayak, Seversk was a major former Soviet Union weapon production and fuel reprocessing facilities site. Since the international agreement on reducing strategic armaments was reached, activities on the site have gradually been shifted away from military towards civil applications.

At Seversk, releases of radionuclides in the environment are due to accidents and to waste handling practices (Alexakhin et al., 2004). Most of the radioactive wastes at Seversk are in liquid form (Liquid Radioactive Waste, LRW). Until 1982, LRW was disposed into at least two surface reservoirs that contain an estimated total inventory of 4.7×10^{18} Bq. These reservoirs have been now covered. However, since 1963, LRW has been disposed of by borehole injection into Cretaceous strata at a depth of 280−400 m at a distance of about 10−20 km east of the Tom River. Around 4×10^{19} Bq of long-lived nuclides is reported to have been disposed of by these means (Lgotin and Makushin, 1998) and a total volume of disposed wastes may reach 8 of millions of cubic meters (Bradley et al., 1996).

The inventory of accidents with atmospheric releases is difficult to set out at Seversk, which is still a closed city. However, from local information sources it is usually accepted that since the time nuclear activities started in 1952−1955, at least 23 accidents occurred resulting in release of radioactivity to the environment.

The main accident that has been documented occurred on April 6, 1993. An explosion happened at the U−Pu extraction line unit in a vessel containing 4 m^3 of nitric acid solution with Pu, Np and U. The total α-activity (mainly Pu: 79% and U: 20%) of the solution was about 7.4×10^{11} Bq and total β-activity was about 1.85×10^{13} Bq (Tcherkezian et al., 1995). Part of this solution has been ejected to the atmosphere during the explosion. Tcherkezian et al. (1995) reported that Pu contamination in soil due to the 1993 accident ranges between 10 and 13 Bq kg^{-1} with maximum up to 17 Bq kg^{-1} and that the accumulation in topsoil layer was up to 1100 Bq m^{-2} by that time. The $^{238}Pu/^{239+240}Pu$ ratio in soil of 0.35 ± 0.07 suggests that it corresponds to partially burn nuclear fuel. This accidental release involves also gamma emitters such as ^{95}Zr, ^{95}Nb, ^{106}Ru, ^{125}Sb and ^{137}Cs (Tcherkezian et al., 1995; IAEA, 1998). However, neither ^{134}Cs nor ^{241}Am was reported as radionuclides related to this accident.

Bradley et al. (1996) suggest that the largest release of radioactive waste into the Russian environment has occurred as a result of inadequate management of radioactive waste resulting from the reprocessing of spent fuel. Most of the areas showing major radioactive contaminations due to military activities in Russia are located in Siberia and Urals. In this region, Seversk and Mayak are considered as the two major sources of radioactive contaminants affecting the Ob River and its estuary (Paluszkiewicz et al., 1997; Cochran et al., 2000). The total amount of radioactivity released from these sources into the Ob aquifer has been estimated to be 4.63×10^{19} Bq (Bradley et al., 1996).

However, because of the lack of information on the radiochemical characters of each source, it is still difficult to determine their respective importance to the radioactive pollution of the Ob River and its estuary. In this study, we present new data on the region of Tomsk−Seversk in order to precise the isotopic composition of actinides and Cs in various soil profiles and a peat bog. These data allow to follow the process of radioactive contamination of the environment since the beginning of the nuclear operation in 1952.

2. Sampling location and procedure

Three zones have been selected for sampling around the Siberian Chemical Combine (SCC) (Fig. 1). The first zone, the Petropavlovskiy Ryam is a peat bog located NW from the SCC. The second zone is a forest soil (Soil-S) located 7 km North from Seversk and the third zone is the bank of the Romashka canal that comes from the SCC and serves as cooling water and discharge facility of radionuclides into Tom River and Ob River.

Sampling of the Petropavlovskiy Ryam peat was performed using a 380 cm length drilling device located in the center of the bog. Until a depth of 23 cm, samples of 2 cm thickness were collected, then 5 cm thick samples until 53 cm deep and finally, two samples were collected at depths of 73 and 183 cm. In the Soil-S profile, eight samples were collected up to a depth of 40 cm. This soil consists of loess sediments mainly made of fine-grained quartz and clays. In the bank of the Romashka canal, three profiles located at three different levels relative to the stream of the canal were taken. From the water level to the top of the bank the profiles are Rc, R and Rb (Fig. 2). According to the very low ^{137}Cs activities, only three samples from the top of the R and Rc profiles were analysed for actinides whereas 16 samples from the Rb soil were analyzed both for Cs and actinides, because here it was possible to measure the entire radioisotope inventory. In this profile, samples were collected every 3 cm.

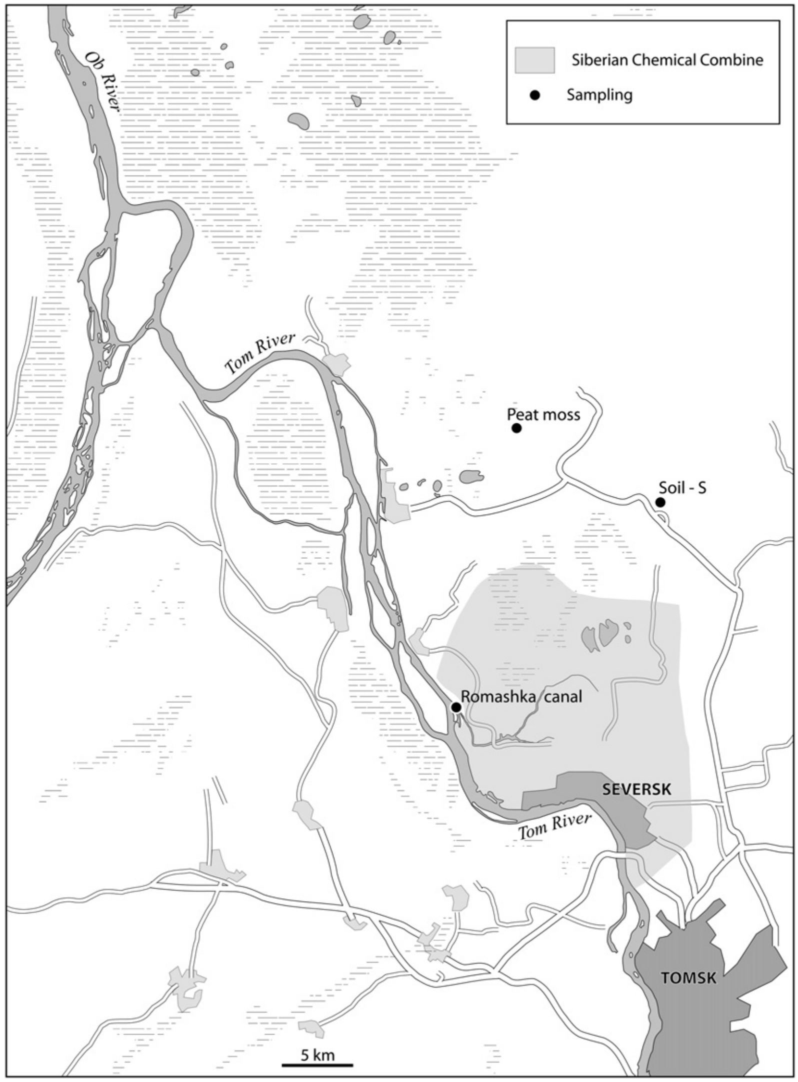

Fig. 1. Location of the SCC and of the sampled areas: forest soil, peat and Romashka canal.

