

Inter laboratory comparison for ^{241}Am determination in environmental samples

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Abstract. ^{241}Am is an alpha emitter which originates from the successive neutron capture reactions by plutonium isotopes in a nuclear reactor : $^{239}\text{Pu} (n,\gamma) \rightarrow ^{240}\text{Pu} (n,\gamma) \rightarrow ^{241}\text{Pu} \xrightarrow{\beta} ^{241}\text{Am}$. Activation products like ^{241}Am may contribute significantly to human exposure in case of an accidental release in the environment. Therefore, this isotope is monitored together with plutonium isotopes in environmental matrices near nuclear facilities. In addition, because of its ^{241}Pu filiation, contribution of ^{241}Am in the environment will increase in future years.

^{241}Am in sands was measured by three different techniques : direct gamma ray spectrometry, alpha spectrometry and sector field ICP-MS. Prior to alpha and ICP-MS measurements by isotope dilution, americium was separated from major components of the matrix and other transuranic elements by anion exchange and extraction chromatography. The results of two different laboratories are compared.

1. INTRODUCTION

Release of ^{241}Am into the environment is due to different human activities. The main source is the reprocessing of nuclear plant spent fuel. As an example, in 1997 50 GBq were released into the sea by Sellafield reprocessing plant and more recently in 1999 3.5 GBq of ^{241}Am were released into the sea by La Hague reprocessing plant. Industrial and medical uses are the other source of the presence of ^{241}Am in the environment. For instance, Americium is the main component of fume ionic detectors and pace makers. Sealed sources of ^{241}Am are also routinely used to calibrate measurement devices.

Americium in the environment presents different chemical forms more or less soluble, which can enter the food chain. Therefore, americium, which may contribute to the human exposure, is monitored in environmental matrices in the surroundings of nuclear facilities. In addition, because of its ^{241}Pu filiation, contribution of ^{241}Am in the environment will increase in the future. The analytical techniques most commonly used to quantify americium are gamma and alpha spectrometry. Gamma spectrometry requires only a homogenous dried or ashed sample whereas alpha spectrometry requires separation of the element of interest from the rest of the matrix and above all from any other potential interfering element.

Because alpha spectrometry is limited by its slow throughput, alternative methods were developed. ICP-MS is one of the faster of the alternative methods proposed for long live radionuclide determination [1].

Recent developments in high resolution or Sector Field ICP-MS (HR ICP-MS, SF ICP-MS), leading to higher transmission and lower background levels, have decreased detection limits to only a few $\text{pg}\cdot\text{kg}^{-1}$ [2]. ICP-MS offers substantial advantages over conventional radiometric techniques for radionuclides with half lives longer than 10^4 years [3]. Although ^{241}Am is not the most favourable isotope for ICP-MS determination because of its relatively short half-life (432 y), this isotope can be concentrated sufficiently in some samples to be rapidly detected by SF ICP-MS [4].

The present paper will present the comparison carried out by two laboratories for the determination of ^{241}Am in two sand samples originating from Republic of Belarus.

2. EXPERIMENTAL SECTION

2.1 Laboratories

- Laboratory N°1 (Lab N°1) or « Laboratoire de Mesure de la Radioactivité de l'Environnement (LMRE) »
- Institute for Nuclear Safety and Protection (IPSN), Bois des Rames 91400 ORSAY, France
- Laboratory N°2 (Lab N°2) or Veterinary Laboratories Agency (VLA), Radiochemistry Unit, Woodham Lane, New Haw, Addlestone, Surrey_KT15 3NB, United Kingdom

2.2 Samples

One aliquot of 100 g of two sand samples, identified as sample 1B and 2A, collected in Republic of Belarus after the Chernobyl accident (collected 1998) were provided to each laboratory. Sample 1B represents 5 to 10 cm depth of the core and sample 2A the first 5 centimeters. Both samples were sieved at 500 μm . The aliquots given to each laboratory could be sub sampled to process the assays.

2.3 Methods

2.3.1 Gamma spectrometry

^{241}Am is determined by direct gamma spectrometry from the dried sand samples. In both laboratories, gamma spectrometry is performed with N-type germanium detectors equipped with 0.5 mm thick beryllium window and allowing a relative efficiency higher than 50% (Lab N°1) and 24 % (Lab N°2) in respect to ^{60}Co energy (1.33 MeV). Detector crystal volumes are respectively higher than 200 and 115 cm^3 . Measurements are carried out in the 25 keV-2.5 MeV energy range [5].

