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^{241}Pu and ^{241}Pu/$^{239+240}$Pu activity ratio in environmental samples from Finland as evaluated by the ingrowth of ^{241}Am

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The activity concentrations of ^{241}Pu and the ^{241}Pu/$^{239+240}$Pu activity ratios were determined from environmental samples to complete the study of transuranium nuclide distribution in environment in Finland. The activity of ^{241}Pu was determined by measuring the activity of its decay product, ^{241}Am, using α-spectrometry. The activity concentrations of ^{241}Pu in lichens from 1967–1976 were from 2.3 to 93 mBq g^{-1} (in the sampling year) and from < 0.7 to 686 mBq g^{-1} in lichen, peat, and grass samples from 1986 (on 1 May 1986). The ^{241}Pu/$^{239+240}$Pu activity ratios for the corresponding samples were from 4.1 to 167. The comparison of the ^{241}Pu results obtained in the present study and those from earlier analyzes (liquid scintillation counting) indicated a fairly good agreement. Regression analysis indicated that the activity concentrations of mainly Chernobyl-derived nuclides ^{238}Pu, ^{241}Pu, and ^{242}Cm correlated significantly with each other in the samples of 1986, the r^2 values being 0.89 and 0.67 for ^{241}Pu/$^{239+240}$Pu *vs.* ^{238}Pu/$^{239+240}$Pu and ^{242}Cm/$^{239+240}$Pu *vs.* ^{241}Pu/$^{239+240}$Pu, respectively.

Introduction

The ^{241}Pu isotope was introduced into the environment from explosions of nuclear weapons, controlled emissions from nuclear fuel reprocessing facilities, and from accidents that released nuclear reactor fuel, such as the 1986 Chernobyl nuclear power plant disaster and, most recently, the 2011 Fukushima catastrophe (Zheng *et al.* 2012). As compared with other Pu isotopes in the environment that are α-emitting and long-lived, ^{241}Pu is a short-lived isotope with a half-life of 14.35 years and it decays by a low-energy (20.8 keV) β emission. However, ^{241}Pu decays to the α emitter ^{241}Am, that has a much longer half-life (432.2 years) and is highly radiotoxic when it is incorporated into either the human or animal body. The ^{241}Pu concentration and activity in a sample can be determined either directly by using β emission or mass spectrometry, or indirectly by determining the ingrowth of ^{241}Am.

The ^{241}Pu isotope has been studied less extensively than the α-emitting Pu isotopes for several reasons. Activity concentration of ^{241}Pu cannot be determined from the same α spectrum as the Pu isotopes 238, 239, and 240, and extra effort is needed in order to analyze ^{241}Pu concentration of a sample. Actually, ^{241}Pu emits α particles, but they have so low probability (0.002%) that ^{241}Pu cannot be measured directly by α-spectrometry

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from samples with environmental activity levels. Concentrations of ^{241}Pu are generally very low in environmental samples. However, relatively high amounts of ^{241}Pu were released into the environment from the Chernobyl accident in 1986. Since then, ^{241}Pu has decayed into ^{241}Am over a period equivalent to nearly two half-lives, thus approximately 30% of Chernobyl-derived ^{241}Pu remained in the environment in 2011.

Liquid scintillation counting (LSC) with (ultra) low background is often used for determination of ^{241}Pu (Paatero *et al*. 1994a, Mietelski *et al*. 1999, Payne *et al*. 2008, Varga and Tarján 2008, Corcho Alvarado *et al*. 2011). However, the background count rate is often high as compared with the actual count rates of ^{241}Pu in low-activity environmental samples, because of the instrumentation or the composition of liquid scintillation cocktail. The counting efficiency of LSC for ^{241}Pu is typically ~35% and varies from 30% to 43%, depending on the quenching (Paatero *et al*. 1994a, Varga and Tarján 2008, Corcho Alvarado *et al*. 2011). The detection limit of LSC for ^{241}Pu is 7 mBq per sample (50 g of peat, dry weight) (Paatero *et al*. 1994a) or 0.1–1.1 mBq g^{-1} (dried soil) (Varga and Tarján 2008, Corcho Alvarado *et al*. 2011) with ultra-low background counters. With conventional LSC, the detection limit is between 73 and 90 mBq per 0.5 or 40 g sample of soil (dry weight), respectively (Mietelski *et al*. 1999, Payne *et al*. 2008), or 2.25 mBq g^{-1} (40 g of soil, dry weight) (Mietelski *et al*. 1999).

It is also possible to use a gas-flow proportional counter for the determination of ^{241}Pu, and the background count rate is often even lower with gas-flow proportional counting than with the LSC method (Rosner *et al*. 1992, Hou and Roos 2008). On the other hand, the counting efficiency of a gas-flow proportional counter for determining ^{241}Pu is poor, due to the absorption of low-energy β particles emitted by ^{241}Pu into the air or into the counter window. The detection limit for ^{241}Pu was 10 mBq per sample using a gas-flow proportional counter (Rosner *et al*. 1992). Therefore, the method is comparable to LSC.

In recent years, Inductively Coupled Plasma-Mass Spectrometry (ICP-MS) has also been used for determination of ^{241}Pu (Ketterer *et al*. 2004, Donard *et al*. 2007, Varga *et al*. 2007). However, this method sets high demands for the radiochemical separation procedure and for the instrument, due to short half-life of ^{241}Pu and any possible isobaric interference especially from ^{241}Am. With sector focusing ICP-MS, the detection-limit range for ^{241}Pu is from 2 to 54 mBq g^{-1} (Kenna 2002, Donard *et al*. 2007, Varga *et al*. 2007, Röllin *et al*. 2009).

The measurement of the ingrown ^{241}Am by α-spectrometry is a convenient method for determination of ^{241}Pu, provided that the ingrowth time is sufficiently long and the original activity concentration of ^{241}Pu in the sample is high enough. The detection limit for ^{241}Pu by α-spectrometry of ^{241}Am from an archived Pu α sample is rather high as compared with other methods discussed above, as the detection limit depends greatly on background counts (Currie 1968). The presence of the ^{238}Pu peak ($E\alpha$ = 5.499 MeV) in the α spectrum in the same energy region as the ^{241}Am peak ($E\alpha$ = 5.486 MeV) causes extra background counts increasing the detection limit of ^{241}Am. The detection limit using this indirect method for determining ^{241}Pu is about 200 mBq per sample (Rosner *et al*. 1992), or ranges between 10 and 90 mBq g^{-1} (Mietelski *et al*. 1999).

The ^{241}Pu/$^{239+240}$Pu activity ratio can be used as an indicator of Pu contamination source in environment as other ratios of Pu isotopes and especially in the measurement of the input from the Chernobyl accident. The ^{241}Pu/$^{239+240}$Pu activity ratios originating from different sources are given in Table 1. The Soviet authorities reported the ^{241}Pu/$^{239+240}$Pu activity ratio to be 67 in the cumulative discharge from the damaged Chernobyl reactor on 26 April, and 82 on 6 May 1986. Nowadays, the ^{241}Pu/$^{239+240}$Pu activity ratio is < 2 in global fallout from atmospheric nuclear weapons testing, and ~28 from Chernobyl-derived fallout, calculated from the value of 94.8 determined in Finland (Paatero *et al*. 1994a). Another ^{241}Pu isotope ratio, namely ^{241}Pu/^{239}Pu mass ratio, is also a sensitive tool for determining Chernobyl-derived Pu contamination (Ketterer *et al*. 2004).