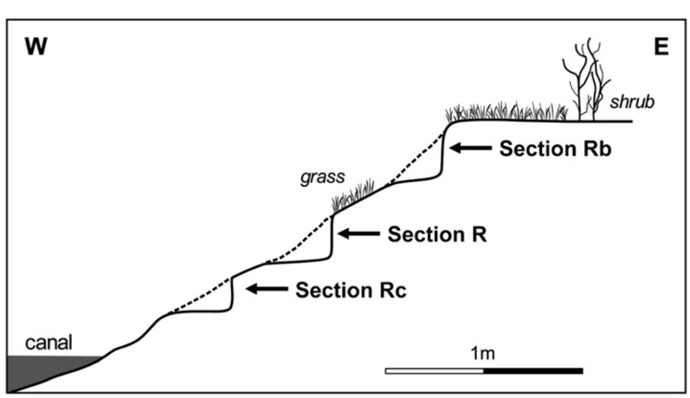

Fig. 2. Location of the R, Rb and Rc profiles at the Romashka canal.

3. Analytical methods

Peat soils were dried at a temperature of 40 °C and crushed prior to the analytical procedure. For determination of ^{210}Pb and ^{214}Pb (as a measure for ^{226}Ra) peat samples were placed in small cylindrical measuring containers of 17 ml volume and weighted, which allowed to estimate their density. The measurements of those radioactive lead isotopes were carried out on a γ-spectrometer with a low-level HPGe detector in ''Laboratoire de Mesure de la Radioactivité dans l'Environnement (IRSN, Orsay, France)''. The detector construction ensures a high sensitivity for lower γ energies, which is necessary for the determination of ^{210}Pb (46.5 keV) and ^{214}Pb (351.9 keV). Counting time of up to 8000 s was required to detect the very low activity of naturally occurring ^{238}U decay series nuclides. Especially, self-attenuation effects in the sample were taken into account.

^{239}Pu, ^{240}Pu and ^{241}Pu concentrations were determined by using ICP-MS, in ''Laboratoire de Mesure de la Radioactivité dans l'Environnement (IRSN, Orsay, France)'', following analytical protocol described by Agarande et al. (2004). Briefly, 30–70 g of ash sample was added to a known amount of a certified ^{242}Pu spike certified for its concentration and isotopic composition before leaching for a few hours in concentrated HNO_3/HCl mixture. Actinides were concentrated by coprecipitation with iron hydroxide (pH 8) or calcium oxalate (pH 1.5). The precipitate was dissolved in 8 M HNO_3 to perform Pu separation from other actinides (U, Am, and Th). The Pu fraction obtained was further purified. In addition to the elimination of the major matrix elements, U should also be as efficiently as possible eliminated in the final Pu fraction to avoid peak overlap on ^{239}Pu peak by the $^{238}U^1H^+$ poly-atomic species. To allow determination of the ^{238}Pu/$^{239+240}$Pu activity ratio samples were additionally analyzed at PSI and measured by means of high resolution alpha spectrometry using intrinsic Si semi-conductive detectors.

Plutonium isotopes activity was acquired with a VG axiom single collector mass spectrometer placed in class 1000 clean room. Liquid samples are introduced into the ICP ion source by means of a peristaltic pump and directed to a nebulizer system where the liquid is converted into an aerosol. Here a micro-concentric nebulizer system was used allowing the uptake of 50×10^{-6}–100×10^{-6} l min^{-1} of solution (MCN 6000 or l PFA 50 nebulizer). The detection limit, defined as three times the standard deviation of blank solution (5% HNO_3), is 1.4 fg ml^{-1} for ^{239}Pu (0.003 mBq ml^{-1}). Starting from a 100 g sample, the final Pu fraction being collected into 3 ml of 5% HNO_3, and with a 65% average chemical recovery, the minimum detectable activity (MDA is 10 times the detection limit) is 1.4 mBq kg^{-1}.

4. Distribution of radionuclides in peat

As shown in Fig. 3, in the peat profile Pu increases from top to the bottom. The lowest concentration is found in moss taken from the surface (0.5 Bq kg^{-1} for $^{239+240}$Pu) while the highest value is found at a depth of about 20 cm (Pu activity concentration of 10.5 Bq kg^{-1}, Table 1A and Fig. 3). In the overall profile, accumulation of ^{239,240}Pu is very high, up to 421 Bq m^{-2} (Table 2), compared to global fallout predicted in this region: 40–50 Bq m^{-2} according to Olivier et al. (2004) or 50–60 Bq m^{-2} according to Hardy et al. (1973). Only very contaminated areas such as in the vicinity of Sellafield, Chernobyl, Mayak or in Semipalatinsk exhibit such high Pu anomalies. It seems therefore likely that most of this plutonium have a local source, probably the SCC discharge.

^{241}Am shows the same general trend along the profile than Pu, with an important positive anomaly (5.7 Bq kg^{-1}) at the depth of 19–21 cm. The total accumulation of ^{241}Am in this profile was calculated to the amount of 161 Bq m^{-2}. This is much more than accumulation due to the decay of ^{241}Pu from global fallout in the Northern Hemisphere, which has been estimated to be 21 Bq m^{-2} (Duffa, 2001). Here again such accumulation must be related to local sources.

In the layers below 5 cm, ^{238}Pu/$^{239+240}$Pu ratios range between 0.02 and 0.04, i.e. values that are commonly predicted for global fallout at this latitude, i.e. 0.03 according to Hardy et al. (1973). However, the high Pu concentrations cannot be explained solely by such source. Furthermore, the scatter of the ^{238}Pu/$^{239+240}$Pu ratios with values as low as 0.012 also suggests the occurrence of various events having different Pu isotope compositions. At the top of the profile, a very high ^{238}Pu/$^{239+240}$Pu value of 0.104 was measured. This ^{238}Pu/$^{239+240}$Pu anomaly in the moss is also related to a very high ^{241}Am/$^{239+240}$Pu ratio of 1.41 ± 0.69. At the deeper part of the profile, ^{241}Am/$^{239+240}$Pu and ^{238}Pu/$^{239+240}$Pu ratios show similar variations but differ significantly in comparison with the topsoil layer. This suggests the occurrence of a special event where radioactive particles were released into the atmosphere that were highly enriched with respect to ^{241}Am and ^{241}Pu.

