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ANALYTICAL METHODS FOR WIDE AREA ENVIRONMENTAL SAMPLING (WAES) FOR AIR FILTERS

Finnish support to IAEA

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Abstract

The Finnish Nuclear Verification organization (FINUVE) performed a field trial in Kazakhstan in order to test sampling and analytical methods developed in Finland. The effect of pre-filtration was studied using two samplers, one with and one without pre-filtration located next to each other. The sampling period was one year from April 2000 to April 2001. Dust, ^7Be , ^{137}Cs , U and Pu-isotopes were measured from the filters. Radiochemical separation techniques were developed for the separation of U, and Pu from the air filters and from each other. γ -spectrometry was used for the measurement of ^7Be and ^{137}Cs , α -spectrometry and liquid scintillation spectrometry for ^{238}Pu , $^{239,240}\text{Pu}$, ^{241}Pu and ICP-MS for total uranium and the $^{235}\text{U}/^{238}\text{U}$ -ratio. Tracer studies were performed to study the effectiveness of the chemical separation.

Certified reference materials were analyzed to validate the accuracy of the measurement techniques. U and Pu concentrations of soil samples from same area in Kazakhstan were also analyzed.

The variation in the content of solid material and the ^7Be concentrations are rather high. ^{137}Cs can only be detected in a part of the samples. The total uranium varies considerably between the samples, while the $^{235}\text{U}/^{238}\text{U}$ -ratio can be determined with a precision of $\pm 2-10\%$. The ratio $^{238}\text{Pu}/^{239,240}\text{Pu}$ is a more sensitive indicator for plutonium than the $^{239,240}\text{Pu}$ concentration.

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1 Introduction

The international Wide Area Environmental Sampling (WAES) working group concluded in its report that air and aerosol sampling combined with the determination of ^{85}Kr , ^{137}Cs , ^{134}Cs , ^{90}Sr , ^{129}I , as well as Pu isotopes and their radioactivity ratios are the most promising signatures for reprocessing and the $^{235}\text{U}/^{238}\text{U}$ -ratio for enrichment [1]. It was also concluded that the report contains considerable uncertainty, which should be tested by field trials.

Kurchatov in Kazakhstan was chosen as the site for the field trial for several reasons. The Semipalatinsk test site in Kazakhstan has a long history of different kinds of nuclear activities. It has been a nuclear weapons test site both above and below ground. There are also research reactors on the site. The sediment in different parts of the Semipalatinsk test site contains highly variable Pu concentrations and plutonium isotope ratios. $^{239+240}\text{Pu}$ values varying from 0.2 to 128,000 Bq/kg and $^{238}\text{Pu}/^{239}\text{Pu}$ -ratios varying from 0.0145 to 0.447 have been reported [2,3,4]. Therefore, one could expect to find varying concentrations in the aerosol filters. The environmental conditions resemble those of Iraq in some respects. In the Kazakhstan field trial, two weekly parallel aerosol samples were collected. The sampling was conducted in Kurchatov close to the Semipalatinsk area. Aerosol particles were collected with Petrianov FPP-15-1.5-type filters, which are made of polyvinyl chloride, and thus contain negligible amounts of uranium compared to typical glass-fibre filters. The collection area of the filter is 42 cm × 24 cm. The filter face velocity is 0.4 m/s when the sampling flow rate is 150 m³/h. According to tests carried out in laboratory conditions, aerosol particles with a diameter of 0.2 μm are collected with an efficiency of more than 80%. The amount of collected air during a week is about 25,000 m³.

One of the two samplers placed next to each

other was equipped with an impactor-type pre-separator. There have been indications that uranium in the form of UF_4 or UCl_6 is originally released in very small particles. Plutonium has also been shown to exist predominantly in small particles in some sediments [5]. The cut-diameter of the pre-separator, defined as a diameter of particles that are removed with 50% efficiency, is 13 μm. During the last part of the field trial, one half of the pre-separator holes were covered to decrease the cut-diameter to 9 μm.

