

# Analytical method for the determination of gross beta, $^{90}\text{Sr}$ , $^{226}\text{Ra}$ and Pu isotopes in environmental samples

Hyuncheol Kim<sup>1</sup>  · Jong-Myoung Lim<sup>1</sup> · Kun Ho Chung<sup>1</sup> · Wannoo Lee<sup>1</sup>

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**Abstract** This study presents analytical methods for the determination of gross beta,  $^{90}\text{Sr}$ ,  $^{226}\text{Ra}$  and Pu isotopes using samples in the IAEA-TEL-2015-04 ALMERA Proficiency Test exercise. Samples for gross beta were prepared by evaporation and then analyzed using a gas proportional counter.  $^{90}\text{Sr}$  in the liquid sample was concentrated as  $\text{SrCO}_3$  precipitates and purified by Sr resin. Pu isotopes and  $^{90}\text{Sr}$  in the soil sample were extracted from the sample by mineral acid leaching and separated using TEVA and Sr resin, respectively. Pu isotopes were determined by alpha spectrometry and  $^{90}\text{Sr}$  were determined with a liquid scintillation counter. Radium in the soil sample was extracted by  $\text{LiBO}_2$  fusion, and the radon-emanation method using LSC was applied for the determination of  $^{226}\text{Ra}$ .

**Keywords** Gross beta ·  $^{90}\text{Sr}$  · Pu · Radium · Environmental sample · Proficiency test

## Introduction

Environmental radiation originates from both artificial and naturally occurring materials. Nuclear weapons testing and some nuclear power plant accidents are the main sources of man-made radionuclides in the environment, such as  $^{90}\text{Sr}$ ,

Pu isotopes and  $^{137}\text{Cs}$  [1, 2]. The global average exposure is  $2.4 \text{ mSv yr}^{-1}$ , and public exposure to natural sources accounts for more than 98% of the radiation dose, with the rest being medical exposure [3, 4]. A reliable assessment of environmental radioactivity is necessary with respect to health physics. Therefore, IAEA-TEL (International Atomic Energy Agency-Terrestrial Environment Laboratory) has hosted international proficiency tests and inter-laboratory comparisons to demonstrate the analytical capabilities of ALMERA (Analytical Laboratories for the Measurement of Environmental Radioactivity) to network members every year [5]. This proficiency test (PT) is an appropriate exercise for estimating and monitoring the analytical methods of participating laboratories [6–8].

This study demonstrates the analytical methods and results for gross beta,  $^{90}\text{Sr}$ ,  $^{226}\text{Ra}$  and Pu isotopes from participation in the IAEA-TEL-2015-04 PT. In this PT, the identification of gamma-emitters, alpha/beta radionuclides in spiked water, biota, and soil was carried out, with 82 laboratories participating [5].

Gross beta, together with gross alpha, is a useful indicator for estimating the radioactivity of drinking water; therefore, the WHO recommends screening levels below  $1 \text{ Bq L}^{-1}$  for gross beta activity in drinking water [9].

Radon accounts for more than 50% of radiation exposure by natural sources and is a significant progeny of  $^{226}\text{Ra}$ . Hence, data pertaining to the concentration of  $^{226}\text{Ra}$  radioactivity in the environment have been a major issue for the public. Radiostrontium and Pu isotopes are also serious public issues, as both radionuclides are very harmful when released to the environment. The results of the IAEA-TEL-2015-04 PT will be presented in this study, and detailed analytical methods, including pretreatment, chemical separation of samples, and instructions on how to conduct the analyses will be provided.

✉ Hyuncheol Kim  
hckim3@kaeri.re.kr

<sup>1</sup> Environmental Radioactivity Assessment Team, Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Yuseong-gu, Daejeon 34057, Korea

