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Intercomparison for separating Pu and Am from environmental samples by a joint radioanalytical method

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Intercomparison for separating Pu and Am from environmental samples by a joint radioanalytical method

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Abstract

In a previous NKS project “OPTIMETHOD” (2018-2019), an optimised method was presented for separation of Pu, Am and Cm isotopes from NPP reactor water samples. This separation method was tested in an intercomparison organised in 2020 with two solid sample matrices, lichen sample and IAEA-384 reference material, for expanding its usability. The goal of the RAD-MERDE project was to provide an alternative, novel method to previously used fairly established methods for separating actinide isotopes from e.g. environmental and nuclear decommissioning samples. The tested separation method includes iron hydroxide co-precipitation of actinide isotopes and radiochemical separation of Pu and Am with extraction chromatography resins TEVA and DGA. The obtained practical experiences and analytical performance for Pu and Am are discussed in this report and the findings suggest that the development and testing of the separation method should be continued with more solid sample materials.

Key words

Intercomparison; actinides; extraction chromatography; separation method; environmental sample; plutonium; americium

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Final Report from the NKS-B RAD-MERDE [AFT/B(20)4]

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1. Introduction

In the previous NKS project “OPTIMETHOD” (2018-2019), analytical experiences were obtained from several radiochemical separation methods used for determining actinides from NPP water samples. Performance of different radioanalytical methods for separating Pu, Am and Cm isotopes was compared. One of the used method produced high radiochemical recoveries for Pu and Am+Cm and alpha spectra of good quality. Furthermore, no significant problems were reported related to using the separation method. This particular separation method included iron hydroxide co-precipitation and column separations with TEVA and DGA extraction chromatography resins (Hou et al., 2019, 2020).

In the next step, the fore mentioned promising radiochemical separation method for determining Pu, Am and Cm was decided to test with solid sample materials. The aim was to evaluate the functionality of the separation method with sample matrices like nuclear decommissioning materials and environmental samples. An up-to-date, improved separation method for actinides in solid matrices was sought as an alternative to more traditional ion exchange based separation schemes and quite established combinations of UTEVA, TRU and TEVA extraction chromatography resins (Eichrom, 2014a, 2014b; Lemons et al., 2018). This independent continuance of “OPTIMETHOD” was named as “RAD-MERDE” and the project including an intercomparison exercise was executed during 2020 by partners that had worked together in “OPTIMETHOD” already. Originally, before Covid19 pandemia was started, a seminar was planned to be held in connection with the project. The seminar would have been focused on alpha spectrometry and directed to researchers, students and experts working with alpha spectrometric measurements.

2. Experimental

The original idea was to use both solid environmental samples and some NPP or nuclear decommissioning sample materials in the intercomparison. However, due to temporarily difficult in availability of solid NPP or decommissioning samples (mostly due to transportation orders and shipment costs for radioactive samples) it was decided to focus on environmental samples only, and exclude NPP and decom samples from the intercomparison. Two different environmental samples were analyzed and isotopes ^{238}Pu , $^{239+240}\text{Pu}$, and ^{241}Am were determined in these matrices in each laboratory by alpha spectrometry. One laboratory analyzed ^{239}Pu and ^{240}Pu separately by ICP-MS.

One sample material was “Lichen 500/65”. This was mixed lichen (Cl. species) which was collected at Veikkola, Southern Finland (60° 16′ N, 24° 25′ E), on 1st September 1965. Due to early sampling date, global fallout might be the only anthropogenic source of radionuclides in this lichen sample. The lichen sample for intercomparison was prepared in-house by grinding and ashing lichen at 600 °C for 24 h. The ashed sample was ground again and well mixed, assumed to be homogeneous for the investigated radionuclides. The measured ash/dry mass ratio is: 0.0126. Then the ashed sample was divided to equal portions and delivered to intercomparison participants. It has to be noted that throughout this report, the weight of the lichen sample is not for dry lichen, instead, it is given for ash. Therefore, the activity concentration results for the lichen are mBq/gram of ash. 0.10–0.25 grams of lichen ash/subsample was used for radiochemical analysis.

IAEA-384 certified reference material (sediment) was selected as other sample material for intercomparison, This sediment was collected in 1996 in Fangataufa Lagoon, French Polynesia (22° 14′ S, 138° 44′ W), where 137 underground nuclear devices explosions in Mururoa and Fangataufa in 1975-1996, together with 10 safety trials were conducted, and the sediment was contaminated (Povinec & Pham, 2000). This reference material was chosen to intercomparison because it contains practical activity concentrations of investigated radioisotopes (IAEA) considering required sample

mass for analysis, and the material has been mainly considered to be homogeneous. Subsamples of IAEA-384 sediment were delivered to intercomparison participants. 0.4– 1 grams of sediment/subsample was used for radiochemical analysis.

The radiochemical separation method used for determining ^{238}Pu , $^{239+240}\text{Pu}$ and ^{241}Am is presented in Figure 1. The amount of the chemicals used in the procedure are not given here, because the detailed method is unpublished and it might be later published as an article. Sample digestion method was not defined in the analytical instructions and each participating laboratory used their own in-house method for decomposing the sample. Part of the labs ashed IAEA-384 sediment sample prior to wet digestion, and other part of the labs used the sediment sample directly without ashing step. The digestion methods used among participants are slightly different and discussed in section “Modifications done to the original method”. All laboratories used ^{242}Pu and ^{243}Am as radiochemical yield tracers for plutonium and Am/Cm isotopes, respectively.