Details of the field trial are presented elsewhere [6,7,8,9]. Two similar aerosol samplers were placed close to each other on the roof of a low building. They were run in parallel and weekly samples of about 25,000 m³ were taken. The filters from both samplers were always changed on the same day. The second sampler was equipped with the pre-filtering unit.

The sample filters were weighed before and after sampling so as to determine the weight of solid material collected onto the filter. Then the samples, one half of each filter, were measured by γ -spectrometry in order to determine the ^7Be and fission products. After that, the sample was ashed, the ash dissolved and the uranium and plutonium were separated chemically into separate fractions. The total uranium and uranium $^{235}/^{238}$ ratio were determined using inductively coupled plasma mass spectrometry (ICP-MS) and plutonium isotopes using α -spectrometry.

Previously, glass-fibre filters have been used for sampling fission products from air. In this work, uranium and plutonium were determined as well. A chemical separation was performed. In order to increase the rate and efficiency of dissolution of the air filter samples, organic filters were used instead of the glass-fibre filter. Since the effectiveness of collecting small particles varies considerably with filter type, different filters were tested and Petrianov FPP-15-1.5 was chosen [6]

2 Analytical methods

2.1 Gamma-spectrometry

Two HPGe-detectors were used for the gamma-spectrometric measurements. The relative efficiencies of the detectors are 39% and 86%. The detectors were placed in low-background shields made of 12-cm-thick old lead rings, which were lined with cadmium and copper to reduce the effect of X-rays.

The filters were placed into plastic beakers (P105) to provide a suitable counting geometry for the gamma-spectrometric analysis. The beaker was cylindrical, with a radius of 37 mm and a height of 26 mm. The samples were measured for about 24 hours' measurement time.

The spectra were analyzed with the GAMMA-program developed in STUK. As output, GAMMA produces a list of found peaks and identified radionuclides with activity concentrations and a picture of the spectrum. The program estimates true coincident summing and sample self-absorption corrections.

The Nuclide-specific Minimum Detectable Concentration MDC ($\mu\text{Bq}/\text{m}^3$) depends on the filtered air volume, the activity of background nuclides in the sample, the measuring time and the decay time before the measurement. The typical MDC for the ^{137}Cs was $1.6 \mu\text{Bq}/\text{m}^3$.

2.2 Chemical separation of Pu and U

2.2.1 Analytical procedure

The determination of U and Pu isotopes in an organic air filter consisted of the following steps:

1. Ashing in 400°C over night. After ashing, ^{242}Pu and ^{236}U tracers were added to the samples.
2. Leaching with HNO_3 for 6 hours, then filtration, after which the filtrate was evaporated to a volume of few millilitres.
3. Extraction chromatography.

Resins

Two extraction chromatography resins were used: UTEVA and TRU, which were obtained from Eichrom Industries. The particle sizes of the resins were 100–150 μm . 0.7 g of both resins was weighed and allowed to equilibrate with deionized water over night. The resins were then packed into polypropylene columns. The columns were conical, 0.8×4 cm and contained up to 2 ml of resin and 10 ml of eluent. The UTEVA column was preconditioned with 5 ml of 3 M HNO_3 and the TRU column with 5 ml of 2 M HNO_3 .

Sample preparation

The evaporated sample was dissolved in 10 ml of 3 M HNO_3 + 0.5 M $\text{Al}(\text{NO}_3)_3$. Aluminum nitrate was used to increase the efficiency of the separation procedure.

2 ml of 0.6 M ferrous sulfamate was added to the sample. Ferrous sulfamate reduces plutonium to Pu(III).

200 mg of ascorbic acid was added to the sample and the sample was allowed to stand for 10 minutes. Ascorbic acid was used to keep Fe in the 2+ state.

Uranium separation using UTEVA resin

- The sample was loaded into a UTEVA column and the eluate was collected for Pu separation.
- 2×5 ml of 3 M HNO_3 was added into the column and the eluate was collected for Pu separation.
- 4 ml of 9 M HCl was added into the column in order to convert the resin to the chloride system.
- 20 ml of 5 M HCl -0.05 M oxalic acid was added into the column. This rinse removed Np, Th and residual Fe.
- U was eluted with 15 ml of 0.01 M HCl .

