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Concentrations of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu in the surface air in Finnish Lapland in 1963

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Activity concentrations of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu (measured both directly by liquid scintillation counting and indirectly by buildup of ^{241}Am) were determined with the aid of archived air filters sampled at Sodankylä (67°22´N, 26°39´E) in 1963. Atmospheric concentrations of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu were < 0.1–3.6(± 0.5) μBq m^{-3}, 0.4(± 0.1)–95(± 3) μBq m^{-3} and < 16–1730(± 70) μBq m^{-3}, respectively. The plutonium concentrations varied seasonally, being highest in spring and summer due to the springtime-enhanced transportation of radioactive aerosols from the stratosphere to the troposphere. Contrary to some earlier assumptions, the $^{239+240}$Pu activity concentration in the air was practically at the same level in northern and southern Finland. The ^{238}Pu/$^{239+240}$Pu activity ratio in the air was 0.014(± 0.003)–0.32(± 0.11), typically close to that of global fallout from the nuclear weapons testing. The ^{241}Pu/$^{239+240}$Pu activity ratio varied between 7.7(± 1.2) and 41(± 8). According to these isotope ratios, plutonium in Sodankylä originated mainly from global nuclear test fallout, even though the effect of the Soviet tests at the end of 1962 were observed at the beginning of 1963. However, direct atmospheric transport from Novaya Zemlya to Finnish Lapland could not be detected.

Introduction

Global radioactive fallout in 1963 originated from the atmospheric nuclear weapons tests of the 1950s and 1960s conducted mainly by the USA and Soviet Union and to a much lesser extent by Great Britain and France (UNSCEAR 2000). A partial nuclear test ban started at the beginning of 1963, but the deposition of the artificial radioactivity released in the weapons testing reached its maximum value in the same year. This was due to the intense atmospheric weapons testing between September 1961 and December 1962. The radioactive debris from these tests descended from the stratosphere to the troposphere, especially in spring 1963. The most powerful atmospheric nuclear test "Tsar Bomba" or "Big Ivan" (50–58 megatons depending on the reference) was carried out on 30 October 1961 at the height of 4000 m above Sukhoy Nos Cape, Novaya Zemlya (approximate coordinates 73.85°N, 54.50°E). During 1958–1962 two main trajectories of radioactive plumes from the Novaya Zemlya nuclear tests spread south towards the Caspian Sea and thousands of kilometers southeast towards the Okhotsk Sea (Khalturin *et al*. 2005).

The intense atmospheric testing of nuclear weapons in the late 1950s led many countries to initiate surveillance programmes for environmental radioactivity. In Finland a nationwide network for airborne radioactivity monitoring was developed within the national weather service, the Finnish Meteorological Institute (FMI). This development gave the opportunity to take into account the influence of atmospheric processes on the concentration and deposition of airborne radioactivity. The air filters collected within FMI's monitoring programme have been archived, excluding the very first years. Such archived air filters collected at Sodankylä, Finnish Lapland, in 1963 were chosen for this study because there are only a few observations of radioactivity in the air from the 1960s at such high latitudes (67–68°N). Activity concentrations of ^{137}Cs, ^{90}Sr, ^{238}Pu, $^{239+240}$Pu and ^{241}Pu in the air during the year of the maximum deposition would produce important information about the behaviour of artificial radioactivity in the boreal atmosphere. Especially of interest were the immediate effects of the Novaya Zemlya experiments on the radioactivity situation at Sodankylä, about 1000 km southwest of the Novaya Zemlya nuclear test site. The possibility of direct atmospheric transport of ^{137}Cs or total beta activity from Novaya Zemlya to Norway has already been investigated at the northernmost Norwegian sampling site, Vadsø, located only about 800 km west of Novaya Zemlya (Bergan 2002). However, no clear evidence of such transport was obtained in Norway. The beta emitter ^{241}Pu was analyzed in this study to get additional information about the isotopic composition of airborne plutonium in Finnish Lapland during 1963. Two methods were utilized in determining ^{241}Pu: indirectly by measuring the alpha activity of ingrown ^{241}Am in a sample, or directly by dissolving NdF_3 precipitate (containing plutonium isotopes) of the alpha counting preparates and measuring the beta particle emissions of ^{241}Pu by liquid scintillation counting (LSC). The total activity of ^{241}Am in the nuclear test fallout in Finland during the 1960s was less than one percent of the total activity of ^{241}Pu (Salminen *et al*. 2005). Therefore, it was assumed that all ^{241}Am in a sample originated from the decay of ^{241}Pu after the sampling.