The data for ^{137}Cs plotted vs. depth show a very steep profile ranging from about 100 Bq kg^{-1} in the uppermost layer to maximum 216 Bq kg^{-1} at 20 cm. With further depth the activity concentration decreases rapidly within

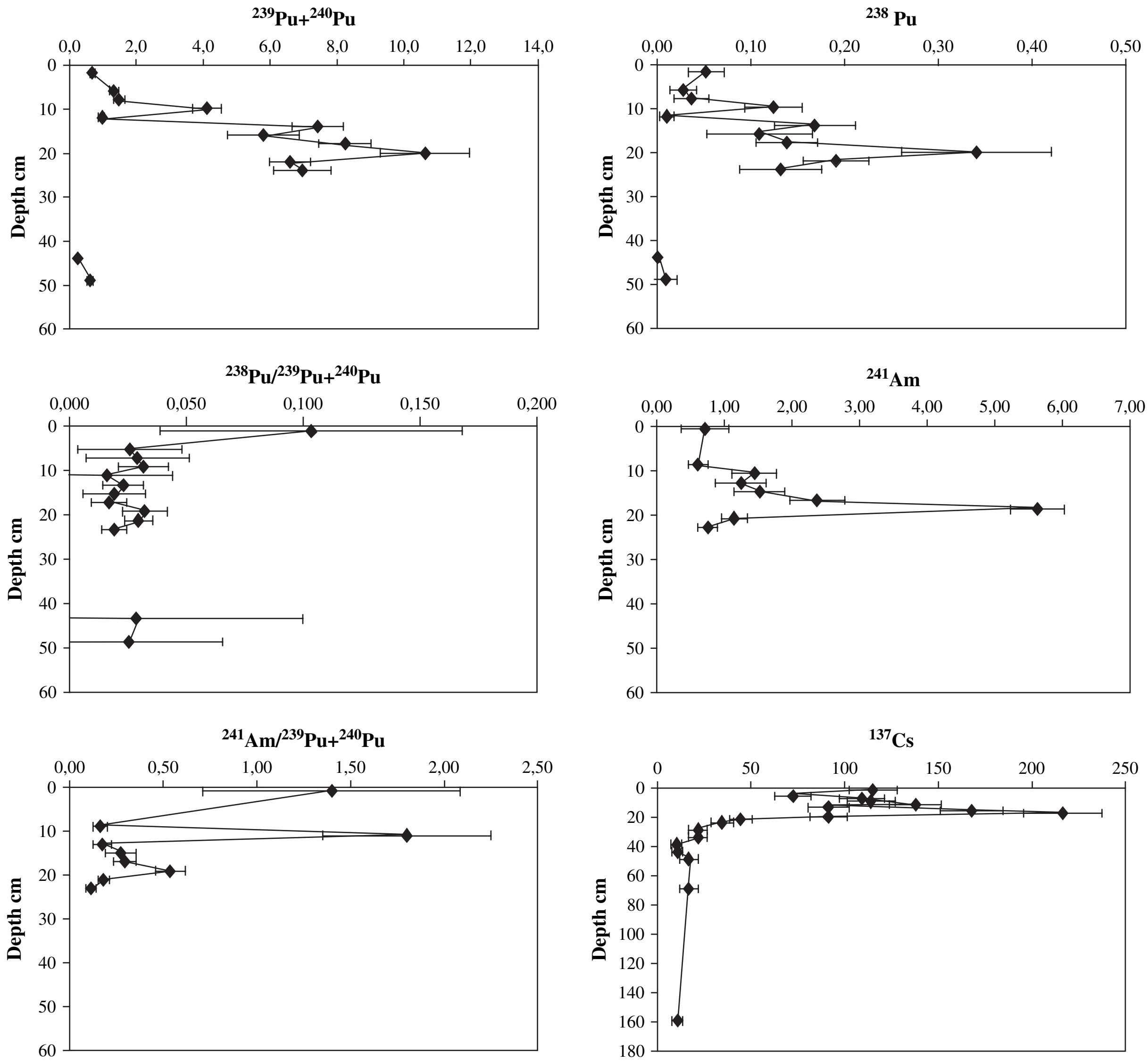

Fig. 3. Distribution of activities (Bq/kg) of $^{239+240}$Pu, ^{238}Pu, ^{241}Am and ^{137}Cs and ^{238}Pu/$^{239+240}$Pu and ^{241}Am/$^{239+240}$Pu ratios in the peat profile.

a few centimeters to values below 30 Bq kg^{-1}. However, significant activity of ^{137}Cs (i.e. larger than 10 Bq kg^{-1}) can be observed below 30 cm, which is accounted by downward diffusion of this element in the peat. MacKenzie et al. (1997) and Kudelsky et al. (1996) reported the high mobility of this element in peats from Scotland and from the catchment of the Pripyat River (Belarus). The total accumulation of ^{137}Cs in the peat profile reaches 11,246 Bq m^{-2}. The surface activity recorded in the peat profile is higher than the accumulation of radiocesium from global fallout at the Tomsk latitude: 3430 Bq m^{-2} according to UNSCEAR (2000) to 5040 Bq m^{-2} according to Aoyama et al. (2006). Besides ^{137}Cs no other man-made gamma emitters, especially no short lived isotopes like ^{60}Co (5.2 y), ^{134}Cs (2.0 y) or europium isotopes (half-life of ^{152}Eu and ^{154}Eu are 13.6 and 8.8 y, respectively) have been measured in the peat samples. This shows that the peat soil did not undergo any recent deposition of man-made radionuclides.

^{214}Pb was not detected above the background in any sample, so the total ^{210}Pb activities were taken to represent fully unsupported ^{210}Pb. The regular decrease of the ^{210}Pb activity with depth allows the use of a Constant Rate of Supply (CRS) of unsupported ^{210}Pb, which is a simplifying but reasonable assumption over time spans in the order

Table 1A
Activity and activity ratios of natural (^{210}Pb and ^{214}Pb) and man-made radionuclides including $^{239+240}Pu$, ^{238}Pu, ^{241}Am, ^{137}Cs (in Bq kg^{-1} of dry matter), $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ in the peat profile (reference date: 01. 01. 2004)