## Experimental

### Reagents and instrument

All reagents were obtained from Sigma-Aldrich and were of analytical grade, including  $\text{HNO}_3$ ,  $\text{LiBO}_2$ ,  $\text{LiBr}$ ,  $\text{BaCl}_2$ ,  $\text{K}_2\text{CO}_3$ ,  $\text{H}_2\text{SO}_4$ , and reagents for the electrodeposition steps. These reagents were used after dilution with deionized water (DIW, MilliQ-Plus, 18 M $\Omega$ ). IAEA-TEL distributed six types of samples. Among them, three were analyzed in this study: sample codes 1 and 2 (both spiked water) along with 5 (Syrian soil). The calibration sources for  $^{90}\text{Sr}$ ,  $^{226}\text{Ra}$ ,  $^{239}\text{Pu}$  and  $^{240}\text{Pu}$  were obtained from Eckert & Ziegler (Georgia, USA). The  $^{238}\text{Pu}$  and  $^{242}\text{Pu}$  sources were obtained from NIST. A gas proportional counter (XLB, Canberra, USA) was used for the determination of gross beta. A liquid scintillation counter (LSC, Quantulus 1220, Perkin Elmer) was used for the analyses of  $^{90}\text{Sr}$  and  $^{226}\text{Ra}$ . The Pu isotopes were analyzed using an alpha spectrometer (Ortec, USA). The soil sample was completely digested with a fusion apparatus (K<sub>2</sub> PRIME, Katanax, Canada) for  $^{226}\text{Ra}$ .

### Preparation of a method-standard solution

All analyses were done with a method-standard solution with a target radionuclide. For gross beta and  $^{90}\text{Sr}$ , 5.0 Bq of  $^{90}\text{Sr}$  was added to 20 g of DIW. The method-standard solution for Pu isotopes was prepared with  $^{242}\text{Pu}$  (30 mBq),  $^{238}\text{Pu}$  (20 mBq),  $^{239}\text{Pu}$  (20 mBq) and  $^{240}\text{Pu}$  (20 mBq) in 60 mL of 14 M  $\text{HNO}_3$ . The method-standard solution for  $^{226}\text{Ra}$  was prepared with a molten solution from  $\text{LiBO}_2$  fusion including 5 Bq of  $^{226}\text{Ra}$ . The uncertainties associated with the known of  $^{90}\text{Sr}$ ,  $^{242}\text{Pu}$ ,  $^{238}\text{Pu}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ , and  $^{226}\text{Ra}$  added were 1, 1.7, 1.6, 0.3, 0.34, and 0.7% at 95% confidence level, respectively. The molten solution came from  $\text{LiBO}_2$  fusion without a sample (*sample pretreatment section*). All method-standard solutions followed the entire procedure for its target radionuclide, from pretreatment to analysis.

### Sample pretreatment

Evaporation or precipitation was applied as the pretreatment method for the liquid samples. Digestion with mineral acid or a fusion method was applied for the soil, depending on the analyte.

For sample codes 1 and 2, a certain amount of liquid sample (2–30 g) was taken into a Teflon beaker and the sample solution was evaporated to be dryness for the reduction of its volume. The remaining solution was transferred to a planchet ( $\Phi$  50 mm, stainless steel) and the washing solution of the beaker was added to it with de-

ionized water (DIW). This was evaporated under an infrared lamp, followed by analysis using a gas proportional counter (GPC).

For the analysis of  $^{90}\text{Sr}$  in sample code 1 with liquid, 5 mg of stable strontium was added as a carrier and tracer to approximately 100 g of the sample. This was precipitated as  $\text{SrCO}_3$  at pH 10 with 3 mL of a 2 M  $\text{Na}_2\text{CO}_3$  solution and 1 mL of a Ca solution (50 mg mL<sup>-1</sup>). This was centrifuged at 3500 rpm for 5 min, after which the supernatant was removed. The precipitate was dissolved in 10 mL of 8 M  $\text{HNO}_3$ . This was followed by chemical separation using Sr resin.

For  $^{90}\text{Sr}$  and Pu in soil in sample code 5, an amount of 10 g of the sample was taken into a glass beaker and dried at 110 °C for 2 h. This dry-mass of the sample is an important value, as the activity concentration unit needed to be reported as the dry-mass of the sample. It was then ashed at 450 °C overnight to decompose the organic material, which can interfere with the subsequent chemical separation process. An amount of 5 mg of stable Sr was added to the sample as the carrier and yield tracer.  $^{242}\text{Pu}$  (30 mBq) was used as the tracer, and 200 mg of  $\text{K}_2\text{S}_2\text{O}_5$  was added to reduce all Pu to Pu(III). This was leached with 60 mL of 14 M  $\text{HNO}_3$  on hot plate set at 180 °C for 4 h, filtered through a glass fiber filter (GF/F, Whatman), and then washed with DIW. The pH of the leaching solution was increased to 7 using ammonia solution to separate Sr and Pu.