Materials and methods

Samples and preliminary analyses

Daily aerosol samples were collected by filter sampling at the Sodankylä meteorological observatory of FMI (67°22´N, 26°39´E, 179 m a.s.l.), about 100 km north of the Arctic Circle. The air filters (Whatman 42) had an effective filtering area of 269 cm^2, the air flow rate was approximately 20.4 m^3 h^{-1} and the filtered air volume was typically 490 m^3. The total beta activities of the aerosol samples were measured in the FMI laboratory five days after the end of sampling, when the short-lived ^{222}Rn progeny had decayed into ^{210}Pb and the ^{220}Rn progeny had decayed into stable lead. In 1963, the measured total beta activity consisted mainly of artificial beta emitters and ^{210}Pb with its daughter nuclide ^{210}Bi (Helminen and Mattson 1964). After the measurements, the filters were stored in the sample archive of FMI. The daily aerosol sampling programme at Sodankylä is still continuing (Paatero *et al*. 1998). Daily air filter halves from year 1963 were retrieved from the sample archive and analysed with HPGe gamma spectrometry to determine ^{137}Cs. Next, the samples were combined to 176 samples, each covering 1–3 days. This pooling of samples was a trade-off between sufficient time resolution for subsequent meteorological analysis and laboratory sample throughput.

Plutonium, americium and strontium analyses

The method used for separating plutonium, americium and strontium was based on Eichrom's TRU® resin and Sr-resin®, and modified and combined from methods used by Sidhu (2004), Nygren *et al*. (2001) and Spry *et al*. (2000) (Fig. 1). The analytical procedure was kept as simple as possible due to the large sample number. Paper filters are a relatively easy sample matrix, which enabled a simplified separation procedure. The separation method produced samples of sufficient purity for alpha spectrometry, but for plutonium measurements by mass spectrometry an extra U/Pu-separation might be necessary. Before the

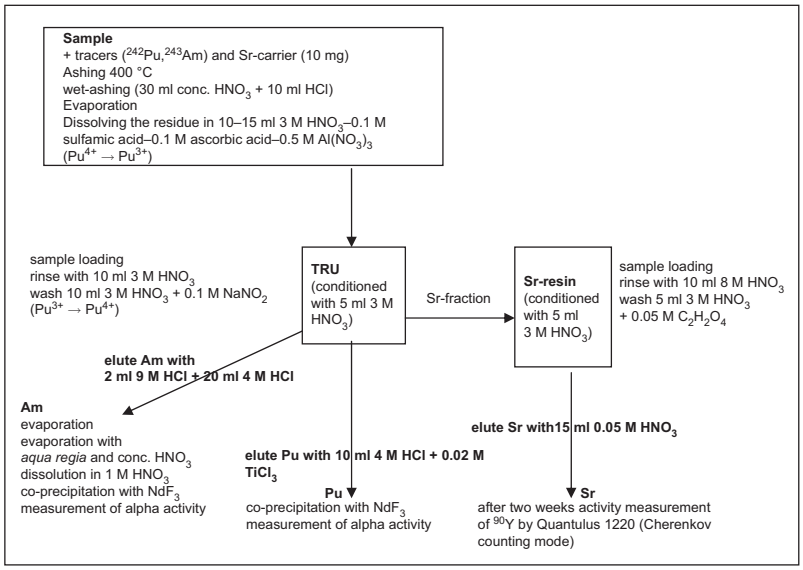

Fig. 1. Separation scheme for the analysis of plutonium isotopes, ^{241}Am and ^{90}Sr.

air filter sample analyses, the method was tested with a standard reference sediment IAEA-135.

After addition of the ^{242}Pu and ^{243}Am tracers and the strontium carrier and decomposition of filters by ashing and wet-ashing, the separation of plutonium, americium and strontium was accomplished by extraction chromatography. Plutonium and americium were co-precipitated with NdF_3 onto a membrane filter for alpha counting by the method of Hindman (1983). Activities of ^{238}Pu, $^{239+240}$Pu and ^{241}Am were measured with PIPS alpha detectors (nominal resolution 20 keV for 5.486 MeV peak of ^{241}Am) *in vacuo* using counting times of between five and seven days.

For measuring ^{241}Pu with LSC, ten plutonium alpha counting preparates were transferred to plastic scintillation vials. A membrane filter containing the NdF_3 precipitate was wetted with 0.6 M H_3BO_3 + 0.1 M HCl and after at least four hours a liquid scintillation cocktail (Ultima Gold LLT) was added. The activity of ^{241}Pu was determined using a liquid scintillation spectrometer Quantulus 1220 (Wallac Ltd.) with a counting time of ten hours.

