

CALIBRATION OF NATURAL GAMMA-RAY SPECTROMETER FACILITY AT THE ERL

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الخلاصة :

يُستخدم مطياف أشعة جاما الموجود في مختبر أبحاث الطاقة لتحديد وقياس النشاط الإشعاعي الطبيعي الصادر من البوتاسيوم (^{40}K) واليورانيوم (^{238}U) والثوريوم (^{232}Th) في التربة وعينات صناعية مختلفة. ويتكوّن المطياف من كاشف $5'' \times 5'' \text{ NaI(Tl)}$ وتجهيزات اليكترونية موصلة بحاسوب لتجميع وتحليل النتائج التجريبية. ولقد تمّ معايرة مطياف أشعة جاما باستخدام اثنتا عشر عينة قياسية معتمدة من (IAEA) في فينا و (NIST) و (NBL) في أمريكا. ولقد وُجد بأن حساسية هذه النظام تصل إلى جزء من المليون لعنصر اليورانيوم وعنصر الثوريوم و ١٥٠ جزءاً من المليون لعنصر البوتاسيوم على اعتبار أن وزن العينة ١٠٠ غم. كما أنه تم تعيين منحنيات المعايرة الخطية لهذا النظام من خلال عمل بعض القياسات لعناصر البوتاسيوم واليورانيوم والثوريوم.

ABSTRACT

Gamma-ray spectrometer facility at the Energy Research Laboratory (ERL) is used for the determination of natural radioactivity from potassium (^{40}K), uranium (^{238}U), and thorium (^{232}Th) in soil, core, and various other industrial samples. The facility consists of a $5'' \times 5'' \text{ NaI(Tl)}$ detector and the electronic setup coupled to a PC-based data acquisition and analysis system. The gamma-ray spectrometer facility was calibrated using 12 certified standard samples from IAEA (Vienna), NIST, and NBL (U.S.A.). The sensitivity of the setup was found to be better than 1 ppm for U and Th and 150 ppm for K considering 100 g sample. Linear calibration curves were established from measurements carried out for potassium, uranium, and thorium.

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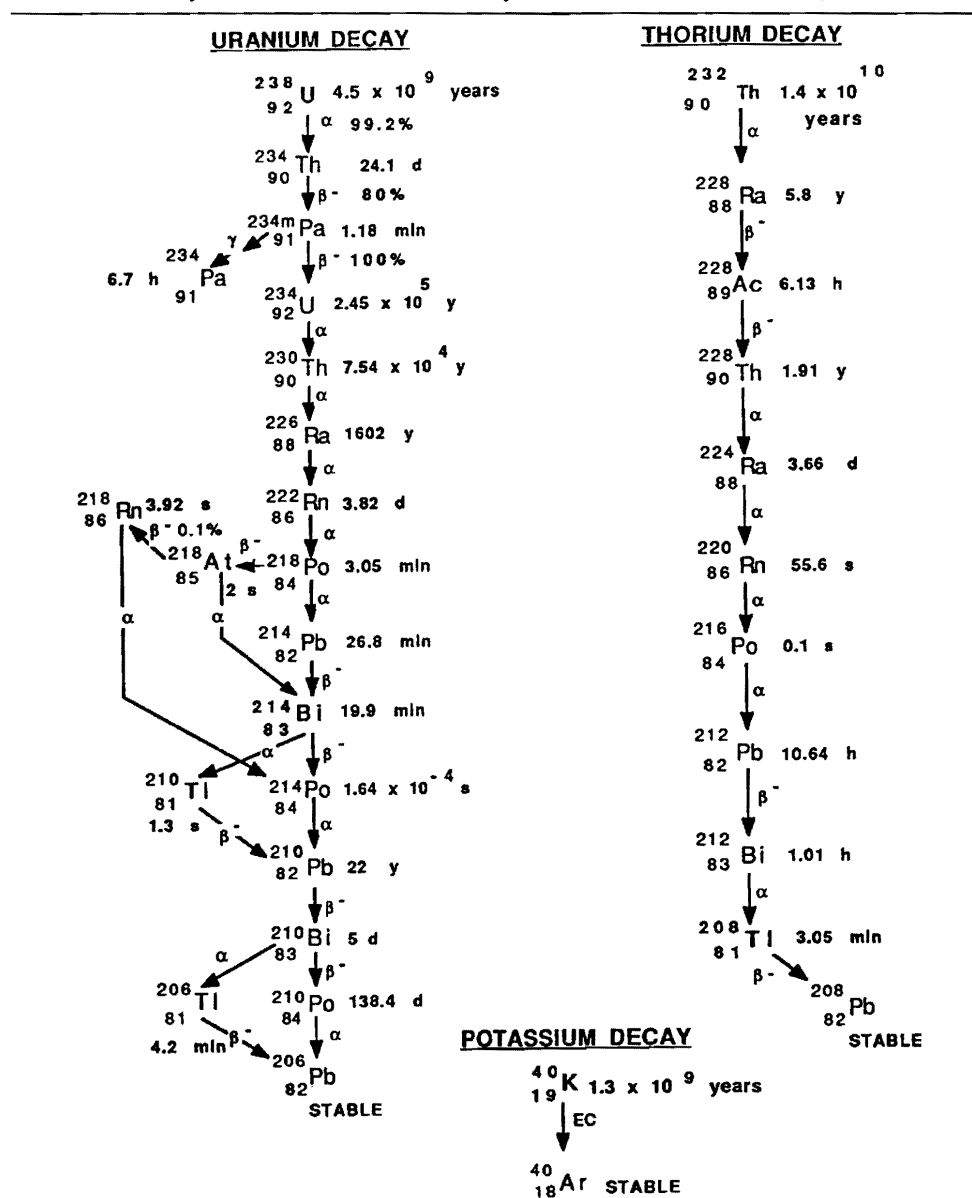
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1. INTRODUCTION