In this solution, Sr exists as an ion, but Pu was co-precipitated with  $\text{Fe}(\text{OH})_3$ . Since the amount of Fe was sufficient, additional Fe was not added.  $\text{Sr}^{2+}$  in the supernatant was precipitated as  $\text{SrCO}_3$  with 50 mg of a Ca solution and 10 mL of a 2 M  $\text{Na}_2\text{CO}_3$  solution. After centrifugation, the precipitate was dissolved in 15 mL of 8 M  $\text{HNO}_3$ , followed by chemical separation using Sr resin. Pu co-precipitated with  $\text{Fe}(\text{OH})_3$  was dissolved with 14 M  $\text{HNO}_3$ , resulting in a final concentration of  $\text{HNO}_3$  of approximately 6 M to stabilize Pu(IV). It was loaded onto a TEVA column [10, 11].

For  $^{226}\text{Ra}$  in sample code 5, 0.5 g of the soil sample was completely dissolved through fusion [12]. During this process, 1 g of  $\text{LiBO}_2$  and 0.25 g of  $\text{LiBr}$  were mixed with the soil sample, after which the fusion step proceeded at 1000 °C, taking about 30 min. The melts obtained from the fusion were transferred into a Teflon vessel of the digestion apparatus containing 50 mL of 0.4 M  $\text{HNO}_3$ . In addition, 1 mL of Ba carrier (10 mg mL<sup>-1</sup>) was added to the molten solution as a carrier and tracer, followed by chemical separation.

### Chemical separation

The chemical separation using Sr resin and TEVA resin were performed on an automated separation system [13, 14].

$^{90}\text{Sr}$  in the 8 M  $\text{HNO}_3$  solution prepared after the pre-treatment of sample codes 1 and 5 was purified using Sr resin (4 mL, bed volume, BV) by the following steps: (1) Conditioning of the Sr resin with 30 mL of 8 M  $\text{HNO}_3$  at  $3 \text{ mL min}^{-1}$ ; (2) loading of the sample at  $2 \text{ mL min}^{-1}$ ; (3) rinsing with 24 mL of 8 M  $\text{HNO}_3$  at  $2 \text{ mL min}^{-1}$ ; (4) Elution with 24 mL of DIW at  $2 \text{ mL min}^{-1}$  [14]. The  $\text{Sr}^{2+}$  from the elution step was precipitated as  $\text{SrCO}_3$  with 5 mL of a 2 M  $\text{Na}_2\text{CO}_3$  solution at pH 10. This was centrifuged, and the supernatant was removed. The remaining precipitates were evaporated in an oven and  $\text{SrCO}_3$  was finally dissolved in 10 mL of 0.1 M  $\text{HNO}_3$ . This was followed by the preparation of the source for the LSC.

The Pu in 6 M  $\text{HNO}_3$  was loaded onto TEVA resin (4 mL, BV) pre-conditioned with 30 mL of 8 M  $\text{HNO}_3$ . Uranium, thorium and americium were removed from the TEVA column with 40 mL of 1 M  $\text{HNO}_3$ , and 40 mL of 9 M HCl was then used to remove the technetium. Pu isotopes were eluted from the TEVA column with 40 mL of a mixture of 0.1 M  $\text{NH}_4\text{I}$  and 9 M HCl [10, 14]. The elution solution was evaporated to near dryness on a hot plate set at  $180^\circ\text{C}$ . This was followed by the preparation of the source for alpha spectrometry.

Ra in the molten solution was co-precipitated as  $\text{Ba(Ra)SO}_4$  with  $\text{H}_2\text{SO}_4$ .  $\text{Ba(Ra)SO}_4$  is not soluble in common mineral acid and was therefore converted into  $\text{BaCO}_3$  through a metathesis approach [12, 15], which is highly soluble in a weak acid. Ten mL of  $\text{H}_2\text{SO}_4$  (98%) was added to precipitate the Ra as  $\text{BaSO}_4$ , and this was then stirred for 30 min. It was centrifuged at 3500 rpm for 5 min, and the precipitate was subsequently washed with DIW until the pH of the supernatant reached approximately 7. The precipitate was transferred into a 20 mL glass vial, mixed with 1 mL of  $\text{K}_2\text{CO}_3$  (50%, w/w), and then heated to  $200^\circ\text{C}$  until it was completely evaporated. It was washed with DIW and subjected to centrifugation until the pH of the supernatant was reduced to 7. The precipitate was evaporated and dissolved in 10 mL of 0.2 M  $\text{HNO}_3$  in a 20 mL glass vial for LSC. The concentration of Ba in the solution was analyzed by ICP-OES (inductively coupled plasma-optical emission spectroscopy) to determine the recovery of Ba(Ra). Finally, it was mixed with 10 mL of a water-immiscible cocktail (Maxilight, Hidex, Finland) for LSC [16].