After individually selected sample dissolution step, iron hydroxide co-precipitation was implemented, for concentrating actinide isotopes to the $\text{Fe}(\text{OH})_2$ precipitate. NaHSO_3 was used for reducing Pu to Pu^{3+} before co-precipitating Pu and Am. Pu^{3+} was converted to Pu^{4+} just before sequential column separation with first TEVA and then DGA column. Part of the laboratories used stacked pre-packed resin cartridges and a vacuum box for the separation and other part used self-packed resin columns and flow rate of the solutions was controlled by gravity. The effluent from TEVA column containing Am was collected and loaded to the DGA column. After a rinse with 0.2 M HNO_3 to remove U and Po, Am was eluted from DGA column with 0.5 M HCl . TEVA column was treated further first optionally with 8 M HNO_3 (for removal of Po, this step is not included in Figure 1 since it was alternative), then with 6 M HCl (for removal of Th) and finally Pu was eluted with $\text{NH}_2\text{OH}\cdot\text{HCl} + 2$ M HCl .

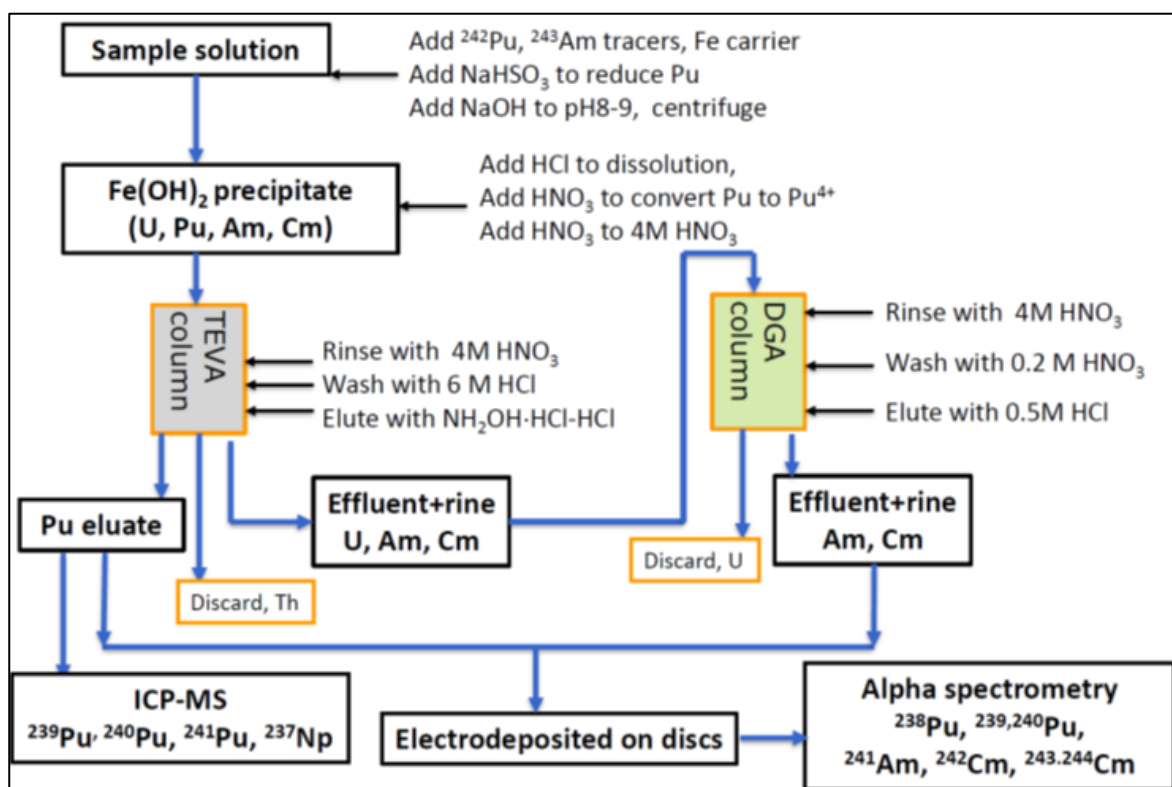


Figure 1. Radioanalytical method used in intercomparison for separating Pu and Am, originally published by Hou et al., (2019, 2020).

Each laboratory prepared alpha targets containing Pu and Am isotopes either by electrodeposition (Talvitie, 1972; Hallstadius, 1984) or by micro-co-precipitation (Sill & Williams, 1981; Hindmann, 1983). The samples were measured with semiconductor alpha detectors for several days. One laboratory also measured ^{239}Pu and ^{240}Pu by ICP-MS in a fraction of the separated Pu samples.

