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Characterisation of the IAEA-375 Soil Reference Material for radioactivity

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HIGHLIGHTS

- Upgrade of the IAEA-375 Soil Reference Material (RM).
- Massic activity values traceable to the SI units.
- ^{40}K , ^{134}Cs , ^{137}Cs , ^{212}Pb , ^{212}Bi , ^{214}Pb and ^{214}Bi measured by γ -ray spectrometry.
- Chemical separation of Sr by extraction chromatography.
- ^{90}Sr assessed by liquid scintillation counting.
- The RM was used for the 2010 EC Interlaboratory Comparison on Radionuclides in Soil.

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ABSTRACT

The Joint Research Centre Institute for Reference Materials and Measurements (JRC-IRMM) participated in a research project initiated by the International Atomic Energy Agency (IAEA) to upgrade some of its existing reference materials (RMs). The aim of the work described in this article was to determine the activity concentration of a series of radionuclides in the IAEA-375 soil RM with values traceable to the SI units. The radionuclides ^{40}K , ^{134}Cs , ^{137}Cs , ^{212}Pb , ^{212}Bi , ^{214}Pb and ^{214}Bi were measured by γ -ray spectrometry after drying the sample and placing it in a suitable container. The ^{90}Sr was assessed by liquid scintillation counting after dissolution of the soil by wet digestion and chemical separation of Sr by extraction chromatography. This soil RM was used later as basis for the 2010 EC Interlaboratory Comparison on Radionuclides in Soil.

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1. Introduction

Certified reference materials (CRM) are useful for evaluating and comparing radiochemical separation and measurement procedures and for calibrating instruments. In addition, reference materials are needed for the quality control and the comparability of measurement results among laboratories, especially those working in surveying and monitoring radioactive materials in the environment and the foodstuff.

Radioactivity levels in foodstuff and the environment are of particular concern; the dose to which humans are exposed depends directly on the level of the radioactivity in the environment they live in and the food they consume. Soil is a natural matrix important in environmental monitoring, as it affects humans both

directly and via the food chain. Few reference materials of environmental matrices, certified for their radioactivity contents, exist and even fewer with their values traceable to the SI units (BIPM, 1998).

The JRC-IRMM participated at an International Atomic Energy Agency (IAEA) – organised Coordinated Research Project (CRP) to upgrade existing reference materials and in this case the IAEA-375 Soil RM, in order to assign property values traceable to the International System of Units (SI). The material was collected for the Agency's Laboratories Seibersdorf AQCS (Analytical Quality Assurance Services) program from a field in the Brjansk region, Russia, in July 1990. The top soil to a depth of 20 cm was collected, dried, milled, sieved (0.3 mm) and shipped to the IAEA. The material was homogenised, bottled and sterilised by γ -ray irradiation to a total dose of 25 kGy using a ^{60}Co source (Strachnov et al., 1996; IAEA, 2000).

Randomly selected bottles, containing 250 g of soil each were sent to the CRP participating laboratories for assay. The JRC-IRMM

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received 6 bottles and participated at the CRP by determining the activity concentrations in the soil RM of the radionuclides ^{40}K , ^{134}Cs , ^{137}Cs , ^{212}Pb , ^{212}Bi , ^{214}Pb and ^{214}Bi measured by γ -ray spectrometry and that of ^{90}Sr by liquid scintillation counting after chemical separation of Sr. The measurement of the activity concentration of ^{226}Ra , ^{235}U , ^{234}U , ^{238}U , ^{238}Pu , $^{239+240}\text{Pu}$, ^{230}Th and ^{232}Th by α -particle spectrometry after chemical separation of the corresponding elements will be the subject of a future article. More details can be found by Altitzoglou et al. (2006). The laboratories were encouraged to use their own routine procedures, so that a variety of analytical methodologies is used.

2. Measurements by high-resolution γ -ray spectrometry

One of the advantages of high-resolution γ -ray spectrometry is that it can be used to assess γ -ray emitting radionuclides in the samples without any prior chemical treatment. In that way, the source preparation is very rapid and simple and in the majority of cases non-destructive. The content of each of the 6 bottles of the soil received from the IAEA was shaken vigorously, using a 3D Turbula mixer (type T2C, Willy A. Bachofen AG Maschinenfabrik, Basel, Switzerland) and was dried in an oven for 48 h at 105 °C to constant weight. During this process, the soil samples lost between 1.7% and 1.9% of their weight.

After cooling to room temperature in a desiccator, the samples were weighed using an analytical balance, calibrated with a calibrated weight set traceable to the IRMM kilogram, which is directly traceable to the BIPM (Bureau International des Poids et Mesures, Sèvres, France) kilogram by regular direct comparisons. One sample was prepared in cylindrical polypropylene containers from each bottle supplied by the IAEA, with the sample mass ranging from 39.4 to 42.0 g. The containers were tapped and placed directly on top of the detector end-cap and measured for 4–12 days each. A 36% relative efficiency, high-purity germanium (HPGe) co-axial detector system (Model GC3518, Canberra Industries, Inc., Meriden, CT, USA) was used for the measurements. The detector was housed in a 10-cm thick Pb shield of circular cross-section, lined with 1 mm Cd and 1 mm Cu; the inner 2 cm of the Pb shield was made of high radiopurity Pb.