### Preparation of calibration sources for the analysis

#### GPC

An aliquot of  $^{90}\text{Sr}$  ( $\sim 5 \text{ Bq}$ ) in equilibrium with  $^{90}\text{Y}$  was put into the planchet and then evaporated under an infrared (IR) lamp. This was used to determine the counting efficiency of GPC for the beta emitter.

#### LSC

An aliquot of  $^{90}\text{Sr}$  ( $\sim 5 \text{ Bq}$ ) in equilibrium with  $^{90}\text{Y}$  was transferred into a 20 mL plastic vial. A solution of 0.1 M  $\text{HNO}_3$  was added until the final volume reached 10 mL. It was analyzed by the Cerenkov counting mode.

An amount of 5 Bq of  $^{226}\text{Ra}$  was put into a 20 mL glass vial, and a 0.2 M  $\text{HNO}_3$  solution was added until the final volume of the solution was 10 mL. This was then mixed with 10 mL of a water-immiscible cocktail (Maxilight). It was measured after  $^{222}\text{Rn}$  was in equilibrium with  $^{226}\text{Ra}$ .

#### Alpha spectrometer

An aliquot of Pu isotopes including  $^{242}\text{Pu}$  (30 mBq),  $^{238}\text{Pu}$  (20 mBq),  $^{239}\text{Pu}$  (20 mBq), and  $^{240}\text{Pu}$  (20 mBq) was put into a Teflon beaker and then evaporated. The source for alpha spectrometry was prepared from dry residue using the following electrodeposition procedure [17]. It was mixed with 2.5 mL of 5%  $\text{NaHSO}_4$ , 5 mL of DIW, and 5 mL of 15% (w/w)  $\text{Na}_2\text{SO}_4$ . The solution was transferred into an electrodeposition cell, after which 1 mL of 0.02% (w/w) ammonium oxalate was added. Pu isotopes were electrodeposited on stainless steel for 2 h at 880–890 mA. Subsequently, the cell was disassembled and the disc was rinsed with a 5% (w/w)  $\text{NH}_4\text{OH}$  solution and ethanol. The back of the disc was then heated with a gas torch and analyzed by alpha spectrometry.

## Results and discussion

### Gross beta activities in sample codes 1 and 2

GPC is a suitable analytical method for separately determining gross alpha and gross beta. It has a very low background at  $0.04 \pm 0.02 \text{ cpm}$  (1 SD, standard deviation, counts per minute) and  $0.8 \pm 0.1 \text{ cpm}$  (1 SD) for alpha and beta emitters, respectively. Before the measurement, a calibration disc source of  $^{90}\text{Sr}$  was used to determine the optimal plateau for gross beta in GPC. The calibration disc source of  $^{210}\text{Po}$  was used to estimate the beta spillover of the alpha emitter. The beta spillover rate was found to be 1.8%.

As sample 2 provided by IAEA contained gamma emitters, LSC was not a proper instrument. Gamma rays can produce photons due to a reaction with the scintillation cocktail. When the sample was mixed with the scintillation cocktail, the effect of the gamma emitters could not be removed from the measured count rate in LSC.

Samples with 10–30 g of sample code 1 and 2–4 g of sample code 2 were used (Table 1). They were counted by

**Table 1** The analytical results of gross beta in sample code 1 and 2

Sample code	Sample mass (g)	Analyst value <sup>a</sup>	Recommended value <sup>b</sup>	Relative error (%)	Z-score <sup>c</sup>
1	10–30	111 ± 8	104 ± 13	7.0	0.56
2	2–4	53 ± 2	51 ± 11	5.1	0.24
Method-standard solution	n.a.	51.6 ± 1.6	52.9 ± 0.9 <sup>a</sup>	–2.4	n.a. <sup>d</sup>

<sup>a</sup> Average ± SD (standard deviation, Bq kg<sup>–1</sup>)

<sup>b</sup> Robust mean ± robust SD by IAEA TEL (Bq kg<sup>–1</sup>)

<sup>c</sup> Calculated by IAEA TEL

<sup>d</sup> Not available

means of GPC for 300 min. For sample code 1, the beta count rate was 17–47 cpm, and for sample code 2 it was 2–4 cpm. The alpha count rates were less than 0.08 cpm for both sample codes 1 and 2. Thus, the beta spillover resulting from the alpha emitter was negligible in both cases.

