Concentrations of ^{239}Pu and ^{240}Pu and Their Isotopic Ratios Determined by ICP-MS in Soils Collected from the Chernobyl 30-km Zone

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Soil samples collected from three forest sites within the 30-km zone around the Chernobyl reactor were analyzed for ^{239}Pu and ^{240}Pu by ICP-MS The average $^{240}Pu/^{239}Pu$ atom ratio in contaminated surface soil samples, values of which are scarce in the literature, was 0 408 There were almost no differences in the $^{240}Pu/^{239}Pu$ ratios between the individual samples analyzed, although the $^{239+240}Pu$ levels varied very widely (i e from 6 3 to 1430 Bq kg^{-1} dry weight) depending on the distance from the reactor and on the soil layers investigated This result corresponded to area-related activities for $^{239+240}Pu$ between 1 1 kBq m^{-2} and 13 3 kBq m^{-2} It was estimated that about half of the Pu migrated from the organic layers to the underlying mineral layers The $^{240}Pu/^{239}Pu$ ratio observed in the Chernobyl area was much higher than that attributed to weapons fallout (ca 0 18) The high ratio was related to the high burn-up grade of the reactor fuel The $^{240}Pu/^{239}Pu$ ratio observed might be used as a 'fingerprint' in identifying the distribution of Chernobyl-derived Pu in the environment and in distinguishing it from other sources e g global fallout Relationships between the concentrations of Pu and those of ^{137}Cs, ^{60}Co, and ^{125}Sb were also discussed

Introduction

Plutonium is one of the most important anthropogenic radionuclides in relation to nuclear industries Its two isotopes, ^{239}Pu (half-life 24110 y) and ^{240}Pu (6564 y), are the most abundant of Pu isotopes in the environment In the Chernobyl reactor accident, it was estimated that about $8\,7 \times 10^{15}$ Bq of $^{239+240}Pu$ were released (*1*) Several papers (*2*−*5*) have already reported the levels of Pu in environmental samples collected from areas around Chernobyl after the accident The most commonly used method for the Pu analysis is α-spectrometry However, this method cannot resolve the ^{239}Pu and ^{240}Pu peaks because of their similar energies (5 15 and 5 16 MeV) Analytical results obtained by α spectroscopy are therefore expressed as the total activity of ^{239}Pu and ^{240}Pu (i e $^{239+240}Pu$)

Information on Pu isotopic compositions in the environment should be very useful in understanding the source of the contamination because the composition depends on a number of factors e g burn up grade of nuclear fuels type of nuclear weapons etc There are several studies (e g refs *3*−*5*) in which $^{239+240}Pu$ and ^{238}Pu (half life 87 7 y) were determined by α spectrometry for samples contaminated with the Chernobyl fallout The $^{238}Pu/^{239+240}Pu$ ratios in the Chernobyl samples are known to be higher than those in global fallout However, there are only a few publications which report reliable data on the ratio of the most abundant two Pu isotopes (i e $^{240}Pu/^{239}Pu$ ratios) in environmental samples, including those contaminated by the Chernobyl accident Therefore, it is worthwhile to use methods other than α spectrometry for providing such information The $^{240}Pu/^{239}Pu$ ratios are also very useful for estimating Pu isotopic composition in the fuel at the time of the accident and for understanding the distribution of the Chernobyl Pu in the environment

Recently we have developed a method for the determination of ^{239}Pu and ^{240}Pu in environmental materials by means of ICP-MS (inductively coupled plasma mass spectrometry) and reported the $^{240}Pu/^{239}Pu$ ratios in IAEA standard reference materials and in some Japanese soil samples (*6*) In the present study, we have applied this technique in order to determine ^{239}Pu and ^{240}Pu and their ratios in soil samples collected within the 30-km zone around the Chernobyl reactor