3. Results

3.1 IAEA-384, Pu isotopes

The activity concentrations of ^{238}Pu , $^{239+240}\text{Pu}$, ^{239}Pu and ^{240}Pu in IAEA-384 sediment determined by different labs are presented in Table 1. The reported radiochemical yields for Pu and type of uncertainty are also given. Reference values for activity concentrations of plutonium isotopes were decay-corrected to 1st Aug. 2020 (relatively short half-life of ^{238}Pu , 87.7 years, will cause a difference for different date). In general, the reported activity concentrations of ^{238}Pu and $^{239+240}\text{Pu}$ by the participating labs correspond well with the reference values given by IAEA. The reported activity concentrations of ^{238}Pu are 32.7 (± 2.3) - 41.4 (± 3.6) mBq/g (uncertainties for the lowest and highest values are one sigma and k=2, respectively), whereas $^{239+240}\text{Pu}$ activity concentrations are 98 (± 3) - 107 (± 8) mBq/g (uncertainties for the lowest and highest values are one sigma and k=2, respectively). There is a relative higher variation in the results for ^{238}Pu compared to $^{239+240}\text{Pu}$. This difference between two (or three) Pu isotopes can be seen in Figures 2 and 3 as well. The reported ^{239}Pu and ^{240}Pu activity concentration are 96.6 \pm 9.8 and 16.4 \pm 3.2 mBq/g, respectively, which fit excellently to the information value ranges given by IAEA.

Table 1. Activity concentrations of ^{238}Pu , $^{239+240}\text{Pu}$, ^{239}Pu and ^{240}Pu in IAEA-384 determined in the intercomparison. # corrected to 1st August 2020. * information value.

Sample No.	A ^{238}Pu (mBq/g)	A $^{239+240}\text{Pu}$ (mBq/g)	Pu yield (%)	Uncertainty info	A ^{239}Pu (mBq/g)	A ^{240}Pu (mBq/g)
1	30.8 \pm 1.4	100 \pm 4	88 (average)	k=2		
3	41.4 \pm 3.6	107 \pm 8		k=2	96.6 \pm 9.8	16.4 \pm 3.2
4	38.1 \pm 1.6	98 \pm 3	101	1 sigma		
5	32.7 \pm 2.3	105 \pm 7	43	1 sigma		
6	33.5 \pm 1.7	106 \pm 4	27	1 sigma		
Reference value	32.3 (31.9-32.8) #	107 (103-110)		95% confidence interval	98 (85-105)*	17.5 (15.1-18.7)*

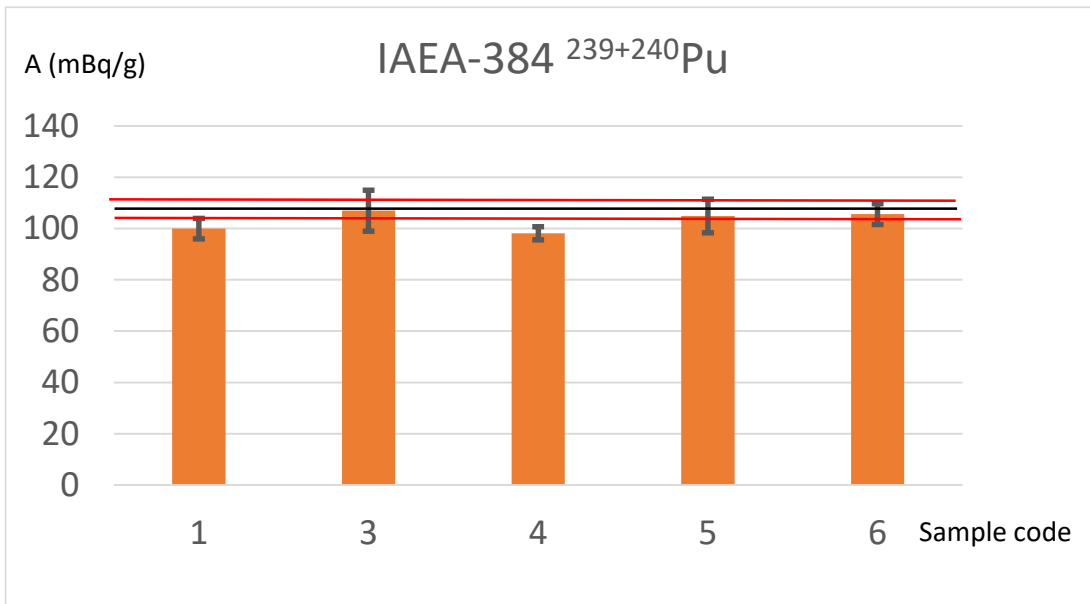


Figure 2. The activity concentration of $^{239+240}\text{Pu}$ in IAEA-384 reference sediment, determined in the intercomparison.