The activity concentrations of the samples were calculated using determining the efficiency and weight of the samples, with Eq. (1). The counting efficiency using the <sup>90</sup>Sr calibration source was 46.9 ± 1.0% (1 SD). The method-standard sample (*n* = 2) was prepared with 20 g of DIW and the known activity of <sup>90</sup>Sr. It was then analyzed with the samples together to obtain more reliable data. The concentration of gross beta was 111 ± 8 Bq kg<sup>–1</sup> (1 SD), and the corresponding values were 54 ± 2 Bq kg<sup>–1</sup> (1 SD) for sample codes 1 and 2, respectively. The activity concentration of gross beta in sample codes 1 and 2 had a relative error within 7% compared to the value recommended by IAEA TEL. The relative error of the method-standard was less than 3%.

$$GB = \frac{N_G - N_B}{\varepsilon_{GB} \times W_{smp}} \quad (1)$$

GB: gross beta activity (Bq kg<sup>–1</sup>), *N<sub>G</sub>*: gross count rate (cps), *N<sub>B</sub>*: background count rate (cps), *ε<sub>GB</sub>*: counting efficiency, *W<sub>smp</sub>*: mass of sample (kg).

### <sup>90</sup>Sr in sample codes 1 and 5

Stable Sr was used as a carrier and tracer. The initial concentration of Sr in the sample was a very important parameter to avoid underestimating the activity

concentration, as the measured recovery value was higher than the actual value. Sample 1 contained 0.16 mg of Sr<sup>2+</sup> L<sup>–1</sup>, which was directly analyzed by ICP-OES. The stable strontium of approximately 100 g of sample 1 was very low relative to the amount of stable strontium added (~5 mg). For this reason, no correction was done.

To determine the stable Sr in sample code 5, 5 g of sample was leached under the conditions described in the pretreatment section above. Sample 5 contained 0.12 ± 0.01 mg of Sr per g of dry soil (1 SD). The initial concentration of stable Sr in the samples should be considered when estimating the activity concentration for <sup>90</sup>Sr, as 10 g of sample 5 contained about 20% of the Sr carrier (~5 mg) used in this study.

The counting efficiency for <sup>90</sup>Sr + <sup>90</sup>Y was 66 ± 1% (1 SD) in the Cerenkov counting mode. The LSC measurement for <sup>90</sup>Sr was delayed until <sup>90</sup>Y reached equilibrium with <sup>90</sup>Sr, which took nearly 14 days. The triplicates in 10 g of sample 5 were analyzed, and the recovery ranged from 74 to 86% (Table 2). The activity concentration of <sup>90</sup>Sr in sample 1 was 28.4 ± 1.2 Bq kg<sup>–1</sup> (1 SD) with a relative bias of –4% for the target value. Sample 5 had 36.2 ± 2.7 Bq kg<sup>–1</sup> of <sup>90</sup>Sr (1 SD), which is identical to the target value suggested by IAEA-TEL. The relative error of the method-standard (*n* = 2) was –4% and its recovery ranged from 90 to 94%.