Natural gamma-rays occur in nature mainly from potassium (^{40}K), and the series of uranium (^{238}U) and thorium (^{232}Th). There is also some minor radioactivity from ^{235}U but it is very little due to the isotopic $^{235}\text{U}/^{238}\text{U}$ ratio of 0.72%. The ^{40}K radioisotope decays to stable nucleus ^{40}Ar by emitting 1461 keV γ -rays while ^{238}U and ^{232}Th have long decay series to produce the stable isotopes of lead. Table 1 shows the decay series of these nuclei. With the assumption of secular equilibrium the 1765 keV γ -rays of bismuth (^{214}Bi) is used for the determination of parent ^{238}U as well as the 2615 keV line of thallium (^{208}Tl) for ^{232}Th . While the 1461 keV gamma ray is used for the determination of ^{40}K . Natural gamma-ray spectroscopy has several advantages: the high energy γ -rays are capable of penetrating a few inches of rock or soil samples, the analysis method is nondestructive, large samples can be analyzed and both NaI(Tl) detector with high efficiency and High Purity Germanium Detector (HPGe) with high energy resolution can be used for the analysis. Therefore, natural γ -ray measurements are widely used in well logging and in the analysis of both geological and environmental samples [1–6].

Table 1. The Decay Series of Natural Gamma Ray Emitters: Uranium, Thorium, and Potassium.



Natural γ -ray measurements result in spectral and total radioactivity of K, U, and Th. The results are used for mineral identification and determination of petrophysical parameters such as volume and type of shale, *etc.*, [3–4]. Such information helps in characterizing the geological environment of the area or the reservoir being studied [5]. Especially in well logging, the results of the gamma logs need to be verified by laboratory measurements of core samples which result in more accurate measurements of the γ -ray activities of the samples and clarifies if there were any depth shift in well logging data [5, 6]. The setup can also be used to measure natural γ -rays from soil, phosphate, gas mantle, and environmental samples [7, 8]. Therefore the accurate calibration of a natural gamma-ray spectrometer setup is important which implies measurements of certified standards of K, U, and Th of similar matrix as the samples. In this respect our procedure is one of the best detector calibration methods done so far for a setup using a large size (5" $\times$ 5") NaI(Tl) detector.

2. EXPERIMENTAL METHOD

The natural gamma-ray spectrometry facility at the ERL consists of a 5" $\times$ 5" NaI(Tl) detector with 7.3% energy resolution for 662 keV line of ^{137}Cs , signal processing electronics and a PC-based data acquisition and analysis system. The detector is well surrounded with lead shielding which cuts the room background by 90%. The signal is processed through an ORTEC spectroscopy amplifier and ADCAM multichannel buffer. The scheme of the measuring setup is shown in Figure 1. Spectra are collected in 4096 channel analyzer and calibrated with standard γ -ray sources (^{22}Na and ^{60}Co). The energy range of the spectrum extended from 100 keV to 3000 keV.

