Dear Editor, April 6th 2020

Referring to Your request to check the manuscript once more by a native English speaker, the text has been processed accordingly. Please see the grammatical corrections as “track changes”. We hope that the revision is now adequate considering the acceptance of the manuscript.

Best regards,

Susanna Salminen-Paatero & co-workers

Measurements and modeling of airborne plutonium in Subarctic Finland between 1965 and 2011

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ABSTRACT

The activity concentrations of ^{238,239,240}Pu and ^{241}Am (for determining its mother nuclide ^{241}Pu) as well as activity ratios ^{238}Pu/$^{239+240}$Pu, ^{241}Pu/$^{239+240}$Pu, $^{239+240}$Pu/^{137}Cs, and mass ratio ^{240}Pu/^{239}Pu were determined from air filter samples collected in Rovaniemi (Finnish Lapland) in 1965–2011. The origin of plutonium in surface air was assessed based on this data from long time series. The most important Pu sources in the surface air of Rovaniemi were atmospheric nuclear weapons testing in the 1950's and 1960's, later nuclear tests ~~on~~ in 1973–1980, and the SNAP-9A satellite accident in 1964, whereas the influence from the 1986 Chernobyl accident was only minor. Contrary to the alpha emitting Pu isotopes, ^{241}Pu from the Fukushima accident in 2011 was detected in Rovaniemi. Dispersion modeling results with the Silam model indicate that Pu contamination in northern Finland due to hypothetical reactor accidents would be negligible in the case of a floating reactor ~~at~~ in the Shtokmann natural gas field and relatively low in the case of an intended nuclear power plant in western Finland.

Key words: Plutonium, isotope ratio, Chernobyl, nuclear weapons testing, nuclear accident, dispersion modeling

1. INTRODUCTION

The distribution of anthropogenic radionuclides in global fallout from nuclear weapons testing is uneven, and even more inhomogeneous is their distribution in regional and local fallout from different sources. It is known that Subarctic and Arctic regions have received radionuclide deposition with ~~different~~ radioactivity levels and composition ~~than~~ different to the more temperate areas of the Earth. Subarctic and Arctic ecosystems have a special combination of harsh climate, often sparse vegetation, lack of nutrients and, in the case of humans, dependence on traditional ~~lines of business~~ livelihoods and life ~~-~~styles like hunting, fishing, reindeer herding, and ~~collection~~ gathering ~~of~~ mushrooms and berries. Consequently, these Nordic ecosystems are highly vulnerable to toxic agents, including radionuclides. Still, there are only a few contiguous long-term radioactivity data series from Subarctic and Arctic areas where the changes in concentration levels and isotope ratios ~~could~~ can be followed and ~~different~~ nuclear events identified as contamination sources in a particular environment.

In total, radionuclides ^{137}Cs, ^{90}Sr, total beta activity, ^{238,239,240}Pu and ^{241}Am were determined from the air filter samples that were collected in Rovaniemi (Finnish Lapland) in 1965~~-~~–2011. ^{241}Am (t½ 432.2 a) was analyzed for calculating the activity concentration of its mother nuclide, the relatively short-lived beta emitter ^{241}Pu (t½ 14.35 a). The major part of ^{241}Am in the samples originates from the decay of ^{241}Pu after the sampling and only a minor part of ^{241}Am originates directly from nuclear events. The results for ^{137}Cs, ^{90}Sr, and total beta activity have been reported elsewhere (Salminen-Paatero et al. 2019). The activity ratio ^{238}Pu/$^{239+240}$Pu and the mass ratio ^{240}Pu/^{239}Pu in Rovaniemi have been presented pictorially with other global ratio values in ~~the~~ an article by Thakur et al. (2017), which did not, however, discuss ~~but~~ the ratio values of Rovaniemi ~~were not discussed~~ in detail ~~there~~.

In this ~~work~~study, radionuclide concentration and isotope ratio data from 1965~~-~~–2011 has been used for estimating nuclear contamination sources in the surface air of the Finnish Subarctic ~~during~~ over almost five decades. ~~The only~~ Few long time series of atmospheric radioactivity exist ~~from~~ in Subarctic and Arctic regions, especially of Pu isotopes, and even ~~less~~ fewer data ~~has~~ have been published about atmospheric transuranium concentrations in these high northern latitudes after the Chernobyl and Fukushima accidents. Furthermore, the atmospheric dispersion of one real and one hypothetical nuclear events has been modeled for ~~finding out~~ establishing the potential transport of Pu isotopes and effect of these nuclear events on atmospheric radioactivity levels in Finnish Lapland. Atmospheric dispersion modeling completed the experimental data by providing risk estimates and

reference values for ~~the~~ future accidental releases of nuclear material in and close to Arctic regions, as well as ~~it pointed out~~ indicating the importance of the accurate source term in calculating the amount of ~~released~~ radioactivity released into the atmosphere ~~with~~ after~~the~~ Fukushima ~~case~~.

2. EXPERIMENTAL

2.1 Sampling and procedures for the air filters before any chemical treatment

The air filter samples were collected at the Finnish Meteorological Institute's ~~[~~(FMI)~~]~~ Rovaniemi monitoring station, 66°34'~~'~~N, 25°50'~~'~~E, elevation 198 m above sea level ~~[~~(a.s.l.~~]~~. ~~).~~ The weekly volume of sampled air ~~volume~~ was ~1,000 m^3. First, total beta activity was measured from the filters five days after the end of sampling. Then the filters were combined into suitable sets for the gamma measurement and determination of ^{137}Cs concentration. The details of air sampling, combining air filters and measurements for the gamma activity of ^{137}Cs and total beta activity have been given ~~in~~ by Salminen et al. (2019).

2.2 Radiochemical separation of Pu, Am, and Sr from air filters

~~The~~ A detailed description ~~about~~ of the radioanalytical separation procedure and the radionuclide measurements is given elsewhere (Salminen-Paatero and Paatero, submitted to MethodsX). ^{238,239,240}Pu, ^{241}Am, and ^{90}Sr were separated from dissolved air filter sample sets containing filters from three months to five years. The separation method included extraction chromatography and anion exchange steps and it was modified from the original method designed for the air filters of 1~~–~~3 days' sampling time~~,~~ presented in Salminen and Paatero (2009). The radiochemical separations were performed in 2013~~–~~2014, i.e. two~~–~~three years after the last air filter sample set of 2011 was taken.

2.3 Measurement of ^{238,239,240}Pu, ^{241}Am, ^{90}Sr, and ^{240}Pu/^{239}Pu in the air filter samples

The activity concentration of ~~alpha~~ alpha-emitting Pu isotopes ^{238}Pu and $^{239+240}$Pu in the air filter samples was determined from the separated Pu fractions by Alpha Analyst spectrometer (Canberra).

From the separated Am fractions the activity concentration of ^{241}Am was also measured by alpha spectrometry~~, for calculating~~ to calculate the activity concentration of its mother nuclide, beta emitter ^{241}Pu in each air filter sample set ~~of the period~~ from 1965~~–~~2011 for the time of sampling. The alpha measurements were performed soon after the radiochemical separations in 2013-2014.

The activity concentration of ^{90}Sr was measured by Quantulus 1220 liquid scintillation counter (LSC) via the activity concentration of the daughter nuclide ^{90}Y. Finally, after an additional purification step of the Pu alpha counting samples, the mass ratio ^{240}Pu/^{239}Pu was determined by SF-ICP-MS (Sector-Focusing Inductively Couple Plasma-Mass Spectrometry), ELEMENT XR (Thermo Scientific). A more detailed description of the measurements is given in Salminen-Paatero and Paatero (MethodsX, in review).