$$A_{90Sr} = \frac{N_G - N_B}{R_{Sr} \times \varepsilon_{90Sr} \times W_{smp}}$$

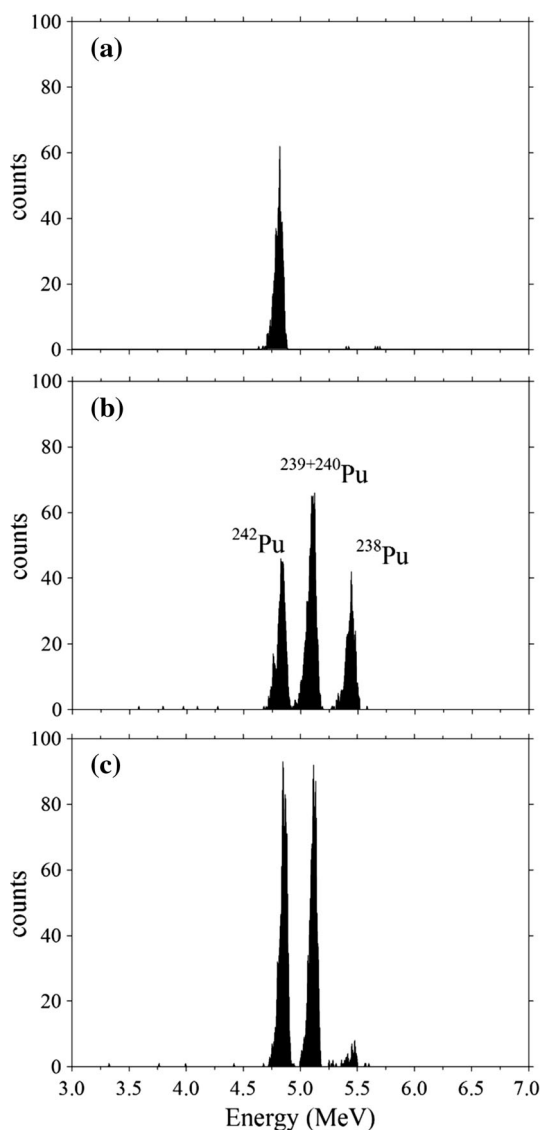
*A<sub>90Sr</sub>*: activity concentration of <sup>90</sup>Sr (Bq kg<sup>–1</sup>), *R<sub>Sr</sub>*: recovery of stable Sr, *ε<sub>90Sr</sub>*: counting efficiency of <sup>90</sup>Sr + <sup>90</sup>Y in Cerenkov counting mode.

**Table 2** The analytical results of sample code 1 and 5 for <sup>90</sup>Sr

Sample code	Sample mass (g)	Recovery (%)	Analyst value <sup>a</sup>	Target value <sup>a</sup>	Relative error (%)	Z-score <sup>b</sup>
1	110–130	91–92	28.4 ± 1.2	29.6 ± 0.8	–4.0	–0.4
5	10	74–86	36.2 ± 1.6	36.2 ± 2.7	0.0	0.0
Method-standard solution	n.a.	90–94	5.1 ± 0.2	5.3 ± 0.2	–4.1	n.a.

<sup>a</sup> Average ± SD (Bq kg<sup>–1</sup>)

<sup>b</sup> Calculated by IAEA TEL



**Fig. 1** Alpha spectrum of method blank (a), method-standard (b), and sample code 5 (c)

**Pu isotopes and <sup>226</sup>Ra in sample 5**

For the analysis of the Pu isotopes using the alpha spectrometer, <sup>237</sup>Np should be separated from the sample. <sup>242</sup>Pu has two main alpha radiation with 4.901 MeV (77.5%) and 4.857 MeV (22.4%). <sup>237</sup>Np emits alpha radiation with 4.788 MeV (48%), 4.771 MeV (25%), and 4.766 MeV (8%) [18]. In alpha spectrometry, the peak of <sup>242</sup>Pu is shown as a single line between 4.6 MeV and 4.9 MeV, then could be overlapped with the peak of <sup>237</sup>Np. Since <sup>242</sup>Pu was used as a tracer, the recovery of plutonium would be overestimated without the removal of <sup>237</sup>Np.

The behavior of Np in the TEVA column is very similar to that of Pu. Even in the washing step, Np still remained in TEVA together with Pu. Accordingly, NH<sub>4</sub>I-9 M HCl was used as the eluent. Using this eluent, the Pu(IV) in TEVA was reduced to Pu(III), which was very easily separated from the TEVA. However, Np remained in the TEVA column.

The method blank was prepared using <sup>242</sup>Pu in 60 mL of 14 M HNO<sub>3</sub>, and the method-standard was prepared with <sup>242</sup>Pu, <sup>238</sup>Pu, <sup>239</sup>Pu and <sup>240</sup>Pu in 60 mL of 14 M HNO<sub>3</sub>, after which the entire procedure with sample 5 followed. Figure 1 shows the alpha spectrum of the method blank, the method-standard solution, and sample 5. The analytical results are shown in Table 3. The recovery rate of Pu ranged from 78 to 90% for sample 5. The activity concentration of Pu isotopes were 2.6 ± 0.2 Bq kg<sup>-1</sup> for <sup>239+240</sup>Pu (1 SD) with a relative error of -3.3% and 0.14 ± 0.02 Bq kg<sup>-1</sup> for <sup>238</sup>Pu (1 SD). The value of <sup>238</sup>Pu was not compared with that of IAEA-TEL, as this value was not known. The recovery rate of Pu for the method-standard solution (n = 3) ranged from 76 to 85%. The relative error of the method-standard solution was 4.8% for <sup>239+240</sup>Pu and 5.1% for <sup>238</sup>Pu with the recovery rate ranging from 76 to 85%.