A set of 12 standard samples were used for the calibration: 6 from the International Atomic Energy Agency (IAEA, Vienna), 4 from the National Institute of Standard and Technology (NIST, U.S.A.) and 2 from New Brunswick Lab. (NBL, U.S.A.). The standards were soil, rock, clay, sediment, ash, and ore types. The list of the standards with the concentrations of U and Th in parts per million (ppm) and K in weight% (wt.%) is shown in Table 2.

Table 2. Natural Gamma-Ray Energies of ^{214}Bi (from ^{238}U), ^{208}Tl (from ^{232}Th), and ^{40}K [14].

^{214}Bi		^{208}Tl		^{40}K	
$E\gamma$ (keV)	(Intensity, %)	$E\gamma$ (keV)	(Intensity, %)	$E\gamma$ (keV)	(Intensity, %)
609	46.9	277	6.8	1461	10.7
768	5.0	511	21.6		
934	3.3	583	86.0		
1120	15.3	860	12.0		
1238	6.1	2615	100.0		
1378	4.1				
1408	2.5				
1765	16.1				
2204	5.1				
2448	1.6				

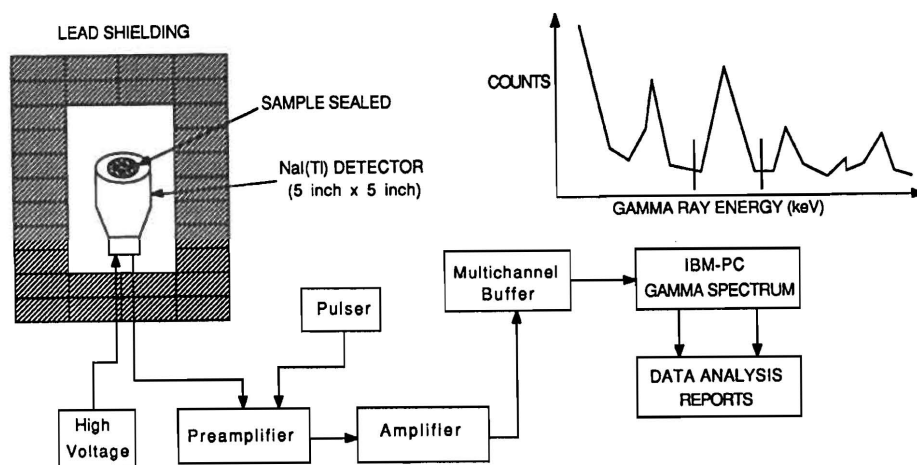


Figure 1. The scheme of the Natural Gamma-Ray Spectrometer Setup at the ERL.

The decay series of ^{238}U passes through radon (^{222}Rn) which is a noble gas having a half-life of 3.8 days. In order to prevent the escape of the radon gas, each sample was placed in a plastic container of 3.5" diameter and 0.5" thickness and was sealed with rubber silicon and placed in a plastic bag which was then air-tight sealed. The sample was left for 3 weeks to allow radon gas decay to reach equilibrium with other daughters of the series [2]. Then, each sample was counted for 24 h to reach a good statistics (around 1%).

Room background was measured using quartz samples (SiO_2) which were free from natural radioactivity [9]. The background of the quartz sample having the same geometry as the sample was also measured for 24 h. A pulser was used to check any gain shifts before each run. To have similar spectra, gain adjustments were made before each run using the pulser so that pulses were collected at the same channel. After counting, the spectra were stored and data analyses were carried out off-line using Maestro-II software package from ORTEC.