3. RESULTS AND DISCUSSION

3.1 ~~The~~ Activity concentrations of ^{238}Pu, $^{239+240}$Pu, and ^{241}Pu in the surface air of Rovaniemi, ~~in~~ 1965~~–~~2011

3.1.1 The activity concentration of ^{238}Pu

In the period studied, 1965–2011, the activity concentration of ^{238}Pu had the highest value of 259±13 nBq m^{-3} in 1968 ~~during the investigated time period 1965-2011~~ (Table 1, Fig. 1). The years of the highest concentrations of ^{238}Pu around 1968 are a consequence ~~from~~ of the destruction of the SNAP-9A satellite nuclear power unit ~~of the satellite~~ re-entering the atmosphere in 1964. Since 1968, the activity concentration of ^{238}Pu in the surface air of Rovaniemi has been decreasing and is now ~~being nowadays~~ below or close to the detection limit. The concentration of ^{238}Pu was also under the detection limit ~~also during~~in the months after the Chernobyl accident, ~~in~~ April~~–~~December ~~in~~ 1986.

3.1.2 The activity concentration of $^{239+240}$Pu

The activity concentration of $^{239+240}$Pu in the surface air of Rovaniemi has been dropping from the highest value 2,270±40 nBq m^{-3} (in 1965) and has been ~~, being~~ a few nBq m^{-3} since 1996 (Table 1, Fig. 1). Two years before the sampling was started, in 1963, ~~was~~ saw the deposition maximum from

atmospheric nuclear tests ~~that were~~ performed before the Partial Test Ban Treaty. For example, at Sodankylä, Finnish Lapland, 120 km north of Rovaniemi, the average $^{239+240}$Pu activity concentration was ~~17~~ 17,000 nBq m^{-3} in 1963 (Salminen & Paatero 2009). Slight peaks in $^{239+240}$Pu concentration can be seen in 1974, 1978 and 1981, evidently due to the atmospheric nuclear tests performed by the People's Republic of China between 1973 and 1980. The effect of these nuclear tests on the radionuclide concentration level in Finnish Lapland has been already observed in the concentration variation of ^{137}Cs (Salminen-Paatero et al. 2019). ~~Like~~ As with ^{238}Pu, the concentration of $^{239+240}$Pu was below the detection limit on April-June 1986 following the Chernobyl accident. For comparison, the concentration of $^{239+240}$Pu was 32 µBq m^{-3} in the surface air in Nurmijärvi (southern Finland)~~, in~~ on 28 April~~,~~ 1986 (Jaakkola et al. 1986).

Based on the extremely low activity concentrations of both ^{238}Pu and $^{239+240}$Pu in the surface air of Rovaniemi ~~during~~ in April~~–~~–December 1986, hardly any plutonium ~~was~~ migrated to Finnish Lapland from the destroyed Chernobyl nuclear reactor after 26th April~~,~~ 1986. This conclusion is ~~also~~ supported by the high concentration of ^{137}Cs (1294±7 µBq m^{-3}) and the low concentration of ^{90}Sr (5.2±1.1 µBq m^{-3}) in the same air filter samples in April~~–~~–June 1986 (Salminen-Paatero et al. 2019). It has been suggested that the initial contamination plume from the destroyed Chernobyl reactor contained intermediate (^{90}Sr) and refractory elements (Pu isotopes) and that the plume passed over central and southern ~~parts of~~ Finland, while the volatile elements ~~like~~ such as ^{137}Cs were mostly in the later contamination plumes which also reached ~~also~~ Lapland (Saxén et al. 1987). However, the observations of ^{241}Pu/$^{239+240}$Pu activity ratio discussed in a later paragraph show some possibility of Chernobyl-derived plutonium in Finnish Lapland.

3.1.3 The activity concentration of ^{241}Pu

The concentration of ^{241}Pu was calculated via ingrowth of ^{241}Am, and ~~like~~ as with $^{239+240}$Pu, the activity concentration of ^{241}Pu ~~had the~~ reached its highest value in 1965, ~~38~~ 38,198±711 nBq m^{-3}, ~~and~~ since ~~then~~ which its concentration has been decreasing, except for small peaks in 1974, 1978, and 1981 (Table 1, Fig. 2). ~~Similarly with~~ In a similar manner to the activity concentration changes of $^{239+240}$Pu, these peaks in the activity concentration of ^{241}Pu are presumably caused by nuclear tests ~~executed~~ in the People's Republic of China. The atmospheric activity concentration of ^{241}Pu was ~~lower than~~ below the detection limit in April~~–~~–June 1986, and since July~~–~~–December 1986, the amount of ^{241}Pu was returned ~~again~~ to the same pre-Chernobyl level ~~as it was before the Chernobyl accident~~ in the surface air of Rovaniemi. Based on the ^{241}Pu concentration ~~only~~alone, there is no evidence ~~about~~ of any Chernobyl-derived ^{241}Pu in Rovaniemi.

Interestingly, Anthe increase in the activity concentration of ^{241}Pu is seen in 2011, unlike with ^{238,239,240}Pu. The activity concentration of ^{241}Pu in 2011, 602±131 nBq m^{-3}, is higher than above the concentration level in Rovaniemi during the last decades before 2011, and it is probably due to the Fukushima accident in of 11th March 2011. The activity of ^{241}Pu has been reported being as much higher than the activity of $^{239+240}$Pu in the emissions from the destroyed Fukushima NPP, with the activity ratio ^{241}Pu/$^{239+240}$Pu having a value of 108 in soil and litter samples (Zheng et al. 2012). The activity concentrations of Pu isotopes were 25 25,000 nBq m^{-3} for ^{241}Pu, 130 nBq m^{-3} for ^{239}Pu and 150 nBq m^{-3} for ^{240}Pu in the air filters sampled at 120 km from Fukushima on 15th March, 2011 (Shinonaga et al. 2014).

It is unfortunate that there is The existence of but only one combined air filter sample of Rovaniemi for the year 2011 is unfortunate:, because the annual concentration is an average of the weekly concentrations in 2011, and now the signal from the Fukushima accident has been diluted under the excess effect of global fallout in the air filters. It would have been interesting to analyze Analysis of plutonium isotopes in weekly filters separately from March 2011, to for determincing Fukushima-derived ^{241}Pu concentration and isotope ratios in Finnish Lapland, would have been of interest.

3.2 The activity ratios ^{238}Pu/$^{239+240}$Pu, ^{241}Pu/$^{239+240}$Pu, $^{239+240}$Pu/^{137}Cs, total beta activity/$^{239+240}$Pu, and mass ratio ^{240}Pu/^{239}Pu in the air filters

3.2.1 ^{238}Pu/$^{239+240}$Pu activity ratio

The activity ratio ^{238}Pu/$^{239+240}$Pu was 0.022±0.003-0.444±0.023 in Rovaniemi in 1965–2011, the with values under below the detection limit excluded (Table 2, Fig. 3). The variation in the activity ratio values is 200-fold. The activity ratio ^{238}Pu/$^{239+240}$Pu in the surface air can vary greatly even in a short time, e.g.for example due to stratospheric-tropospheric exchange, resuspension and introduction of several contamination sources. For example, the activity ratio ^{238}Pu/$^{239+240}$Pu varied from 0.014±0.003 to 0.32±0.11 in Sodankylä, Finnish Lapland, during in one year in 1963 alone;, still, the most typical value was ~0.03, which that represents the activity ratio for the global fallout (Salminen and Paatero 2009). The ratio started to increase in 1966 in Rovaniemi, reaching a maximum in 1967 due to the previously aforementioned SNAP-9A satellite accident in 1964. Previously, an increased ^{238}Pu/$^{239+240}$Pu activity ratio due to the SNAP-9A accident has been found in lichens both in Subarctic Finland (Jaakkola et al. 1978) and Sweden (Holm and Persson 1975) a couple of years after 1964.