Strontium fractions eluted from Sr-resin® columns were set aside for two weeks (ingrowth of ^{90}Y and formation of ^{90}Y/^{90}Sr equilibrium) and the activity of ^{90}Y was measured with a Quantulus 1220 spectrometer using the Cherenkov counting mode. Recovery of strontium was determined with ion chromatography. The activity contents of ^{137}Cs and ^{90}Sr in the air filter samples will be reported in the future.

Results and discussion

Functionality of the separation method

Samples of the reference sediment IAEA-135 were analyzed prior to the air filter samples to confirm the functionality of the separation procedure especially for plutonium. Blank samples (unexposed air filter samples) were analyzed to detect possible contamination from reagents, glassware, etc. during analysis. Alpha count rates of plutonium and americium blank samples were negligible. Results for the reference samples and blank analyses are summarized in Table 1.

Activity concentrations of ^{238}Pu and $^{239+240}$Pu in the IAEA-135 sediment agreed well with the values recommended by IAEA. On the other hand, activity concentrations of ^{241}Am in IAEA-135 samples exceeded the confidence interval. However, a probable reason for this is the increased activity of ^{241}Am in the reference sediment since its reference date (1 January 1992) due to ingrowth from ^{241}Pu.

The average recoveries of plutonium and americium were 65% and 60%, respectively, for

the reference samples and for the blank samples the corresponding recoveries were 93% and 60%. With the actual air filter samples, median recoveries for plutonium and americium were 90% and 65%, respectively.

Activity concentrations of plutonium isotopes, and $^{238}Pu/^{239+240}Pu$ and $^{241}Pu/^{239+240}Pu$ activity ratios in air filter samples

The minimum, maximum and median values for activities of plutonium isotopes and ^{241}Am in the air of Sodankylä are summarized in Table 2. Elevated activity concentrations in spring and summer were observed concomitantly with the total beta activity (Helminen and Mattson 1964), ^{238}Pu, $^{239+240}Pu$ and ^{241}Am (^{241}Pu) activity concentrations. The spring maximum was caused by the increased transport of radionuclides from the stratosphere down to the troposphere where the radionuclides were susceptible to dry and especially wet deposition. Activity concentrations of ^{238}Pu and $^{239+240}Pu$ were < 0.1–3.6(± 0.5) μBq m^{-3} and 0.4(± 0.1)–95(± 3) μBq m^{-3}, respectively, decay-corrected to year 1963 (Figs. 2 and 3). The quarterly mean values for activity concentration of $^{239+240}Pu$ at Sodankylä were 13, 29, 21 and 5 μBq m^{-3}. For comparison, activity concentrations of $^{239+240}Pu$ in the air in Helsinki (southern Finland) during the same year has been reported to be 11, 34, 25 and 7 μBq m^{-3}, in other words, the highest value corresponding with April–June 1963, too (Jaakkola *et al*. 1979). Thus, there are no significant differences in the average $^{239+240}Pu$ content of the air in northern and southern Finland. The variation of individual values is, however, large and they can deviate significantly from the average values.

Due to one third less precipitation in northern Finland than in southern Finland, it has been assumed that the deposition from global fallout in northern Finland was only two thirds of that in southern Finland 1945–1978 (Mussalo-Rauhamaa *et al*. 1984). Furthermore, this difference

Table 1. Activity concentrations of $^{239+240}Pu$, ^{238}Pu and ^{241}Am for reference samples and recoveries for reference and blank samples. The reference date for the values recommended by IAEA is 1 Jan. 1992. * = information value only.

	Activity concentrations			Pu recovery (%)	Am recovery (%)
IAEA-135	$^{239+240}Pu$ (mBq g^{-1})	^{238}Pu (mBq g^{-1})	^{241}Am (mBq g^{-1})		
Sample 1	220 ± 6	45 ± 3	453 ± 7	66	59
Sample 2	201 ± 5	41 ± 2	482 ± 8	73	54
Sample 3	187 ± 6	46 ± 3	446 ± 6	62	63
Sample 4	229 ± 7	43 ± 3	499 ± 8	51	52
Sample 5	291 ± 5	50 ± 2	500 ± 8	75	73
Average	226 ± 6	45 ± 3	476 ± 8	65	60
Recommended value	213	43	318*		
Confidence interval	205–225.8	41.6–45	310–325*		
Blank (6 samples)	negligible	negligible	negligible	89–95	57–66

Table 2. Minimum and maximum activity concentrations of ^{238}Pu (1963), $^{239+240}Pu$ (1963), ^{241}Pu (1963) and ^{241}Am (2007), and activity ratios $^{238}Pu/^{239+240}Pu$ (1963) and $^{241}Pu/^{239+240}Pu$ (1963). ^{241}Pu was determined by ^{241}Am ingrowth. 1σ errors were calculated using the method of Currie (1968).