Table I. Determinations of U and Pu isotopes in reference material, IAEA-135 Sediment. The error is one sigma error of activity. The confidence intervals were calculated using statistical tables for distribution-free sample population and represent symmetrical two-sided intervals at a significance level of 0.05.

	Sample 1	Sample 2	Recommended value of IAEA-135 (Bq/kg)	Confidence interval of IAEA-135 (Bq/kg)
U recovery (%)	64±4	29±2		
Pu recovery (%)	60±2	10±0.4		
²³⁴ U (Bq/kg)	15±2		14*	11.7–18.5 (n)
²³⁸ U (Bq/kg)	14±2		13*	11.3–18.1 (n)
γ-spectrometry				
²³⁸ U (Bq/kg)	13±2	12±2	13*	11.3–18.1 (n)
ICP-MS				
^{239,240} Pu (Bq/kg)	211±7	221±13	213	205–225.8
²³⁸ Pu (Bq/kg)	40.7±2	37±4	43	41.6–45

*information value only, (n) normal dissolution techniques

Plutonium separation using TRU resin

- Pu solutions collected in the uranium separation were loaded into a TRU column.
- 5 ml of 2 M HNO₃ was added into the column.
- 5 ml of 2 M HNO₃+0.1 M NaNO₂ was added into the column. NaNO₂ oxidizes Pu (III) to Pu(IV).
- 5 ml of 0.5 M HNO₃ was added into the column.
- 3 ml of 9 M HCl was added into the column.
- 20 ml of 4 M HCl was added into the column. This rinse removes Am.
- 25 ml of 4 M HCl+0.1 M HF was added into the column. This washing step removes any residual Th present.
- Pu was eluted with 10 ml of 4 M HCl+0.02 M TiCl₃

50 µg of Nd was then added as a non-isotopic carrier and Pu was co-precipitated with NdF₃ and the precipitate was filtrated.

2.2.2 Reliability tests

Four blank analyses were made. In the *filter blank analysis*, a piece of the same filter fabric as used in the real air filters was ashed. Then the tracers were added and normal procedure (leaching, UTE-VA and TRU) followed. Three *reagent blank analyses* were otherwise the same, but because the filter fabric was not used the filtration was unnecessary after leaching. Both the plutonium and uranium contents of the blank analyses were below the detection limit.

2.3 Pu measurement

Alpha activity was measured with a Canberra passivated implanted planar silicon (PIPS) detector. After the alpha measurement, the membrane filter and precipitate were introduced into a 20-ml liquid scintillation vial and wetted with 0.6 M H₃(BO₃)₄ and finally mixed with scintillation cocktail. ²⁴¹Pu activity was measured with a low-level liquid scintillation counter, Quantulus 1220 for 600 minutes.

2.4 Uranium measurement

The analysis of total uranium and uranium isotope ratio after chemical separation was performed using the ICP-MS-technique. The instrument used was a VG Plasma Quad 2+. ²³⁶U added to the samples was used for yield correction for the chemical separation. The influence of the small amount of ²³⁵U in the tracer solution was corrected in the isotope ratio measurements.

2.4.1 Standard reference samples

Recovery of the extraction chromatography was tested with tracers ²⁴²Pu and ²³⁶U and the recoveries were found to be close to 100% for both U and Pu.

To simulate real air filter samples, 0.3 g sediment (reference material, IAEA-135) was added to the air filter material before ashing. The sample was leached with HNO₃ + HCl for 6 hours after ashing. Extraction chromatography was carried out as explained in Section 2.2.1. The results of the two experiments are shown in Table I.

Table II. $^{235}\text{U}/^{238}\text{U}$ ratio in NIST and IAEA standard reference materials.