Lichens have been used globally as bioindicators for different atmospheric pollutants due to their ability to adsorb substances from air and precipitation (Tuominen and Jaakkola 1973).

Lichens and also peats have been investigated extensively in Finland for four decades in order to estimate deposition of transuranium nuclides that originated from nuclear weapons testing and the Chernobyl accident (Tulikoura *et al*. 1974, Jaakkola *et al*. 1978, 1981, Hakanen *et al*. 1984, Reponen *et al*. 1993, Paatero *et al*. 1994a, 1998).

The aim of this study was to determine transuranium nuclides in previously investigated environmental samples (Jaakkola *et al*. 1978 and 1986, Reponen *et al*. 1993, Paatero *et al*. 1994a and 1998, Salminen *et al*. 2005, Salminen-Paatero *et al*. 2012) by measuring and analyzing the activity concentrations of ^{241}Pu and the ^{241}Pu/$^{239+240}$Pu activity ratios in the samples. In addition, the present (year 2011) activity concentrations of ingrown ^{241}Am, in the samples of two groups with different age and contamination source, was sought. Archived Pu α-counting samples were re-measured by α-spectrometry to determine the ingrowth of ^{241}Am from ^{241}Pu during the 34-year period since the operation of separated Pu fractions for α-counting. Originally the samples were lichens collected during the inclusive period of 1967–1976. In addition, other old Pu α-counting samples were redissolved and ^{241}Am was separated from these samples by extraction chromatography. The separated fractions of ^{241}Am co-precipitated with neodymium(III)fluoride were measured by α-spectrometry in order to determine the ingrown ^{241}Am content. These samples (peat, air filter, grass, lichens, and hot particles) were collected immediately after the Chernobyl accident in 1986. The comparison between α-spectrometry (indirect method for determining ^{241}Pu) and LSC (direct method) was also carried out for the peat samples.

Experimental methods

Samples

Lichen samples from 1967–1976

Twelve lichen (*Cladonia* sp.) samples were collected from southern Finland and Lapland during 1967–1976. The samples were analyzed in 1977 (Jaakkola *et al*. 1978). Plutonium isotopes were separated from the lichen samples by anion exchange and electrodeposited on the platinum discs. The activity concentrations of ^{238}Pu and $^{239+240}$Pu in lichen were determined by α-spectrometry. These electrodeposited samples were re-measured directly using α-spectrometry in 2011, in order to determine the activity concentrations of $^{239+240}$Pu and ^{238}Pu+^{241}Am in the samples. Subsequently, the activity concentration of ^{241}Pu in the lichen samples in the sampling year was calculated from the ingrowth of ^{241}Am during the 34-year-long storage of samples separated for α-counting.

Table 1. ^{241}Pu/$^{239+240}$Pu activity ratio in plutonium from different sources.

^{241}Pu/$^{239+240}$Pu activity ratio	Date	Pu source (sample)	Reference
15	1963–1972	Atmospheric nuclear weapons testing (fresh fallout)	Perkins and Thomas (1980)
0.75–7.5	1945–1974	Weapons-grade Pu	Irlweck and Hrnecek (1999)
85	1 May 1986	The Chernobyl fallout (air filters, soil)	Holm *et al.* (1992), Mietelski *et al.* (1999)
74.6 ± 5.7	1986	The Chernobyl fallout (air filters)	Irlweck and Wicke (1998)
94.8	1 May 1986	The Chernobyl fallout (lichen, peat)	Paatero *et al.* (1994a)
67	26 April 1986	The Chernobyl fallout	USSR State Committee on the Utilization of Atomic Energy (1986)
82	6 May 1986	The Chernobyl fallout	USSR State Committee on the Utilization of Atomic Energy (1986)

Environmental samples from 1986

Six peat samples were collected from peat-producing peatland sites between 12 and 14 May 1986 (Jantunen *et al*. 1987). Five lichen samples (*Cladonia alpestris*) and a meadow fescue grass sample (*Schedonorus pratensis*) were gathered in southern and central Finland during the summer and autumn of 1986 (Paatero *et al*. 1998). An air filter sample was collected in Nurmijärvi, southern Finland, on 28 April 1986 (Jaakkola *et al*. 1986). Two hot particles were found on Scots pine (*Pinus sylvestris*) twigs in Uusikaupunki, on the southeastern coast of Finland, on 29 April 1986 (Raunemaa *et al*. 1987, Saari *et al*. 1989). Pu was separated from these samples by anion exchange and by co-precipitations, and then it was finally co-precipitated with NdF_3 onto membrane filters. The activities of ^{238}Pu and $^{239+240}$Pu were determined by α-spectrometry, and for peat samples the activity of ^{241}Pu was also determined by liquid scintillation counting from a separate sample fraction.

The membrane filters onto which the Pu isotopes were co-precipitated for α-counting could have been measured directly to determine ^{241}Pu in the samples via the ingrowth of ^{241}Am in the α-counting samples. However, in this study we also determined the ^{240}Pu/^{239}Pu mass ratio in the same samples, *see* Salminen-Paatero *et al*. (2012). Therefore, the dissolution of the α-counting samples and further radiochemical purification was necessary for that purpose.

Radiochemical separation of ^{241}Am from the samples of 1986

The α-counting samples of Pu in the NdF_3-precipitates were re-dissolved in hot concentrated nitric acid. ^{243}Am tracer was added to the samples at the beginning of the dissolution for determination of the ^{241}Am yield. Then a small amount of boric acid was added to the solutions during the dissolution procedure to decompose the complexes of plutonium trifluoride (PuF_3) (Moreno *et al*. 1998, Mietelski and Gaca 2002). The solutions were filtered and evaporated. The dry residues were dissolved in 1 M $Al(NO_3)_3$ + 3 M HNO_3 for separation procedure with UTEVA® and TRU® resins (Eichrom Technologies) (Salminen-Paatero *et al*. 2012). The americium and plutonium fractions were collected separately. Americium was co-precipitated with NdF_3 and filtered onto membrane filters for α-counting. Plutonium fractions were measured by ICP-MS to determine the ^{240}Pu/^{239}Pu mass ratio in the samples (Salminen-Paatero *et al*. 2012).

Determination of ^{241}Am, $^{239+240}$Pu, and ^{238}Pu with α-spectrometry

The activities of the isotopes ^{241}Am, $^{239+240}$Pu, and ^{238}Pu + ^{241}Am were measured for five to seven days using an Alpha Analyst (Canberra) spectrometer fitted with PIPS (planar implanted passivated silicon) detectors whose area was 450 mm^2 (Fig. 1). The nominal resolution for the 5.486 MeV peak of ^{241}Am was 20 keV, the actual resolution for the same peak being 19–75 keV for α-counting samples co-precipitated with NdF_3. The mean detection limit (DL) for ^{241}Am was 0.14 mBq per sample, calculated with the method of Currie (1968). ^{243}Am tracers often contain some ^{241}Am as an impurity. The radioactivity of ^{241}Am in the ^{243}Am tracer used in the present study was 2.5% of the total activity in the tracer, and thus this additional activity of ^{241}Am in the samples within the tracer was subtracted from the yield- and efficiency-corrected activity of ^{241}Am in the samples.

