

## Consensus evaluation of radioactivity-in-soil reference materials in the context of an NPL Environmental Radioactivity Proficiency Test Exercise

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### ABSTRACT

The development of two radioactivity-in-soil reference materials is described – one for peat and one for soil with high sand content. Each bulk material was processed, subdivided and measured before being sent to participants in an NPL Environmental Radioactivity Proficiency Test Exercise. Activity concentrations of radionuclides in each material were determined by ‘consensus’ evaluations of participants’ results using two weighted mean methods. The project demonstrated the use of such exercises in delivering reference materials to the user community.

### 1. Introduction

Radiochemical analyses of solid matrices are often required in the context of nuclear site clearance and environmental monitoring. Appropriate reference materials are needed to validate analytical methods and to serve as quality control materials. An industry working group convened by the National Physical Laboratory (NPL) in 2012 concluded that, in the UK, there was a need in particular for reference materials for analyses of concrete, metals, soils and oils, preferably prepared from ‘real’ contaminated samples rather than from artificially spiked matrices. In an earlier paper, Harms and Gilligan (2010) described the development of a concrete reference material at NPL. This was achieved by (i) processing and homogenising a bulk core sample taken from a nuclear reactor bioshield, (ii) testing the stability and homogeneity of the processed material, (iii) issuing sub-samples of the material to participants in a radioactivity measurement Proficiency Test Exercise (PTE), and finally (iv) certificating nuclides present in the bulk material by statistical analysis of the results submitted by the participants.

In the current study, NPL has applied this methodology to the evaluation of samples of sandy soil and peat prepared at another UK research laboratory approximately 30 years ago. The preparation of the original samples, their treatment, measurement and subdivision at NPL, and the analysis of the PTE participants’ reported data using two statistical methods, are described. All uncertainties quoted herein are standard uncertainties with a coverage factor of  $k = 1$ , providing a level of confidence of approximately 68%.

### 2. Sample preparation

Bulk samples of ‘Fyfield’ sandy soil and ‘Thetford’ peat were prepared in 1984 by the National Radiological Protection Board (now Public Health England, Didcot, U.K.) by the addition of nominal amounts of  $^{90}\text{Sr}$ ,  $^{137}\text{Cs}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  to the soils to support studies of radionuclide uptake in plants. Each bulk sample was regularly mixed and the activity concentrations periodically measured; the concentrations (all radionuclides combined) were of the order of  $4 \text{ Bq g}^{-1}$  for the sandy soil and  $15 \text{ Bq g}^{-1}$  for the peat. In 2013, samples of both soil types (10 kg nominal) were coarsely sieved to remove large soil aggregates and stones and dispatched to NPL.

The sandy soil sample was inspected at NPL to check for any remaining soil aggregates, stones or other materials; none were found. The sample was divided between four bottles (each 2 kg nominal), heated overnight at  $110^\circ\text{C}$  and recombined into a single container. The sample was again divided into two bottles and each bottle was separately mechanically mixed for 2–3 h. The contents of the bottles were recombined and mixed for a further 2–3 h. Finally, the sample was divided into 54 sub-samples (150 g nominal each) in 100 ml plastic bottles. The bulk sample of peat was treated in the same way as the sandy soil. The final mass of peat after heating was significantly reduced, being approximately 6.7 kg. A total of 65 sub-samples were prepared (nominal mass 100 g nominal).

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### 3. NPL measurements

#### 3.1. Homogeneity testing

This was carried out on each of the two sets of sub-samples using the method described by Harms and Gilligan (2010). For each soil type, the sub-samples were each measured once by high-resolution gamma-spectrometry. The decay-corrected count rate per unit mass of the 59.5 keV Full-Energy Peak (FEP) from the decay of  $^{241}\text{Am}$  was determined for each measurement. This dataset was used to calculate the variance between the sub-samples ( $u_{bb}^2$ ) and the squared mean of the relative uncertainties ( $u_{int}^2$ ). One sample from each soil type was re-measured a further nine times (for a total of ten measurements) and the measurement variance ( $u_{meas}^2$ ) of the decay-corrected counts per unit mass of the 59.5 keV FEP was determined. The homogeneity uncertainty was calculated using either:

$$u_{\text{hom}}^2 = u_{bb}^2 - u_{\text{int}}^2 \quad (1)$$

or

$$u_{\text{hom}}^2 = u_{bb}^2 - u_{\text{meas}}^2 \quad (2)$$

whichever gave the smaller value. The homogeneity uncertainty was determined as 8% for sandy soil and 4% for peat.

#### 3.2. Activity concentrations

Sub-samples of both soil types were measured to confirm the approximate activity concentration values provided by the supplier. Two methods were used: (i) gross alpha and beta counting of dried soil, and (ii) radiochemical separation of  $^{239}\text{Pu}$  and  $^{241}\text{Am}$  followed by alpha spectrometry.