Standard	$^{235}\text{U}/^{238}\text{U}$ certified	$^{235}\text{U}/^{238}\text{U}$ measured	Deviation from certified value %
U-005	0.04919	0.00502	2.1
U-030	0.03143	0.03194	1.6
U-200	0.25126	0.24923	0.8
NIST 1633 fly ash	0.00725	0.00726	0.1
NIST 1633 fly ash	0.00725	0.00742	2.3
NIST 1633 fly ash	0.00725	0.00725	0.0
NIST 1633 fly ash	0.00725	0.00750	3.3
IAEA Soil 1	0.00725	0.00715	1.4

The $^{235}\text{U}/^{238}\text{U}$ ratio was determined in several standard reference materials to control the accuracy of the ICP-MS technique in these determinations. The results are presented in Table II.

2.4.2 Kurchatov samples

The samples were ashed in an oven at a temperature of 400°C, leached with acid and purified with

Eichrom's UTEVA and TRU resins as described in Section 2.2.1. ^{236}U and ^{242}Pu tracers were added to the air filters after ashing.

The Pu recoveries varied from 40% to 100%, median value was 74%. The uranium recoveries were between 46% and 100%, with the median value of 78%.

3 Results and discussion

3.1 Results of the parallel air filter samples

Normally one half of the filter was used for the analysis. To further confirm the results of isotope ratios and activities seven air filters were selected for a new analysis from the other half of the same filter. The results of these analyses are shown in Table III.

For samples A49, A51, A63 and B92, the changes in isotope ratio were relatively small whereas samples A61, B86, and B88 show greater differences in isotope ratios between the two halves of the same filter. There is still a great difference in $^{239,240}\text{Pu}$ activities between the first and the second analysis of samples A49 and A51, but also the second analysis shows exceptionally high $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratios. Highly varying Pu activities and isotope ratios between the different air filters and between the halves of the same filter are probably caused by hot particles.

3.2 Impurities and their elimination

3.2.1 Polonium

^{210}Po disturbed the ^{239}Pu determination when eluting Pu with $\text{NH}_4\text{HC}_2\text{O}_4$ from the TRU column and two methods were tested to solve this problem. In the first method, crushed pieces of silver were used to remove the polonium from the solution before making the preparation by electro deposition for alpha counting. In the second method, the plutonium was eluted from the TRU column with TiCl_3 solution in order to separate the polonium. This method was more convenient to use because it produced greater yields than the first method. The TiCl_3 elution was selected in order to prevent Po from contaminating the Pu samples.

3.2.2 Thorium

^{228}Th has one alpha-peak very close to that of ^{238}Pu and the concentration of thorium in environmen-

Table III. Analysis of the parallel filters halves. $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratios and $^{239,240}\text{Pu}$ activities in the first half of an air filter and in the second half of the same filter. The error is one sigma error of activity.

Sample	A ($^{239,240}\text{Pu}$) Bq/kg		A (^{238}Pu)/A($^{239,240}\text{Pu}$)	
	First half of filter	Second half of the filter	First half of filter	Second half of the filter
A49	111±4	12±1	0.021±0.004	0.079±0.0026
A51	228±7	7.4±1.0	0.015±0.002	0.11±0.04
A61	2.8±0.4	4.4±0.7	2.4±0.4	0.53±0.12
A63	4.2±0.5	4.5±0.6	1.9±0.3	1.9±0.3
B86	1.9±0.3	5.7±0.6	0.88±0.20	0.24±0.06
B88	1.1±0.2	0.67±0.21	1.6±0.4	1.4±0.6
B92	1.8±0.4	1.8±0.3	1.9±0.5	1.9±0.4

tal samples is usually orders of magnitude higher than the concentrations of plutonium isotopes. Thus, the possible leaking of Th to the Pu fraction will interfere with the determination of ^{238}Pu and especially the activity ratio of $^{238}\text{Pu} / ^{239,240}\text{Pu}$. The separation of Th from Pu by the method used in the present work was checked by several experiments.

1. Two-air filter samples were measured twice in order to see if ^{228}Th causes the high isotope ratios. ^{224}Ra , the daughter nuclide of ^{228}Th , could be seen in the plutonium spectrum if Th leaked into the Pu fraction. ^{224}Ra was not seen in the Pu spectrum.
2. Two analyses of lichen. The lichen was collected in 1963, before the SNAP explosion in 1964 which released ^{238}Pu activity into the atmosphere. The $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratio was 0.031 ± 0.003 in Sample 1, and 0.023 ± 0.003 in Sample 2. The results indicate that the method used for separating plutonium from the lichen samples totally removes traces of thorium from the plutonium fraction.
3. A tracer study with ^{229}Th tracer showed that there was no counts do to ^{229}Th in the alpha spectrum of the Pu fraction.