Results and discussion

^{241}Pu content of the environmental samples

Lichens of 1967–1976

The activity concentrations of ^{241}Pu in the lichen samples collected during 1967–1976 varied between 2.3 ± 0.2 and 93 ± 2 mBq g^{-1} (Table 2). The reference date for the activity was 1 July in the sampling year. There was almost a 100-fold variation in the activity concentration of ^{241}Pu among the samples, depending on the sampling site and whether the samples comprised the top or the bottom part of the lichen. As compared

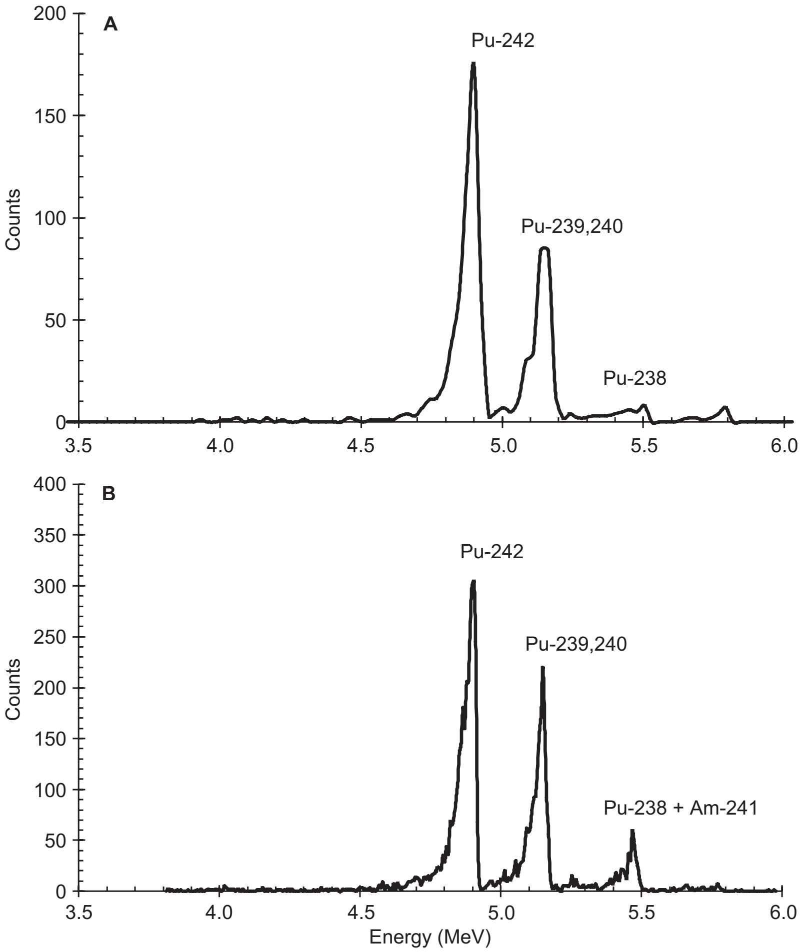

Fig. 1. Typical α spectrum of Pu isotopes in a peat sample. The α-counting sample was prepared by co-precipitating Pu with NdF_3 onto a membrane filter. (**A**) The sample spectrum in 1990 (counting time 2150 min), and (**B**) the corresponding spectrum measured 21 years later (counting time 10020 min), showing the ingrowth of ^{241}Am in the peak region of ^{238}Pu.

with the α-emitting Pu isotopes, the activity concentration of ^{241}Pu was about 100-fold that of ^{238}Pu, and 10-fold that of $^{239+240}$Pu in these samples. The activity concentration of ^{241}Pu was higher in the lichen samples taken from southern Finland than in those that originated from Lapland. The concentration of transuranium nuclides from nuclear weapons testing has decreased constantly in the surface air of the northern hemisphere since 1962, and due to the downward migration of old nuclear fallout from lichens to soil, activity concentrations of ^{241}Pu in lichens from Lapland sampled in 1976 were lower as compared with those in lichens from southern Finland sampled in 1967–1968. In those lichens taken during the 1960s–1970s, the activity concentration of ^{241}Pu was higher in the bottom part of the lichen than in the top part. A similar distribution was found for ^{238}Pu and $^{239+240}$Pu in the same samples (Table 2), and all Pu activity concentrations show migration of old nuclear-test fallout downwards the lichen. In the present study, ^{241}Pu was found in lichen taken from Inari, Lapland. However in Paatero *et al.* (1994a), the activity concentration of ^{241}Pu was below the detection limit in lichen collected from Inari in 1987. The deposition level in Lapland due to the Chernobyl accident was relatively low, and nowadays the activity level of ^{241}Pu, and also other anthropogenic radionuclides, is extremely low in that area. Previously, activity concentrations of ^{241}Pu in Finnish lichens during the period 1960–1977 were determined to be from 22 ± 1 to 123 ± 6 mBq g^{-1} (Hakanen *et al.* 1984). Holm and Persson (1978), found similar activity concentrations of ^{241}Pu in Swedish lichens (surface layer of 0–3 cm): from 22 ± 4 to 118 ± 11 mBq g^{-1} for the 1966–1975 period.

Table 2. The activity concentrations of ^{241}Pu, $^{239+240}$Pu, and ^{238}Pu in lichens, peat, and other environmental samples. The reference date for the lichen samples from 1967–1976 is 1 July in the sampling year, and for samples collected in 1986 the reference date is 1 May 1986. The error is 1-σ propagated counting error. The activity concentrations of $^{239+240}$Pu and ^{238}Pu have been previously published by Jaakkola *et al.* (1978) and (1986), Reponen *et al.* (1993) and Paatero *et al.* (1998).