	^{238}Pu (μBq m^{-3})	$^{239+240}Pu$ (μBq m^{-3})	^{241}Am (μBq m^{-3})	^{241}Pu (μBq m^{-3})	$^{238}Pu/^{239+240}Pu$	$^{241}Pu/^{239+240}Pu$
Minimum	< 0.1	0.4 ± 0.1	< 0.5	< 16	0.014 ± 0.003	7.6 ± 1.2
Maximum	3.6 ± 0.5	95 ± 3	47 ± 2	1725 ± 74	0.32 ± 0.11	41 ± 8
Median	0.63	15	7.8	292	0.045	18

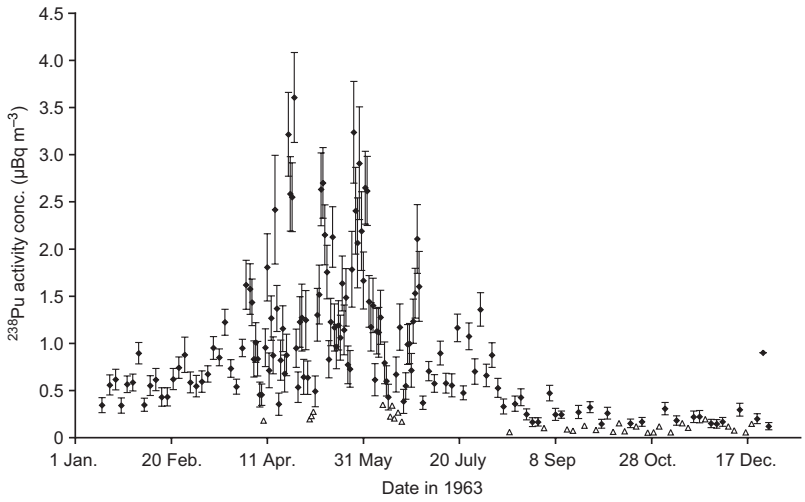

Fig. 2. Activity concentration of ^{238}Pu in the air of Sodankylä in 1963. White triangles without error bars are halves of the MDA values.

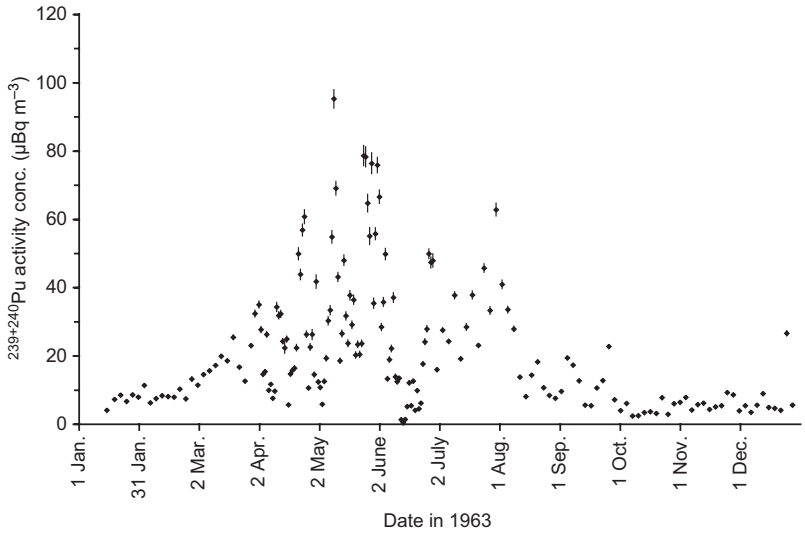

Fig. 3. Activity concentration of $^{239+240}$Pu in the air of Sodankylä during 1963.

has been assumed to exist also in the case of atmospheric concentrations and thus exposure to inhaled radionuclides. However, from data for Helsinki and Sodankylä it is clear that there is no significant difference in the activity concentration of plutonium in air between southern and northern Finland, at least during 1963, despite the different precipitation amounts. In 1963 the annual averages of total beta activity concentrations were 80 000 and 94 300 μBq m^{-3} in northern and southern Finland, respectively. In other words, the difference in atmospheric concentrations of the total beta activity between the southern and northern parts of Finland was 15% (Helminen and Mattsson 1964). In Norway, the deposition of ^{137}Cs, ^{131}I and ^{90}Sr, representatives of long-lived radionuclides studied, did not decrease from south to north (Bergan 2002). This is explained by varying precipitation amounts at sampling sites, which in this case did not systematically decrease from south to north.