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This ~~over two year~~ delay of over two years after the accident indicates ~~how~~ the slowness of the interhemispheric transport of stratospheric radionuclides ~~is~~ (Fabian et al. 1968).

The activity ratio $^{238}Pu/^{239+240}Pu$ cannot be determined for the period immediately after the Chernobyl accident because the activity concentrations of ^{238}Pu and $^{239+240}Pu$ were below the detection limit ~~during~~ in April ~~–~~ December 1986. This finding is in agreement with the previous assumptions about ~~hardly~~ barely any Chernobyl-derived refractory elements in Finnish Lapland (Salminen-Paatero et al. 2019). ~~Due~~ Because ~~to~~ the activity concentrations of ^{238}Pu and $^{239+240}Pu$ ~~being~~ were below the detection limit, the activity ratio $^{238}Pu/^{239+240}Pu$ cannot be determined for the year of the Fukushima accident, 2011, either. For comparison, both ^{238}Pu and $^{239+240}Pu$ were detected in Lithuania, ~~–~~ 1,300 km south ~~from~~ of Rovaniemi, soon after the Fukushima accident (Lujanienė et al. 2012). The combined air filter sample set in the Lithuanian study contained the sampled air volume of ~2 x 10^6 m^3 ~~during~~ from 23 March ~~to 23~~ – 15 April ~~15~~ 2011, the activity concentration of $^{239+240}Pu$ being 44.5±2.5 nBq m^{-3}, and the activity concentration of ^{238}Pu being 1.2 times higher than of $^{239+240}Pu$. The resulting activity ratio $^{238}Pu/^{239+240}Pu$ in Lithuania was 1.2, clearly deviating from the activity ratio values in the Chernobyl fallout and global fallout from nuclear weapons testing.

3.2.2 $^{241}Pu/^{239+240}Pu$ activity ratio

The activity ratio $^{241}Pu/^{239+240}Pu$ varied between 8.2±0.7 and 79±17 in the surface air of Rovaniemi in 1965 – 2011, except April – December 1986 and 2011, when the concentration of one or both isotopes (either $^{239+240}Pu$ or ^{241}Pu) was ~~under~~ below the detection limit (Table 2, Fig. 4). These two periods following the accidents of Chernobyl and Fukushima would have interesting $^{241}Pu/^{239+240}Pu$ activity ratio values for determining the Pu contamination source in Rovaniemi. Unfortunately, the concentration of $^{239+240}Pu$ in the surface air of Finnish Lapland was extremely low during those periods.

The $^{241}Pu/^{239+240}Pu$ activity ratio values of Rovaniemi were mainly due to atmospheric nuclear weapons testing in 1965 – March 1986 and ~~for the years~~ 1987 – 2005. The, ~~an~~ influence ~~from~~ of the Chernobyl accident can be seen as elevated ratio values. The $^{241}Pu/^{239+240}Pu$ activity ratio was determined to be 15 in fresh nuclear fallout in 1963 – 1972 (Perkins and Thomas 1980) and the corresponding ratio values in the fallout from the Chernobyl accident have been 85 in Sweden and Poland (Holm et al. 1992; Mietelski et al. 1999), and 95 in Finland (Paatero et al. 1994). The published $^{241}Pu/^{239+240}Pu$ activity ratio values for the Fukushima-derived contamination are also high, e.g. 89 in

air filters (calculated from the individual isotope concentrations in Shinonaga et al. (2014)), and 108 in soil and litter samples (Zheng et al. 2012).

3.2.3 $^{240}Pu/^{239}Pu$ mass ratio

The mass ratio $^{240}Pu/^{239}Pu$ was 0.117±0.009-0.278±0.093 in 1965-2011 (Table 2, Fig. 5) and the majority ~~part of the~~ ratio values corresponds to the value ~0.18 for global fallout from atmospheric nuclear weapons testing in the northern hemisphere (Beasley et al. 1998), taking into account the relative measurement uncertainties. The highest mass ratio value occurred in April–June 1986, while the activity concentrations of ^{238}Pu, $^{239+240}Pu$ and ^{241}Pu were under detection limit by alpha spectrometry. Therefore, it was possible to determine ^{239}Pu and ^{240}Pu by mass spectrometry even from the samples with very low Pu-concentration (April–December 1986, 2011, etc.) although the relative measurement uncertainties ~~by~~ of ICP-MS are much higher for these samples with very low Pu-concentration ~~compared with~~ than the measurement uncertainties of samples with a higher Pu-concentration level.

The mass ratio $^{240}Pu/^{239}Pu$ is higher in the emissions from the destroyed Chernobyl reactor than~~,~~ ~~compared with~~ the global fallout value. For example, ~~the~~ a mass ratio value 0.408±0.003 has been determined from ~~the~~ samples of the Chernobyl-contaminated soil layer (Muramatsu et al. 2000)~~,~~ and two hot particles that migrated to Finland from Chernobyl had the mass ratios 0.33±0.07 and 0.53±0.03 (Salminen-Paatero et al. 2012). The air filters sampled in Rovaniemi in April-June and July-December 1986 seem to have elevated mass ratios, 0.278±0.093 and 0.254±0.073 respectively, but with consideration of ~~taking into account~~ their high measurement uncertainties, these post-Chernobyl ratio values might be close to the global fallout ratio 0.18 after all.

~~Similarly~~ In a similar manner to ~~with~~ the refractory element emissions from the Chernobyl accident, the ~~released~~ fuel particles released from the Fukushima accident have significantly higher mass ratio $^{240}Pu/^{239}Pu$ than the global fallout value 0.18. Dunne et al. (2018) have compared the mass ratios $^{240}Pu/^{239}Pu$ in soil, sediment and vegetation samples collected at the surroundings of Fukushima with the known mass ratios in global fallout and in the destroyed nuclear reactors of Fukushima NPP. The mass ratio $^{240}Pu/^{239}Pu$ for the Fukushima reactor units was obtained ~~by~~ using ORIGEN code, being 0.344 for Reactor 1, 0.320 for Reactor 2, and 0.356 for ~~the~~ Reactor 3, respectively (Nishihara et al. 2012). All investigated environmental samples from the proximity of Fukushima had ~~the~~ $^{240}Pu/^{239}Pu$ atom ratios between the global fallout value and the value for the

Reactor Unit 3 calculated by ORIGEN, with the exception of one deviating value (Dunne et al. 2018).

~~It was highlighted in~~ The same study highlighted that the concentration level of Pu isotopes and the mass ratio $^{240}Pu/^{239}Pu$ varies greatly in the environment of Fukushima, and that they do ~~n't~~ not necessarily correlate with each other. ~~Also~~ The lowest mass ratio values in Fukushima have also been at the global fallout level. ~~This variety of isotope concentrations and isotope ratios has been noticed in other~~ Other Fukushima-related investigations have also noted this variety of isotope concentrations and isotope ratios~~as well~~. ~~From~~ In a litter and soil sample set collected ~~at~~ 20–32 km from Fukushima, three samples had high ^{241}Pu concentrations and mass ratios 0.303-0.330 that can be considered as representing contamination from the destroyed reactors of Fukushima (Zheng et al. 2012). The rest of the soil and litter samples from the proximity of Fukushima in (Zheng et al. 2012) had low ^{241}Pu concentrations and the $^{240}Pu/^{239}Pu$ mass ratios were at the northern hemisphere global fallout level. In another study, the air filter samples collected at 120 km from Fukushima formed two groups: one having low ^{239}Pu concentration and fairly similar mass ratio to global fallout (0.141±0.002) and another having high ^{239}Pu concentration and mass ratio clearly deviating from global fallout (≥ 0.3) (Shinonaga et al. 2014).