Sample type and identity	Sampling location	Sampling time	A ^{241}Pu (mBq g^{-1})	A $^{239+240}$Pu (mBq g^{-1})	A ^{238}Pu (mBq g^{-1})
Top lichen 26/67	Helsinki (60.2°N, 24.9°E)	1967	30 ± 1	2.7 ± 0.1	0.11 ± 0.02
Bottom lichen 27/67	Helsinki (60.2°N, 24.9°E)	1967	93 ± 2	8.0 ± 0.4	0.33 ± 0.04
Top lichen 44/67	Loppi (60.7°N, 24.4°E)	1967	16.3 ± 0.9	1.28 ± 0.06	0.11 ± 0.02
Bottom lichen 45/67	Loppi (60.7°N, 24.4°E)	1967	57 ± 2	4.5 ± 0.3	0.15 ± 0.03
Top lichen 87/68	Loppi (60.7°N, 24.4°E)	1968	11.4 ± 0.7	1.1 ± 0.08	0.13 ± 0.03
Bottom lichen 88/68	Loppi (60.7°N, 24.4°E)	1968	40 ± 1	4.0 ± 0.2	0.21 ± 0.03
Top lichen 99/68	Virolahti (60.6°N, 27.7°E)	1968	7.9 ± 0.4	0.77 ± 0.04	0.09 ± 0.03
Bottom lichen 100/68	Virolahti (60.6°N, 27.7°E)	1968	25.9 ± 0.9	2.5 ± 0.1	0.17 ± 0.03
Top lichen 4/76Y	Inari (68.9°N, 27.0°E)	1976	2.3 ± 0.2	0.31 ± 0.02	0.015 ± 0.004
Bottom lichen 4/76A	Inari (68.9°N, 27.0°E)	1976	4.1 ± 0.2	0.48 ± 0.03	0.044 ± 0.007
Top lichen 12/76Y	Inari (68.9°N, 27.0°E)	1976	2.6 ± 0.2	0.35 ± 0.03	0.019 ± 0.007
Bottom lichen 12/76A	Inari (68.9°N, 27.0°E)	1976	8.0 ± 0.3	1.01 ± 0.06	0.09 ± 0.01
Top lichen 303/86	Kalanti (60.8°N, 21.5°E)	17 July 1986	686 ± 10	5.7 ± 0.3	3.0 ± 0.2
Bottom lichen 303/86	Kalanti (60.8°N, 21.5°E)	17 July 1986	184 ± 9	1.1 ± 0.2	0.8 ± 0.2
Top lichen L328	Huittinen (61.2°N, 22.8°E)	13 Sep. 1986	71 ± 4	0.87 ± 0.04	0.36 ± 0.02
Bottom lichen T328	Huittinen (61.2°N, 22.8°E)	13 Sep. 1986	30 ± 1	0.80 ± 0.05	0.13 ± 0.02
Top lichen 323/06	Pärnäsaari, Päijänne (61.2°N, 25.6°E)	21 Aug. 1986	< 4	0.18 ± 0.01	0.048 ± 0.007
Peat 22	Leivonmäki (61.7°N, 25.9°E)	12–14 May 1986	< 0.7	0.27 ± 0.01	0.007 ± 0.002
Peat 37	Juva (61.8°N, 27.2°E)	12–14 May 1986	4.6 ± 0.3	1.13 ± 0.03	0.033 ± 0.003
Peat 42	Punkaharju (61.5°N, 29.0°E)	12–14 May 1986	7.9 ± 0.5	1.53 ± 0.05	0.040 ± 0.004
Peat 60	Kiuruvesi (63.3°N, 26.3°E)	12–14 May 1986	64 ± 3	1.08 ± 0.07	0.43 ± 0.03
Peat 64	Sonkajärvi (63.8°N, 27.2°E)	12–14 May 1986	35 ± 1	0.80 ± 0.04	0.18 ± 0.01
Peat 143	Valtimo (63.7°N, 28.4°E)	12–14 May 1986	77 ± 4	1.35 ± 0.09	0.29 ± 0.03
Grass N307	Kalanti (60.8°N, 21.5°E)	17 July 1986	33 ± 1	0.35 ± 0.03	0.14 ± 0.02
Air filter *	Nurmijärvi (60.5°N, 24.8°E)	28 Apr. 1986	3.1 ± 0.2	0.033 ± 0.004	0.016 ± 0.004
Hot particle U37(1) **	Uusikaupunki (60.8°N, 21.4°E)	29 Apr. 1986	1005 ± 24	9.4 ± 0.4	6.5 ± 0.3
Hot particle U37/2(1) **	Uusikaupunki (60.8°N, 21.4°E)	29 Apr. 1986	62 ± 6	0.64 ± 0.08	0.27 ± 0.06

* The activity concentration in air (mBq m^{-3}). ** The activity of the whole sample (mBq).

Lichens, peats, grass, air filter, and hot particle samples of 1986

In the miscellaneous environmental samples from 1986, the activity concentrations of ^{241}Pu were from < 0.7 to 686 ± 10 mBq g^{-1} (reference date 1 May 1986) (Table 2), and showed a similar variation to that found in lichens collected between 1967 and 1976. In this study, the activity concentration of ^{241}Pu was orders of magnitude higher than that of ^{238}Pu or $^{239+240}$Pu in the Chernobyl-contaminated samples taken in 1986, as was previously found by Holm *et al*. (1992), Buzinny *et al*. (1996) and Mietelski *et al*. (1999). Nevertheless, there was no systematic magnitude difference between concentrations of Pu isotopes in our Chernobyl-contaminated samples as compared with our older lichen samples contaminated only by global fallout. The activities of ^{241}Pu were 62 ± 6 mBq and 1005 ± 24 mBq in the two hot-particle samples obtained from the twigs of Scots pine.

The activity concentration of ^{241}Pu was higher in the top parts of the lichens than in their bottom parts for lichen samples 303/86 (Kalanti) and 328 (Huittinen) (Table 2). This difference was due to the accumulation of fresh Chernobyl-deposition onto the lichen top before sampling a few months after the accident in 1986. In lichen sample 323/06 (Pärnäsaari), the activity concentration of ^{241}Pu was below the detection limit. Lichen samples from the same sites (Kalanti and Huittinen) were also taken in 1987–1988, and in these samples ^{241}Pu was unsystematically distributed between the top and bottom parts of the lichens (Paatero *et al*. 1994a), the activity concentrations of ^{241}Pu being between 10.6 ± 0.9 and 204 ± 11 mBq g^{-1} (reference date 1 May 1986) (Paatero *et al*. 1994a). These values were similar to those found in the lichen samples taken in 1986. Elevated activity concentrations of ^{241}Pu were found in other studies performed in Chernobyl-contaminated areas: the activity concentrations of ^{241}Pu were from < 90 to 254 ± 43 mBq g^{-1} in surface soil samples in northeastern Poland (Mietelski *et al*. 1999), 234 ± 18 mBq g^{-1} in ashed fodder from Pest, Hungary (decay-corrected value of Varga and Tarján 2008), and 15.6–5300 mBq g^{-1} in soil and plant samples from Ukraine (Buzinny *et al*. 1996), the reference date for all these values was April–May 1986. Excluding the highest activity concentrations of ^{241}Pu in some Ukrainian plant samples, the activity concentrations of ^{241}Pu were quite similar in surface soils, plants, lichens, and peats in areas of the heaviest Chernobyl deposition. Today, due to radioactive decay the activity of Chernobyl-derived ^{241}Pu is approximately 1/3 of the original activity in 1986.

Comparison of the LSC and the method based on the ingrowth of ^{241}Am

^{241}Pu activity concentrations in six peat samples measured in this study by the ingrowth of ^{241}Am and α-spectrometry were earlier determined by LSC (Paatero *et al*. 1994a) (Table 3). The activity concentration values obtained with these two methods are in fairly good agreement, with the exception of sample 143, which had the highest ^{241}Pu activity concentration. Mietelski *et al*. (1999) found a slight difference between the activity concentrations of ^{241}Pu in 16 parallel samples as determined by these two methods. The LSC method possibly gave higher ^{241}Pu activity concentration values than the α-spectrometry method, but similar was not found in comparable results of our six peat samples. More parallel samples would be needed to find possible systematic differences between activity concentrations of ^{241}Pu as obtained by these two methods. Despite both methods giving

Table 3. Comparison of activity concentrations of ^{241}Pu in six peat samples determined directly by liquid scintillation counting (LSC) of ^{241}Pu (Paatero *et al.* 1994a) and by the ingrowth of ^{241}Am and α-spectrometry (this work). The reference date is 1 May 1986.

Peat sample	A ^{241}Pu (mBq g^{-1})*	A ^{241}Pu (mBq g^{-1})**
22	1.03 ± 0.09	< 0.7
37	5.0 ± 0.3	4.6 ± 0.3
42	5.9 ± 0.3	7.9 ± 0.5
60	57 ± 5	64 ± 3
64	27 ± 2	35 ± 1
143	45 ± 3	77 ± 4

* LSC (Paatero *et al.* 1994a), ** α-spectrometry via ^{241}Am.

comparable and reliable results, the lower detection limits and reduced uncertainty of radioactivity concentration are the benefits given by LSC over that of the indirect method based on the ingrowth of ^{241}Am (Mietelski *et al*. 1999).