Sample	Depth	^{210}Pb	^{214}Pb	$^{239+240}Pu$	^{238}Pu	$^{238}Pu/^{239+240}Pu$	^{241}Am	$^{241}Am/^{239+240}Pu$	^{137}Cs
2	1–5	630 ± 90	<26	0.5 ± 0.1	0.06 ± 0.02	0.104 ± 0.06	0.7 ± 0.4	1.41 ± 0.69	114 ± 13
2–2	5–7	450 ± 80	<23	1.2 ± 0.1	0.03 ± 0.01	0.026 ± 0.02			71 ± 10
3	7–9	450 ± 60	<26	1.3 ± 0.2	0.04 ± 0.02	0.030 ± 0.02			108 ± 12
4	9–11	520 ± 100	<31	4.0 ± 0.4	0.13 ± 0.05	0.032 ± 0.01	0.6 ± 0.2	0.17 ± 0.04	113 ± 13
5	11–13	330 ± 70	<14	0.8 ± 0.1	0.01 ± 0.01	0.012 ± 0.01	1.5 ± 0.3	1.81 ± 0.45	137 ± 14
6	13–15	400 ± 80	<20	7.3 ± 0.8	0.17 ± 0.04	0.024 ± 0.01	1.3 ± 0.3	0.18 ± 0.05	90 ± 11
7	15–17	280 ± 40	<23	5.7 ± 1.1	0.11 ± 0.05	0.020 ± 0.01	1.6 ± 0.3	0.28 ± 0.08	167 ± 17
8	17–19	230 ± 60	<26	8.1 ± 0.8	0.14 ± 0.03	0.017 ± 0.01	2.4 ± 0.4	0.30 ± 0.06	216 ± 21
9	19–21	190 ± 60	<16	10.5 ± 1.3	0.34 ± 0.06	0.033 ± 0.01	5.7 ± 0.3	0.54 ± 0.08	90 ± 10
10	21–23	280 ± 40	<13	6.4 ± 0.6	0.19 ± 0.06	0.030 ± 0.01	1.2 ± 0.3	0.18 ± 0.03	43 ± 6
11	23–28	290 ± 60	<15	6.8 ± 0.9	0.14 ± 0.10	0.020 ± 0.01	0.8 ± 0.3	0.12 ± 0.03	33 ± 6
12	28–33	393 ± 60	<15						20 ± 5
13	33–38	66 ± 31	<32						20 ± 5
14	38–43								9 ± 3
15	43–48			0.1 ± 0.02	0.003 ± 0.003	0.029 ± 0.07			9 ± 3
16	48–53			0.5 ± 0.09	0.012 ± 0.012	0.026 ± 0.04			15 ± 5
20	68–73								15 ± 5
28	158–183								9 ± 4

of years. Given this assumption, the ^{210}Pb ages of the peat layers can be estimated (CRS dating model from Appleby, 2001) as well as the peat accumulation rate and furthermore the accumulation rates of deposited artificial radionuclides (Table 1B and Fig. 4). The strongest deposition of Pu occurred soon in the fifties (i.e. about 10 mBq cm^{-2} y^{-1}) followed by a slow decrease in the following decades (6 mBq cm^{-2} y^{-1} in the seventies and 2–5 mBq cm^{-2} y^{-1} in the eighties). An important decrease of the Pu flux occurred during a short period of time around 1980. The 1993s accident is shown by a plateau of the curve.

5. Distribution of radionuclides in Soil-S profile

The results of the radioisotope measurements of soil profile ‘‘S’’ are given in Table 3 and the total accumulations in Table 2. The ^{137}Cs, $^{239+240}Pu$ and ^{241}Am accumulations in the soil profiles reach very high values up to 33,811 Bq m^{-2}, 1316 Bq m^{-2} and 439 Bq m^{-2}, respectively. Such values are again too high to be related solely to

Table 1B
Peat ages and peat and plutonium accumulation rates

Depth	^{210}Pb (Bq kg^{-1})	Calendar age	Peat density (g cm^{-3})	Peat accumulation rate (g cm^{-2} y^{-1})	$^{239+240}Pu$ (mBq cm^{-2} y^{-1})
1–5	630 ± 90	2004	0.21	0.10	1
5–7	450 ± 80	1992	0.15	0.10	2
7–9	450 ± 60	1989	0.15	0.09	3
9–11	520 ± 100	1985	0.15	0.07	5
11–13	330 ± 70	1981	0.18	0.09	1
13–15	400 ± 80	1977	0.16	0.07	6
15–17	280 ± 40	1972	0.19	0.08	5
17–19	230 ± 60	1967	0.19	0.09	6
19–21	190 ± 60	1962	0.20	0.09	10
21–23	280 ± 40	1957	0.16	0.05	10
23–28	290 ± 60	1950	0.19	0.04	11
28–33	393 ± 60	1940	0.12	0.02	
33–38	66 ± 31	1875	0.08	0.02	
38–43	33	1836	0.05	0.01	
43–48	8	1784	0.05	0.01	

Table 2
Inventory of artificial radionuclides in studied soils (Bq m^{-2})

	Peat	Soil-S	Soil Rb
$^{239+240}Pu$	421	1316	5903
^{241}Am	161	439	1220
^{137}Cs	11,246	33,811	68,544

global fallout and as for the peat bog, other sources for these radionuclides have to be considered. The distribution of radionuclides along the profile shows diffusion curves, which reach "normal" concentration at a level deeper than 20 cm (Fig. 5). However, some variations of actinides' ratios can be observed in the profile. In samples located in the first 15 cm from the surface, $^{238}Pu/^{239+240}Pu$ activity ratio shows values (0.01 ± 0.003) which are lower than those expected for global fallout (0.03) (Table 3). It is also interesting to note that this ratio increases to a value as high as 0.18 at the bottom of the profile. A decrease of $^{241}Am/^{239+240}Pu$ ratio is observable along the profile from 0.39 to 0.21.

Activities determined using ICP-MS are in good agreements with activities determined from alpha spectrometry, excepted for the sample located at the surface of the profile, just below the litter (Table 4). In this sample, $^{239+240}Pu$ concentrations are much more higher when analyzed by ICP-MS (66 ± 1.7 Bq kg^{-1}) than by alpha counting (11.9 ± 0.3 Bq kg^{-1}). In order to confirm such results, the solution analyzed by ICP-MS has also been analyzed by alpha counting giving the same result (58.0 ± 18 Bq kg^{-1}). Such difference is then interpreted as resulting to the heterogeneity of the Pu distribution at the surface of the soil. Tcherkezian et al. (1995) have described "hot particles" around Seversk that deposited after the 1993 accident. Therefore, when hot particles are involved the analysis is likely to yield not agreeing results between two aliquot samples.

The graphical illustration of the activity concentration vs. depth is presented in Fig. 5 for ^{238}Pu, $^{239+240}Pu$, ^{241}Am, ^{137}Cs and their isotopic ratios. The concentration data show again smooth profiles with maximum values close to the surface and a strong decrease of the measured activities with depth. Such patterns are typical for soil diffusion profiles of strongly adsorbing species onto clay minerals.

In order to distinguish between global fallout and further sources of radionuclide emission into the environment, normalized three isotope diagrams are useful. For instance, mixture of two components in such *X*/*Z* vs. *Y*/*Z* diagrams plot on a straight line between the two end members. Such a situation is presented in Fig. 7 with $^{241}Pu/^{239}Pu$ vs. $^{240}Pu/^{239}Pu$. This figure shows that most of the samples have constant values (0.00075 and 0.05). Higher values of $^{241}Pu/^{239}Pu$ ratio (exceeding 0.001 in samples S3, S7 and S8) are arguable due to the high uncertainty of this ratio (>50%). Such very low $^{240}Pu/^{239}Pu$ values, with respect to stratospheric fallout of nuclear tests (0.18) or civil nuclear fuel (0.408 given by Chernobyl fallout, see Ketterer et al., 2004), are comparable with the signature of nuclear weapons accidents occurring in Palomares (0.056 ± 0.003) and in Thule (0.033 ± 0.004) (Mitchell et al., 1997).