##### 3.2.1. Gross alpha and beta counting

Five samples of each soil type were prepared for counting by dispensing a weighed mass of ground soil (2 g nominal) to a planchet (diameter 2"). The planchets were counted in a Berthold Technologies LB 790-2 gross alpha and beta proportional counter at an operating voltage of 1450 V. Alpha and beta detection efficiencies had not been established for soils in this geometry, so values based on dispensing of weighed drops of standard solutions of  $^{90}\text{Sr}$  and  $^{241}\text{Am}$  to planchets were applied (the values were 47% and 20% respectively). Consequently, these measurements were regarded as indicative only. The results are given in Table 1.

##### 3.2.2. Radiochemical separation and alpha spectrometry

Four samples (25 g each) of each soil type were dried at 100 °C in an oven for 24 h.

The peat samples were transferred to ceramic crucibles and placed in a furnace to be ashed (this was necessary due to the high organic matter content). The furnace temperature was increased at a rate of 100 °C per hour until the temperature had reached 500 °C, after which the samples were ashed overnight.

Each sandy soil sample and each peat sample was treated as follows:

1. The sample was placed in a round-bottomed flask with hydrochloric acid (6 M, 100 ml).
2. The solution was refluxed for 2–3 h before being filtered using glass-

fibre paper; the filter was transferred to a PTFE beaker.

3. Hydrofluoric acid (40–60%, sufficient to cover the filter) was added to the beaker and the solution was warmed to dryness; this process was repeated three times.

4. The precipitate was dissolved in hydrochloric acid (6 M) and combined with the filtrate from step (2).

An aliquot of each of two of the samples of dissolved peat was combined with  $^{241}\text{Am}$  and  $^{242}\text{Pu}$  yield tracers (approximately 1 Bq each). Two of the samples of dissolved sandy soil were treated in the same way. The actinides in each of the four spiked samples were separated using AG1-X8 ion-exchange resin.

In nitric acid, plutonium (IV) forms negatively charged complexes such as  $[\text{Pu}(\text{NO}_3)_6]^{2-}$  and is strongly absorbed onto anion-exchange resin from nitric acid (8 M) with a distribution coefficient  $K_d > 102$ . Americium, on the other hand, like most cations in oxidation state III is not retained by the resin from nitric acid solutions. Plutonium (IV) is selectively eluted from the column by reduction to plutonium (III) in hydrochloric media.

Using this approach, a separation using AG1-X8 resin was carried out as follows:

1. The spiked sample was evaporated and redissolved in nitric acid (8 M) in a beaker.
2. A pre-packed AG1-X8 column was preconditioned with nitric acid (8 M, 10 ml) and the sample was loaded.
3. The beaker was rinsed with nitric acid (8 M) which was added to the column.
4. The column was rinsed with further nitric acid (8 M, 20 ml) and the  $^{241}\text{Am}$  fraction was collected.
5. The column was rinsed with hydrochloric acid (9 M, 20 ml) in the presence of ammonium iodide (0.1 M) to elute  $^{239}\text{Pu}$ .
6. The americium and plutonium fractions were electroplated onto steel disks and the activities were measured by alpha spectrometry.

The separation process is summarised in Fig. 1. The results of the alpha spectrometry measurements are given in Table 2.

### 4. PTE participants' data

Sub-samples of sandy soil (21) and peat (12) were issued to participants in the 2013 NPL Environmental Radioactivity Proficiency Test Exercise (Dean et al., 2014). These exercises are voluntary and are run by NPL on an approximately annual basis. They play a major role in laboratory Quality Assurance for the environmental monitoring sector and support accreditations in this field granted by the United Kingdom Accreditation Service (UKAS). Most sample types prepared for these exercises are aqueous solutions spiked with standardised radioactive solutions and their traceability to national standards of radioactivity

**Table 1**  
Gross alpha and beta data.

	Peat	Sandy soil
Combined alpha and beta activity concentration ( $\text{Bq g}^{-1}$ )	3.70	0.88
Nominal concentrations from supplier's data ( $\text{Bq g}^{-1}$ )	16	4
Ratio NPL value/supplier's value	0.23	0.22

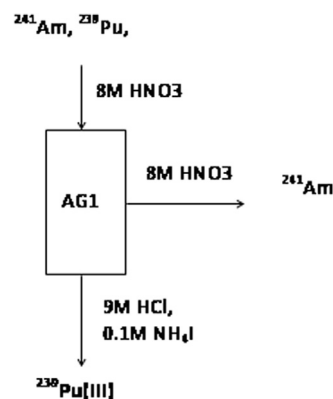


Fig. 1. Schematic view of AG1 separation procedure.

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