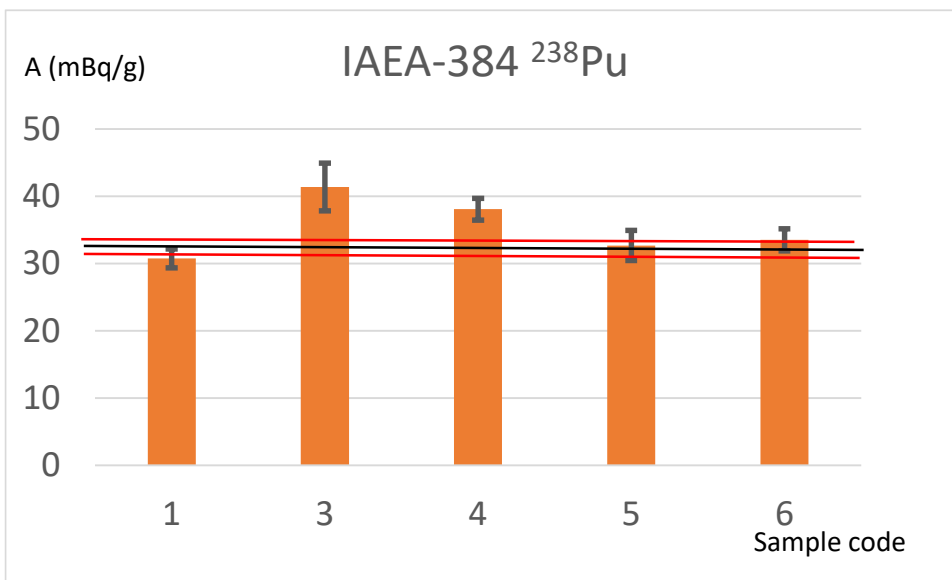


Figure 3. The activity concentration of ^{238}Pu in IAEA-384 reference sediment, determined in the intercomparison.

3.2 IAEA-384, ^{241}Am

The activity concentration of ^{241}Am in IAEA-384 reference sediment is presented in Table 2 and Figure 4. It has to be noted that the ingrowth of ^{241}Am from ^{241}Pu has to be taken into account when comparing the results with the reference value published by IAEA, 7.1 (6.7-7.4) mBq, in 1996. In other words, since 1996 there has been additionally ingrown ^{241}Am in the reference material, as well as decay of ^{241}Am in consideration its half-life of 432 years. In Table 2, a correction has been performed including additional ^{241}Am from the decay of ^{241}Pu and decay of ^{241}Am in 1996-2020 has been included in the reference value and the range of reference values.

The determined values range from 7.6 ± 0.6 to 9.9 ± 0.8 mBq/g (uncertainties for the lowest and highest values are one sigma errors), refer to 1st Aug. 2020. This range is broader than the range of IAEA 7.3-

8.6 mBq/g in 2020, taking into account the additional ingrowth from ^{241}Pu in the sediment since 1996. Nevertheless, the results are close to the recommended range and it has to be noted that the wide range of ^{241}Pu activity concentration, 41-69 mBq (1st August 1996), produces fluctuation in the fraction of additional ingrown ^{241}Am in the reference material.

Table 2. The activity concentration of ^{241}Am (reference date: 1st Aug. 2020) in IAEA-384 determined in the intercomparison exercise.

Sample No.	A ^{241}Am (mBq/g)	Am yield (%)	Uncertainty info
1	8.8 ± 0.8		k=2
3	7.8 ± 1.3		k=2
4	9.9 ± 0.8	99	1 sigma
5	7.6 ± 0.6	64	1 sigma
6	No results were obtained due to low yield and unclear alpha spectra of separated Am fractions		
Reference value	$8.0 (7.3-8.6)\#$		95% confidence interval

In-growth from ^{241}Pu during 1996-2020 in the reference material has been taken into account and added to the reference value and range given by IAEA, for 1st August 1996.

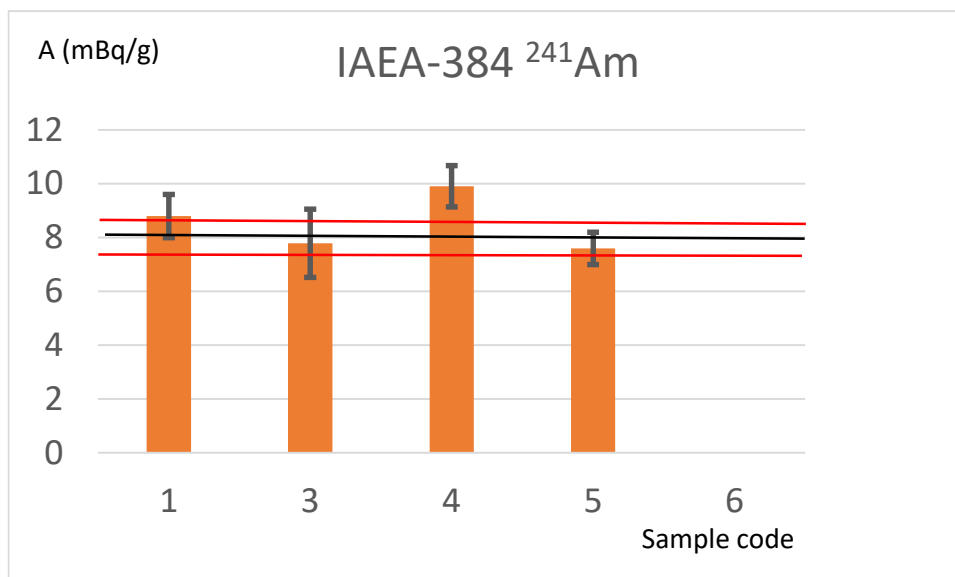


Figure 4. The activity concentration of ^{241}Am in IAEA-384 reference sediment, determined in the intercomparison.