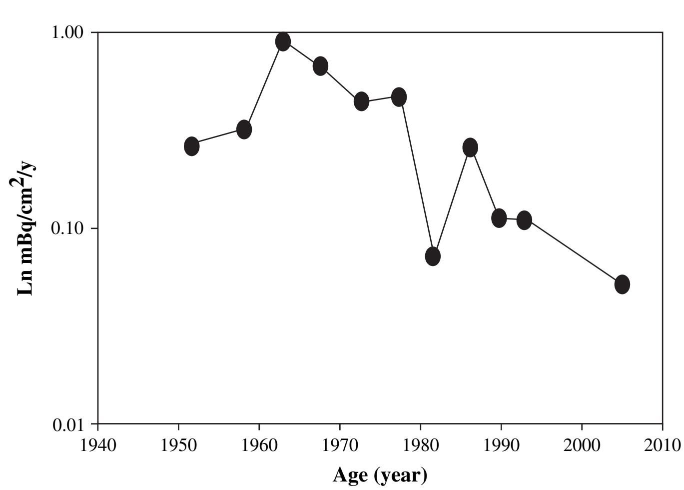

Fig. 4. Flux of $^{239+240}Pu$ vs. age in the peat bog.

Table 3
Activity and activity ratios of man-made radionuclides including $^{239+240}Pu$, ^{238}Pu, ^{241}Am, ^{137}Cs (in Bq kg^{-1} of dry matter), $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ ratios in the Soil-S profile (reference date: 01. 01. 2002)

Sample	Depth (cm)	$^{239+240}Pu$	^{238}Pu	$^{238}Pu/^{239+240}Pu$	^{241}Am	$^{241}Am/^{239+240}Pu$	^{137}Cs	^{134}Cs
S1 (litter)	0	11.30 ± 0.3	0.15 ± 0.03	0.013 ± 0.003	4.4 ± 0.8	0.39 ± 0.07	290 ± 10	<2
S2 (soil)	0–3	11.90 ± 0.3	0.15 ± 0.03	0.013 ± 0.003	4.9 ± 0.8	0.41 ± 0.07	375 ± 11	<2
S3 (soil)	3–6	11.70 ± 0.3	0.18 ± 0.03	0.015 ± 0.003	2.5 ± 0.7	0.21 ± 0.06	331 ± 10	<2
S4 (soil)	6–11	7.20 ± 0.2	0.09 ± 0.02	0.013 ± 0.003	2 ± 0.6	0.28 ± 0.08	155 ± 6	<2
S5 (soil)	11–18	1.80 ± 0.1	0.02 ± 0.01	0.011 ± 0.006	<0.5	0.17 ± 0.05	39 ± 3	<2
S6 (soil)	18–25	0.49 ± 0.05	0.01 ± 0.01	0.020 ± 0.021	<0.5		11 ± 2	<2
S7 (soil)	25–32	0.18 ± 0.03	0.02 ± 0.01	0.111 ± 0.059	<0.5		4 ± 2	<2
S8 (soil)	32–40	0.14 ± 0.03	0.03 ± 0.01	0.214 ± 0.085	<0.5		3 ± 2	<2

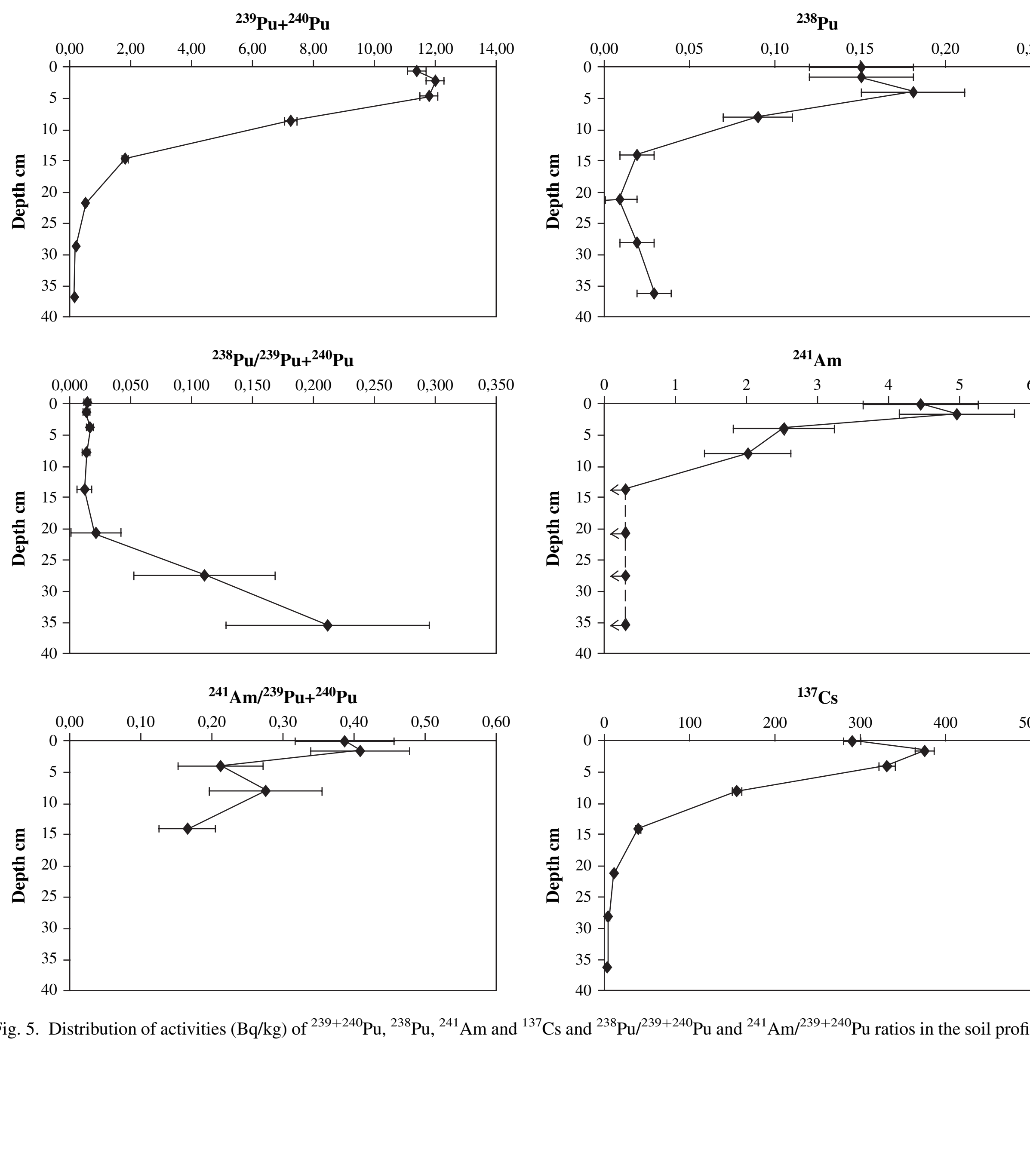

Fig. 5. Distribution of activities (Bq/kg) of $^{239+240}Pu$, ^{238}Pu, ^{241}Am and ^{137}Cs and $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ ratios in the soil profile S.