^{241}Pu/$^{239+240}$Pu activity ratio in studied samples

The lichen samples of 1967–1976

In the lichen samples taken during 1967–1976, the ^{241}Pu/$^{239+240}$Pu activity ratios were from 7.3 ± 0.7 to 12.8 ± 0.9, i.e. typical activity ratios in global fallout during the mid-1960s, a few years after the most intensive period of atmospheric testing of nuclear weapons (Table 4). Similar values (from 7.2 ± 0.9 to 9.1 ± 1.1) for the period of 1966–1971 were measured from the Finnish lichens by Hakanen *et al*. (1984). Holm and Persson (1978) obtained the values that were almost identical to ours: in surface lichens in the period of 1966 to 1975, the ^{241}Pu/$^{239+240}$Pu activity ratios ranged from 7 ± 1 to 14 ± 2.

There was no evident difference in the ^{241}Pu/$^{239+240}$Pu activity ratios between top and bottom parts of the same lichens for the 1967–1976 period, but the ^{238}Pu/$^{239+240}$Pu activity ratios differed between the lichen top and bottom in every pair comparison (except for the pair top lichen 26/67 and bottom lichen 27/67). Global fallout was the only source of ^{241}Pu and $^{239+240}$Pu in Finland in the 1960s–1970s, whereas introduction of “extra ^{238}Pu activity” released into the atmosphere by the SNAP-9A satellite accident in 1964 was seen as a change in the ^{238}Pu/$^{239+240}$Pu activity ratio in lichen samples. ^{238}Pu first accumulated in the top parts in the samples of 1967–1968, and then migrated downwards to the bottom parts in the 1976 samples (Table 4). The same change in the ^{238}Pu/$^{239+240}$Pu activity

Table 4. Activity ratios of ^{238}Pu/$^{239+240}$Pu and ^{241}Pu/$^{239+240}$Pu in selected environmental samples. The activity ratios of ^{238}Pu/$^{239+240}$Pu in the samples have been published earlier by Jaakkola *et al.* (1978), Jaakkola *et al.* (1986), Reponen *et al.* (1993) and Paatero *et al.* (1998).The reference dates for lichen samples taken in 1967–1976 inclusive is 1 July in each sampling year, and for the samples collected in 1986, 1 May.

Sample type and identity	Sampling year	A ^{238}Pu/A $^{239+240}$Pu	A ^{241}Pu/A $^{239+240}$Pu
Top lichen 26/67	1967	0.043 ± 0.008	11.3 ± 0.7
Bottom lichen 27/67	1967	0.044 ± 0.006	11.5 ± 0.7
Top lichen 44/67	1967	0.09 ± 0.01	12.8 ± 0.9
Bottom lichen 45/67	1967	0.033 ± 0.008	12.7 ± 0.9
Top lichen 87/68	1968	0.12 ± 0.02	10 ± 1
Bottom lichen 88/68	1968	0.052 ± 0.008	9.9 ± 0.5
Top lichen 99/68	1968	0.12 ± 0.04	10.3 ± 0.8
Bottom lichen 100/68	1968	0.07 ± 0.01	10.2 ± 0.7
Top lichen 4/76Y	1976	0.05 ± 0.02	7.3 ± 0.7
Bottom lichen 4/76A	1976	0.09 ± 0.02	8.5 ± 0.7
Top lichen 12/76Y	1976	0.05 ± 0.02	7.5 ± 0.7
Bottom lichen 12/76A	1976	0.09 ± 0.01	7.9 ± 0.5
Top lichen Pu 303/86	1986	0.52 ± 0.05	120 ± 7
Bottom lichen 303/86	1986	0.7 ± 0.2	167 ± 31
Top lichen L328	1986	0.42 ± 0.03	82 ± 6
Bottom lichen T328	1986	0.16 ± 0.02	37 ± 3
Top lichen 323/06	1986	0.27 ± 0.05	–
Peat 22	1986	0.027 ± 0.006	–
Peat 37	1986	0.029 ± 0.003	4.1 ± 0.3
Peat 42	1986	0.026 ± 0.003	5.2 ± 0.4
Peat 60	1986	0.40 ± 0.04	60 ± 5
Peat 64	1986	0.22 ± 0.02	44 ± 3
Peat 143	1986	0.22 ± 0.02	57 ± 5
Grass N307	1986	0.40 ± 0.07	95 ± 9
Air filter	1986	0.50 ± 0.13	98 ± 14
Hot particle U37(1)	1986	0.69 ± 0.04	107 ± 5
Hot particle U37/2(1)	1986	0.43 ± 0.10	96 ± 15

ratio in lichens after 1966 caused by the SNAP-9A accident was previously found by Holm and Persson (1975) and Jaakkola *et al.* (1981). The mean residence time of Pu in lichens is about six years (Holm and Persson 1975), and two to four years for the biological half-life (Holm and Persson 1975, Jaakkola *et al.* 1981), which covers the movement of SNAP-9A-derived ^{238}Pu from top to bottom parts of lichen during 1967–1976.

Samples after the Chernobyl accident, 1986

In the environmental samples of 1986, the ^{241}Pu/$^{239+240}$Pu activity ratio was 4.1 ± 0.3–167 ± 31 (1 May 1986) (Table 4). Large variations in the activity ratios indicate contamination of the samples from both global fallout and nuclear fuel with high burnup from the Chernobyl accident. In 1986 (i.e. before the Chernobyl accident), the ^{241}Pu/$^{239+240}$Pu activity ratio for global nuclear fallout in the northern hemisphere was about 3.9, when calculated using the corresponding value of 12.3 for the early 1960s (UNSCEAR 1982). Four lichen samples of this sutdy had high ^{241}Pu/$^{239+240}$Pu activity ratios, from 37 ± 3 to 167 ± 31, which indicated a strong Chernobyl contamination. Lichen sample 303/86 (Kalanti) had a higher activity ratio in its top part than it did in the bottom part, whereas lichen sample 328 (Huittinen) had a lower activity ratio in its top part than in its bottom part. Activity concentrations, and activity and mass ratios of transuranium nuclides in the lichen obtained from Kalanti were abnormal. This anomaly was probably due to hot particles in the lichen sample (Paatero *et al.* 1998, Salminen-Paatero *et al.* 2012). In a previous study (Paatero *et al.* 1994a) with different lichen samples collected from Huittinen in 1987 and 1988, the ^{241}Pu/$^{239+240}$Pu activity ratios in the top and bottom parts were from 41 ± 9 to 98 ± 12. The activity ratios and their ranges in the lichen samples obtained from Huittinen were similar in the current study and in the study by Paatero *et al.* (1994a). Moreover, the activity ratios were higher in the top parts in all three top–bottom lichen sample pair comparisons.