Publications on the activity concentrations of plutonium isotopes in air in the early 1960s have been scarce. In spring 1963, atmospheric concentrations of $^{239+240}$Pu and ^{238}Pu in Hanford Nuclear Reservation, Richland, were 11 and 0.6 μBq m^{-3}, respectively (Pan and Stevenson 1996). The annual mean $^{239+240}$Pu activity concentration was 26 μBq m^{-3} in Chilton, UK (Cambray *et al.* 1974). Therefore, atmospheric concentrations of

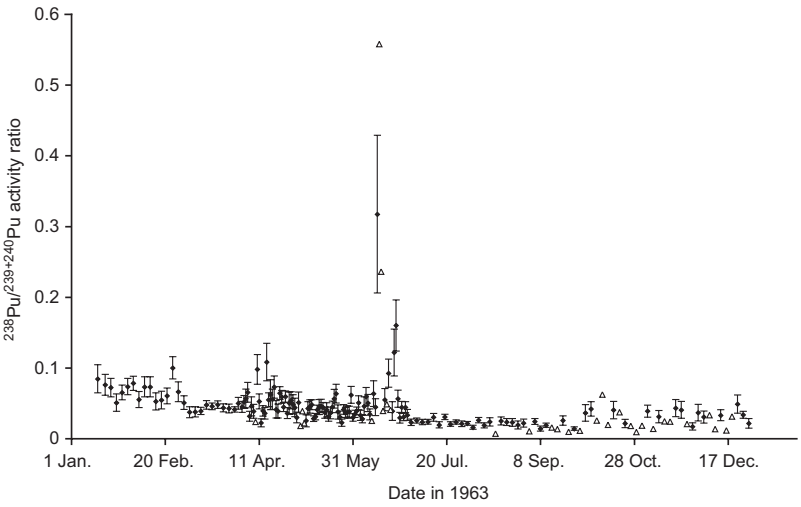

Fig. 4. $^{238}Pu/^{239+240}Pu$ activity ratio in Sodankylä air filters, 1963. The white triangles without error bars represent samples where the ^{238}Pu concentration remained below the detection limit. In these cases a value of half the MDA for ^{238}Pu divided by $^{239+240}Pu$ activity was used.

$^{239+240}Pu$ were at the same level in USA, UK and Finland in 1963.

The isotopic composition of the weapons-grade and the fallout plutonium differ from each other because of the neutron-induced nuclear reactions in plutonium during the detonation of the weapon. The $^{238}Pu/^{239+240}Pu$ activity ratio in air filter samples collected in Helsinki was 0.016(± 0.004)–0.048(± 0.027) (Jaakkola *et al*. 1979), while the ratio in air filters from Sodankylä had a wider variation with some higher values, 0.014(± 0.003)–0.32(± 0.11). The main part of the samples had a $^{238}Pu/^{239+240}Pu$ ratio of ~0.03 (Fig. 4), corresponding to the ratio of 0.024 in global nuclear testing fallout before 1964 (Hardy *et al*. 1973). Some low values for the ratio could indicate traces of weapons-grade plutonium, but this has to be confirmed with other methods discussed in the following sections. The high activity ratios, on the other hand, are connected to very low plutonium concentrations, and subsequently to large counting errors, observed in the middle of June 1963.

The $^{238}Pu/^{239+240}Pu$ activity ratios in global nuclear test fallout and weapons-grade plutonium are nowadays so similar that by using this isotope ratio it is difficult to reliably identify the source of plutonium. Therefore, other methods are needed to confirm the source of plutonium in a sample. The $^{240}Pu/^{239}Pu$ mass ratio by ICP-MS is a better indicator for origin of the plutonium from global fallout or weapons-grade plutonium, since the $^{240}Pu/^{239}Pu$ ratios from nuclear testing (~0.18) and weapons-grade plutonium (approximately 0.05) differ significantly (Warneke *et al*. 2002).

In 1963, the activity concentration of ^{241}Pu in the air of Sodankylä determined by the ingrowth of ^{241}Am was < 16–1730(± 70) μBq m^{-3} (Fig. 5 and Table 2). The highest activity concentration values of ^{241}Pu occurred during the spring and the summer similarly to alpha emitting plutonium isotopes. The $^{241}Pu/^{239+240}Pu$ ratio extrapolated to 1963 varied between 7.6(± 1.2) and 41(± 8) (Fig. 6 and Table 2). The median value obtained in this study was 18, representing plutonium from the global fallout because the $^{241}Pu/^{239+240}Pu$ activity ratio in the fresh nuclear test fallout and the weapons-grade plutonium was 16 and 4, respectively (Holm 1995). Values for the weapons-grade $^{241}Pu/^{239+240}Pu$ ratio of 0.75–7.5 from nuclear weapon tests conducted in 1945–1963 have been summarized by Irlweck and Hrnecek (1999). Our results are in agreement with the US authorities' results (Hardy 1973). They reported that in 1963 the activity ratio in the stratospheric air at the latitude 70°N, i.e. the same latitude that crosses northernmost Finnish Lapland, was 19.2. Nuclear tests performed by the USA in the 1950s produced a higher $^{241}Pu/^{239+240}Pu$ activity ratio than tests conducted by the USSR at the beginning of the 1960s (26 and 12–14, respectively; Koide and Goldberg 1981). The high values observed in this work could be partially explained