Materials and Method

Soil samples were collected from three forests in the 30-km zone around the Chernobyl reactor (NNP) by a group from the Federal Office of Radiation Protection, Germany, in 1994 and 1995 Locations of the sampling places are as follows Kopachy 2 (K2), 6 0 km to the southeast of the NPP Dityatki 1 (D1), 28 5 km to the south of the NPP, and Dityatki 3 (D3) 26 km to the south of the NPP

Lux et al (*3*) describes some details of the sampling places (types of soils, plants, etc) as well as the levels of several nuclides measured in previously collected samples in these same areas during 1992 and 1993 In the present study we prepared composite samples, e g mixtures of L (litter) Of (fermented organic layer), and Oh (humificated organic layer) based on the area-related density of each layer In addition to the organic layers, the underlying Ah horizon (transition layer) and B horizon (mineral layer) were also used in the analysis

The procedure used for the analysis of Pu was based on a method reported previously (*6*) Only a brief description is given here Samples (1−10 g, depending on the concentration level of Pu) were mixed with a known amount of ^{242}Pu (CRM 130, New Brunswick Laboratory) as a spike Each sample was placed in a Pyrex beaker, and 8 M (or more concentrated) nitric acid (30−80 mL) was added The beaker was covered with a watchglass and heated on a hot plate (180−200 °C) for about 5 h The warm leachate was filtered through a glass fiber filter The residue in the beaker was boiled again with 8 M nitric acid for about 30 min and then filtered All the filtrates were collected in a beaker and heated on a hot plate until a thick wet paste was obtained The wet paste was dissolved by adding nitric acid and then it was diluted with deionized water to adjust the acidity to 8 M

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TABLE 1 Analytical Results for ^{239}Pu and ^{240}Pu and Their Atomic Ratios in Chernobyl Soil Samples Together with Area Related Activity[a]

sampling points	soil layers	^{239}Pu (Bq kg^{-1})	^{240}Pu (Bq kg^{-1})	$^{239+240}Pu$ (Bq kg^{-1})	$^{240}Pu/^{239}Pu$ (atom ratio)	density[b] (kg m^{-2})	$^{239+240}Pu$ (Bq m^{-2})
			10 km Zone (Kopachy)				
K2	L/Of/Oh 94	572 ± 10	858 ± 19	1430 ± 22	0 408 ± 0 003	5 2	7431
K2	L/Of/Oh 95	451 ± 12	680 ± 17	1130 ± 21	0 411 ± 0 002	5 2	5881
K2	Ah/B 95	30 8 ± 0 5	45 9 ± 0 7	76 7 ± 0 9	0 406 ± 0 004	86 5	6630
			30-km Zone (Dityatki)				
D1	L/Of/Oh 94/95	55 3 ± 1 2	81 6 ± 1 8	137 ± 2	0 403 ± 0 001	4 4	602
D1	Ah/B 94/95	2 6 ± 0 1	3 7 ± 0 1	6 3 ± 0 2	0 386 ± 0 013	86 9	544
D3	L/Of/Oh 94	69 8 ± 1 2	106 ± 3	175 ± 3	0 412 ± 0 008	4 4	771
D3	L/Of/Oh 95	68 8 ± 1 5	103 ± 3	172 ± 3	0 408 ± 0 004	4 4	756
D3	B 94/95	5 5 ± 0 1	7 9 ± 0 2	13 4 ± 0 2	0 390 ± 0 001	37 1	498
ratio av for all samples (*8*)					0 403 ± 0 009		
ratio av for samples excluding Ah and B layers (*5*)					0 408 ± 0 003		