The $^{240}Pu/^{239}Pu$ mass ratio was only 0.145±0.091 in the surface air of Rovaniemi ~~during~~ in the year of the Fukushima accident, 2011. Again, the activity concentrations of both ^{239}Pu and ^{240}Pu were extremely low in Rovaniemi ~~during~~ in that year and the uncertainty of the mass ratio is therefore high, suggesting that the ratio value in 2011 is probably due to global fallout ~~though~~.

3.2.4 $^{239+240}Pu/^{137}Cs$ activity ratio

The activity ratio $^{239+240}Pu/^{137}Cs$ varied between 0.0005±0.0001 and 0.0393±0.0038 in the surface air of Rovaniemi in 1965–2011, excluding the samples of April–December 1986 and 2011, when the concentration of $^{239+240}Pu$ fell below the detection limit (Table 2). The lowest value for the activity ratio occurred in 2006–2010, when the activity concentration of both radionuclides ($^{239+240}Pu$ and ^{137}Cs) in the surface air ~~has~~ had been constantly decreasing ~~in the surface air~~ for decades. The range of the values in Rovaniemi is in agreement with the previous studies of surface air in Finland. The activity ratio $^{239+240}Pu/^{137}Cs$ was 0.0020±0.0008–0.029±0.010 in Sodankylä ~~(Finnish Lapland) during~~ in 1963 (Salminen-Paatero and Paatero 2012) and 0.005±0.002–0.012±0.004 (range of annual mean values) in Helsinki (southern Finland) in 1962–1977 (Jaakkola et al. (1979).

Bossew et al. (2007) have calculated the reference values for $^{239+240}Pu/^{137}Cs$ activity ratio in global fallout and the Chernobyl accident, obtaining 0.0180±0.0024 (data from Bunzl and Kracke, 1988) and 6.6 x 10^{-6} (data from Irlweck and Khademi, 1993), respectively. The values ~~of~~ for Rovaniemi are higher than ~~the value~~ those for Chernobyl contamination, and some values ~~of~~ for Rovaniemi are even higher than the value for global fallout.

~~On the contrary to~~ In contrast with high $^{239+240}Pu/^{137}Cs$ ratio values in the surface air of Rovaniemi and in global fallout, very low $^{239+240}Pu/^{137}Cs$ activity ratios have been observed in the Fukushima environment. Among all litter and soil samples of Fukushima in the study by Zheng et al. (2012), the three samples that represent the Fukushima-derived contamination, i.e., have both ~~the~~ high ^{241}Pu concentration and ~~the~~ high $^{240}Pu/^{239}Pu$ mass ratio, had the $^{137}Cs/^{239+240}Pu$ activity ratios 4 x 10^{-8}, 2 x 10^{-7}, and 5 x 10^{-6} in 2011.

3.2.5 Total beta activity/$^{239+240}Pu$ activity ratio

The ratio between total beta activity (Salminen-Paatero et al. 2019) and $^{239+240}Pu$ remains rather constant during the atmospheric nuclear testing era (Fig. 6). The ratio reflects the produced nuclide composition after fission and activation reactions in the detonating devices. Following the Chernobyl accident, the ratio increases by almost three orders of magnitude. After the initial explosion plume, the emissions from the burning reactor were dominated by volatile fission products, which explains the high total beta activity/$^{239+240}Pu$ activity ratio. After the decay of short-lived fission products, the ratio soon returns ~~close~~ to near the pre-Chernobyl level. Towards the end of the 20th century, the ratio starts to gradually increase~~ing~~. This is explained by the decreasing amount of plutonium in the atmosphere, while the total beta activity remains on a constant level due to natural atmospheric radioactivity, mainly ^{210}Pb.

3.3 Effect of actual and hypothetic nuclear detonations on the surface air of the Finnish subarctic

At least two new nuclear facilities in or close to the European Arctic region are under preparation. ~~A~~ Construction of infrastructure for a new nuclear power plant at Pyhäjoki, western Finland, has ~~been started~~begun. The Shtokmann natural gas field is located in the Barents Sea ~~about~~ between northern Finland and Novaya Zemlya. The plans indicate that future gas extraction production facility will be

powered by a floating nuclear power plant ~~in the plans~~. ~~In case of hypothetical accidents in these halfway plants,~~ The atmospheric dispersion of plutonium contamination in the event of accidents in these future plants was assessed with atmospheric transport modeling. In this study, ADM (atmospheric dispersion modeling) provided risk estimates and reference contamination levels related to future nuclear activities in and close to Arctic regions, which can be compared with earlier actual releases.

^{241}Pu dispersion in the atmosphere was simulated with the SILAM model (Sofiev et al., 2006; 2008). The model runs were based on the meteorological forecast data of the European Centre for Medium-Range Weather Forecasts (www.ecmwf.int) with a horizontal resolution of 0.25 degrees and with 9 vertical levels up to ~~the~~ a height of 7,700 m. Transport and dispersion calculations for both sites were made for each day in the year 2010. The average activity concentrations of ^{241}Pu in the surface air during the first 48 hours after the release were calculated.

The following accident conditions, previously listed in Paatero et al. (2014), for the Pyhäjoki ~~power~~ reactor, 64°32'N, 24°15'E, were used:

- a pressurized water reactor with ~~a~~ thermal power of 4,000 MW,
- the end of the refueling interval,
- an immediate release after shutdown with an effective release height of 200 m above sea level, and
- a ^{241}Pu inventory of 6.2x10^{17} Bq, release fraction of 0.1%, and a release of 6.2x10^{14} Bq.

The following accident conditions for the case of the Shtokmann gas field, ~~the~~ Barents Sea, 73°N, 44°E, were used (previously used by Paatero et al. 2014):

- an ice breaker reactor with a fuel burnup of 466,000 MWdays T^{-1} HM,
- an immediate release two hours after shutdown,
- a radionuclide inventory according to Reistad and Ølgaard (2006),
- an effective release height of 100 m above sea level, and
- a ^{241}Pu inventory of 3.2x10^{14} Bq, release fraction of 0.2%, and a release of 6.4x10^{11} Bq.

Varying meteorological situations have a decisive effect on ~~the~~ atmospheric plutonium transport following accidental emissions from a nuclear reactor. The wind direction determines the path of the emission plume. The wind speed sets how quickly the emission plume is advected. However, the wind speed also affects the turbulence that disperses the plume vertically and horizontally. This influences the plutonium concentrations in the air. Precipitation, for ~~one's~~ its part, efficiently ~~scavenges~~ brings plutonium-bearing particles from the atmosphere to the surface, which affects the deposition of plutonium and furthermore its transfer to food webs.

From the Rovaniemi region ~~point of view~~perspective, the worst of the calculated 365 dispersion cases would have caused ~~in ground-level air~~ an average ^{241}Pu activity concentration of less than 1 kBq m^{-3} in ground-level air ~~during~~ in the first 48 hours after the release (Fig. 7). This equals an annual average ^{241}Pu exposure of 5 Bq m^{-3}. For comparison, ~~due to~~ the atmospheric nuclear tests caused the ^{241}Pu activity concentration to vary ~~varied~~ between a few dozen~~s~~ and some 1,700 µBq m^{-3} in 1963 in northern Finland, or in other words ~~on~~ several orders of magnitude lower ~~level~~ (Salminen and Paatero 2009). In practice, the human exposure to ^{241}Pu *via* inhalation would remain on a clearly lower level because the civil ~~defence~~defense authorities would order the population to stay indoors with ~~the~~ ventilation systems ~~closed~~ turned off and doors and windows sealed.