3.3 Lichen, Pu isotopes

The activity concentrations of ^{238}Pu and $^{239+240}\text{Pu}$ in the ashed lichen “500/65” obtained in the intercomparison exercise are given in Table 3, and Figures 5 & 6. The activity concentrations of ^{238}Pu are $6.7 (\pm 0.9) - 13.0 (\pm 1.5)$ mBq/g (uncertainties for the lowest and highest values are one sigma errors) and of $^{239+240}\text{Pu}$ $216 (\pm 6) - 552 (\pm 18)$ mBq/g (uncertainties for the lowest and highest values are one sigma errors). There was a high variation in the activity concentrations and the results seem to fall into two groups, one with lower and another with higher values. The lichen was sampled in 1965 and assumed to be homogenous, containing only radionuclides from global fallout without hot particles. The discussion of sample homogeneity continues in section “Discussion”.

Table 3. The activity concentrations of ^{238}Pu , $^{239+240}\text{Pu}$, ^{239}Pu and ^{240}Pu in lichen “500/65” determined in the intercomparison.

Sample No.	A ^{238}Pu (mBq/g)	A $^{239+240}\text{Pu}$ (mBq/g)	Pu yield (%)	Uncertainty info	A ^{239}Pu (mBq/g)	A ^{240}Pu (mBq/g)
1	10.2 ± 1.4	506 ± 29	65	1 sigma		
2	6.8 ± 2.8	289 ± 35		k=2	161.5 ± 17.7	117.2 ± 25.7
3	11.3 ± 0.9	467 ± 16	88 (average)	k=2		
4	6.7 ± 0.9	216 ± 6	112	1 sigma		
6	13.0 ± 1.5	552 ± 18	49	1 sigma		

3.4 Lichen, ^{241}Am

The analytical results of ^{241}Am in lichen “500/65” are presented in Table 4 and Figure 7. The activity concentration range of ^{241}Am was from 87 ± 4 to 564 ± 48 mBq/g (uncertainties for the lowest and highest values are one sigma errors). The variation in activity concentration was even higher with ^{241}Am compared to Pu isotopes. The results for ^{241}Am further confirms the observation that the lichen sample from 1965 is not homogeneous and it might contain particles with high radioactivity content.

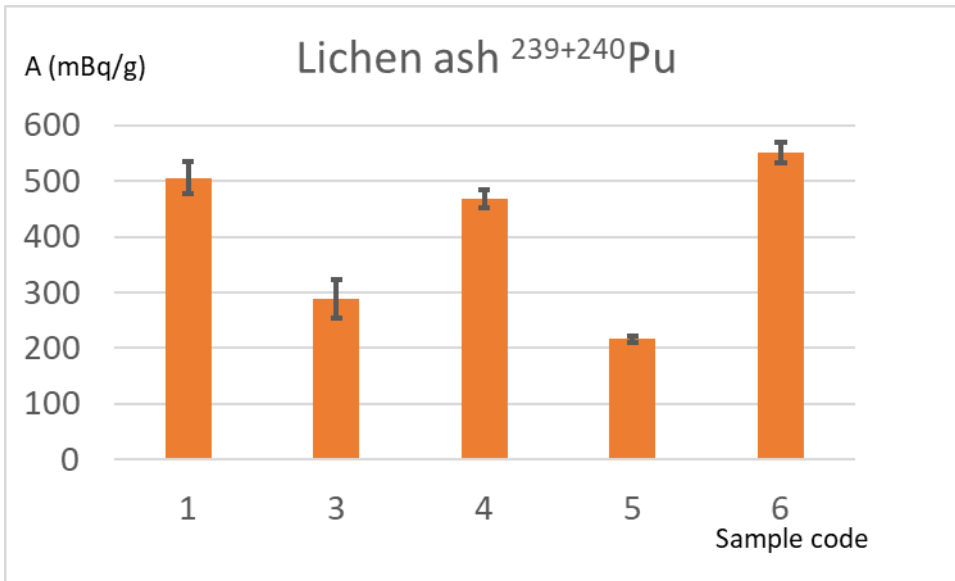


Figure 5. The determined activity concentration of $^{239+240}\text{Pu}$ in “Lichen 500/65”.

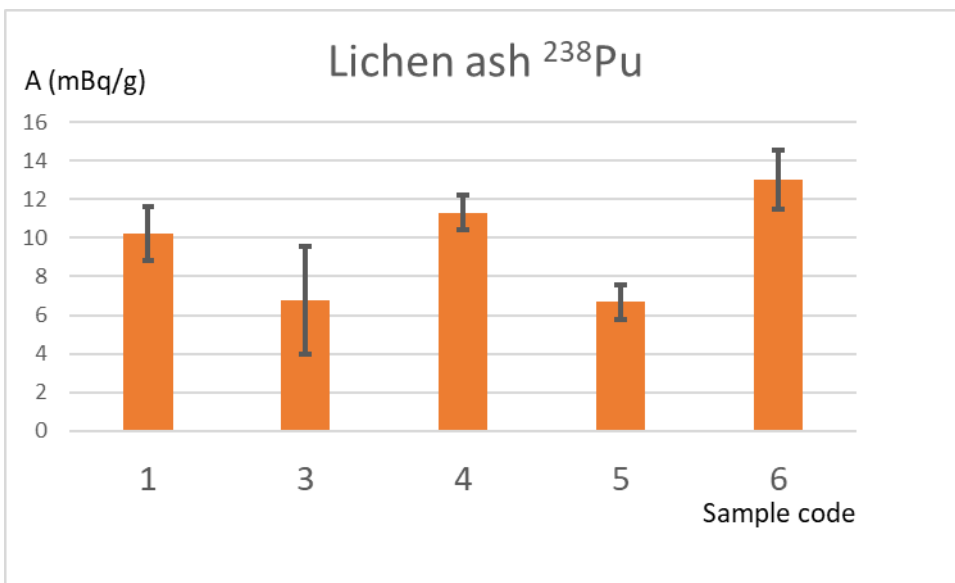


Figure 6. The determined activity concentration of ^{238}Pu in “Lichen 500/65”.