3.2.3 Soil samples

Even though Th experiments confirm the high $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratios, some soil samples from Kazakhstan were also analyzed; the results are presented in Table IV.

The isotope ratios in the Table IV indicate that in soil sample 2 from Kazakhstan, the activity ratio of ^{238}Pu to $^{239,240}\text{Pu}$ is relatively high. This observation supports the results of the air filter analyses, where high activity ratios of plutonium isotopes were found.

Fractionation according to the particle size was performed for sample 5 using the sedimentation method. The method was adopted from the study of Livens and Baxter [5]. The results in Table V show that Pu and U are evenly distributed in particles $< 63 \mu\text{m}$.

3.3 Results of air filter samples

The results of the Kazakhstan samples are shown in graphical form in Figs. 1–7. K1 is the normal air particle sampler and K2P is the sampler with the pre-filtering unit. The first figure shows the

Table IV. $^{238}\text{Pu} / ^{239,240}\text{Pu}$ ratios and $^{239,240}\text{Pu}$ activity of soil samples from Kazakhstan. The error is one sigma error of activity.

Sample	A (^{238}Pu) / A ($^{239,240}\text{Pu}$)	A ($^{239,240}\text{Pu}$) Bq/kg
1	0.051 ± 0.015	0.40 ± 0.03
2	0.031 ± 0.007	0.72 ± 0.03
3	Under detection limit	0.08 ± 0.01
4	Under detection limit	0.36 ± 0.03
5	0.22 ± 0.02	1.06 ± 0.04

amount of solid material per m^3 of collected air. ^7Be can be determined in all the samples, as shown in Fig 2.

Figure 1 shows the amount of dust per m^3 of air collected onto the filters. The variation is significant, depending on the time of year, wind direction, velocity and the presence of precipitation. The Be values are usually used to show the integrity of the sampler. From the values in Fig. 2, one can see that the values vary within a factor of about three. Therefore, only very large discrepancies can be detected with this method. The analytical error is much smaller than the variation in the concentrations. Consequently, nothing can be gained by improving the analytical accuracy.

All the other results are indicated as concentration per unit of solid material on the filter, including the error bars. ^{137}Cs results are below the detection limit in most samples. All the results above the detection limit are indicated in Fig. 3. The rest of the results were below the detection limit.

The background in the Semiplatinsk test site is low. By measuring the whole filter, or two weekly filters, all or more of the results might be above the detection limit. There is a rather high variation in the concentrations. It would be easier to interpret the data and find discrepancies from the trend, if all results were above the detection limit. Since a significant part of the background originates from re-suspended material, the authors think that it is easier to interpret the results when indicated as a fraction of the solid material, rather than a concentration in air.

Both total uranium, and the $^{235}\text{U}/^{238}\text{U}$ ratio, and $^{239+240}\text{Pu}$ and the $^{238}\text{Pu}/^{239+240}\text{Pu}$ ratio have been measured. The results are shown in the Figs. 4–7.