Compared with the Pyhäjoki accident scenario, the consequences after a hypothetical accident in a floating nuclear reactor in the Barents Sea would ~~remain~~ be much less significant from the northern Finnish ~~point of view~~perspective. This ~~is~~ would be due to ~~the~~ smaller emissions, greater distance and favorable climatic conditions, namely prevailing ~~wind directions from the~~ westerly and south-westerly winds. Only dispersion calculation ~~one case out~~ of 365 ~~dispersion calculations~~ produced an atmospheric transport pattern that reached the northernmost part of Finland (Fig. 8). The ground-level ^{241}Pu activity concentrations would have been less than 0.01 Bq m^{-3} ~~during~~ in the first 48 hours, corresponding to an annual average concentration of 55 µBq m^{-3}. This is similar to the activity concentrations occurring in the early 1960s.

3.4 Case "Fukushima 2011 and ^{241}Pu"

~~In~~ An earlier work by Paatero et al. (2012), ~~it was~~ observed that the Silam model simulates the temporal behavior of the Fukushima emission plume in the High Arctic well. The calculated activity concentration levels, however, were an order of magnitude lower than the observed ones. This

deviation was attributed to ~~the~~ inaccuracies in the source term. From the same model dataset, the ^{137}Cs activity concentration in the surface of Rovaniemi was extracted. The level of these values was then corrected by adjusting them to the observed weekly ^{137}Cs activity concentration of 170 µBq m^{-3} between 28 March and 4 April 2011 (Salminen-Paatero et al. 2019). From these values, the ^{241}Pu activity concentrations were obtained by multiplying with the ^{241}Pu/^{137}Cs activity ratio of 7.81x10^{-6}. This activity ratio was found in hot particles close to the Fukushima Daiichi NPP by Igarashi et al. (2019). The calculated hourly ^{241}Pu activity concentration reaches a maximum level of 0.01 µBq m^{-3} for two short periods (Fig. 9). The calculated peak activity concentrations are very low, six orders of magnitude lower, ~~compared with~~ than daily ^{241}Pu activity concentrations observed in northern Finland in 1963 (Salminen and Paatero 2009). However, there is a discrepancy between this assessment and the annual observed ^{241}Pu activity concentration of 0.6 µBq m^{-3} (Fig. 2). If we assume that the background ^{241}Pu activity concentration due to the atmospheric nuclear tests and the Chernobyl accident ~~would be~~ were 0.03 µBq m^{-3}, then the average activity concentration should be 9.3 µBq m^{-3} between 27 March and 17 April~~;~~ in other words, a thousand times higher. An obvious explanation is that the ^{241}Pu/^{137}Cs activity ratio we used (7.81x10^{-6}) ~~we used~~ is not valid. The value may not be representative ~~to~~ of the bulk emission mixture of the destroyed reactors. Zheng et al. (2012) found out that the ^{137}Cs/^{239,240}Pu activity ratio in environmental samples varied over four orders of magnitude. In addition, the hot particles were found close to the source, and fractionation processes were possible during the atmospheric transport of over 10,000 km ~~long atmospheric transport could occur too~~.

4. CONCLUSIONS

Based on the activity concentrations of ^{238,239,240,241}Pu, hardly any refractory elements from the exploded Chernobyl reactor reached Finnish Lapland in 1986. Previously Chernobyl-derived ^{137}Cs, a more volatile isotope, has been detected from the same air filter samples, whereas there was no increased concentration of ^{90}Sr in the samples after March 1986. The influence from the Fukushima Daiichi accident is seen as ~~the~~ increased concentration of ^{241}Pu in the air filters. Nuclear weapons testing in the 1950's and 1960's, later nuclear tests on 1973–1980, SNAP 9A-satellite accident in

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1964, and the Fukushima accident in 2011 have been the main sources of Pu in the surface air in Finnish Lapland during 1965-2011.

Overall, the mass ratio $^{240}Pu/^{239}Pu$ is a more sensitive contamination source indicator than the activity ratios $^{238}Pu/^{239+240}Pu$ or $^{241}Pu/^{239+240}Pu$ ~~due to~~ because of the lower detection limit of ICP-MS, compared with alpha spectrometry and LSC. However, it is always useful to analyze more than one isotope ratio or activity ratio, and single isotope concentrations when characterizing the origin of Pu contamination. In this case, the contribution of the Fukushima accident in Rovaniemi would not have been observed without analyzing the concentration of ^{241}Pu in the air filter samples.

Dispersion modeling results with the atmospheric dispersion model Silam indicate that Pu contamination in northern Finland would be negligible due to a hypothetical accident in a floating nuclear reactor at the Shtokmann natural gas field in~~,~~ the Barents Sea. The Pu contamination risk would be higher in ~~case~~ the event of a severe accident at the intended nuclear power plant at Pyhäjoki, western Finland, due to the ~~bigger~~ larger, closer reactor ~~and shorter distance~~. The modeling of the Fukushima case demonstrated ~~how important is~~ the importance of ~~the~~ accurate source term data for predicting the activity concentrations of the radionuclides in the air following an atmospheric release of radioactivity.

ACKNOWLEDGEMENTS

We acknowledge Emil Pesonen's help in ~~is acknowledged for help with~~ cutting the air filter samples before ashing, and Ilia Rodushkin's (ALS Scandinavia Luleå laboratory) help in ~~for~~ measuring the Pu samples with ICP-MS. This work belongs to "Collaboration Network on EuroArctic Environmental Radiation Protection and Research (CEEPRA)". The project was funded by EU Kolarctic ENPI CBC 2007-2013 program~~me that was~~ managed by the Regional Council of Lapland. The authors ~~want~~ would like to thank the EU~~-~~ project ''TOXI Triage'' (Project ~~id.~~ID 653409) for additional support.

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Table captions

1. The atmospheric activity concentrations of ^{238}Pu, $^{239+240}$Pu and ^{241}Pu in Rovaniemi, Finnish Lapland. The activity values have been decay-corrected to the middle point of the sampling period.

2. The activity ratios ^{238}Pu/$^{239+240}$Pu, ^{241}Pu/$^{239+240}$Pu, $^{239+240}$Pu/^{137}Cs, and the mass ratio ^{240}Pu/^{239}Pu in the air filters collected in Rovaniemi. The uncertainty is 1 sigma error for the activity ratios and 2 sigma error for the mass ratio. – means that one or both isotopes had concentration below the detection limit.

Figure captions

1. The activity concentration of ^{238}Pu (thin line, nBq m^{-3}) and $^{239+240}$Pu (thick line, nBq m^{-3}) in the surface air of Rovaniemi in 1965-2011. Values below the detection limit have been depicted as half the MDA value (Table 1). The black circles indicate the times of atmospheric nuclear tests (UNSCEAR 2000).

2. The activity concentration of ^{241}Pu (nBq m^{-3}) in the surface air of Rovaniemi (thick line 1965-2011 left vertical scale, thin line 1982-2011, right vertical scale). Values below the detection limit have been depicted as half the MDA value (Table 1).

3. The activity ratio ^{238}Pu/$^{239+240}$Pu in the surface air of Rovaniemi as a function of time.

4. The activity ratio ^{241}Pu/$^{239+240}$Pu ~~the~~ in the surface air of Rovaniemi as a function of time.

5. The mass ratio ^{240}Pu/^{239}Pu in the surface air of Rovaniemi as a function of time.

6. The ratio of total beta activity (Salminen-Paatero et al. 2019) and $^{239+240}$Pu activity content in the surface air in Rovaniemi in 1965-2011. $^{239+240}$Pu values below the detection limit have been replaced with half the MDA values (Table 1).