Table 4. The activity concentration of ^{241}Am in lichen sample “500/65” determined in the intercomparison.

Sample No.	A ^{241}Am (mBq/g)	Am yield (%)	Uncertainty info
1	217 ± 12	99	1 sigma
2	289 ± 28		k=2
3	210 ± 8		k=2

4	87 ± 4	92	1 sigma
6	564 ± 48	33	1 sigma

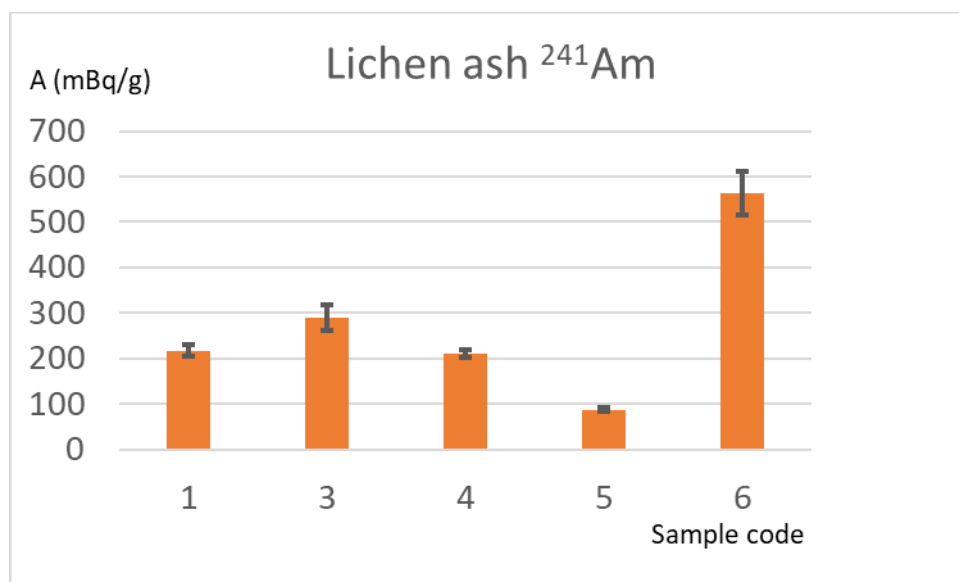


Figure 7. The determined activity concentration of ²⁴¹Am in “Lichen 500/65”.

4. Discussion

4.1 Modifications on the analytical method

Although the radiochemical separation procedures used for determining Pu and Am in solid environmental samples were quite uniform among different labs and the purpose in the intercomparison was to follow the original method as carefully as possible, some individual modifications were performed to the basic method in the labs. The most variable step in the separation scheme is probably sample dissolution method. In three labs, leaching using *aqua regia* under heating on a hot plate was used for sample dissolution. Two labs used microwave oven digestion for leaching plutonium and americium, one lab with concentrated HNO₃ and another with combination of HNO₃ + HCl + HF. Although all these sample digestion methods are considered to be effective for releasing actinides from the sample to the acid solution, there might still be small differences between the dissolving efficiency.

Some labs used NH₃ in Fe(OH)₃ co-precipitation step, instead of NaOH. One lab did not perform Fe(OH)₃ co-precipitation step at all. Some additional in-house steps were used for oxidation state adjustment of Pu before column separation. Pu was then stabilized either as Pu⁴⁺ by NaNO₂ and heating the sample solution, or by a sequential addition of ferrous sulphamate, ascorbic acid and NaNO₂. One lab used 0.1 M HCl + 0.05 M HF + 0.03 TiCl₃ for eluting Pu from TEVA column, instead of NH₂OH·HCl + 2 M HCl. Two labs prepared their alpha counting samples by electrodeposition and three labs by micro-co-precipitating actinides with NdF₃ or CeF₂.

Maybe the clearest difference in user experiences came from the form of used extraction chromatography resins. Part of the labs used pre-packed cartridges/columns and a vacuum box for

controlling the flow rate. These participants were more satisfied with separation speed. With a vacuum box system, separation with TEVA+DGA chromatography is fast and takes only a couple of hours. Participants who used self-packed resin columns and gravity-based flow rate complained about the slowness in the step using DGA column. It took almost a full working day to perform the purification of Am with DGA column. Also prehandling (soaking) of DGA resin for a self-packed column requires several days, due to floating/hydrophobic nature of this resin, and probably centrifugation of the resin is always needed for a complete soaking of the resin before loading it to the column. Based on these experiences, the use of a vacuum box and pre-packed cartridges is more favorable in case of using DGA resin for radiochemical separations and some participants aim to take them into use in their future works.