In this study, five peat samples had ^{241}Pu/$^{239+240}$Pu activity ratios that ranged from 4.1 ± 0.3 to 60 ± 5. Peat samples 37 and 42 had low activity ratios due to global nuclear fallout, whereas peat samples 60, 64, and 143 had high activity ratios that were attributed to Chernobyl fallout. In previous studies, in which the same peat samples were used, an uneven regional distribution of Chernobyl-derived transuranium deposition was found (Reponen *et al.* 1993, Paatero *et al.* 1994a, Salminen *et al.* 2005). A study on soil samples in Poland revealed local Chernobyl-derived ^{241}Pu contamination among other transuranium nuclides, which were in the form of hot particles (Broda *et al.* 1989, Mietelski *et al.* 1999) (Table 5). In contrast to those found in our samples, low ^{241}Pu/$^{239+240}$Pu activity ratios associated with low activity concentration of ^{241}Pu were found in soil and moss samples collected in Hungary in 1998 (Varga and Tarján 2008). High ^{241}Pu/$^{239+240}$Pu activity ratio with relatively high activity concentration of ^{241}Pu were found only in a Hungarian ashed fodder sample, the ratio in 1998 was 68.5 ± 6.7 (Varga and Tarján 2008).

An air filter sample from Nurmijärvi had a ^{241}Pu/$^{239+240}$Pu activity ratio of 98 ± 14. This value was in excellent agreement with a previously-determined value 94.8 for the fresh Chernobyl-derived contamination in Finland (Table 1) (Paatero *et al.* 1994a). It was also comparable with the values of 85 ± 20 and 86 ±

Table 5. ^{241}Pu/$^{239+240}$Pu activity ratios in environmental samples collected in Chernobyl-contaminated areas.

A ^{241}Pu/A $^{239+240}$Pu	Sample type	Country	Reference
9.6 ± 2.9 to 69 ± 11	Soil	Poland	Mietelski and Was (1995)
86 ± 47	Soil	Poland	Mietelski *et al.* (1999)
68.5 ± 6.7	Ashed fodder	Hungary	Varga and Tarján (2008)
85 ± 20	Air filter	Sweden	Holm *et al.* (1992)
115 ± 14	Soil	Russia	Carbol *et al.* (2003)
102 ± 15	Hot particles	Poland	Broda *et al.* (1989)

47 determined in surface air in Sweden (Holm *et al*. 1992) and in soil in Poland (Mietelski *et al*. 1999) (Table 5), respectively, and 67–82 given by the USSR State Committee on the Utilization of Atomic Energy (1986) (Table 1). Moreover, grass and hot-particle samples had high ^{241}Pu/$^{239+240}$Pu activity ratios, typical for Pu deposition from Chernobyl fallout. Some of our samples had exceptionally high (> 100) ^{241}Pu/$^{239+240}$Pu activity ratios, as compared with the ratio in Chernobyl-contaminated samples quoted in the literature. A very high ^{241}Pu/$^{239+240}$Pu ratio of 115 ± 14 (in April 1986) was also found by Carbol *et al*. (2003) in soil in Bryansk, Russia. Another high ^{241}Pu/$^{239+240}$Pu ratio value of 102 ± 15, was determined in hot particles found in Poland after the Chernobyl accident (Broda *et al*. 1989).

The ^{241}Pu/$^{239+240}$Pu activity ratios in our environmental samples taken in 1986 and measured in 2011 were from 1.22 ± 0.09 to 50 ± 9. The lowest values are similar to the activity ratio of < 2 attributed to global fallout from nuclear weapons testing today, whereas the highest values were even higher than ~28 [the ^{241}Pu/$^{239+240}$Pu ratio in Chernobyl-derived fallout in 2011 calculated by us from the data of Paatero *et al*. (1994a)].

Practically all Chernobyl-originated ^{241}Pu will have decayed to ^{241}Am by the year 2059, when the ^{241}Am/$^{239+240}$Pu activity ratio would be 2.8 excluding the ^{241}Am present in original Chernobyl fallout (Paatero *et al*. 1994a). In our samples of 1986 analysed in 2011, the activity concentration of ^{241}Am and the activity ratio ^{241}Am/$^{239+240}$Pu (^{241}Am exclusively from decay of Chernobyl-derived ^{241}Pu) were from < 0.02 to 15.6 ± 0.9 mBq g^{-1} and from 0.093 ± 0.007 to 3.8 ± 0.7, respectively. The median value of the ^{241}Am/$^{239+240}$Pu ratio for 2011 was 1.9, i.e. already 68% of the value 2.8 predicted for the year 2059. In these samples, the ^{241}Am/$^{239+240}$Pu activity ratio will be between 0.14 ± 0.01 and 5.5 ± 1.0 in 2059. In our lichen samples from 1967–1976, the activity concentrations of ^{241}Am ingrown from ^{241}Pu present in global nuclear fallout were in 2011 from 0.060 ± 0.006 to 2.60 ± 0.15 mBq g^{-1}, while for the same samples the ^{241}Am/$^{239+240}$Pu activity ratios were from 0.19 ± 0.02 to 0.36 ± 0.03.

The ^{242}Cm activity concentration and ^{242}Cm/$^{239+240}$Pu activity ratio in the environmental samples from 1986 were determined in 1986–1988 by Jaakkola *et al*. (1986) and Paatero *et al*. (1998). Linear regression analysis made by us revealed that the relation between the ^{241}Pu/$^{239+240}$Pu and ^{238}Pu/$^{239+240}$Pu activity ratios was strong (*see* Fig. 2), and that between the ^{241}Pu/$^{239+240}$Pu and ^{242}Cm/$^{239+240}$Pu activity ratios fairly strong (*see* Fig. 3). ^{238}Pu and especially ^{241}Pu and ^{242}Cm are considered to be important Chernobyl-derived transuranium isotopes, so it is interesting to see if there are any differences resulting from the origin of these nuclides (global fallout *vs*. Chernobyl accident) in the same samples. However, due to different sources of transuranium isotopes, ^{238}Pu/$^{239+240}$Pu and ^{242}Cm/$^{239+240}$Pu have much stronger correlation with ^{241}Pu/$^{239+240}$Pu than ^{240}Pu/^{239}Pu has with ^{241}Pu/$^{239+240}$Pu. ^{239}Pu and ^{240}Pu are predominantly originating from global fallout, unlike ^{238}Pu, ^{241}Pu, and ^{242}Cm. Linear regression analysis made by us revealed that the dependence between the ^{240}Pu/^{239}Pu mass ratio and ^{241}Pu/$^{239+240}$Pu activity ratio was moderate (Fig. 4). The data point of ‘Top lichen 303/86’ (Kalanti) deviates from the line fitting, which distorts the correlation. As previously mentioned, this sample had anomalous activity concentrations and isotope ratios that could probably be attributed to hot particles.