[a] Note average of two separate analyses (duplicate determinations) with standard deviation (±) [b] Soil density per unit area used for the analyses (kg m^{-2}) Concentrations of $^{239+240}Pu$ per unit area (Bq m^{-2}) were calculated from the analytical results and the density data

nitric acid (40−100 mL) With the addition of sodium nitrite to the sample solution, Pu was converted to tetravalent Pu-(IV), which is the only retainable form in chromatography columns The sample solution was loaded onto the column containing 2 mL of Dowex 1 × 8 at a speed of <2 mL m^{-1} The resin was washed with 8 M nitric acid (40 mL) and then with 10 M hydrochloric acid (40 mL) Finally, ammonium iodide (5%)−10 M hydrochloric solution (40 mL) was added to reduce Pu(IV) to Pu(III) which is not retained in the resin To volatilize iodine completely, nitric acid (4 mL) and hydrogen peroxide (1 mL) were added, and the solution was heated to dryness The residue was dissolved with 4% nitric acid to make the final solution Concentrations of Pu isotopes were determined with a quadruple type ICP-MS (Yokogawa PMS 2000) using a conventional liquid nebulizer (Meinhard type) Concentrations of ^{239}Pu and ^{240}Pu were calculated from the results of isotopic ratios relative to the ^{242}Pu spike (isotope dilution method) Atom ratio was obtained from direct measurement data (rather than being calculated from the individual concentration values of ^{239}Pu and ^{240}Pu) A Pu isotope standard solution (NIST-947) with a known $^{240}Pu/^{239}Pu$ ratio was used to check the accuracy

During the ICP MS measurements, three determinations of the Pu isotopes were made for each sample solution, and the mean values were calculated The relative standard deviations (RSDs) for the three determinations were in most cases less than 4% Better RSDs of 0 7−1 8% were found in determinations of the $^{240}Pu/^{239}Pu$ ratio, because the measurement of the ratio did not require any normalization to the ^{242}Pu isotope spike count rate In other words, the influence of the sensitivity fluctuation was less in direct measurement of the isotope ratio ($^{240}Pu/^{239}Pu$) than in the absolute measurement for the individual nuclides, which were normalized to the ^{242}Pu spike Two separate analyses were performed for all samples The errors for the duplicate analyses were on average about 7% for the concentration measurement and 1 2% for the measurement of the $^{240}Pu/^{239}Pu$ ratio

Concentrations of ^{137}Cs and some other γ-emitting nuclides were determined by counting with a Ge-detector for 80 000 s The decay correction was made at the beginning of the measurement for this series of samples (June 1997) Details on the γ spectrometry method used in this study were described in our previous paper (*8*)

Results and Discussion

Levels of ^{239}Pu and ^{240}Pu Analytical results of the concentrations of ^{239}Pu and ^{240}Pu and their isotopic ratios obtained for eight soil samples collected from the Chernobyl area are shown in Table 1 Reproducibilities of the duplicate analyses for each sample are satisfactory In two soil samples (L+Of+Oh layers) collected from Kopachy (6 km from the NPP), very high $^{239+240}Pu$ concentrations (mean value 1280 Bq kg^{-1} dry weight (DW)) are found The concentrations in the three samples (L+Of+Oh layers) from Dityatki (28 5 or 26 km) are on average 161 Bq kg^{-1} DW, which is about one-eighth of that from Kopachy The levels of $^{239+240}Pu$ found in this study agree well with the values reported by Lux et al (*3*) for samples collected from the same areas in 1992 and 1993 Compared to the concentrations in the composite samples of the L+Of+Oh layers, the underlying Ah/B layers show markedly lower $^{239+240}Pu$ concentrations (i e 77 Bq kg^{-1} DW for Kopachy (K2) and 9 9 Bq kg^{-1} DW for Dityatki (mean of D1 and D3)), suggesting that Pu deposited on forest soil is retained by the organic layers very effectively However, if we consider density and thickness of the Ah/B layers, a considerable amount of Pu was observed to be distributed in the mineral layers (see the following discussion) Area related activities which correspond to the inventory for both organic and mineral layers are also given in Table 1 At K2, the measured area related activity (sum of results for the organic and the mineral layers) is 13 kBq m^{-2}, whereas at D1 and D3 the values are 1 1 kBq m^{-2} and 1 3 kBq m^{-2}, respectively We think it is interesting to note that for all three sites, roughly the same proportion of Pu can be found in the organic horizons (L+Of+Oh) and the mineral horizons (Ah+B) i e the proportion of the inventory is about 1 1 indicating the possible migration of Pu into the deeper layers These results provide useful information in understanding the radionuclide inventory and redistribution of the nuclide in the forest compartment Accordingly we cannot exclude the possibility that part of the initially deposited Pu has already reached even deeper layers