**Table 3** The analytical results of sample code 5 for Pu isotopes

Sample	Analyte	Sample mass (g)	Recovery (%)	Analyst value <sup>a</sup>	Target value <sup>a</sup>	Relative error (%)	Z-score <sup>b</sup>
Code 5	<sup>238</sup> Pu	10	78–90	0.14 ± 0.02	n.a.	n.a.	n.a.
	<sup>239+240</sup> Pu			2.6 ± 0.2	2.7 ± 0.4	-3.3	-0.3
Method-standard solution	<sup>238</sup> Pu	n.a.	76–85	22.4 ± 1.2 <sup>c</sup>	21.3 ± 0.1 <sup>d</sup>	5.1	n.a.
	<sup>239+240</sup> Pu			43.4 ± 0.1 <sup>c</sup>	41.4 ± 1.9 <sup>d</sup>	4.8	n.a.

<sup>a</sup> Average ± SD (Bq kg<sup>-1</sup>)

<sup>b</sup> Calculated by IAEA

<sup>c</sup> mBq

<sup>d</sup> Average ± uncertainty (k = 1, mBq)

**Table 4** The analytical results of sample code 5 for  $^{226}\text{Ra}$ 

Sample	Sample mass (g)	Recovery (%)	Analyst value <sup>a</sup>	Target value <sup>a</sup>	Relative error (%)
Code 5	0.5	70–82	$45.4 \pm 3.1$	$50 \pm 9.3$	9.2
Method-standard solution	n.a.	63–81	$2.71 \pm 0.10^b$	$2.92 \pm 0.02^c$	−7.3

<sup>a</sup> Average  $\pm$  SD ( $\text{Bq kg}^{-1}$ )

<sup>b</sup> mBq

<sup>c</sup> Average  $\pm$  uncertainty ( $k = 1$ , mBq)

Five replicates of 0.5 g of sample 5 were used for the analysis of  $^{226}\text{Ra}$ . The recovery rate ranged from 70 to 82%, and the activity concentration of  $^{226}\text{Ra}$  was  $45.4 \pm 3.1 \text{ Bq kg}^{-1}$  (1 SD). We did not report this value to IAEA-TEL; hence, we asked IAEA-TEL for its target value and then compared this value with our results. The relative error was approximately 9%. Kim et al. [16] used  $^{133}\text{Ba}$  as a tracer, but in this study stable Ba was used as both a carrier and a tracer. The concentration of stable barium in sample 5 was negligible relative to the amount of stable barium added; hence no correction had to be made. The use of stable barium as a tracer was found to be very useful, as shown in the results (Table 4). The relative error of the method-standard ( $n = 3$ ) was  $-7\%$ , and its recovery rate ranged from 63 to 81%.

## Conclusion

We have presented analytical methods and results for measuring  $^{90}\text{Sr}$ ,  $^{226}\text{Ra}$ , gross beta and Pu isotopes in liquid and soil types of samples, as distributed by IAEA-TEL in 2015. For gross beta, GPC is a very useful instrument because it avoids the effect of gamma emitters, which is a major form of interference during beta analyses using LSC.

Gross beta activity can be overestimated due to photons caused by reactions between gamma rays and the scintillation cocktail. Radiostromtium and Pu isotopes are artificial radionuclides and were mostly attached to the surface of the soil sample. Mineral acid leaching was sufficient to extract those radionuclides from soil, as shown in this study. They were separated by extraction chromatography using Sr resin and TEVA resin, respectively, in an automated separation system [13].

In contrast,  $^{226}\text{Ra}$  is a natural radionuclide, and it is very difficult to extract all Ra isotopes using typical mineral acid leaching. The alkali fusion method used in this study is a very simple and reliable pretreatment approach for the analysis of natural radionuclides. The radon emanation method used to determine  $^{226}\text{Ra}$  activity takes three weeks but is applicable to routine analyses which do not require quick results.

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