In Lab N°1, these detectors are set in a room located two storeys below ground level under a 3 m thick boron concrete paving. All the walls of this room are covered with 10 cm thick lead bricks of low background activity, lined with 0.5 cm thick electrolytic copper tiles in order to neutralize lead X-rays. Room materials were chosen for their low radioactivity level. Plexiglas casing swept by gaseous nitrogen insulated each detector and the circulation of filtered air permit to reduce levels of radon and its daughters [6]. In Lab N° 2, the detectors are situated at ground level. Background reduction is achieved by using lead castling of 10 cm thickness. Cadmium then copper sheeting inside the castling provide attenuation of lead X-rays.

Lab N°1 detectors are calibrated with Analytix mixed nuclides, including ^{241}Am , solid sources (water-equivalent density), in the laboratory containers (17 to 380 ml). Lab N° 2 detectors are calibrated with Amersham mixed nuclides, including ^{241}Am , aqueous standard, in laboratory containers typically 30 to 2500 ml.

Sample and standard source measurements are carried out close to the detectors. The counting time is set at 80 000 seconds (Lab N°1), in Lab N°2, for the sands 580 000 seconds and 274 000 seconds were used for samples 1B and 2A respectively.

^{241}Am is characterized by gamma emission at 59,5 keV (emission probability: 35,9 %). For energies higher than 100 keV, self-absorption correction only depends on sample density. Below 100 keV, matrix composition must be taken into account. Commercial software usually proposes an average sample composition, which must be used carefully below 100 keV. Therefore, self-absorption correction factor is also determined experimentally using an external source of ^{241}Am by transmission measurement or generated from Gamma Tool software (Amersham) in order to calculate mass energy-attenuation coefficient in the sample matrix [7].

2.3.2 Alpha spectrometry and ICP-MS

Before alpha spectrometry measurement, americium was chemically isolated from the rest of the matrix elements and other actinides [4]. Each laboratory carried out its own chemical separation method.

2.3.2.1 Chemical separation

The chemical separations carried out by Lab N°1 and 2 are summarized in figure 1. The chemical separation of Lab N°1 was designed to allow the alpha spectrometry measurements as well as the ICP-MS measurements. After alpha counting, the plated sample was dissolved again and iron removed with a 9M HCl AG1 X4 column. Samples were then evaporated to dryness and dissolved in 3 ml of 0,5 M HNO₃ for ICP-MS measurement. Overall, in both laboratories, chemical recoveries varied from 40 to 80 % and samples were plated according to the Talvitie method [9]. Counting time was set to 3 to 14 days in Lab N°1, and to 3 days in lab N°2.

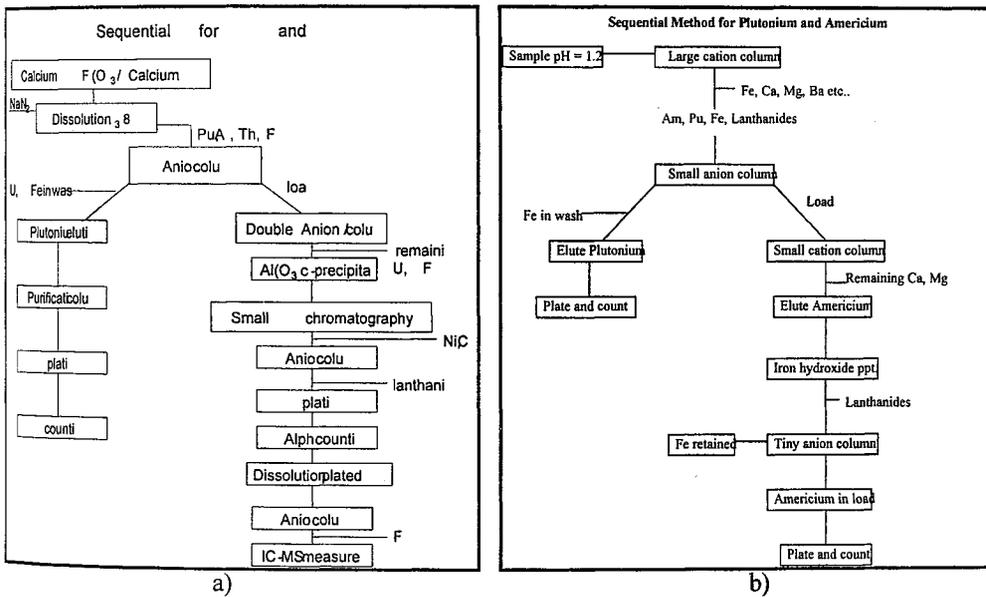


Figure 1: Chemical separation scheme, a) Lab N°1, b) Lab N°2

2.3.2.2 Detection by alpha spectrometry

The alpha detectors are 300 to 450 mm² passivated implanted planar silicon (PIPS) detectors in both laboratories.