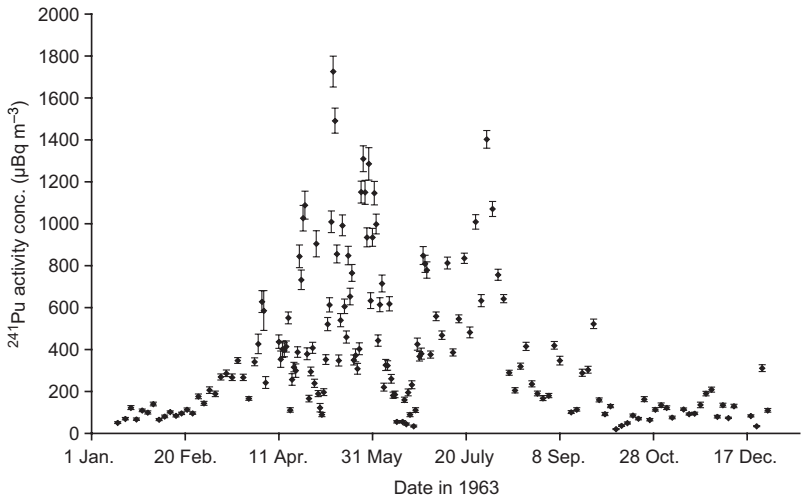

Fig. 5. Activity concentration of ^{241}Pu in the air of Sodankylä, 1963, based on the ingrowth of ^{241}Am between 1963 and 2007.

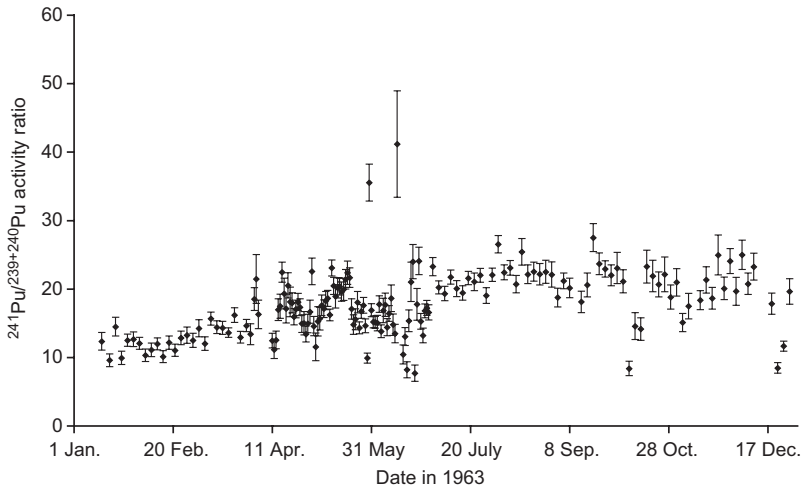

Fig. 6. $^{241}Pu/^{239+240}Pu$ activity ratio in air filters of Sodankylä, 1963.

by the influence from the nuclear tests made by the USA. Some very low $^{241}Pu/^{239+240}Pu$ ratios were observed, and these could be attributed to weapons-grade plutonium if coincident with low $^{238}Pu/^{239+240}Pu$ ratios. But, there is no correlation between low $^{238}Pu/^{239+240}Pu$ and low $^{241}Pu/^{239+240}Pu$ activity ratios. Therefore, it has to be concluded that determination of $^{241}Pu/^{239+240}Pu$ ratios cannot confirm the presence of weapons-grade plutonium suspected from low ratios of $^{238}Pu/^{239+240}Pu$. This study clearly shows the importance of comparative methods for evaluating sources of plutonium.