The Chernobyl-derived fraction of Pu from total Pu in the samples from 1986

The fraction (F) of Chernobyl-derived Pu in total Pu in the environmental samples taken in 1986 was calculated with the formula of Paatero *et al*. (1994a):

$$F = (C_s - C_f)/(C_c - C_s),$$

where C_s, C_f and C_c are the activity (mass) ratios in the sample, in global fallout and in Chernobyl fallout, respectively. The fraction coefficient S for the Chernobyl-derived Pu in total Pu in the sample can then be calculated as:

$$S = F/(F + 1)$$

The Chernobyl-derived fraction of Pu, expressed as S × 100 (%), was calculated in three

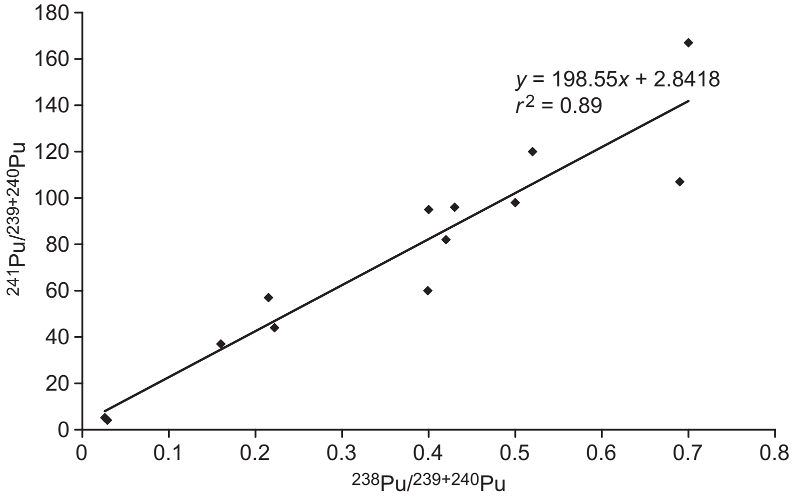

Fig. 2. The ^{241}Pu/$^{239+240}$Pu activity ratio as a function of the ^{238}Pu/$^{239+240}$Pu activity ratio in the selected environmental samples collected in 1986.

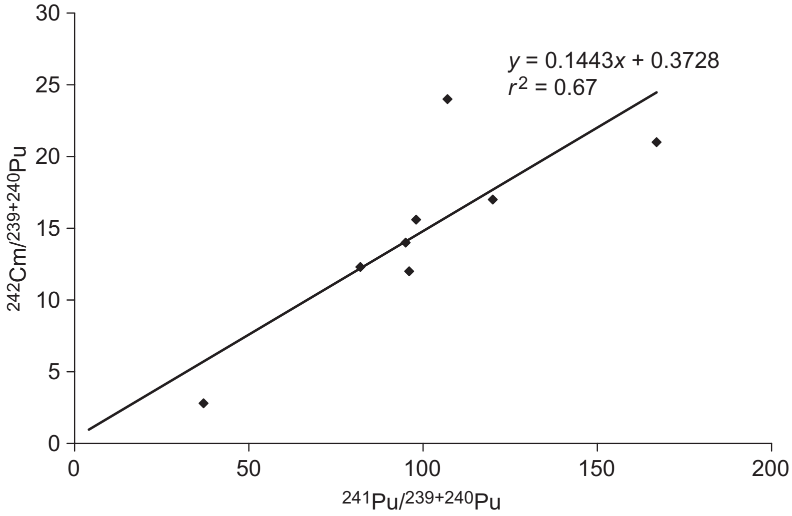

Fig. 3. The ^{242}Cm/$^{239+240}$Pu activity ratios as a function of the ^{241}Pu/$^{239+240}$Pu activity ratios in the selected environmental samples. The ^{242}Cm/$^{239+240}$Pu activity ratios in these samples were published by Paatero *et al.* (1998).

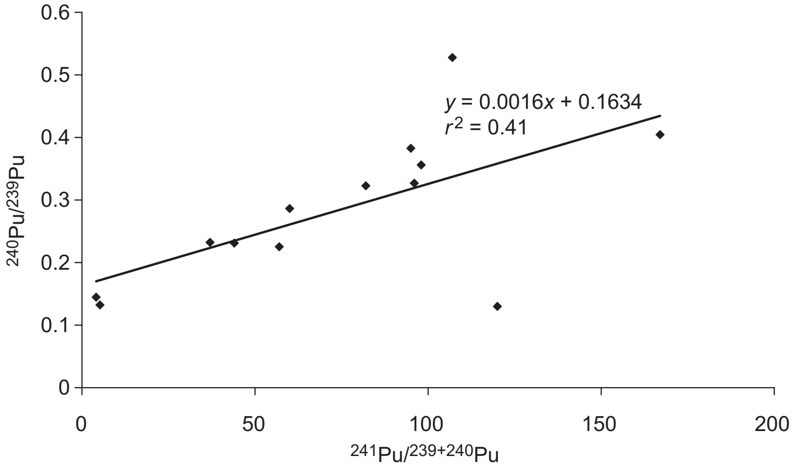

Fig. 4. The ^{240}Pu/^{239}Pu mass ratios *vs.* ^{241}Pu/$^{239+240}$Pu activity ratios in the environmental samples collected in 1986. The ^{240}Pu/^{239}Pu mass ratios in the samples were published in (Salminen-Paatero *et al.* 2012).

ways: from the ^{238}Pu/$^{239+240}$Pu activity ratio, the ^{241}Pu/$^{239+240}$Pu activity ratio, and the ^{240}Pu/^{239}Pu mass ratio. In present study 0.03, 3.9, and 0.18 were used as C_f values for ^{238}Pu/$^{239+240}$Pu activity ratio, ^{241}Pu/$^{239+240}$Pu activity ratio, and ^{240}Pu/^{239}Pu mass ratio, respectively. For C_c the values of 0.5, 94.8, and 0.38 for ^{238}Pu/$^{239+240}$Pu activity ratios, ^{241}Pu/$^{239+240}$Pu activity ratios, and ^{240}Pu/^{239}Pu mass ratios, respectively, were used in the calculations. The Chernobyl-derived Pu fraction in the same sample was roughly at the same level regardless of the way it was calculated (Table 6). However, for the same samples, the three activity and mass ratio values used in this study gave Chernobyl-derived fractions of Pu that deviated inconsistently from each other. Therefore, it seems necessary to evaluate the Chernobyl-derived Pu contamination in a sample with at least two isotope ratios to get more reliable estimation of the magnitude of Chernobyl-derived contamination than when using only one isotope ratio. It is interesting that of the six peat samples analyzed in this study and by Paatero *et al*. (1994a), samples 22, 37, and 42, that were contaminated by global fallout only, had Chernobyl-derived fractions of ~0% regardless of the isotope ratio method used. However, samples 60, 64, and 143 were mainly contaminated by the Chernobyl accident and the fractions of Chernobyl-derived Pu measured in the present study and by Paatero *et al*. (1994a) (direct ^{241}Pu determination by LSC) differed, the percentages being 45%–61% and 92%–97%, respectively. However, samples 60, 64, and 143 were mainly contaminated by the Chernobyl accident and the fractions of Chernobyl-derived Pu (direct ^{241}Pu determination by LSC) measured in the present study and previously differed: 45%–61% in the present study and 92%–97% in Paatero *et al*. (1994).

The 1-σ error for the Chernobyl-derived fraction of Pu in total Pu was calculated from counting statistics only, and these fraction values should be considered an overall estimate magnitude, not exact values, due to difficulties in producing an absolute uncertainty from all reference values and uncertainty factors. Large variations in Chernobyl-derived fractions of Pu in total Pu, in addition to very high S values with high uncertainties, were also reported by Mietelski *et al*. (1999) for forest soil samples with varying levels of Chernobyl-contamination in Poland.

Transuranium isotope composition and fuel burnup of the two hot particle samples

The hot particle sample U37(1) was more active

Table 6. The Chernobyl-derived fraction of Pu (Pu_{Ch}) derived from total Pu in environmental samples collected after the Chernobyl accident in 1986, based on the activity ratios ^{238}Pu/$^{239+240}$Pu (Jaakkola *et al.* 1986, Reponen *et al.* 1993, Paatero *et al.* 1998) and ^{241}Pu/$^{239+240}$Pu (this study) and mass ratio ^{240}Pu/^{239}Pu determinations (Salminen-Paatero *et al.* 2012).