7. The average activity concentration of ^{241}Pu in the surface air during the first 48 hours after a hypothetical reactor accident at Pyhäjoki, assumed release 20 January 2010.

8. The average activity concentration of ^{241}Pu in the surface air during the first 48 hours after a hypothetical accident in a floating reactor at Shtokmann natural gas field, the Barents Sea, assumed release 5 May 2010.

9. Modeled hourly ^{241}Pu activity concentration (μBq m^{-3}) in the surface air of Rovaniemi in March-April 2011.

Table 1. The atmospheric activity concentrations of ^{238}Pu, $^{239+240}Pu$ and ^{241}Pu in Rovaniemi, Finnish Lapland. The activity values have been decay-corrected to the middle point of the sampling period.

Year	**A ^{238}Pu (nBq m^{-3})**	**A $^{239+240}$Pu (nBq m^{-3})**	**A ^{241}Pu (nBq m^{-3})**
1965	68±8	2270±40	38198±711
1966	117±7	1371±21	21182±451
1967	221±10	497±13	7768±236
1968	259±13	969±20	16237±396
1969	245±12	973±20	14585±372
1970	135±9	1040±20	15027±367
1971	76±5	1211±16	15975±387
1972	28±3	325±7	3456±179
1973	26±3	206±7	1701±128
1974	13±2	570±12	7383±261
1975	15±3	250±10	3769±182
1976	6.7±1.2	74±3	804±75
1977	6.9±1.2	297±7	3632±169
1978	13±2	563±10	9106±291
1979	6.1±1.2	175±5	3645±210
1980	2.7±0.9	74±4	1063±92
1981	7.0±1.7	248±9	2137±137
1982-March 1986	0.59±0.16	15.3±0.8	200±19
April-June 1986	< 1.6	< 7.2	< 381
July-December 1986	< 1.1	< 5.2	315±71
1987-1990	2.2±0.3	5.8±0.4	101±15
1991-1995	0.23±0.07	16.9±0.1	73±11
1996-2000	< 0.1	6.5±0.2	39±8
2001-2005	0.37±0.19	1.4±0.3	41±10
2006-2010	< 0.4	0.51±0.14	< 25
2011	< 1.5	< 3.5	602±131

599 Table 2. The activity ratios $^{238}Pu/^{239+240}Pu$, $^{241}Pu/^{239+240}Pu$, $^{239+240}Pu/^{137}Cs$, and the mass ratio
600 $^{240}Pu/^{239}Pu$ in the air filters collected in Rovaniemi. The uncertainty is 1 sigma error for the activity
601 ratios and 2 sigma error for the mass ratio. – means that one or both isotopes had concentration
602 below the detection limit.

Year	**A ^{238}Pu/ A $^{239+240}$Pu**	**A ^{241}Pu / A $^{239+240}$Pu**	**mass ratio ^{240}Pu/^{239}Pu**	**A $^{239+240}$Pu/A ^{137}Cs**
1965	0.030±0.004	16.8±0.4	0.177±0.006	0.0071±0.0001
1966	0.085±0.005	15.5±0.4	0.172±0.003	0.0067±0.0001
1967	0.444±0.023	15.6±0.6	0.170±0.003	0.0079±0.0003
1968	0.267±0.014	16.8±0.5	0.190±0.004	0.0108±0.0003
1969	0.252±0.014	15.0±0.5	0.172±0.005	0.0104±0.0003
1970	0.130±0.009	14.5±0.5	0.186±0.007	0.0087±0.0002
1971	0.063±0.004	13.2±0.4	0.174±0.006	0.0135±0.0002
1972	0.087±0.008	10.6±0.6	0.125±0.007	0.0116±0.0005
1973	0.125±0.015	8.2±0.7	0.131±0.008	0.0182±0.0009
1974	0.022±0.003	12.9±0.5	0.182±0.005	0.0102±0.0003
1975	0.058±0.011	15.1±0.9	0.132±0.008	0.0102±0.0005
1976	0.091±0.016	10.9±1.1	0.138±0.009	0.0130±0.0010
1977	0.023±0.004	12.2±0.6	0.216±0.015	0.0097±0.0004
1978	0.024±0.003	16.2±0.6	0.209±0.011	0.0102±0.0003
1979	0.035±0.007	20.8±1.4	0.209±0.012	0.0107±0.0004
1980	0.036±0.012	14.3±1.5	0.173±0.015	0.0090±0.0006
1981	0.028±0.007	8.6±0.6	0.117±0.009	0.0107±0.0005
1982-March 1986	0.038±0.011	13.1±1.4	0.142±0.011	0.0065±0.0006
April-June 1986	-	-	0.278±0.093	-
July-December 1986	-	-	0.254±0.073	-
1987-1990	0.376±0.056	18±3	0.152±0.026	0.0014±0.0001
1991-1995	0.245±0.082	79±17	0.132±0.091	0.0393±0.0038
1996-2000	-	32±8	0.131±0.066	0.0106±0.0010
2001-2005	0.260±0.142	29±9	0.170±0.082	0.0030±0.0007
2006-2010	-	-	0.194±0.116	0.0005±0.0001
2011	-	-	0.145±0.091	-

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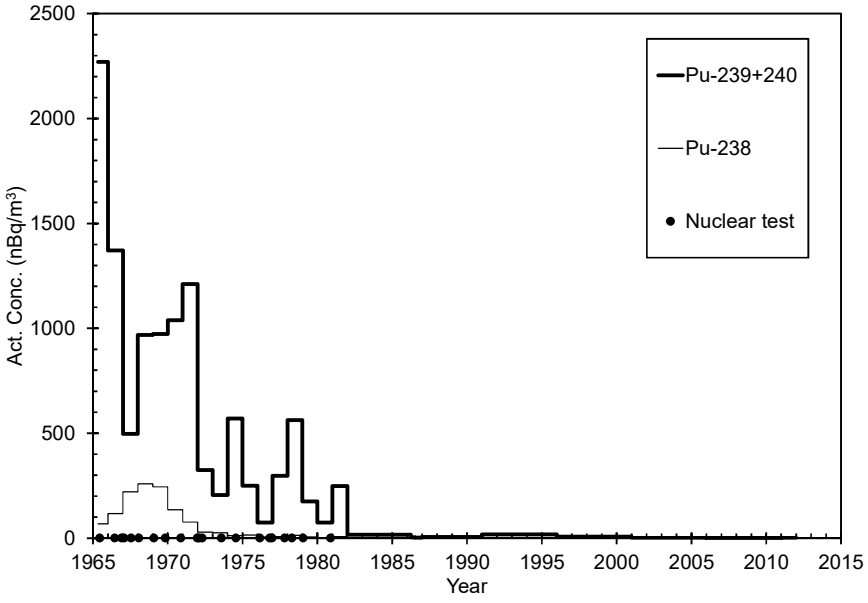


Fig.1. Activity concentration of ^{238}Pu (thin line, nBq m^{-3}) and $^{239+240}$Pu (thick line, nBq m^{-3}) in the surface air of Rovaniemi in 1965-2011. Values below the detection limit have been depicted as half the MDA value (Table 1). The black circles indicate the times of atmospheric nuclear tests in the People's Republic of China (UNSCEAR 2000).