Table 4
Activities (Bq kg^{-1}), concentrations (at/gr) and atomic ratios of some plutonium isotopes (^{239}Pu, ^{240}Pu and ^{241}Pu) measured by ICP-MS

Sample	Depth (cm)	^{239}Pu (Bq kg^{-1})	^{239}Pu (at/gr)	^{240}Pu (Bq kg^{-1})	^{240}Pu (at/gr)	^{241}Pu (Bq kg^{-1})	^{241}Pu (at/g)	^{240}Pu/^{239}Pu	^{241}Pu/^{239}Pu
S1	0	7.9 ± 1.0	8.7E+09 ± 1E+09	1.5 ± 0.2	4.5E+08 ± 6E+07	9.2 ± 2.3	6.00E+06 ± 1E+06	0.052 ± 0.0090	0.00070 ± 0.00019
S2	0–3	55.4 ± 1.4	6.1E+10 ± 2E+09	10.6 ± 0.3	3.3E+09 ± 9E+07	104.7 ± 4.4	6.70E+06 ± 3E+06	0.054 ± 0.0020	0.00112 ± 0.00006
S3	3–6	12.6 ± 0.2	1.4E+10 ± 2E+09	2.5 ± 0.1	7.6E+08 ± 2E+07	17.4 ± 2.0	1.10E+07 ± 1E+06	0.055 ± 0.0010	0.00084 ± 0.00010
S7	25–32	0.2 ± 0.0	1.7E+08 ± 3E+09	0.03 ± 0.006	9.6E+06 ± 2E+06	0.5 ± 0.2	3.20E+05 ± 1E+05	0.055 ± 0.0140	0.00188 ± 0.00086
S8	32–40	0.1 ± 0.0	1.1E+08 ± 2E+09	0.03 ± 0.003	6.0E+06 ± 1E+06	0.3 ± 0.2	2.00E+05 ± 2E+05	0.054 ± 0.0120	0.00177 ± 0.00145
Rb1	0–3	32.9 ± 0.5	3.61E+10 ± 6E+08	6.0 ± 0.1	1.81E+09 ± 3E+07	43.8 ± 2.1	2.75E+07 ± 1E+06	0.050 ± 0.0009	0.00077 ± 0.00004
Rb2	3–6	40.0 ± 1.1	4.28E+10 ± 1E+09	7.3 ± 0.2	2.18E+09 ± 6E+07	52.0 ± 2.5	3.50E+07 ± 2E+06	0.051 ± 0.0013	0.00082 ± 0.00004
Rb4	9–12	33.5 ± 0.7	3.78E+10 ± 8E+08	6.1 ± 0.1	1.83E+09 ± 4E+07	46.5 ± 1.9	3.00E+07 ± 1E+06	0.049 ± 0.0012	0.00080 ± 0.00003
Rb5	12–15	29.5 ± 0.6	3.27E + 10 ± 7E+08	5.4 ± 0.1	1.61E+09 ± 3E+07	40.5 ± 1.7	2.75E+07 ± 1E+06	0.049 ± 0.0011	0.00085 ± 0.00003
Rb7	18–21	14.6 ± 0.3	1.61E+10 ± 3E+08	2.7 ± 0.1	8.03E+08 ± 2E+07	17.7 ± 2.4	1.15E+07 ± 2E+06	0.050 ± 0.0010	0.00072 ± 0.00010

Table 5
Activity and activity ratios of man-made radionuclides including ^{60}Co, ^{134}Cs, ^{137}Cs, ^{152}Eu, ^{154}Eu, $^{239+240}Pu$, ^{238}Pu, ^{241}Am (in Bq kg^{-1} of dry matter), $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ ratios in the Rb soil profile (reference date: 01. 01. 2004)

Sample	Depth (cm)	$^{239+240}Pu$	^{238}Pu	$^{238}Pu/^{239+240}Pu$	^{241}Am	$^{241}Am/^{239+240}Pu$	^{137}Cs	^{134}Cs	^{60}Co	^{152}Eu	^{154}Eu
Rb1	0	27.5 ± 0.6	0.56 ± 0.03	0.02 ± 0.002	18 ± 2	0.65 ± 0.07	100 ± 3	22 ± 2	1025 ± 30	165 ± 4	55 ± 2
Rb2	3	39.8 ± 0.9	0.58 ± 0.03	0.015 ± 0.001	18 ± 2	0.45 ± 0.05	217 ± 5	12 ± 1	540 ± 15	189 ± 4	61 ± 3
Rb3	6	36.1 ± 0.9	0.65 ± 0.03	0.018 ± 0.001	8.3 ± 1.3	0.23 ± 0.04	238 ± 5	7 ± 1	258 ± 10	152 ± 4	40 ± 2
Rb4	9	32.5 ± 0.9	0.51 ± 0.04	0.016 ± 0.002	4.2 ± 0.9	0.13 ± 0.03	233 ± 5	1.3 ± 0.8	131 ± 6	146 ± 4	37 ± 2
Rb5	12	32.6 ± 0.9	0.47 ± 0.04	0.014 ± 0.002	4.1 ± 0.9	0.13 ± 0.03	336 ± 7	1.7 ± 0.8	60 ± 4	105 ± 3	17 ± 1
Rb6	15	31.2 ± 0.9	0.67 ± 0.04	0.022 ± 0.002	3.5 ± 0.8	0.11 ± 0.03	447 ± 8	<1	34 ± 2	55 ± 2	7 ± 1
Rb7	18	17.4 ± 0.9	0.35 ± 0.04	0.02 ± 0.003	1.5 ± 0.7	0.09 ± 0.04	340 ± 6	<1	22 ± 2	30 ± 3	4 ± 1
Rb8	21	5.5 ± 0.3	0.12 ± 0.02	0.021 ± 0.005	0.7 ± 0.4	0.13 ± 0.07	198 ± 5	<1	11 ± 1	11 ± 2	<2
Rb9	24	0.99 ± 0.08	0.02 ± 0.01	0.02 ± 0.012	<0.5		35 ± 2	<1	5 ± 1	<2	<2
Rb10	27	1.11 ± 0.08	0.02 ± 0.01	0.015 ± 0.009	<0.5		14 ± 2	<1	<1	<2	<2
Rb11	30						17 ± 2	<1	<1	<2	<2
Rb12	33	0.86 ± 0.05	0.01 ± 0.01	0.012 ± 0.013	<0.5		16 ± 2	<1	<1	<2	<2
Rb13	36						12 ± 2	<1	<1	<2	<2
Rb14	39	0.75 ± 0.05	0.01 ± 0.01	0.013 ± 0.014	<0.5		2 ± 1	<1	<1	<2	<2
Rb15	42						$<1 \pm 0.1$	<1	<1	<2	<2
Rb16	45						$<1 \pm 0.1$	<1	<1	<2	<2

Thus from $^{240}Pu/^{239}Pu$ the high Pu inventory observed in the S profile (1774 Bq m^{-2}) can be inferred to military fuel produced inside SCC.