Sample	Pu_{Ch}/Pu_{total} (%) by $A^{238}Pu/A^{239+240}Pu$	Pu_{Ch}/Pu_{total} (%) by $A^{241}Pu/A^{239+240}Pu$	Pu_{Ch}/Pu_{total} (%) by ^{240}Pu/^{239}Pu (mass ratio)
Peat 22	0	–	0
Peat 37	0	0	0
Peat 42	0	1.4 ± 0.1	0
Peat 60	78 ± 12	61 ± 7	55 ± 6
Peat 64	41 ± 5	45 ± 4	25 ± 2
Peat 143	39 ± 6	58 ± 7	23 ± 1
Top lichen Pu 303/86	104 ± 14	128 ± 10	0
Bottom lichen 303/86	143 ± 58	179 ± 48	115 ± 21
Top lichen L328	83 ± 8	86 ± 8	70 ± 5
Bottom lichen T328	28 ± 5	37 ± 4	25 ± 2
Top lichen 323/06	51 ± 13	–	75 ± 53
Grass N307	79 ± 19	100 ± 13	100 ± 12
Air filter	100 ± 37	104 ± 20	90 ± 38
Hot particle U37(1)	140 ± 12	113 ± 8	175 ± 14
Hot particle U37/2(1)	85 ± 28	102 ± 22	75 ± 21

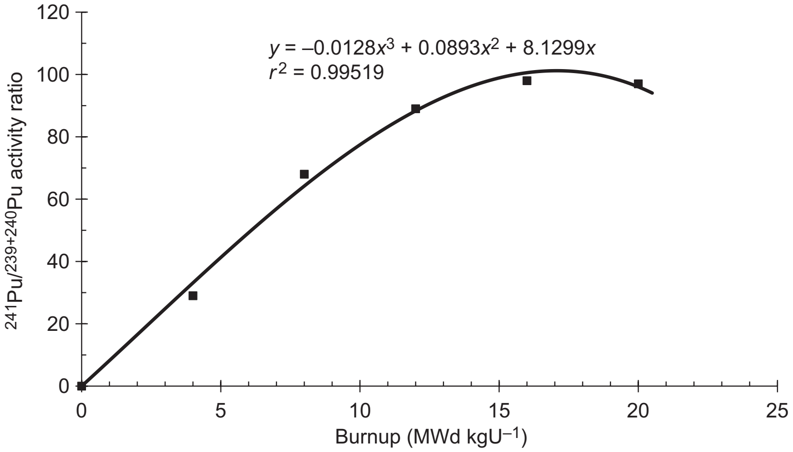

Fig. 5. The ^{241}Pu/$^{239+240}$Pu activity ratios as a function of fuel burnup of an RBMK reactor (Zaritskaya *et al.* 1979).

than sample U37/2(1), as was reported in the previous study of these samples (Table 7). However, both hot particle samples had high activity ratios of ^{238}Pu/$^{239+240}$Pu, ^{241}Pu/$^{239+240}$Pu, and ^{242}Cm/$^{239+240}$Pu in addition to the mass ratio of ^{240}Pu/^{239}Pu, which revealed contamination from nuclear fuel with high burnup of the destroyed 4th reactor of the Chernobyl nuclear power plant. Based on the work of Zaritskaya *et al*. (1979) it can be estimated that the measured ^{241}Pu/$^{239+240}$Pu activity ratios of the two hot particles correspond to burnup values of 16 and 14 MWday kgU^{-1} (Fig. 5). Earlier burnup estimations based on other plutonium isotope ratios were 14 and 9–10 MWdays kgU^{-1} for the hot particles U37(1) and U37/2(1), respectively (Paatero *et al*. 1998, Salminen-Paatero *et al*. 2012). An early Soviet report gave a mean burnup value of 10.5 MWdays kgU^{-1} for the nuclear fuel of the destroyed reactor (USSR State Committee on the Utilization of Atomic Energy 1986). Thus our current results are in reasonable agreement with the earlier reports.

Conclusions

The ^{241}Pu/$^{239+240}$Pu activity ratios gave supporting information for previously determined ^{238}Pu/$^{239+240}$Pu activity ratios and ^{240}Pu/^{239}Pu mass ratios, regarding the origin of Pu contamination in the environmental samples collected in Finland in 1967–1976 and in 1986. The samples taken in 1967–1976 had global fallout as the only contamination source whereas the samples taken in 1986 had global and Chernobyl fallout in varying fractions as contamination sources. High correlation between the activity concentrations of ^{238}Pu, ^{241}Pu, and ^{242}Cm, originating mostly from the Chernobyl accident, were found in environmental samples of 1986.

The activity concentration of ^{241}Am, that originates from ^{241}Pu present in fallout both from nuclear weapons testing and the Chernobyl accident, will increase in the environment during next decades until all Chernobyl-derived ^{241}Pu decays. The activity of ingrown ^{241}Am already

Table 7. Activity and mass ratios of transuranium nuclides in two hot particles.

	Hot particles	
	U37(1)	U37/2(1)
A ^{238}Pu/A $^{239+240}$Pu^{c}	0.69 ± 0.04	0.43 ± 0.10
A ^{241}Am/A $^{239+240}$Pu^{c}	0.43 ± 0.05	0.40 ± 0.13
A ^{242}Cm/A $^{239+240}$Pu^{c}	24 ± 2	12 ± 3
A ^{242}Cm/A ^{241}Am^{c}	56 ± 7	31 ± 11
A ^{241}Pu/A $^{239+240}$Pu	107 ± 5	96 ± 15
A ^{241}Pu/A ^{241}Am	251 ± 32	247 ± 82
A $^{239+240}$Pu (mBq)^{a,c}	9.4 ± 0.4	0.64 ± 0.08
A ^{241}Pu (mBq)^{a}	1005 ± 24	62 ± 6
^{240}Pu/^{239}Pu mass ratio^{d}	0.53 ± 0.03	0.33 ± 0.07
Burnup (MWday kgU^{-1})^{b}	16	14

[a] The activity of the whole sample. [b] Calculated from the ^{241}Pu/$^{239+240}$Pu activity ratio in the samples and from the data published by Zaritskaya *et al.* (1979). [c] From Paatero *et al.* (1998). [d] From Salminen-Paatero *et al.* (2012).

exceeded the activity of $^{239+240}$Pu in some of our Chernobyl-contaminated samples examined in 2011. Considering additional ^{241}Am deposited directly from nuclear weapons testing and the Chernobyl accident, the total amount of ^{241}Am in the environment is even higher. Still, the total annual deposition of natural α and β emitters, ^{210}Pb and ^{210}Po, in Finland is about hundredfold higher than the transuranium deposition from the Chernobyl accident was (Paatero *et al*. 1994b).

Determination of ^{241}Pu in 27 environmental samples completes the previous studies of transuranium deposition (regional and temporal behaviour and nuclide mixture) in Finland during the 20th century. Provided that the original concentration of ^{241}Pu in an environmental sample is high enough and the ingrowth period is sufficiently long, determination of ^{241}Pu from archived Pu α-counting samples via measuring ^{241}Am by α-spectrometry is a relatively simple, non-destructive, and low-cost method.

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