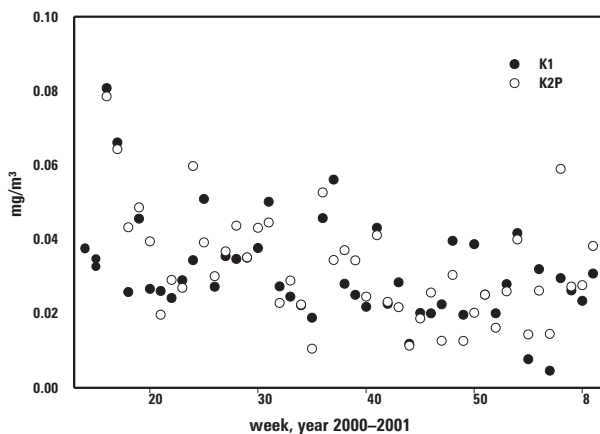
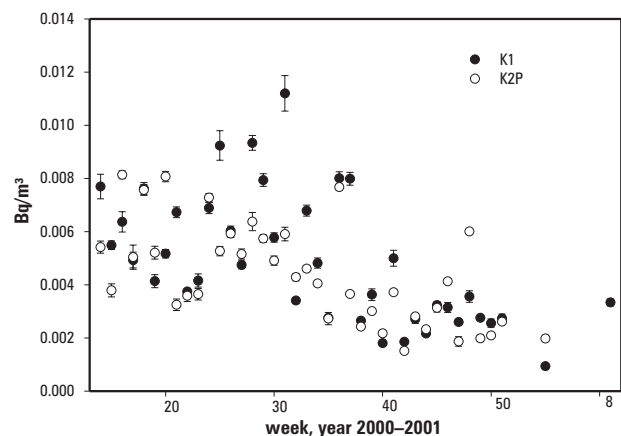
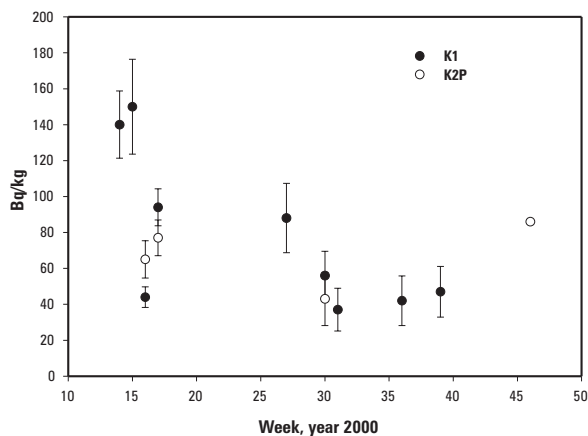
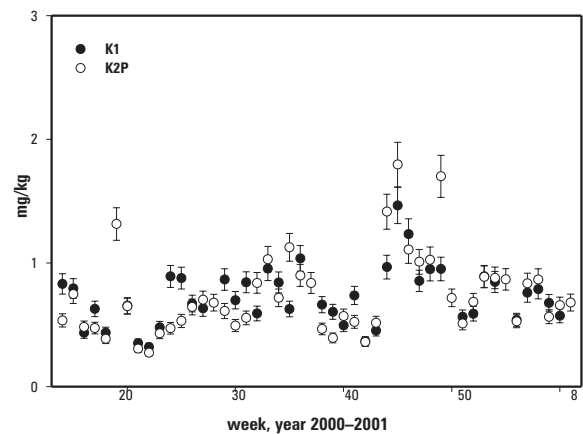
The total uranium concentrations, as seen in Fig. 4, in general show a smooth variation, which depends on the time of year. Within this, there is a

Table V. Uranium and plutonium in fractionated soil sample.

Grain size	Frac. (%)	total U (mg/kg) NAA	soluble U (mg/kg) ICP-MS	$^{239,240}\text{Pu}$ (Bq/kg)
> 2.0 mm	9.95	1.79	0.61	0.016 ± 0.006
0.25–2.0 mm	42.34	0.923	0.207	0.042 ± 0.004
0.125–0.25 mm	16.14	0.975	0.582	0.180 ± 0.015
0.063–0.125 mm	7.77	1.39	0.549	0.347 ± 0.036
0.032–0.063 mm	4.61	1.87	0.588	2.211 ± 0.091
20–32 μm	6.45	2.42	0.65	1.194 ± 0.072
8–20 μm	5.10	1.73	0.541	1.760 ± 0.100
2–8 μm	4.13	1.83	0.634	1.359 ± 0.085
< 2 μm	3.52	1.84	0.813	1.854 ± 0.280

statistical variation, usually within a maximum factor of two, which means about 50% from the median. The few outliers above the average concentrations can be clearly distinguished. An approximate deviation of 100% from the median can

be clearly distinguished. An improvement of the analytical accuracy would not improve the detectability of a deviation significantly, because the maximum deviation of the background, is of the same order as the analytical precision.