4.2 Performance of the separation method

Taking into account the reported uncertainties of the determined activity concentrations, most of the analytical results for $^{239+240}\text{Pu}$, ^{241}Am and even for ^{238}Pu in IAEA-384 sediment by different labs are in a fairly good agreement with corresponding values from other labs and the reference values. Limited number of activity concentration results – five result sets for Pu isotopes and ^{241}Am – together with highly varying concentrations in lichen sample “500/65” make statistical analysis of the results unfeasible. The radiochemical yields for Pu was 27-101% (four values) and 64-99% for Am (two values) for IAEA-384 reference sediment and for lichen “500/65”, the corresponding values were 49-112 (four values) and 33-99% (three values), respectively. Again, no clear conclusions can be drawn from only few reported yield values and comparison to other corresponding radiochemical separation method is difficult, if it is based only on representativity of the determined activity concentrations and radiochemical yields. More sample analyses would be needed for that, and preferably from more sample matrices and homogenous sample materials.

The main findings of this intercomparison were more like qualitative than quantitative, since the obtained data set is limited regarding statistical data analysis, and practical experience of using the novel method was in import part of this intercomparison. The separation method for Pu and Am shared in this exercise was tested first time ever in four labs, where other established separation methods have been in use. Most of the user comments were positive (see the next section where free quotations from the users are summarized) and the method will be used, as such or as customized, in some of the labs in different projects. Therefore, this intercomparison has managed to share knowledge among Nordic partners and disseminate novel separation methods for transuranium elements. Participation to the intercomparison has facilitated implementation of an advanced method greatly, thus improving the method development work in each lab. A close co-operation of Nordic radiochemistry laboratories in nuclear industry, research institutes, universities and authorities was further strengthened by this inter-comparison.

4.3 Comments about the tested method

”I liked to use this method. In our own method there are more time-consuming evaporation steps compared to this one with more precipitation and centrifugation steps. I used plastic labware with this method, as normally we use more glassware in analysis. The use of DGA resin was surprisingly easy after shared tips in our project meeting. We need to develop a vacuum system for further use of the separation method in our lab.”

“I like the method since it seems to be robust. The alpha counting sample discs and alpha spectra look very good.”

”I plan to use this separation method for Pu/Am determination in my forthcoming projects.”

”DGA separation was extremely slow, we need to take vacuum box in use, if we want to perform DGA separations in future”

”My bad yields for Pu and Am by other separation methods were quite same with this new method. I’ve had doubts that some reagent, resin or working step with an old UTEVA+TRU method I’ve been using, would be ruined or incorrect. At least it was now proved that the fault is not in any specific reagent or resin used with old methods.”

4.4 Inhomogeneity of actinides in the analyzed lichen material

While selecting suitable sample materials for this intercomparison, it was assumed that there would be no hot particles in Finnish lichens collected in early 1960’s. At that time the only radioactive source in Finnish environment was global fallout from nuclear weapons testing, which produced uniform radionuclide deposition and only tiny hot particles. Compared to bigger, unevenly deposited hot particles from the Chernobyl accident in Finnish environment, the smaller hot particles from global fallout haven’t been as abundant, or at least they have been rarely detected in Finland (Paatero et al., 2010). The highly varying activity concentration of all investigated radionuclides in mixed lichen sample “500/65” can’t be explained with any other reason than sample inhomogeneity. As the sample was homogenized and ashed to a fine powder, the reason left for inhomogeneity is presence of hot particles in the lichen.

Another possible reason for inhomogeneity of the lichen sample might be the ashing temperature 600 °C since in some studies ashing temperature over 450 °C has caused the formation of refractory plutonium (Wang et al., 2015) which doesn’t necessarily dissolve to acids. This might yield in partial and varying dissolution of plutonium. However, opposite views exist and ashing temperature 500-600 °C has been considered safe even for long times, whereas temperatures 700-1000 °C should be avoided in order to have plutonium in easily leachable form in the sample (Nielsen & Beasley, 1965). Since performance of ashing and dissolution procedures are not undeniable or uniform in different researches, possible effect of ashing temperature on the variation in plutonium isotope concentrations remains open and should be investigated separately.

After intercomparison it was noted that hot particles actually have been detected in fallout samples (aerosol and rain water) in Finland in early 1960s already (Figure 8, Mattsson et al., 1965). This implies that the big variation of the determined results of plutonium isotopes and ²⁴¹Am in the lichen in this work might be attributed to the inhomogeneity of these radionuclides due to the presence of hot particles in this sample. This is unfortunate as the sample materials in a radioanalytical intercomparison should be as homogeneous as possible (Bowen, 1978), and selection of sample matrix turned out to be unsuccessful in this respect.