Data Analyses

Figure 2 shows a γ -ray spectrum from the standard Obsidian Rock (SRM-278 from NIST) after subtracting the sample background. In order to have clearer peaks free from overlapping low energy γ -rays, the high energy γ -rays were used. These were 1461 keV of ^{40}K , 1765 keV of ^{214}Bi (from ^{238}U -series), and 2615 keV of ^{208}Tl (from ^{232}Th -series). These γ -lines are the most convenient energies for the determination of K, U, and Th (see Table 3) [1, 10]. To cover the whole region of interest (ROI) under the peak, the following energy windows were considered: The energy window for the K was around 250 keV (1317–1573 keV), for U around 200 keV (1661–1968 keV) and for Th 400 keV (2417–2807 keV). The integrated number of counts in the ROI of each peak was then evaluated. For K and U the background-subtracted net counts were taken. In the case of Th, it was observed that, by using total gross counts instead of net ones, better results were achieved for the determinations. Reproducibility measurements were carried on by measuring one sample for several runs. The results of the analysis showed that the data were reproducible within $\pm 2\%$. The data were corrected for the dead time, which was less than 1% in all runs. Then the data were normalized to the sample mass. The measurements are based on the relative method using standards of almost the same dimensions and geometries as the sample to be measured. Therefore, no corrections are needed for the scattered gammas.

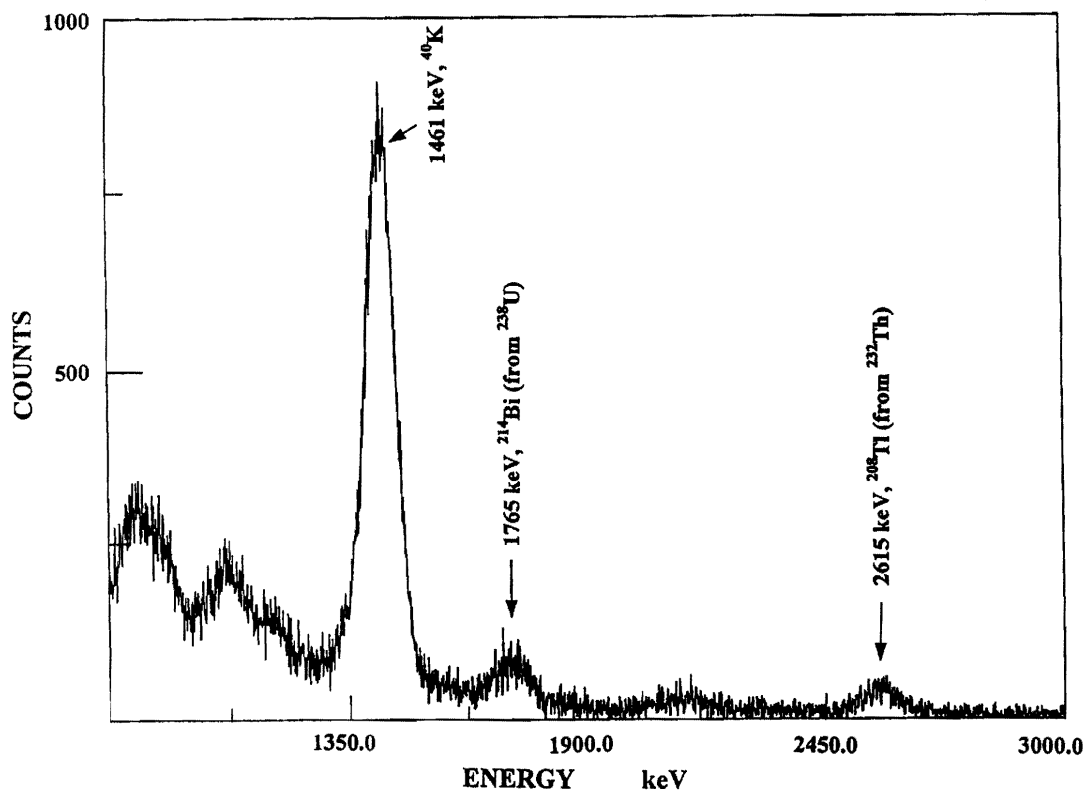


Figure 2. A γ -ray net spectrum from the Obsidian Rock Standard (SRM-278 from NIST). The energy intervals for the K, U, and Th windows were 1317–1573 keV, 1661–1968 keV and 2417–2807 keV, respectively.

The Minimum Detection Limit (MDL) which is the lowest measurable element concentration for each of the elements was determined using the following equation [12-13]:

$$MDL = (m/NC) \times 3.3\sqrt{B} \quad (1)$$

where NC and B are the background subtracted net counts for a sample of mass m and the background counts under the peak of interest, respectively. The sensitivities of the system for these elements were determined for a sample mass of 100 g.