$^{240}Pu/^{239}Pu$ Isotopic Ratios The $^{240}Pu/^{239}Pu$ atom ratios determined in this study are also shown in Table 1 The isotopic ratios obtained for the eight samples are in a relatively narrow range, 0 386−0 412, although the range of the concentration is quite wide (6 3−1430 Bq kg^{-1} DW) The mean value of the $^{240}Pu/^{239}Pu$ atom ratio for eight samples is 0 403 ± 0 009 The mean ratio in five samples from the surface layer (organic layers) which is the most contaminated layer is 0 408 ± 0 003 The $^{240}Pu/^{239}Pu$ ratios observed in the Chernobyl area are markedly higher than the global fallout ratio of 0 176 ± 0 014 due to weapons tests as reported by Krey (9) The value is also much higher than that for samples in other contaminated areas such as Irish Sea sediment (about 0 21) (*6*, *10*), Mururoa sediment (about 0 05) (*6*, *11*), and Semipalatinsk soil (about 0 05) (*10*)

TABLE 2 Literature Survey on ^{239}Pu and ^{240}Pu Inventories in the Chernobyl Reactor and the Calculated Ratio of $^{240}Pu/^{239}Pu$

ref	^{239}Pu	^{240}Pu	$^{240}Pu/^{239}Pu$ atom ratio
(*13*)	0 85 PBq	1 2 PBq	0 38
(*12*)	412 kg	176 kg	0 425
(*1*)	0 96 PBq	1 5 PBq	0 43
(*14*)	4 7 TBq/ton U	9 7 TBq/ton U	0 56

[a] $^{240}Pu/^{239}Pu$ atom ratios were calculated from the reported values of ^{240}Pu and ^{239}Pu

The important reactions producing ^{239}Pu and ^{240}Pu in the reactor are as follows

$$^{238}U + n \rightarrow {}^{239}U \rightarrow {}^{239}Np + e^- \rightarrow {}^{239}Pu + e^-$$

$$^{239}Pu + n \rightarrow {}^{240}Pu$$

With the increase of burn-up time of the fuel in the reactor the $^{240}Pu/^{239}Pu$ ratio becomes higher due to the higher production of ^{240}Pu Some estimates of the inventory of several isotopes, including ^{239}Pu and ^{240}Pu, in the No 4 reactor at the time of the Chernobyl accident have been made For example Belyaev et al (*12*) estimated the core's inventory of the important long-lived radionuclides based on the following data Operation time 715 effective days, total uranium in the fuel 190 2 tons, average core burn-up 11 MWd/kg U They reported the amounts of ^{239}Pu and ^{240}Pu in the reactor were 412 and 176 kg, respectively Using these data, we calculated the $^{240}Pu/^{239}Pu$ atom ratio as 0 425 There are also some other estimations of inventory in the literature (*1*, *12*–*14*) Those data are summarized in Table 2 The $^{240}Pu/^{239}Pu$ isotopic atom ratios calculated from the inventory data are also shown in the table They are in a relatively wide range (i e 0 38–0 56) Our measured values on the $^{240}Pu/^{239}Pu$ ratio (about 0 40) obtained for the contaminated soil near the reactor should be useful in validating the estimation of the nuclides' composition in the burned fuel at the time of the accident

