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Validation of aerosol low-level activities by comparison with a deep underground laboratory

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HIGHLIGHTS

- LARA laboratory in Barcelona validated measurements of ^{40}K and ^{137}Cs in air filters.
- Validation by an exchange programme with an underground laboratory.
- Baselines activities of ^{40}K and ^{137}Cs in Barcelona air of 2005–2010 are presented.

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ABSTRACT

The activities on air filters of the radionuclides ^{40}K and ^{137}Cs were measured in an above ground laboratory and compared with the activities measured in an underground laboratory. The average relative differences were lower than 20% for ^{137}Cs and ^{40}K. The temporal series of these radionuclides in the period 2006–2010 are also presented. These results form the baseline of these radionuclides in the studied period, and are particularly interesting because they correspond to the radioactive background in Barcelona before the Fukushima accident.

1. Introduction

The Radiochemical and Radioactivity Analysis Laboratory (LARA) of the Institute of Energy Technologies of the Technical University of Catalonia has been working in collaboration with the Spanish Nuclear Safety Council since March 2000 as one of the laboratories of the Spanish Sparse Network for Environmental Surveillance (De Cort et al., 2005). Although atmospheric radioactivity is dominated by natural radionuclides ^{7}Be and ^{210}Pb other radionuclides also detected are ^{40}K and ^{137}Cs being the latest ones present at very low activities, near the Minimum Detectable Activity (MDA) of a surface laboratory. Despite the fact that it is fairly well known that measurements near the MDA should be avoided, nevertheless, measurements should be made, and results reported, whenever the case warrants it.

The aim of this work is the validation of the determination of the activities of radionuclides collected on aerosol filters from high-volume air dust samplers including activities close to MDA. For activities greater than the MDA we have followed an in-house protocol based on Eurachem recommendations (Eurachem, 1998) including precision test of repeatability and accuracy. To validate activities close to MDA an exchange exercise with a deep underground laboratory, Laboratori Nazionali del Gran Sasso (LNGS) in Italy, was chosen due to its very low cosmic background. The evaluation of these results is one of the elements of the validation process.

Once the method and the corresponding measurements were validated, the temporal series of the activity concentrations of ^{137}Cs and ^{40}K in the period 2006–2010 are presented. These results are the baseline of these radionuclides in the studied period, and are particularly interesting because they correspond to Barcelona's radioactive background before the Fukushima accident.

2. Materials and methods

2.1. Sampling

Airborne particulate sampling has been carried out using an air dust sampling station (ASS-500) located in Barcelona (Spain).

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G3 $(44 \times 44)\,cm^2$ polypropylene filters were used for the preparation of the calibration standards, the blanks and samples collection.

2.2. Instruments

At LARA for the measurements of filters, Canberra HPGe coaxial detectors model GX3020 and a cryostat with a Be window model GX4020 and a cryostat with a Carbon Epoxy window were used. Their nominal efficiencies are 33% and 41% respectively, and the resolutions are 1.77 and 1.86 keV at 1.33 MeV of ^{60}Co.

In Italy at LNGS, the exchange samples were measured using an ORTEC HPGe detector (p-type coaxial, 85.5% relative efficiency, 0.5 mm Cu entrance window).

2.3. Validation methodology

2.3.1. Preparation of calibration standards and reference material (RM)

The filters standards for calibration of germanium detectors and the reference material were prepared by spiking the filters with Amersham standard solution with gamma ray emitting radionuclides in the energy range 59.5–1332.5 keV and drying it using an infra-red lamp. To get a uniform distribution of the standard solution over the filter, squares of $1 \times 1\ cm^2$ were drawn on the filter and droplets of 0.1 mL were spiked on each square. After preparation the filter was folded and pressed to obtain a surface area of about $8 \times 8\ cm^2$ and a thickness of 1.6 cm and placed in a square plastic box with the active area inwards.

The RM was prepared by the "Nuclear and Radiological Engineering Group" at University of the Basque Country and was used to check the accuracy of the method.

2.3.2. Exchange of environmental samples for validating low-level activities

The exchange sample exercise included measuring the calibration standard, one blank and seven filters collected at Barcelona (Spain) in which the nuclides ^{40}K and ^{137}Cs were detected at activities close to MDA. All the filters were folded and pressed in the same way to get the same geometry. Selection was made covering almost the range of detected ^{137}Cs activities in the period 2006–2010 which were between 0.1 and 1 $\mu Bq/m^3$.