**Figure 1.** Dust in air in samples from Kurchatov, Kazakhstan.**Figure 2.** ^7Be activity in air samples from Kurchatov, Kazakhstan.**Figure 3.** Cs in dust in air samples from Kurchatov, Kazakhstan.**Figure 4.** Uranium concentration in air samples from Kurchatov, Kazakhstan.

The U was determined also in soil samples. The total uranium determined by instrumental neutron activation analysis was 1.6–2.0 mg/kg. The amount of uranium soluble into the mixture of nitric acid-hydrochloride acid was determined by ICP-MS. The analysis shows that 10–15 % of uranium in the soil samples is leached into acids, which corresponds to 0.2 mg/kg in soil. The analyzed uranium concentrations in the collected air dust were higher than the concentration of leached uranium in soil samples. The median value for uranium concentration in dust was 0.66 mg/kg.

The $^{235}\text{U}/^{238}\text{U}$ ratios are shown in Fig. 5. When there is enough material for the analysis, the ICP-MS method has a precision of $\pm 5\%$. Also here, there are sequences where that precision has been reached. However, sometimes the deviation is greater. Therefore, deviations of 10 % or greater from the natural ratio can be distinguished. This means that the isotope ratio is a much better method to detect signatures than the total uranium concentration, as was already stated by the

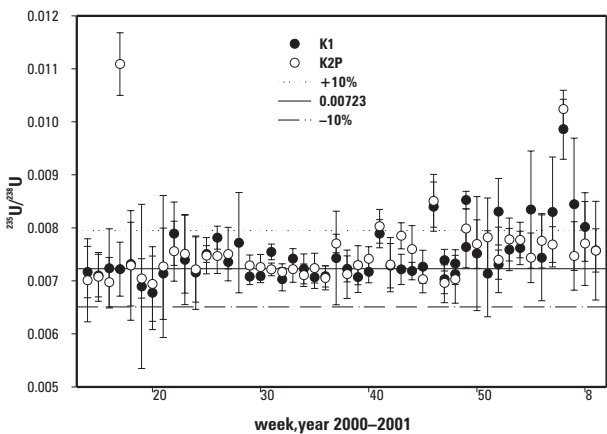


Figure 5. Uranium isotope ratios in air filter samples from Kurchatov, Kazakhstan.

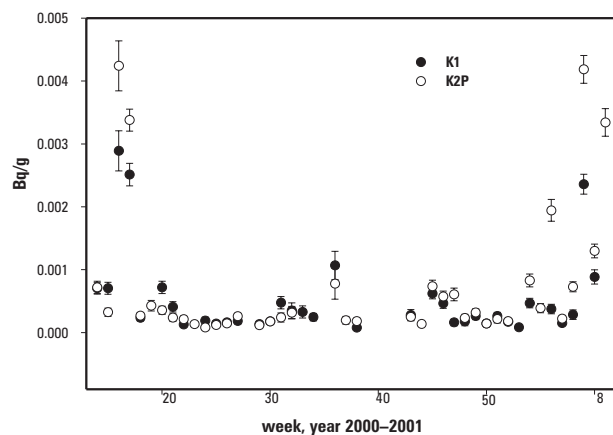


Figure 7. ^{238}Pu in aerosol samples from Kurchatov, Kazakhstan.

WAES group [1]. An improvement of the analytical precision would enhance the ability to detect signatures.

Figure 6 shows the $^{239+240}\text{Pu}$ values and Fig. 7 the ^{238}Pu values. The detection limit D_L is 0.05 mBq/sample for ^{238}Pu and $^{239,240}\text{Pu}$ at a counting time of 10,000 minutes. $^{239,240}\text{Pu}$ activity per m^3 of air collected onto the filters varied between (13 ± 4) nBq and (1665 ± 57) nBq and the median was 80 nBq/ m^3 .