There are only a few studies on the ratios of $^{240}Pu/^{239}Pu$ in environmental samples contaminated by the Chernobyl accident Arnold and Kolb (*15*) reported the isotopic ratio of Pu determined by LX-ray and α-particle measurements in soil samples, collected from different areas including three samples from near Chernobyl They described the activity ratio as $A(^{240}Pu)/A(^{239+240}Pu)$ and its range for the three samples was 0 49–0 57, which corresponds to $^{240}Pu/^{239}Pu$ atom ratios of 0 26–0 36 Erdmann et al (*16*) determined Pu isotopes in some soil samples from the Chernobyl region by resonance ionization mass spectroscopy (RIMS) and found that the abundances of ^{239}Pu and ^{240}Pu were 66 9 (±4)% and 25 3 (±2)%, we calculated the $^{240}Pu/^{239}Pu$ atom ratio as 0 377 ± 0 037 Belyaev et al (*17*) reported the $^{240}Pu/^{239}Pu$ ratios determined by mass spectrometry for three soil samples collected from the 10-km zone (0 350) and 30-km zone (0 303 and 0 300) of Chernobyl Boulyga et al (*18*) analyzed a single hot particle originating from the Chernobyl reactor by means of solid-state nuclear track detectors The isotopic compositions (as %) they reported were 70 ± 3 for ^{239}Pu and 23 ± 3 for ^{240}Pu, corresponding to a $^{240}Pu/^{239}Pu$ atom ratio of 0 33 ± 0 05 The ratios reported in the above-mentioned four publications are all lower than our data We note, however that our results are based on a larger number of samples and they show better uncertainties compared to those given in the literature Additionally, our values for the $^{240}Pu/^{239}Pu$ ratio are in a narrow range, although the samples were collected from three different areas with different Pu levels We have validated our measurements using a Pu isotopic [illegible] sufficiently reliable Alternatively there is a possibility that the variation of the ratios particularly for individual particles would be due to possible inhomogeneity of the isotopes in the reactor Since only a few reports have been made which offer accurate measurements of the $^{240}Pu/^{239}Pu$ isotopic ratios it is necessary to obtain more data on the Pu isotopic composition in areas contaminated by the Chernobyl accident

A closer look at Table 1 reveals that ratios for the underlying Ah and B horizons seem to be somewhat lower than those for surface organic layers Since the concentrations of Chernobyl Pu in deeper layers are low the corresponding ratios might be influenced by preexisting Pu which had its origin in fallout from weapons tests and which has a markedly lower $^{240}Pu/^{239}Pu$ ratio of 0 176 ± 0 014 (Krey) (*9*) Assuming that the ratios measured in the organic layers from K2 are most representative for the Chernobyl fallout, due to the high Pu contamination from Chernobyl fallout we used a mean value of K2 L/Of/Oh 94 and K2 L/Of/Oh 95 (i e 0 410 ± 0 002) as a best estimate for the Chernobyl fallout Using this ratio for the Chernobyl fallout, the ratio for weapons fallout as mentioned above, and the measured ratios for the mineral layers at the three sites from Table 1, we estimated the Pu deposition from weapons fallout in the mineral layers The area related $^{239+240}Pu$ activity due to weapons fallout is estimated as follows

K2 75 ± 61 Bq m^{-2}

D1 50 ± 17 Bq m^{-2} (sum of organic and mineral layers)

D3 29 ± 3 Bq m^{-2}

The mean values for these three places are 51 ± 21 Bq m^{-2} Error ranges are quite large, specifically for sample K2, which has a high level of Chernobyl contamination However it is interesting to compare these results with estimates of global Pu fallout For example, UNSCEAR (*19*) gives an estimate of 58 Bq m^{-2} for the integrated deposition density of $^{239+240}Pu$ for the north temperate zone (40–50°) Bunzl et al (*20*) report for south Bavaria an average $^{239-240}Pu$ deposition of 49 Bq m^{-2} for grassland soil and of 64 Bq m^{-2} for forest soil Though we cannot exclude the possibility that part of the Pu deposited at the investigated sites has already reached deeper layers and though the uncertainties involved with our estimates are considerable, our estimates for the global Pu fallout agree remarkably well with literature values This result indicates that accurate knowledge of $^{240}Pu/^{239}Pu$ ratios can be used in identifying the distribution of the Chernobyl derived Pu in the environment and in distinguishing it from other sources such as Pu from global fallout The $^{240}Pu/^{239}Pu$ ratios may also provide useful information on the dynamics of Pu in forest ecosystems