2.3.2.3 Detection by SF ICP-MS

Measurements were performed in Lab N°1 only with a Sector field ICP-MS, the Axiom single collector from VG Elemental (Winsford, Cheshire, UK). This mass spectrometer is equipped with a double-focusing magnetic sector mass analyser of forward geometry. For sample uptake, a very specific introduction system was used, the MCN 6000. It consists of a microconcentric nebulizer in series with a desolvator membrane (Cetac, Omaha, NE, USA). All ICP-MS experimental conditions have already been described elsewhere [4, 10].

3. RESULTS AND DISCUSSION

Figure 2 represents the gamma spectrum of sample 2A. An example of alpha spectrum of sample 1B is given in figure 3 and figure 4 represents an example of ICP-MS spectrum of sample 2A. All the results relative to sample 1B and 2A are presented in figures 5 and 6 respectively. The two samples have very different ranges of activity.

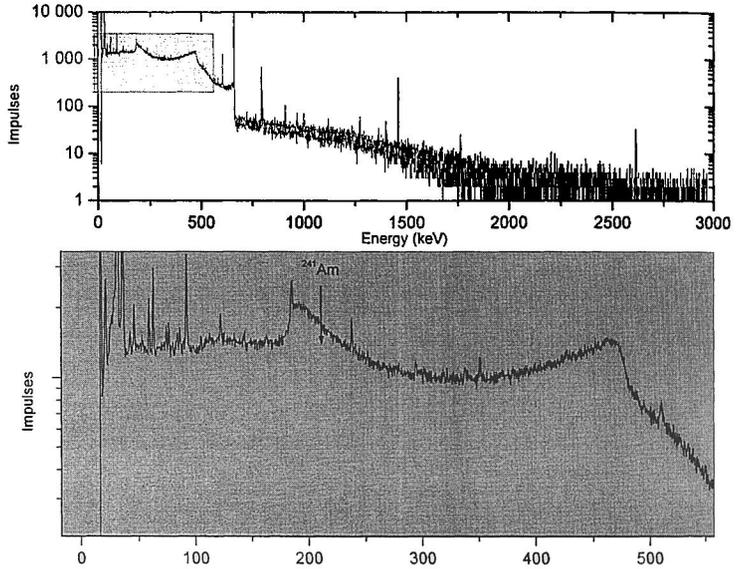


Figure 2: Sample 2A, gamma spectrum

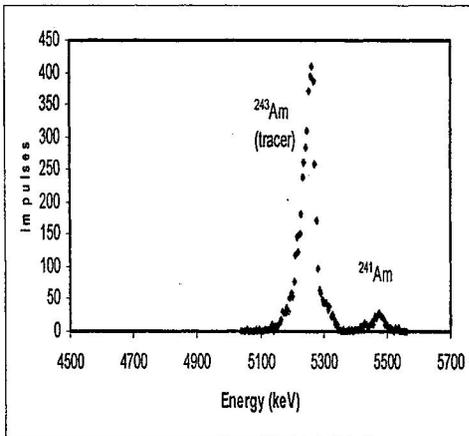


Figure 3: Sample 1B, alpha spectrum

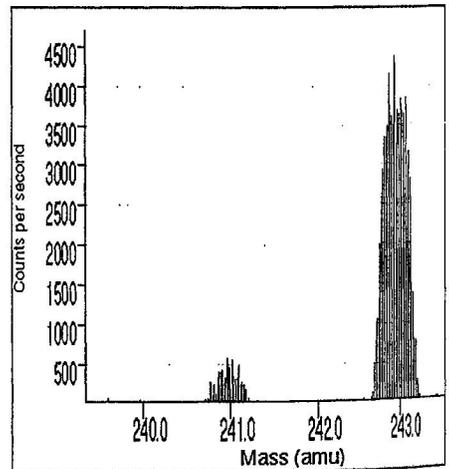


Figure 4: Sample 2A, ICP-MS spectrum

3.1 Alpha spectrometry and ICP-MS

Alpha spectrometry and ICP-MS results of Lab N°1 are relative to the same sub sample as each ICP-MS measurement was performed after dissolution of the alpha plated sample (figure 1a). For sample 1B, more representative of radioactivity levels occurring in the environment, less than 1 picogram in one kilogram of sand, alpha spectrometry and SF ICP-MS results are in good agreement as it has already been showed [4]. Alpha spectrometry results of the two laboratories are similar. For sample 2A with radioactivity levels higher, whatever measurement techniques, results are in good agreement. The results from Lab N°2 show a higher uncertainty because of the lower counting time, 3 days instead of 14 days in Lab N°1.