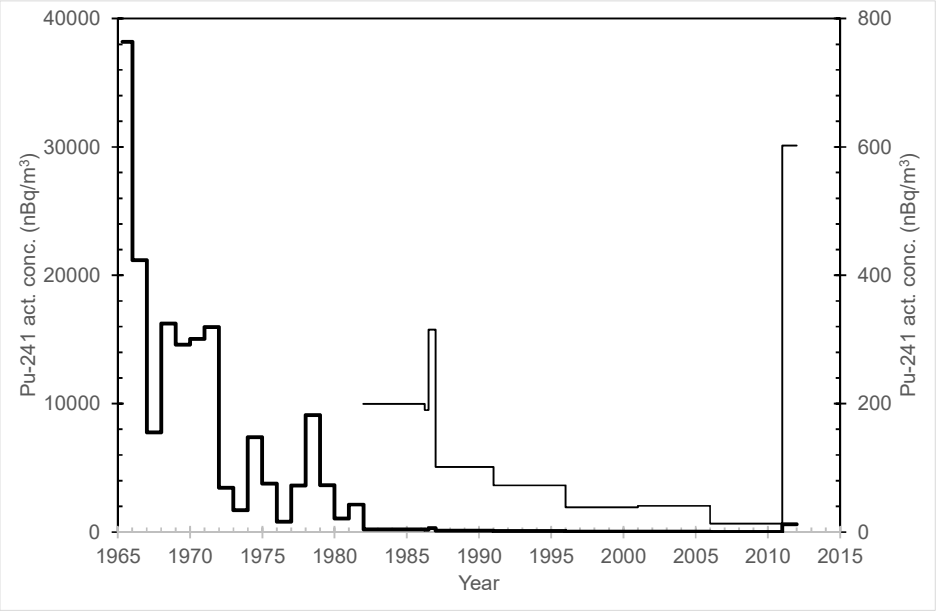


Fig. 2. Activity concentration of ^{241}Pu (nBq m^{-3}) in the surface air of Rovaniemi (thick line 1965-2011, left vertical scale; thin line 1982-2011, right vertical scale). Values below the detection limit have been depicted as half the MDA value (Table 1).

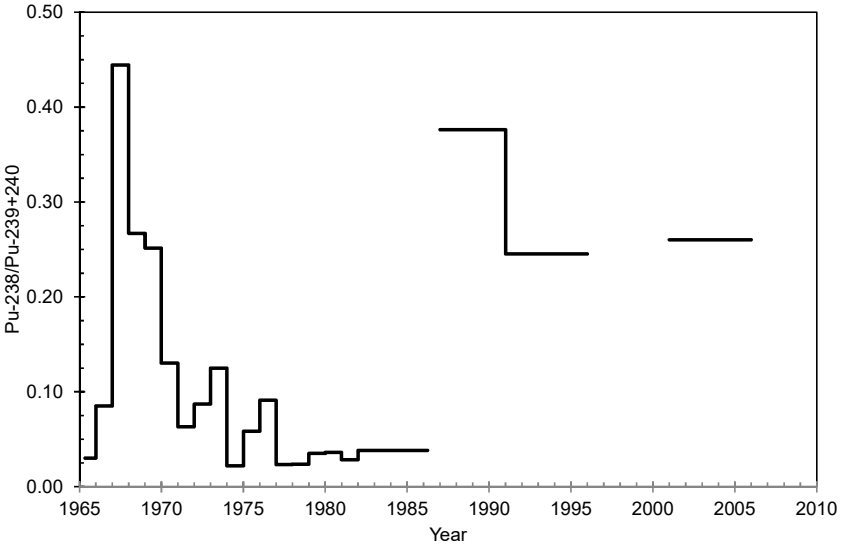


Fig. 3. The activity ratio $^{238}Pu/^{239+240}Pu$ in the surface air of Rovaniemi as a function of time.

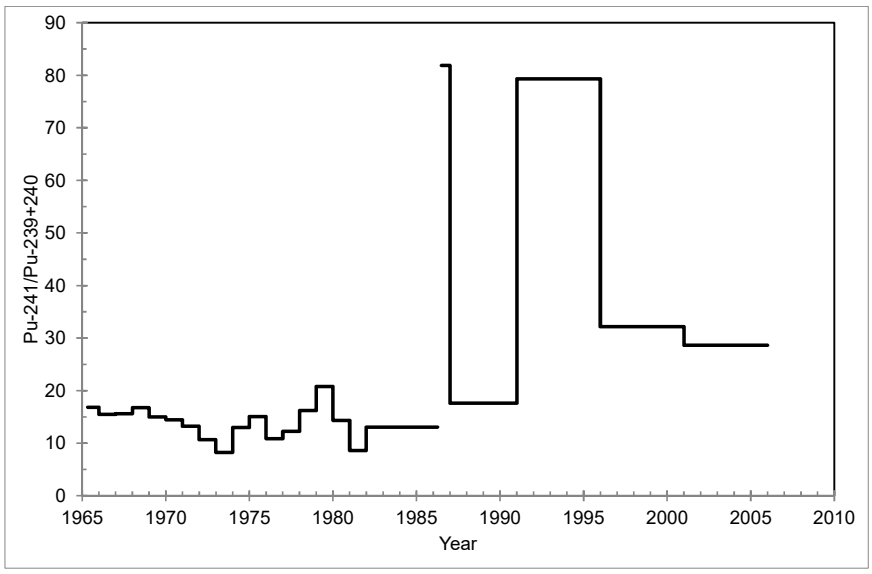


Fig. 4. The activity ratio $^{241}Pu/^{239+240}Pu$ in the surface air of Rovaniemi as a function of time.

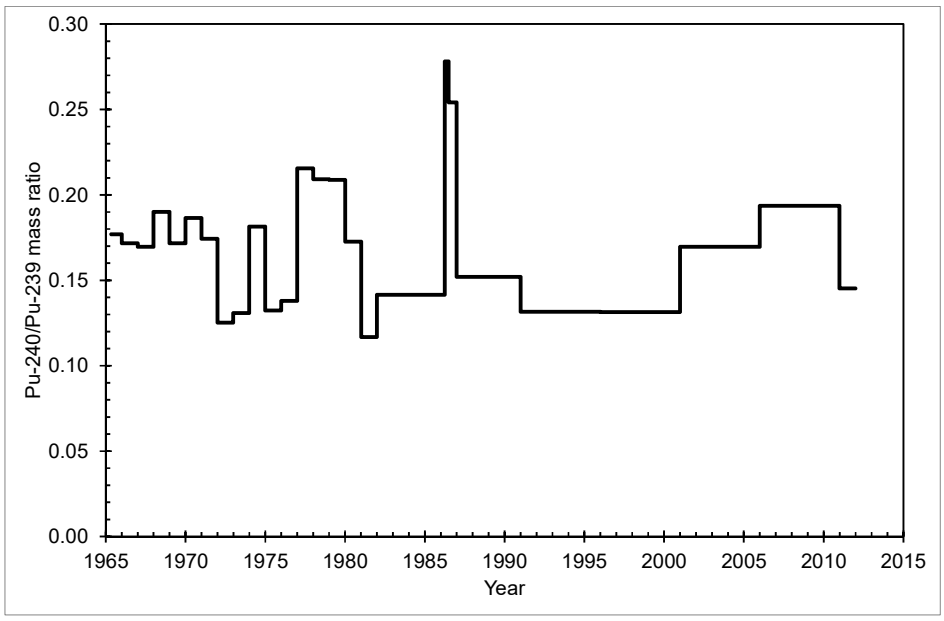


Fig 5. The mass ratio $^{240}Pu/^{239}Pu$ in the surface air of Rovaniemi as a function of time.