These results demonstrate that reliable analytical data for the $^{240}Pu/^{239}Pu$ ratio are useful, allowing it to serve as a fingerprint to identify the origin of Pu from different sources The uncertainties of the above-mentioned cases are rather large due to the very high contamination from the Chernobyl fallout More reliable results can be obtained if contamination levels from two different sources are similar

Comparison to Other Nuclides Concentrations of radionuclides (i e ^{137}Cs ^{125}Sb and ^{60}Co) determined by γ spectrometry and their calculated area related activities (i e inventories in the layers) are shown in Table 3 The concentrations of ^{137}Cs in the organic layers (L/Of/Oh) were more than a magnitude higher than those in the corresponding inorganic layers (Ah/B layers) However data of the area related activities showed that the inventories of the

TABLE 3 Levels of ^{137}Cs ^{60}Co and ^{125}Sb in Soils from the Chernobyl Area and Their Activity Ratio to $^{239+240}Pu$[a]

sampling point	soil layer	^{137}Cs Bq kg^{-1}	^{137}Cs Bq m^{-2}	^{125}Sb Bq kg^{-1}	^{125}Sb Bq m^{-2}	^{60}Co Bq kg^{-1}	^{60}Co Bq m^{-2}	$^{137}Cs/Pu$ ratio	$^{125}Sb/Pu$ ratio	$^{60}Co/Pu$ ratio
				10 km Zone (Kopacky)						
K2	L/Of/Oh 94	177000	920000	1120	5820	129	668	124	0 78	0 09
K2	L/Of/Oh 95	138000	718000	853	4440	132	684	122	0 75	0 12
K2	Ah/B 95	8770	759000	48	4150	3 9	339	114	0 63	0 05
				30-km Zone (Dityatki)						
D1	L/Of/Oh 94/95	14700	64700	82	361	7 7	34	107	0 60	0 06
D1	Ah/B 94/95	645	56100	nd		nd		103		
D3	L/Of/Oh 94	15100	64400	112	493	7 8	34	86	0 64	0 04
D3	L/Of/Oh 95	15400	67800	155	682	28	124	90	0 90	0 16
D3	B 94/95	1420	52700	nd		nd		106		
mean (ratio)								106	0 72	0 09
SD								13%	16%	53%

[a] Decay correction 1 June 1997 nd not detected

nuclide in organic layers in the three locations were comparable to those in the underlying inorganic layers This indicates that about half of the ^{137}Cs migrated from the organic layers to the mineral layers This is similar to the above-mentioned observation on the inventory of Pu, i e , almost the same amount of Pu was distributed between the organic and mineral layers The radionuclide inventory data provide a basis for discussion of radionuclide redistribution in forests

The ratios of the nuclides to $^{239+240}Pu$ are also shown in Table 3 to allow examination of internuclide relationships to Pu No marked difference was found between the ratio of $^{137}Cs/^{239+240}Pu$ in organic and mineral layers This may also indicate that Pu was redistributed similarly to ^{137}Cs in the forest soil, though the chemical properties of these nuclides are very different Linkov et al (*21*) studied the migration of radionuclides in the forest ecosystem and mentioned that the transport mechanism of Pu and ^{137}Cs in the forest would be related to organo-metal complexes which redistribute similarly