Calculation of Absolute Concentrations

To determine the concentration of K, U, and Th in the sample the following equations are used [11]:

$$K_{sa}(\text{wt.}\%) = \frac{(A_{K,sa} - B_K)/W_{sa}}{(A_{K,st} - B_K)/W_{st}} \times K_{st}(\text{wt.}\%) \quad (2)$$

$$U_{sa}(\text{ppm}) = \frac{(A_{U,sa} - B_U)/W_{sa}}{(A_{U,st} - B_U)/W_{st}} \times U_{st}(\text{ppm}) \quad (3)$$

$$Th_{sa}(\text{ppm}) = \frac{(A_{Th,sa})/W_{sa}}{(A_{Th,st})/W_{st}} \times Th_{st}(\text{ppm}) \quad (4)$$

where,

A_K, A_U, A_{Th} = dead time corrected count rate for K, U, and Th, respectively.

B_K, B_U are the background count rate for K and U.

W_{sa} and W_{st} are the mass of sample and standard.

K_{st}, U_{st} , and Th_{st} are the standard certified concentrations.

Therefore, inserting the measured values into the above Equations 2, 3, and 4, the concentrations of U and Th in ppm and K in wt.% can be calculated.

The uncertainties of the measurements are the statistical uncertainties of the measurements with those of the background counts folded in. The uncertainties were calculated from the following equation:

$$\text{Uncertainty, \%} = \sqrt{(N + B)} / \text{Net Counts} \times 100 \quad (5)$$

where N is the integrated counts and B is the background counts under the peak considered.

Table 3. The Concentrations of Potassium, Uranium, and Thorium in the Certified Standards from IAEA, NIST, and NBL Used in the Present Experiments.

Standard name	Code	K (wt.%)	U (ppm)	Th (ppm)
Feldspar	F1	8.30 ± 0.3	2.52 ± 0.26	(1.37)*
Sediment Lake	SL-1	(1.5)	4.02 ± 0.32	14.0 ± 1
Soil	SOIL-7	(1.21 ± 0.06)	2.6 ± 0.6	8.2 ± 0.5
Soil	IAEA-312		16.5 ± 0.9	91.4 ± 10.1
Sediment Stream	IAEA-313		18.2 ± 1.1	77.1 ± 2.3
Sediment Stream	IAEA-314		56.8 ± 3.9	17.8 ± 1.0
Flint Clay	SRM-97B	0.513 ± 0.023		(36)
Obsidian Rock	SRM-278	3.4535 ± 0.0166	4.58 ± 0.04	12.4 ± 0.3
Western Phosphate Rock	SRM-694	0.423 ± 0.0166	141.4 ± 0.6	
Buffalo River Sediment	SRM-2704	2.00 ± 0.04	3.13 ± 0.13	(9.2)
Uranium (Normal) Ore	NBL-74A		1040 ± 20	
Thorium Ore	NBL-80A		40	1005 ± 10

*The values in parentheses are given for information only.

