Applied Radiation and Isotopes ∎ (∎∎∎∎) ∎∎∎–∎∎∎

Contents lists available at ScienceDirect

Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

A review of the nationwide proficiency test on natural radioactivity measurements by gamma spectrometry

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HIGHLIGHTS

- We organized the first proficiency test on radioactivity measurement in Turkey.
- Radioactivity measurement capabilities of the laboratories in Turkey were examined.
- Twelve participant laboratories reported results with 49% acceptable scores.
- Labs using HPGe reported more accurate and precise results than the ones using NaI.
- Participants need more practice and training for their staff.

ARTICLE INFO

Article history:
Received 22 December 2015
Accepted 22 December 2015

Keywords:
Radioactivity in soil
Radioactivity proficiency test
Environmental gamma ray spectrometry
^{226}Ra
^{232}Th
^{40}K

ABSTRACT

This study is the review of the first proficiency test on radioactivity measurement organized in Turkey by Sarayköy Nuclear Research and Training Center (SANAEM) of Turkish Atomic Energy Authority (TAEK) in 2013. The objective of the test was to determine ^{226}Ra, ^{232}Th and ^{40}K activity concentrations in natural soil samples using gamma-ray spectrometry. The bulk material consisting of uranium- and thorium-rich soil and sand was milled, mixed thoroughly and sieved. Homogeneity of the final mix was tested with 6 randomly taken samples. 16 proficiency test samples were distributed to 16 participating laboratories. 12 laboratories reported results. The results were evaluated on the accuracy and precision criteria adopted by the IAEA Proficiency Testing Group. The percentage of acceptable scores was 49%. Some recommendations have been provided to the laboratories to improve the quality of their results. It is planned to extend these proficiency tests periodically for various radionuclides in various matrices.

1. Introduction

Naturally occurring radionuclides are among the main sources of external gamma-ray dose. Human beings are exposed to natural terrestrial radiation dose caused by the radionuclides in soil. Long lived radionuclides such as ^{226}Ra, ^{232}Th and ^{40}K in soil and their corresponding decay products are of special interest in this context. Determination of the activity concentrations of natural radionuclides in soil with high accuracy is important in the assessment of radiation dose by these radionuclides, observing the behavior of natural radioactivity in the eco-system, and monitoring the natural background.

There are many universities and institute laboratories in Turkey currently measuring radioactivity or setting up new measurement systems. They need proficiency tests (PTs) to demonstrate the quantitative measurement capabilities of their laboratories on the identification of natural radionuclides in soil and to ensure the reliability of their measurement and analysis results. Since most of the PTs on radioactivity measurements are organized by institutes which are abroad with limited participation and sometimes with participation fees, laboratories in Turkey cannot regularly participate in these PTs. In addition, participation in PTs with laboratories outside Turkey demand considerable effort from the laboratory due to customs procedures and radiation protection issues. These problems create a lack of willingness in the laboratory to participate in these tests. For these reasons, TAEK, the EC EURAMET delegate on ionizing radiation metrology in Turkey, organized a PT to enable the related laboratories in Turkey to test their analysis methods and help these laboratories in their accreditation processes. This was the first PT arranged in Turkey on ionizing radiation measurement and it is planned to be repeated periodically for various radionuclides in different matrices.

This PT was organized to evaluate the analytical performance of

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http://dx.doi.org/10.1016/j.apradiso.2015.12.053
0969-8043/

the laboratories in universities in Turkey. A total of 16 participants registered for the test. Twelve of the laboratories reported their results. The results of this PT are presented in this work and general recommendations for laboratories to perform better gamma spectrometry analysis are provided.

2. Materials and methods

2.1. Collection and pretreatment of the material

Soil from Manisa province with a relatively high ^{226}Ra activity concentration and a soil–sand mixture from the Aegean coast of Çanakkale province with high levels of ^{232}Th were used to prepare a soil matrix for the proficiency testing. The Manisa site is an area with high levels of uranium and other minerals whereas the Çanakkale coast has some hotspots where ^{232}Th is accumulated with monazite-containing sands at certain points with water currents. These two bulk soil samples from two different regions were subjected to mixing and coarse sieving. The sieved material was milled with the laboratory's ring pulverizer. Then the material was oven-dried at 105 °C for about 48 h until the humidity of the soil was less than 2%. The dried soil was mixed in an industrial mixer for three consecutive days. The mixer was turned off at 2 h periods, the dust was allowed to settle and the stuck material was scraped from the walls of the mixer for a more thorough mixing. The mixed material was then sieved to have a mesh size < 0.15 mm and finally remixed again. A set of 50 plastic containers was filled with 300 g material in each container and sterilized with about 10 kGy ^{60}Co radiation dose at the on-site sterilization plant to prevent microbial growth.