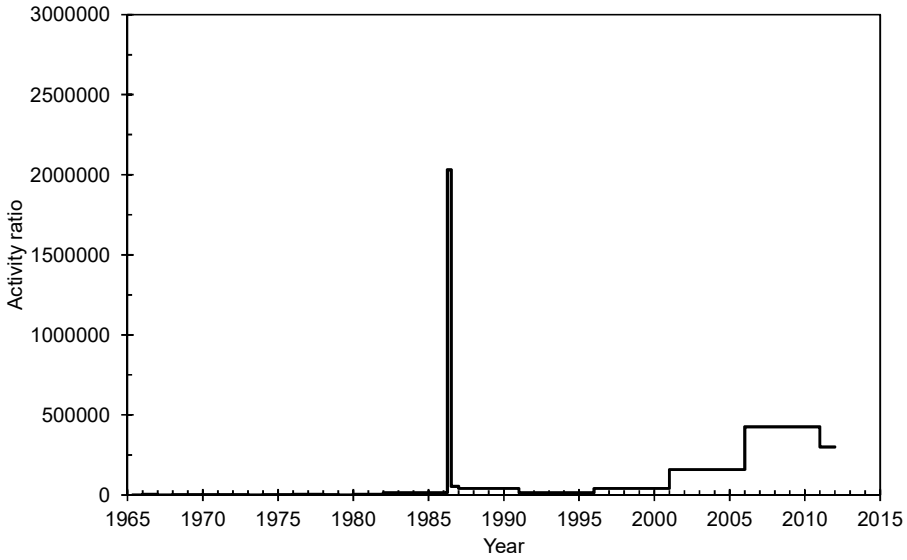
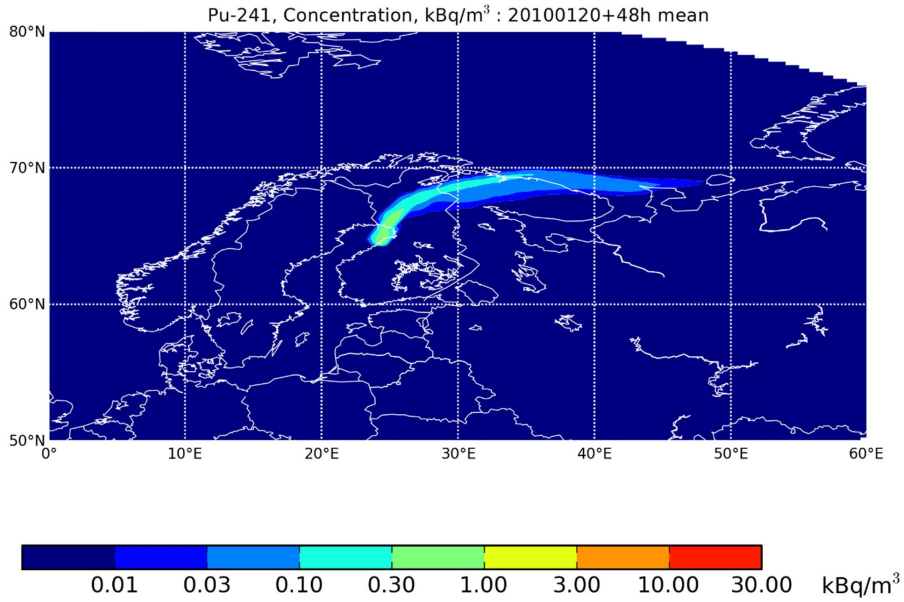


Fig. 6. The ratio of total beta activity (Salminen-Paatero et al. 2019) and $^{239+240}$Pu activity content in the surface air in Rovaniemi in 1965-2011. $^{239+240}$Pu values below the detection limit have been replaced with half the MDA values in the ratio calculation (Table 1).



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719 Fig. 7. The average activity concentration of ^{241}Pu in the surface air during the first 48 hours after a
720 hypothetical reactor accident at Pyhäjoki, assumed release 20 January 2010.

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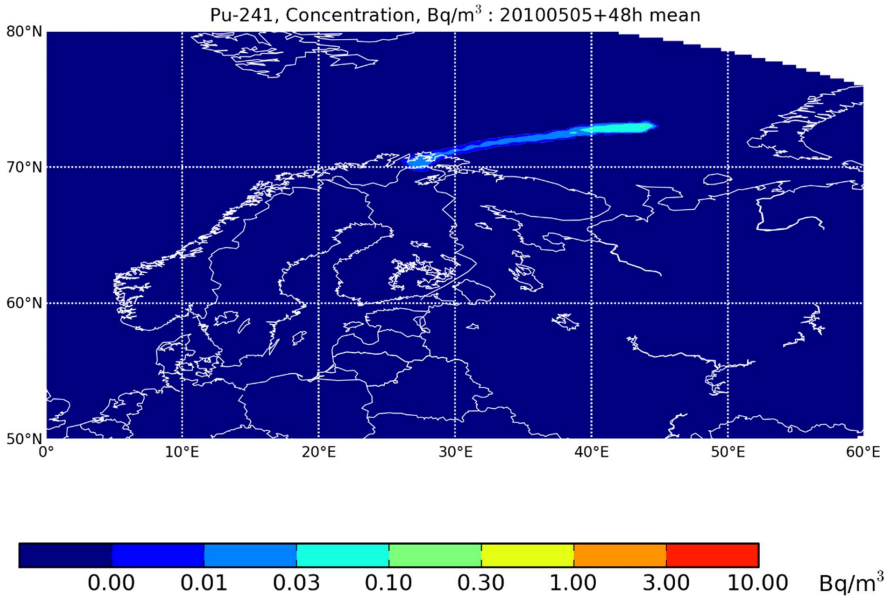
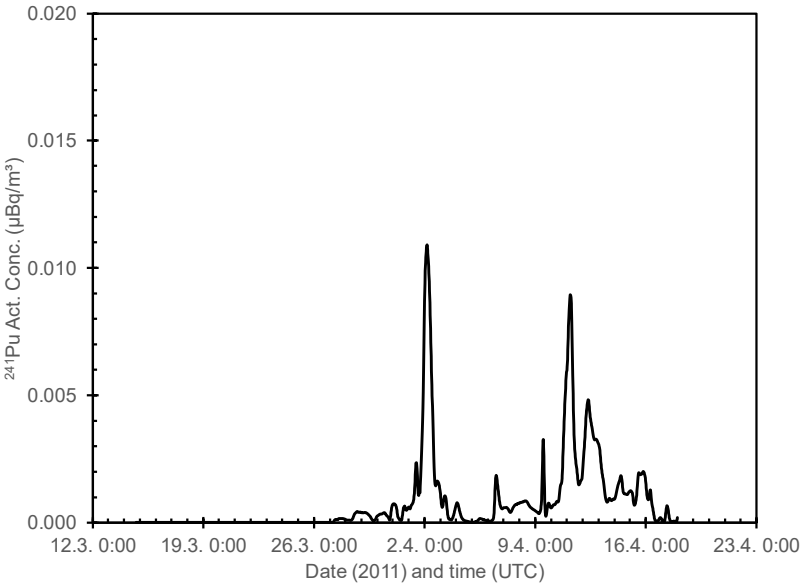


Fig. 8. The average activity concentration of ^{241}Pu in the surface air during the first 48 hours after a hypothetical accident in a floating reactor at Shtokmann natural gas field, the Barents Sea, assumed release 5 May 2010.



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731 Fig. 9. Modeled hourly ^{241}Pu activity concentration (µBq m^{-3}) in the surface air of Rovaniemi in
732 March-April 2011.

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Author contributions

Susanna Salminen-Paatero performed radiochemical analysis and data analysis. Julius Vira produced Silam calculations. Jussi Paatero provided the air filter sampling and sampling data, and planned the accident scenarios. All authors contributed to writing the manuscript.

Data availability

Data will be available ~~at~~ in the University of Helsinki open data system.