Figure 8 shows the Pu isotope ratios. Four groups of results can be identified. In most samples the activity ratio $^{238}\text{Pu}/^{239+240}\text{Pu}$ is at the level of 0.3. Then there is a group of samples with a ratio clearly above that, in the order of 0.6–0.8, which indicates the presence of plutonium from spent fuel. In a few samples the ratio is very low, which indicates the presence of weapons grade plutonium originating from the nuclear weapons tests. There is also a group of very high isotope ratios, between 1–2, indicating the presence of fuel with high burn-up.

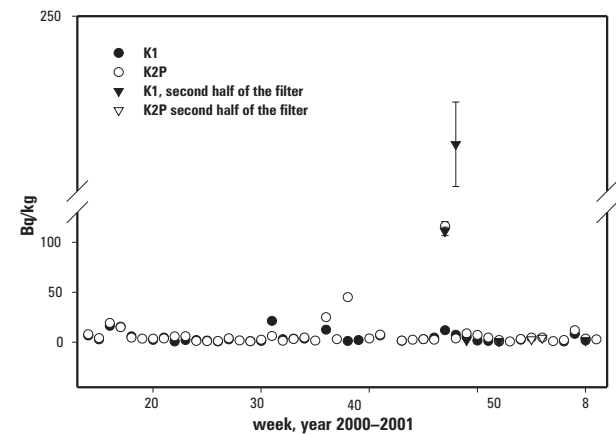


Figure 6. $^{239+240}\text{Pu}$ in aerosol samples from Kurchatov, Kazakhstan.

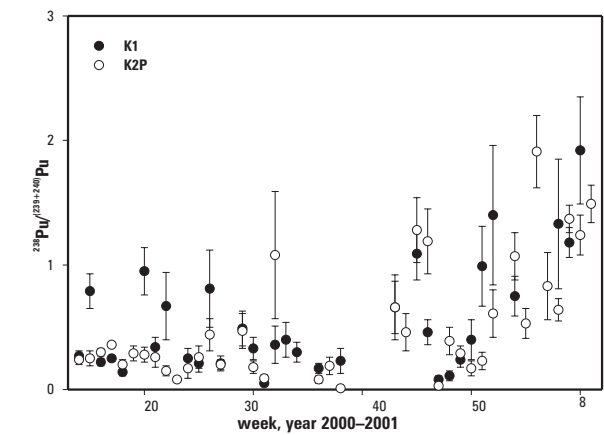


Figure 8. $^{238}\text{Pu} / (^{239+240}\text{Pu})$ in aerosol samples from Kurchatov, Kazakhstan.

4 Conclusions

The determination of total uranium and $^{235}\text{U}/^{238}\text{U}$ ratio in quartz air filters previously used for air filter sampling was problematic. Carbon and oils collected on the filters were not totally removed from the sample solutions in the chemical separation, which increased the non-selective background in the mass spectrum, and thus the uncertainty of the measurement. This was the case especially in isotope ratio measurement where small numbers of counts were collected at the mass 235. By using organic filters, the non-selective background was decreased to 1/3 compared to the solutions prepared from quartz filters; the stability of the background signal was similar with the stability of 1% HNO_3 solution, and the precision of the uranium signal was the same as with standard solutions with the same uranium concentration.

The benefit of extraction chromatography for the analysis of plutonium air filters is that the method is faster than the conventional ion-exchange method. In addition, smaller amounts of

acids are needed in the analysis, and the recoveries are close to 100% in tracer studies

The determination of the radioactivity ratio of ^{241}Pu to $^{239,240}\text{Pu}$ will give additional information on the source and release date of the plutonium found in environmental samples. The activity ratio of ^{241}Pu to $^{239,240}\text{Pu}$ was about 15 in the global fallout in the first half on the 1960s, and according to Paatero et al. [10], the corresponding ratio in the Chernobyl fallout was 95 in 1986. Using the low-background liquid scintillation counter (Quantulus 1220), the detection limit for ^{241}Pu was 8 mBq/sample at a counting time of 600 minutes. In most samples collected from Kazakhstan, the ^{241}Pu concentration was under the limit of detection. To diminish the detection limit and to confirm the spectral purity in the low beta-energy region (β max of ^{241}Pu is 21 keV) some further methodical development for determination of ^{241}Pu by low-background liquid scintillation counting is needed.

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