6. Distribution of radionuclides in the banks of the Romashka canal

Among the three profiles made in the bank of the Romashka canal only the upper one (profile Rb) presents significant radioactive anomalies (Tables 5 and 6 and Fig. 6). Surprisingly, the two lowest profiles Rc and R which are close to the stream of the canal present only low-levels of ^{137}Cs with values as low as 31.0 and 1.4 Bq kg^{-1}, respectively. These values greatly differ from the accumulations observed in the Rb profile where a ^{137}Cs activity as high as 68,544 Bq m^{-2} has been accumulated (Table 2). In this profile, maximum of ^{137}Cs is found at a depth of 15 cm (447 ± 8 Bq kg^{-1}) whereas it decreases to very low values at 25 cm deep (Table 5 and Fig. 5). Among man-made gamma emitters some short lived radionuclides such as ^{60}Co (5.2 y), ^{134}Cs (2.0 y) and europium isotopes (half-life of ^{152}Eu and ^{154}Eu are 13.6 and 8.8 y, respectively) have been measured. This shows that recently deposited of rather short lived man-made radionuclides can be seen in the Rb soil. Furthermore, vertical variations of Cs isotopes and Eu isotopes ratios with depth could indicate that several depositions of discharge wastewater with different isotope compositions occurred at this site.

Plutonium and americium are also strongly accumulated in profile Rb compared to the other profiles. The total accumulations for $^{239+240}Pu$ and ^{241}Am are up to 5903 and 1220 Bq m^{-2}, respectively (Table 2). Pu is mainly present in the first 15 cm from the surface whereas Am is mostly concentrated in the first 5 cm. $^{238}Pu/^{239+240}Pu$ ratio, ranging between 0.012 and 0.022, exhibits less variability than in previous profiles. However, a decrease of $^{241}Am/^{239+240}Pu$ ratio is noticeable from top to the bottom Rb profile. The ranges of $^{241}Pu/^{239}Pu$ (0.00072–0.00085) and $^{240}Pu/^{239}Pu$ (0.0487–0.0512) atomic ratios are close to the ones observed in the Soil-S profile (Table 4).

7. Discussion

Comparison of artificial radionuclides inventories in four zones allows to point out impacted places submitted to chronic and accidental release from SCC. In this respect, the bank of the Romashka canal exhibits the strongest actinides and fission products inventory, probably due to recent aquatic discharges in the canal. In comparison, soils that underwent atmospheric depositions like peat and forest soil exhibit lower activities of actinides and ^{137}Cs. More detailed studies based on large scale monitoring program are however needed to precisely determine to most contaminated places around Seversk.

Actinides inventories reported in this study (Pu ranges between 1774 and 8826 Bq m^{-2}) are too high to be related solely to global fallout and thus the source of plutonium must be discharges from the SCC plant. Such heavily contaminated fields are commonly observed around nuclear facilities where accidental release of radionuclides had occurred in the environment like in Chernobyl (Muramatsu et al., 2000), Sellafield (Jones et al., 1996) or Mayak (Skipperud et al., 2005).

The source of the actinides is confirmed by the plutonium ratios observed in the soils. Thus the very low $^{241}Pu/^{239}Pu$ and $^{240}Pu/^{239}Pu$ atomic ratios with respect to global fallout ratio or civil nuclear fuel are consistent

Table 6
Activity and activity ratios of man-made $^{239+240}Pu$, ^{238}Pu, ^{137}Cs (in Bq kg^{-1} of dry matter), $^{238}Pu/^{239+240}Pu$ ratio in the R and Rc soil profiles (reference date: 01. 01. 2004)

Sample	Depth (cm)	$^{239+240}Pu$	^{238}Pu	$^{238}Pu/^{239+240}Pu$	^{137}Cs
R2	10	0.18 ± 0.03	<0.01	<0.05	1.4 ± 0.5
R3	12.5	0.09 ± 0.02	<0.01	<0.05	1.9 ± 0.5
R4	15	0.04 ± 0.02	<0.01	<0.05	<1.0
Rc1	6	n.a.	n.a.		31 ± 2
Rc2	12	n.a.	n.a.		3 ± 1
Rc3	18	n.a.	n.a.		1.0 ± 0.9
Rc4	24	n.a.	n.a.		<1
Rc5	30	n.a.	n.a.		<1
Rc6	36	n.a.	n.a.		<1