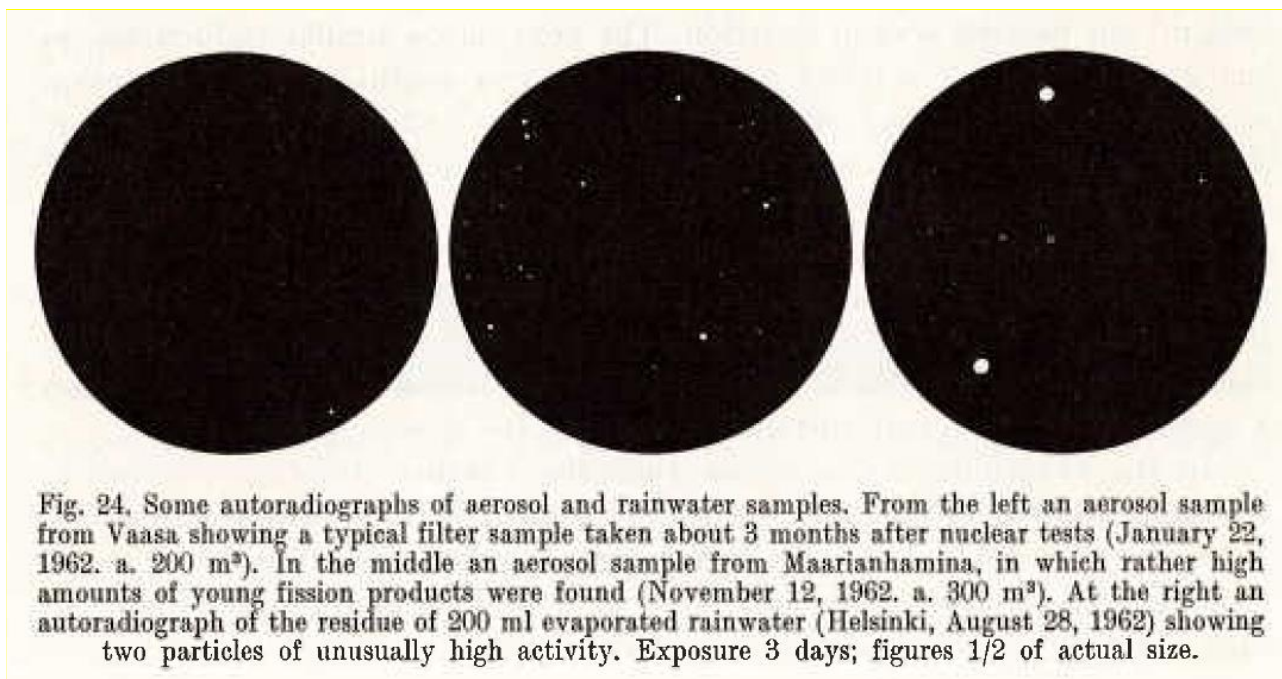


Figure 8. Hot particles found in Finland in 1962, originating from atmospheric nuclear weapons testing. (Mattsson et al., 1965).

5. Summary and future prospects

Two goals were expected in the RAD-MERDE project. First, an intercomparison exercise was arranged, for testing the functionality of the radiochemical separation method for Pu and Am, the same method based on TEVA and DGA extraction chromatography resins was shared among participating laboratories. The findings from the intercomparison were promising, because the obtained activity concentrations for ²³⁸Pu, ²³⁹⁺²⁴⁰Pu and ²⁴¹Am in IAEA-384 were adequately similar to reference values. For most laboratories, the newly adopted method provided better radiochemical yields, faster analysis or more convenient separation procedure compared to their older previously used methods. The general attitude towards the tested method was very positive afterwards and it will be implemented in more laboratories from now on. It can be concluded that this intercomparison increased greatly exchange of experiences and radioanalytical competence of the participating Nordic laboratories.

Obtained widely varying results of plutonium isotopes and ²⁴¹Am in the analysed lichen sample, in turn, proved selection of this sample material unsuccessful. On the other hand, this newly occurred evidence of hot particles in Finland from old global fallout may be beneficial for other radioecological and environmental radioactivity studies in Nordic countries.

This collaboration created a need for further development of tested radioanalytical method. At the next stage, more solid sample matrices should be tested and preferably homogenous environmental and/or NPP or nuclear decommissioning materials. First stage in 2020 proved the vacuum box based extraction chromatography system to be more convenient than gravity-based flow rate control, and it would be useful to continue testing with focus on vacuum box systems. More data, i.e. determined radioactivity concentrations and radiochemical yields, is needed from different sample matrices for eventually validating the novel radioanalytical method in Nordic labs.

Another project goal was to arrange a user workshop or seminar focusing on alpha spectrometry for all interested scientists and students from Nordic countries in 2020. Unfortunately, this goal was not

reach due to Covid-19 and general lockdown during 2020. Consequently, the Nordic alpha spectrometry seminar will be postponed, hopefully in 2022.

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Disclaimer

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Title	Intercomparison for separating Pu and Am from environmental samples by a joint radioanalytical method
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Abstract max. 2000 characters	In a previous NKS project “OPTIMETHOD” (2018-2019), an optimised method was presented for separation of Pu, Am and Cm isotopes from NPP reactor water samples. This separation method was tested in an intercomparison organised in 2020 with two solid sample matrices, lichen sample and IAEA-384 reference material, for expanding its usability. The goal of the RAD-MERDE project was to provide an alternative, novel method to previously used fairly established methods for separating actinide isotopes from e.g. environmental and nuclear decommissioning samples. The tested separation method includes iron hydroxide co-

precipitation of actinide isotopes and radiochemical separation of Pu and Am with extraction chromatography resins TEVA and DGA. The obtained practical experiences and analytical performance for Pu and Am are discussed in this report and the findings suggest that the development and testing of the separation method should be continued with more solid sample materials.

Key words Intercomparison; actinides; extraction chromatography; separation method; environmental sample; plutonium; americium

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