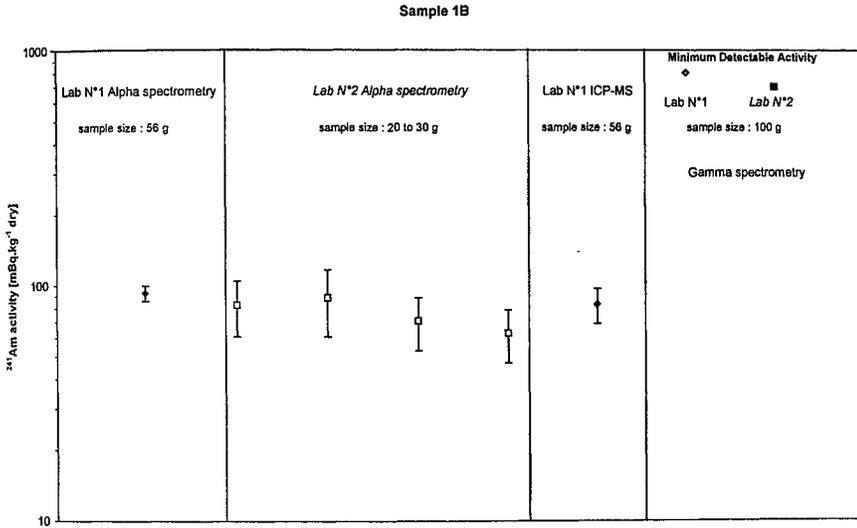


Figure 5: (²⁴¹Am ± 2s) in sample 1B (mBq.kg⁻¹ dry), comparison of all results

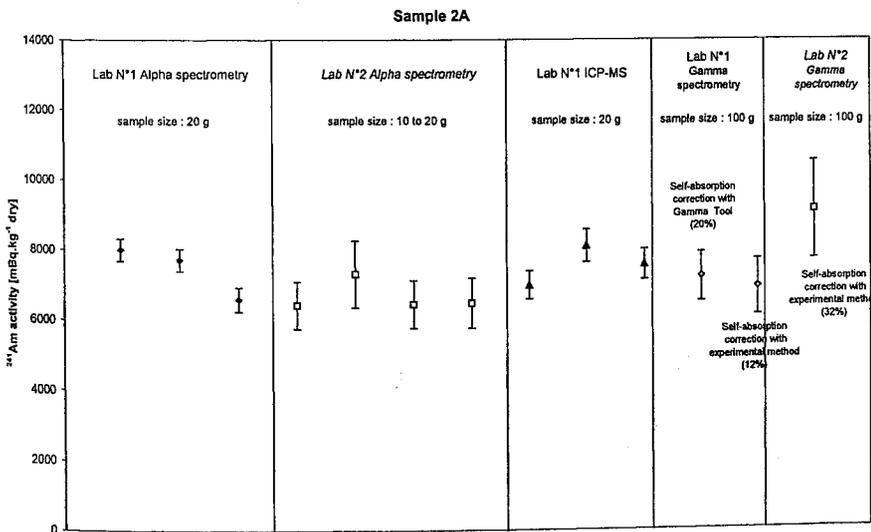


Figure 6: (²⁴¹Am ± 2s) in sample 2A (mBq.kg⁻¹ dry), comparison of all results

3.2 Gamma spectrometry

For sample 1B, results by gamma spectrometry measurement of the two laboratories are below the minimum detectable activity because of the low level of the sample.

For sample 2A, all results of Lab N°1 are in good agreement whichever technique is considered. In figure 6 are presented the results corrected by Gamma Tool and by the experimental self-absorption factor. The difference between the two results of Lab N°1 is due to the limits of the commercial software (Gamma Tool) below 100 keV.

4. CONCLUSION

This paper allowed comparison of three different techniques for ^{241}Am determination, two techniques are well established and conventional, gamma and alpha spectrometry, the other one, SF ICP-MS is rather newer.

Depending on ^{241}Am content, one technique can offer substantial advantages compared to the others. For environmental samples, gamma spectrometry is ideally suited to ^{241}Am determination as a screening method. For low level, two techniques requiring chemical preparation can be chosen : alpha spectrometry or SF ICP-MS. Even if alpha spectrometry measurement gives lower minimum detectable activity (factor of 10 improvement (sample size : 200 g)), results are obtained faster with ICP-MS (counting time : few minutes compared to 14 days for alpha spectrometry) [11]. For the two samples analyzed, these latest techniques are in good agreement. Whereas the chemical preparations of the two laboratories are different, results of the two laboratories are similar.

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