In the acquisition system a calibrated stable quartz oscillator with a frequency of 100 kHz provided the time base of the live-time clock gates. As time base, the legal time in Germany on the basis of Coordinated World Time (UTC), generated at the Physikalisch-Technische Bundesanstalt (PTB, Braunschweig, Germany) by atomic clocks and broadcasted through the LF transmitter DCF77, was used. More details can be found by Altitzoglou et al. (2004).

The high-resolution γ -ray spectrometry method is an indirect method, using the detection system as a comparator of the sample to a reference sample and therefore it must be calibrated, ideally with a calibration standard source traceable to the SI units and of the same geometry as that of the sample. Then, the measurement result is directly linked to the known value of the calibration standard.

The detector system employed in this work was calibrated for peak efficiency using single-nuclide point sources, as well as multi-nuclide liquid standards prepared in the same geometry as the actual samples. In addition, actual samples of soil were spiked with known amounts of standard ^{54}Mn , ^{60}Co , ^{65}Zn , ^{137}Cs and ^{241}Am solutions, mixed thoroughly and measured, in order to obtain information on the matrix self-absorption. The standard radionuclide solutions used to prepare the calibration standards originated from standardisation campaigns – usually Key Comparisons organised by the BIPM Comité Consultatif pour les Rayonnements Ionisants (CCRI(II)).

Wherever a calibration transfer from one geometry to another was done, the calculation of the efficiency was performed using the Monte Carlo computer code GEOLEP (Solé, 1990; Lépy et al., 2001). The same code was used for the calculation of the total efficiency, necessary to calculate the coincidence summing corrections in the case of ^{134}Cs and ^{60}Co . The efficiency calculated using GEOLEP agreed within less than 2.2% with the experimentally measured efficiency for the standard point sources, liquid standards and the spiked soil.

The measured data either for the efficiency or the activity determination, were corrected for background, decay, decay during measurement and, where appropriate (i.e., ^{134}Cs and ^{60}Co), for coincidence summing. For the calculation of the ^{134}Cs activity its most prominent peaks at 605 and 795 keV were used.

3. Measurements by liquid scintillation counting

The measurement of ^{90}Sr requires Sr to be separated from the matrix and from other interfering radionuclides first. The material was first dried in an oven at 105 °C to constant weight. The mass of each sample used was determined gravimetrically, using an analytical balance (model AT21, Mettler-Toledo, Greifensee, Switzerland), calibrated with standard weights traceable to the IRMM kilogram which is traceable to the BIPM kilogram. Sample amounts of the order of 5 g were used and in total six samples, one from each received bottle, were analysed.

The soil was heated to 200 °C for one hour and combusted for at least 4 h at 550 °C to destroy organic compounds before the microwave digestion. The sample mass reduction after ashing was about 10%. After adding the tracer (^{85}Sr) for the chemical recovery determination, wet digestion with concentrated nitric/hydrofluoric acids and hydrogen peroxide was performed with a Mars 5 Digestion System (CEM Corp., Matthews, NC, USA). Finally, this solution was treated with concentrated HCl and 65% HNO_3 until it became visually clear and ready for the chemical separation. Details can be found in Hill et al., (2004).

The digested sample in 3M HNO_3 was passed through an extraction chromatography column (2 mL pre-packed TRU-resin column, density 0.37 g mL $^{-1}$, particle size 100–150 μm , Eichrom Technologies, Inc., Darien, IL, USA). The eluate of the TRU column containing Sr was then passed through a Sr-resin column (2 mL pre-packed, density 0.33 g mL $^{-1}$, particle size 100–150 μm) conditioned with 8 M HNO_3 , to obtain a pure Sr fraction. The final Sr eluate was evaporated and the residue taken up by 6 mL 0.05M HNO_3 was transferred to a 20-mL High-Performance Packard scintillation vial containing 14 mL of InstaGel Plus (PerkinElmer, Boston, MA, USA) LS cocktail.

The samples were measured immediately after separation of Sr and several times later, using a Wallac 1220 Quantulus (PerkinElmer) ultra low-level liquid scintillation spectrometer. Blanks introduced before and after each sample measurement, were prepared by adding 6 mL 0.05 N HNO_3 into 14 mL of Insta-Gel Plus LS cocktail. The data reduction and analysis included the background subtraction, decay correction, decay during measurement correction, correction for the contribution of the tracer (^{85}Sr) and the ingrowth of ^{90}Y .

Since the sample went through digestion and chemical separation, in order to isolate the strontium, a tracer for the chemical recovery calculation was used. We have opted for ^{85}Sr , which we then measured by γ -ray spectrometry and the chemical recovery was calculated as the ratio of the counts under the 514-keV γ -ray peak of the sample to that of a reference source (in the same geometry).

For the counting efficiency calibration of the LSC for ^{90}Sr , the CIEMAT/NIST ^3H efficiency tracing method (Grau Malonda and

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