As shown in Table 3, the ratio $^{137}Cs/^{239+240}Pu$ for eight samples is almost constant, i e 106 ± 14 A relatively constant value is also found for the $^{125}Sb/^{239+240}Pu$ ratio (0.72 ± 0.11) The ratio of $^{60}Co/^{239+240}Pu$ varies very widely, i e 0 04–0 16 (0.009 ± 0.005) Cobalt-60 is produced from the reactor material (e g steel) as an activation product, and therefore its distribution in the fuel (or reactor) and its behavior in the event of a release might differ in comparison to Pu On the contrary, ^{125}Sb and ^{137}Cs were produced as fission products in fuel, and therefore, their distribution in fuel material should be similar to that of Pu However, due to differences in the chemical characteristics of the nuclides involved, their initial composition in fuel material relative to Pu need not necessarily be found in soil

For example, about 33% of the initial core inventory of ^{137}Cs is estimated to be released during the accident, i e about 8.6×10^{16} Bq (*1*) This compares to an estimated 3 5% of fuel material released, corresponding to the release of about 8.7×10^{13} Bq $^{239+240}Pu$ (*1*) The resulting $^{137}Cs/^{239+240}Pu$ ratio of 770, corrected for radioactive decay to June 1997, is markedly higher than the ratio of 106 measured in the investigated soil samples The behavior of Pu at its release from the reactor is expected to be very different from that of more volatile radiocesium It is known that the fuel particles tended to deposit closer to the reactor due to their larger particle size, whereas the proportion of the condensed particles, which are normally finer than the fuel particles, was higher in remote areas than that of the fuel particles (*22 23*) Since the $^{137}Cs/^{239+240}Pu$ ratios in the condensed particles are expected to be higher than those in fuel particles due to the volatility of Cs the ratio in the fallout should be higher for areas far from Chernobyl An alternative explanation that Cs had migrated more readily to deeper layers compared to Pu, seems to be unlikely, since it is known that organic horizons of forest soil also retain radiocesium very effectively (*24*) It is interesting to note that very high ratios of $^{137}Cs/^{239+240}Pu$ were found in far-area from Chernobyl For example, Hotzl et al (*25*) reported that close to Munich, Germany, the ^{137}Cs total deposition relative to $^{239+240}Pu$ was higher by a factor of about 700 compared to the release ratio given above This also indicates that Cs was more readily transported over large distances compared to Pu, whereas Pu was more readily deposited near-field (see above) The $^{137}Cs/^{239+240}Pu$ ratio observed in the investigated soil samples is much higher than the ratio for global weapons fallout of 38 ± 4 (as of June 1994) (*26*) In the case of ^{125}Sb, similar results are observed, i e a $^{125}Sb/^{239+240}Pu$ ratio (again correcting for radioactive decay to June 1997) of 2 1 is estimated from the releases at the accident, but a significantly low ratio of 0 7 is measured in the investigated field soil samples (Table 3)

In this study we have obtained new information on the composition of the most abundant Pu isotopes (^{239}Pu and ^{240}Pu) in soil samples (organic and mineral layers) collected near the Chernobyl reactor We demonstrated that the $^{240}Pu/^{239}Pu$ ratio was useful as a fingerprint to identify the source of the Pu contamination Data obtained for the Pu isotopic composition together with the concentrations of other nuclides could also be used in model assessments such as proposed by Linkov et al (*21*) for the transfer of radionuclides in forest ecosystems Comparison of Pu to other nuclides would also be important in understanding the fate of the nuclides in the environment

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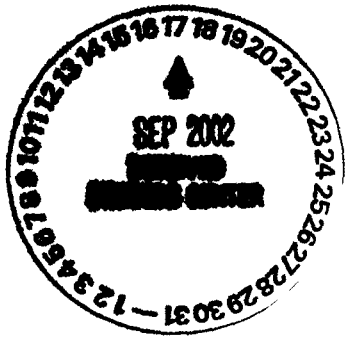

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