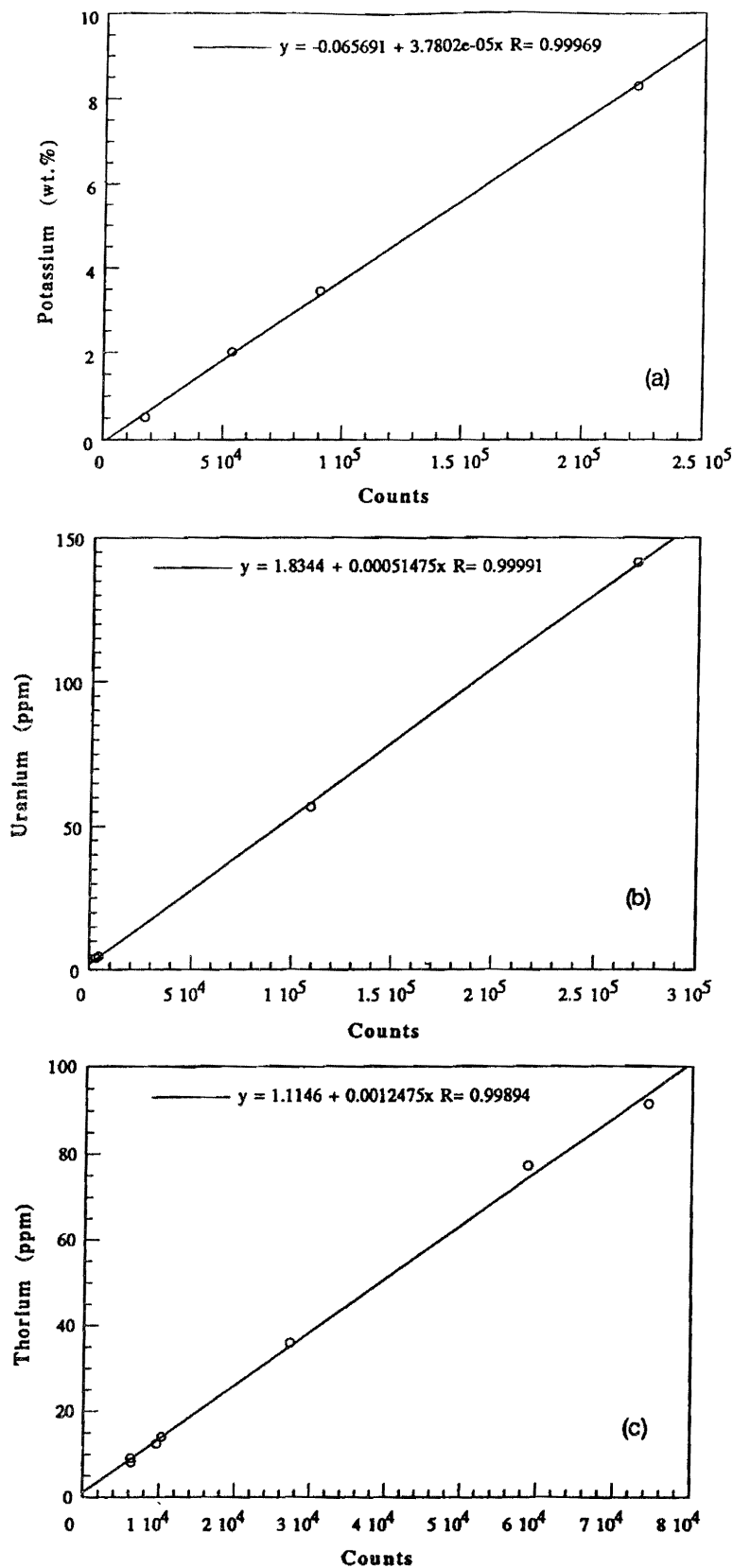


Figure 3. Calibration curve for potassium (a), uranium (b), and thorium (c) determinations by natural gamma-ray spectroscopy. The solid lines are the linear least-square fits.

3. CALIBRATION CURVES

Using Equation (1), the MDL for K, U, and Th were calculated as 16 μg for U; 54 μg for Th; and 15 μg for K, while the sensitivities were smaller than 1 ppm for U and Th and around 150 ppm (or 0.015 wt.%) for K for 100 g sample. Concentrations of an element in the various samples were plotted versus corresponding counts. The data were then fitted with the least-square linear lines. The calibration curves for K, U, and Th are shown in Figures 3(a), 3(b), and 3(c), respectively. The fitting equations are also shown in the figures. The uncertainties were statistical with background uncertainties folded in. The uncertainties of the data calculated as per Equation (5) were 0.2 to 1% for U; 0.2 to 3.3% for Th; and 0.5 to 4.3% for K.

4. SUMMARY

The natural gamma-ray spectrometer facility at the ERL was calibrated for determination of ^{40}K , ^{238}U , and ^{232}Th . The absolute concentration of potassium in wt.%, uranium and thorium in ppm, were determined via measurements of the natural γ -ray activities of 1461 keV of ^{40}K for potassium, 1765 keV of ^{214}Bi for uranium, and 2615 keV of ^{208}Tl for thorium. The measurements were reproducible within $\pm 2\%$. A total of 12 standards from IAEA, Vienna, NIST and NBL, U.S.A., with certified values of U and Th in ppm and K in wt.% were used for the calibration. The sensitivities of the setup for U and Th were better than 1 ppm while for K it was around 0.015 wt.% considering 100 g sample. The uncertainties were statistical with background uncertainties folded in. The uncertainties of the data were 0.2–1% for U, 0.2–3.3% for Th and 0.5–4.3% for K.

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