The $^{238}Pu/^{239+240}Pu$ ratio decreased slightly during 1963, while the $^{241}Pu/^{239+240}Pu$ ratio increased (Fig. 7). Perkins and Thomas (1980) reported slightly higher $^{238}Pu/^{239+240}Pu$ ratios in the first half of 1963 as compared with those in the latter half in the air in Richland, Washington, USA, which is in agreement with this study. The ratio of the total beta activity concentration (Helminen and Mattson 1964) to $^{239+240}Pu$ decreased continuously as the fission products decayed (Fig. 8). The half-life of the fission products was 81 days in January–March while it is almost twice as long, 151 days, in April–December. This can be explained with the change in the origin of the airborne radionuclides. In January–March the fission product mixture was relatively fresh, originating from the nuclear tests made just before the partial test ban treaty became promulgated. Large test explosions by the USA

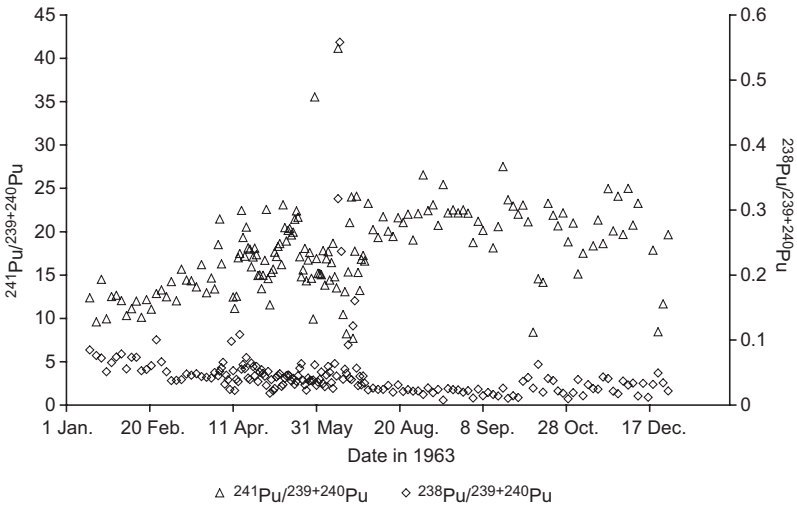

Fig. 7. Trends in activity ratios $^{238}Pu/^{239+240}Pu$ and $^{241}Pu/^{239+240}Pu$ in 1963.

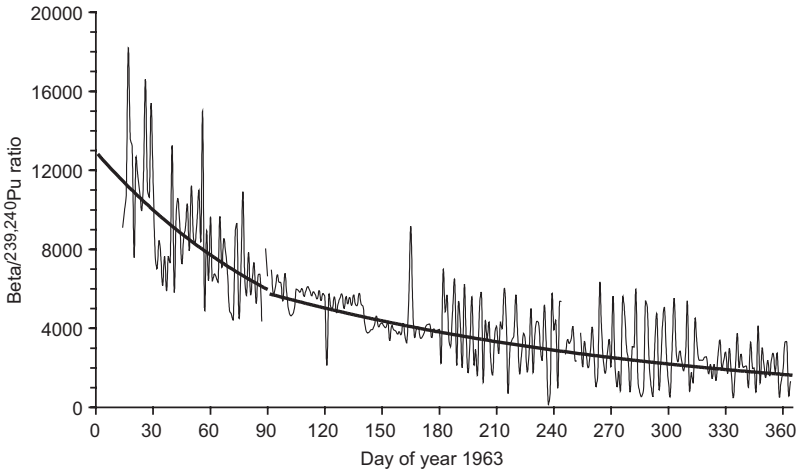

Fig. 8. The ratio of total beta activity to $^{239,240}Pu$ activity concentration in the air of Sodankylä in 1963.

were made in the southern hemisphere and the inter-hemispheric transport of airborne pollutants, whether radioactive or chemical, is slow. The Soviet Union made three megaton-range tests at the Novaya Zemlya test site on Christmas 1962. The tropospheric portion of the debris released in these explosions dominated the artificial radioactivity in the northern air during the first months of 1963. During the arctic night the troposphere is stratified due to the absence of solar radiation causing convection. Also the amount of precipitation causing wet deposition is at its seasonal minimum. These two factors increase the aerosol residence time in the arctic troposphere in winter (Paatero *et al*. 2003). Thus, in spring 1963 the increased stratospheric/tropospheric exchange of air masses and the enhanced vertical mixing in the troposphere brought an aged mixture of radionuclides in large quantities to the ground-level air. In the beginning of the year 1963 the isotopic composition of plutonium is dominated by the Soviet nuclear tests conducted in the latter half of 1962, while later the isotope ratios represent a more general mixture of plutonium originating from various nuclear tests.

In addition to indirect determination of ^{241}Pu by ^{241}Am, ^{241}Pu was directly measured from ten selected samples using LSC (Quantulus 1220) (Table 3). For most samples, the methods gave comparable values, but for samples 5.6. and 8.9 the difference between activity values was unacceptably great: the value from LSC being twice or three times higher than the value from the alpha spectrometric determination.