2.4. Quality assurance

The LNGS is participating regularly to proficiency tests, which show that the results obtained on different geometries for unknown sample activities are well reproduced within 5% to 10%.

LARA has been accredited according to ISO/IEC 17025 requirements (ISO, 2005) in measuring radionuclides in aerosols samples.

3. Results

3.1. Comparison of two standards

Two standards were prepared by different technicians at LARA in the way described in Section 2.3.1. They were measured in two detectors and these results were used to evaluate variations due to the source heterogeneity. The repeatability was calculated by the relative standard deviation (RSD) in percentage. Fig. 1 shows both averages RSD in experimental efficiencies and those calculated using the fitting functions obtained for the radionuclides spiked in the standards. RSD is below 5% for photon energies between 60 and 1332 keV and no differences between detectors are observed. These results are in agreement with those published

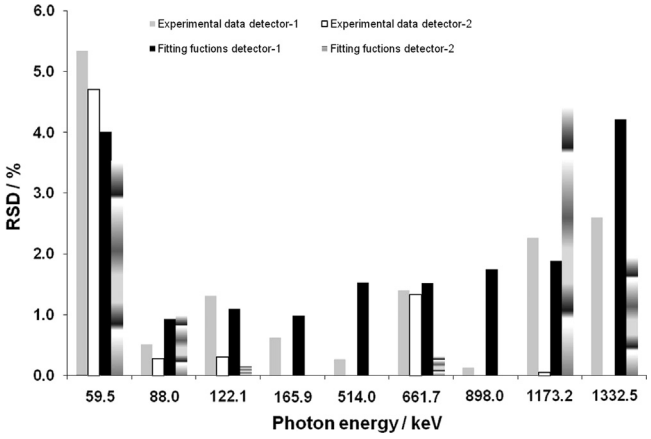

Fig. 1. Comparison of two standards.

Table 1
Accuracy in the above ground laboratory using reference material.

Radionuclide	Energy (keV)	Reference activity (Bq/filter)[a]	Detected activity (Bq/filter)[a]	Relative bias (%)
Am-241	59.5	360 ± 9	383 ± 32	6.5
Cd-109	88.0	141 ± 9	146 ± 9	3.7
Co-57	122.1	0.85 ± 0.01	0.91 ± 0.09	6.5
Cs-137	661.6	298 ± 6	321 ± 12	7.4
Co-60	1332.5	210 ± 3	215 ± 15	2.5

[a] Uncertainties are expanded with a coverage factor $k=2$.

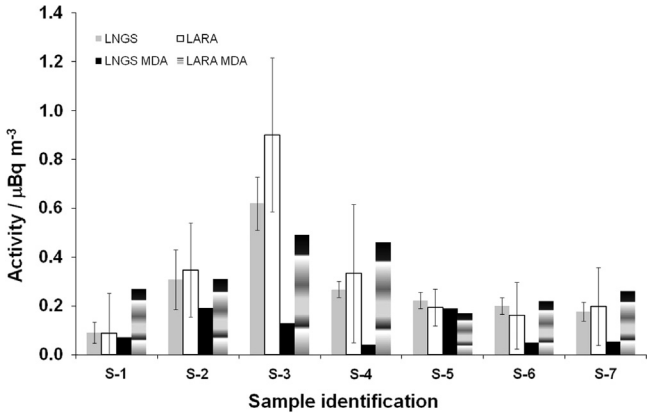

Fig. 2. ^{137}Cs activities and MDA in the exchange samples.

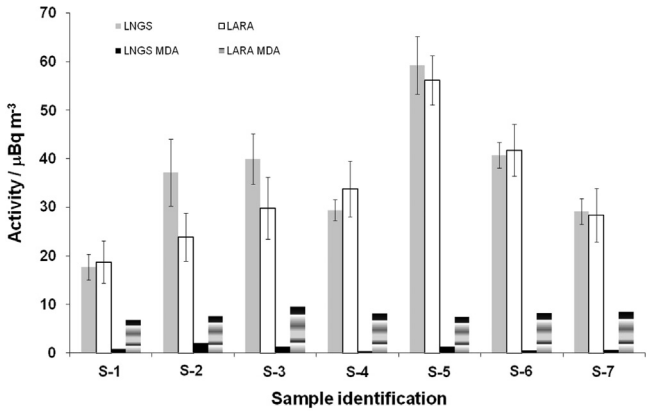

Fig. 3. ^{40}K activities and MDA in the exchange samples.