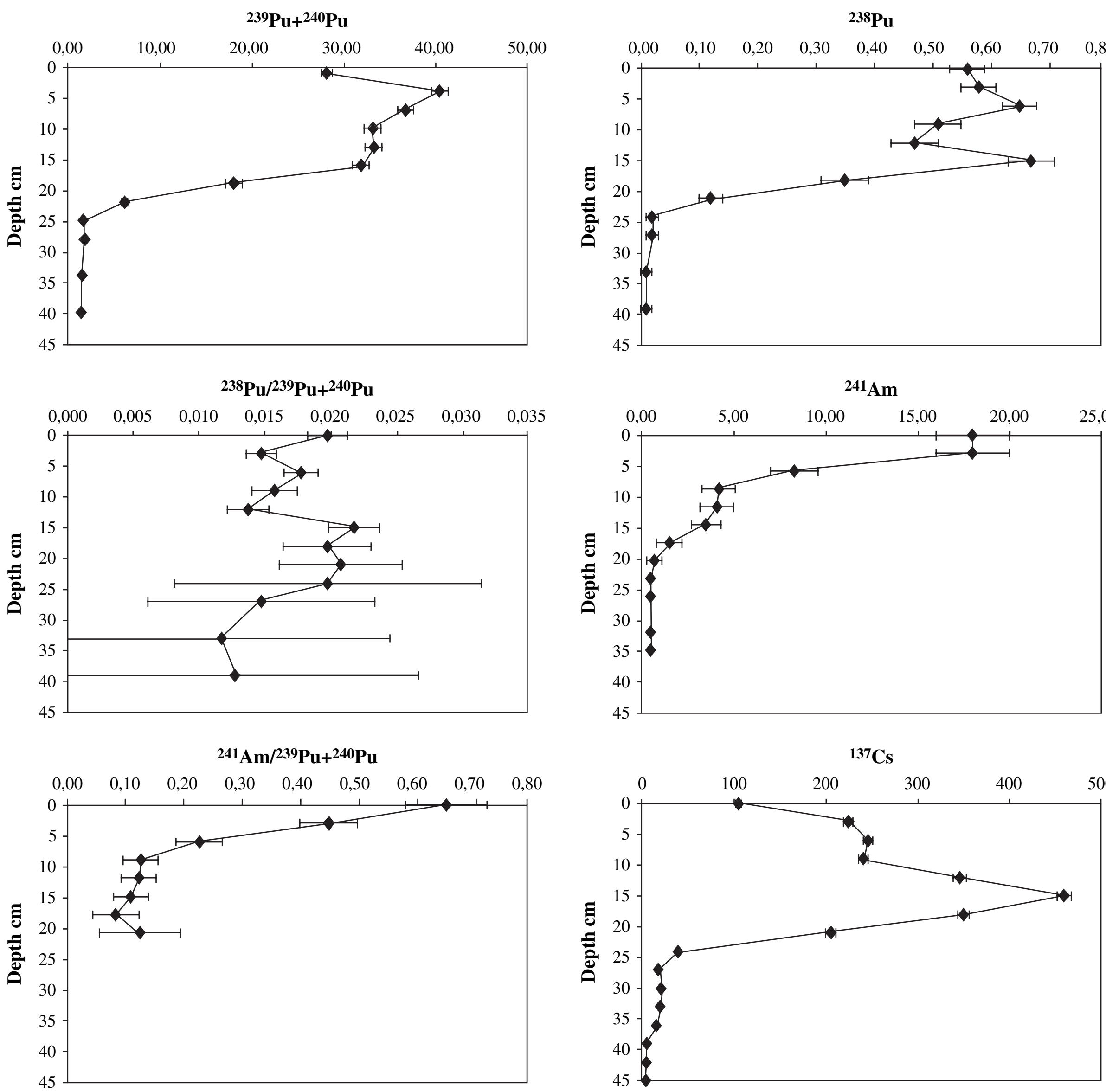

Fig. 6. Distribution of activities (Bq/kg) of $^{239+240}Pu$, ^{238}Pu, ^{241}Am and ^{137}Cs and $^{238}Pu/^{239+240}Pu$ and $^{241}Am/^{239+240}Pu$ ratios in the soil profile Rb located in the bank of the Romashka canal.

with those reported for weapons grade (Figs. 7 and 8). It is noteworthy that nearly the overall plutonium measured in the vicinity of SCC is related to releases from those facilities. The analysis of one water sample collected in the canal Romashka has shown the occurrence of short half-life radionuclides such as ^{239}Np (19 ± 1 Bq l^{-1}) and ^{131}I (0.20 ± 0.05 Bq l^{-1}) (half-life of ^{239}Np and ^{131}I is 2.35 and 8.0 d, respectively) suggesting continuous release of radioactive contaminants from the SCC plant. As the isotopic composition of the discharges remains unknown it was not possible to point out chronic and accidental release. Especially, it was not possible to quantify the total plutonium accidental release occurring in 1993, although the $^{238}Pu/^{239+240}Pu$ ratio of the accident is known (Tcherkezian et al., 1995). Possibly, plutonium chronic discharges are so large that it is not possible to evidence any impact of the accident.

At least three heavily contaminated sites occur inside the Ob–Irtysh catchment basin (i.e. Tomsk–Seversk, Mayak, in Russia and Semipalatinsk, in Republic of Kazakhstan). Thus the release of radioactive contaminants in the rivers by

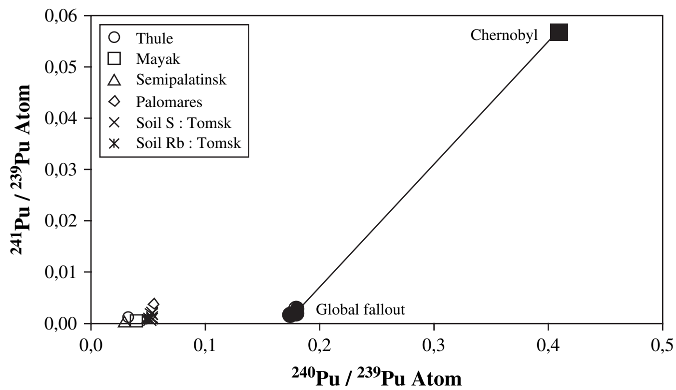

Fig. 7. Plutonium isotopic ratios of some samples from S and Rb profiles and other possible sources of plutonium including Chernobyl fallout (Ketterer et al., 2004), global fallout (Ketterer et al., 2004; Kelley et al., 1999), production and testing of weapons (in Mayak and Semipalatinsk, after Beasley et al., 1998), nuclear weapons (Thule and Palomares accidents, after Mitchell et al., 1997). Samples from Tomsk are away from the mixing line joining the global fallout and Chernobyl values.

nuclear facilities, the drainage of the heavily contaminated ground and underground as well as the drainage of global fallout account for the strong plutonium activity concentrations and the variability of plutonium ratios observed in sediments of the Ob–Irtysh basin (Cochran et al., 2000; Kenna and Sayles, 2002; Beasley et al., 1998; Baskaran et al., 1996). Up to now, the influence of Tomsk–Seversk plutonium discharges was subject to speculation (Cochran et al., 2000; Kenna and Sayles, 2002). Isotopic data from the present study show indeed clearly that plutonium measured in SCC probably constitutes to massive source of plutonium in the aquatic environment (Fig. 8). This is suggested by the mixing line where "military" (including Tomsk, Mayak and Semipalatinsk) and global fallout plutonium constitute the two end members. From this pattern, we estimated the contribution of Seversk plutonium releases in the Ob sediments by using: (1) the mixing equation that had allowed Duffa and Fréchou (2003) to estimate the proportion of Pu released by Marcoule NRP and (2) the isotopic signatures of the two sources, i.e. Tomsk plutonium releases ($^{240}Pu/^{239}Pu = 0.05$ and $^{241}Pu/^{239}Pu = 0.0007$) and plutonium global fallout ($^{240}Pu/^{239}Pu = 0.19$ and $^{241}Pu/^{239}Pu = 0.003$). Thus we estimated that the proportion of plutonium from SCC source can reach 45% for ^{239}Pu and 60% for ^{241}Pu in the Ob River sediments.

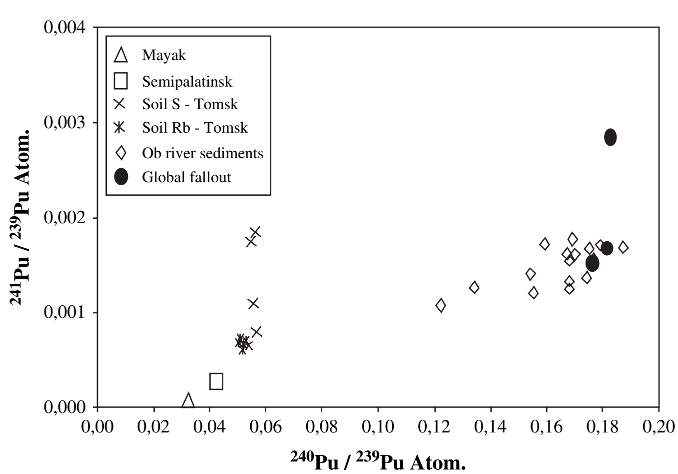

Fig. 8. Plutonium isotopic ratios of the Ob River sediments (after Cochran et al., 2000) and other possible sources of plutonium, including global fallout (Ketterer et al., 2004; Kelley et al., 1999) and soils from strongly contaminated sites located in the Ob–Irtysh catchment: SCC discharges (present study) and Mayak discharges Semipalatinsk test sites (from Beasley et al., 1998).

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