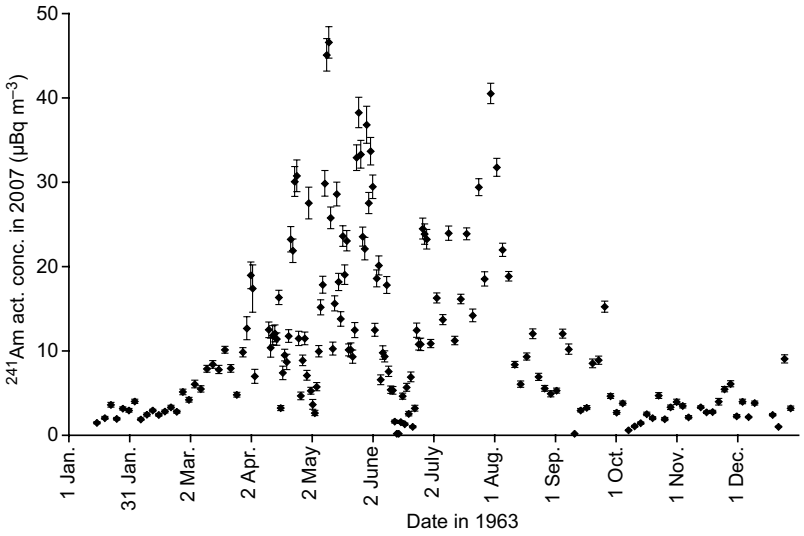

Fig. 9. Apparent activity concentration of ^{241}Am in the air samples taken at Sodankylä in 1963, as measured in 2007.

Rosner *et al*. (1992) compared these two methods, direct and indirect, for determining ^{241}Pu from environmental samples using a proportional counter for ^{241}Pu (β) and a semiconductor detector or an ionization chamber for ^{241}Am (α). Both methods gave similar results indicating no difference in reliability of determining ^{241}Pu. The detection limit is significantly lower with alpha spectrometry (~0.05 mBq/sample) than with LSC (~8 mBq/sample), but the activity level and the age of the sample as well as the time required for the analysis are the most critical factors in determining which method is better for a particular situation. LSC is a faster method compared to alpha spectrometry for detection of ^{241}Pu as there is no need to allow ^{241}Am to grow in. However, if samples containing ^{241}Pu are old enough, as in our case, then it may be more trustworthy to detect ^{241}Pu *via* ^{241}Am and alpha spectrometry because of the lower detection limit.

During the 44 years between sampling and chemical separation, 88% of the ^{241}Pu originally in filters decayed to its daughter nuclide ^{241}Am (Fig. 9). The activity of ^{241}Am in the air filters measured in 2007 would correspond to an activity concentration from < 0.5 to 50(± 2) μBq m^{-3}. It can be assumed that the activity concentration of ^{241}Am in the air compared to alpha-emitting plutonium isotopes is nowadays higher than in these old air filters because of later ^{241}Pu emissions from different sources after 1963 and its decay to ^{241}Am. Due to the atmospheric nuclear tests, the Chernobyl accident and releases from nuclear fuel reprocessing plants discharging ^{241}Pu and ^{241}Am, the inventory of ^{241}Am will increase in the environment in forthcoming decades.

Conclusions

All plutonium isotopes had a spring maximum in the air of Sodankylä in 1963 due to seasonally-enhanced atmospheric transport from the stratosphere to the troposphere. Contrary to earlier assumptions, activity concentrations of $^{239+240}$Pu in the air were practically at the same level in northern and southern Finland in 1963. According to ^{238}Pu/$^{239+240}$Pu and ^{241}Pu/$^{239+240}$Pu ratios,

Table 3. Parallel results for activity concentration of ^{241}Pu in 1963 determined by two independent methods.

Sample	^{241}Pu by LSC (μBq m^{-3})	^{241}Pu by alpha spectometry via ^{241}Am (μBq m^{-3})
22 Jan.	< MDA	120 ± 10
5 Mar.	270 ± 20	210 ± 20
22 Apr.	870 ± 70	840 ± 50
11 May	1410 ± 120	1490 ± 60
5 June	1430 ± 120	720 ± 40
7 July	540 ± 50	470 ± 20
6 Aug.	650 ± 60	760 ± 30
8 Sep.	1040 ± 90	350 ± 20
23 Oct.	< MDA	160 ± 10
13 Dec.	320 ± 30	–

plutonium in Sodankylä originated mainly from the global nuclear test fallout even though the effect of the Soviet tests at the end of 1962 could be observed at the beginning of 1963. However, direct atmospheric transport from Novaya Zemlya to Finnish Lapland could not be detected. Determination of $^{240}Pu/^{239}Pu$ ratios with ICP-MS as well as air mass trajectory analysis will in the future shed more light on the origin of plutonium in the air in Finnish Lapland during the maximum fallout period in 1963.

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