2.2. Homogeneity and activity determination of the PT material

Six of the plastic containers, each having about 300 g sample, were chosen randomly and each bottle was divided into two to test the in-bottle and between-bottle homogeneity of the soil material and to calculate the target values as specific activities for each radionuclide. Each of the sub-samples of about 150 g each were placed in polyethylene beakers. The beakers were completely sealed and waited for more than one month to allow radioactive equilibrium to be reached. This step was needed to ensure that radon gas was confined within the volume and that the daughters of radon would also remain in the sample. After the radioactive equilibrium was reached, each sub-sample was measured by a gamma-ray spectrometric system that consists of an n-type high purity germanium (HPGe) detector with 50% relative efficiency. The gamma-ray spectrometric method used in the analyses was the direct comparison of peak areas, corrected for density and composition, with IAEA RGU-1, RGTh-1 and RGK-1 reference materials. Three gamma-ray spectrometric supplementary methods were also used to check the accuracy. The first method was efficiency transfer from a multi-nuclide reference source for calculation of full energy peak efficiencies for the PT material and then calculation of the coincidence summing correction for ^{226}Ra and ^{232}Th daughters, especially ^{214}Bi, ^{228}Ac and ^{208}Tl. Secondly, the full energy peak efficiencies together with coincidence summing correction factors were calculated with the EGS4 Monte-Carlo code (Nelson et al., 1985). Finally, efficiency calibration was done with LABSOCS (GENIE2000) and the factory-characterized detector from Canberra Industries. Results from all four methods matched very well. The relative biases between the results of the other methods and the direct comparison method were less than 5%. High resolution alpha spectrometry, X-ray fluorescence spectrometry and ICP-MS were used for further accuracy checks with less success due to drawbacks such as small sample intake and tedious sample preparation procedures.

The analysis results were subjected to the Grubbs outlier test (Grubbs, 1969), normal distribution, modality test and ANOVA test (ISO, 2006). There was no reason to exclude any data according to the outlier test. The results were normally distributed which allowed using Gaussian distribution parameters when calculating the uncertainties due to inhomogeneity. Multimodality might be an indicator of inhomogeneity and should be checked during PTs and reference material preparation. All results had single modality which ensured that there was no accumulation of results at two or more values. The ANOVA test was done to evaluate the in-bottle and between-bottle inhomogeneity (Spasova and Vasile, 2010; ISO, 2006).

Final reference value assignment was performed by averaging all the results for each radionuclide. The specific activity of ^{226}Ra was determined from the 295.2 keV and 351.9 keV gamma rays of the ^{214}Pb daughter and the 609.3 keV, 1120.3 keV, 1238.1 keV and 1764.5 keV peaks of the ^{214}Bi daughter. ^{232}Th activities were determined from the following gamma ray peaks: 238.6 keV of ^{212}Pb, 727.3 keV of ^{212}Bi, 338.3 keV, 911.2 keV and 969.0 keV of ^{228}Ac and 277.4 keV, 583.2 keV, 860.5 keV and 2614.5 keV. ^{40}K was determined from its single peak at 1460.8 keV. The final specific activity value for each sample was calculated as the uncertainty weighted average value from all peaks of each radionuclide. The dry weight of the sample material was taken into account in the calculation of the specific activity by considering the moisture content in the sample material.

The uncertainties for the reference values were calculated by the following equation:

$$u_{TV}=k\sqrt{u_{sm}^2+u_{rmm}^2+u_{arm}^2+u_h^2+u_d^2+u_{sts}^2+u_{lts}^2}$$

Here u_{TV} is the expanded uncertainty of the target value, k is the coverage factor for a 95% confidence level, u_{sm} is the sample material counting uncertainty considering background counting, u_{rmm} is the standard reference material counting uncertainty considering background counting, u_{arm} is the uncertainty of the activity of the standard reference material, u_h is the uncertainty due to heterogeneity, u_d is the uncertainty due to density difference between the sample material and the standard reference material and u_{sts} and u_{lts} are the uncertainties due to short- and long-term instability of the material. u_{sts} and u_{lts} were assumed to be negligible in the characterization study since no degradation of the sample is expected in the short- or long-term. The relative uncertainty components due to heterogeneity at $k=1$, u_h, were determined as 1.42%, 1.87% and 2.74% for the activities of ^{226}Ra, ^{232}Th and ^{40}K, respectively. All contributions to the uncertainty budget are

Table 1
Contributions to the uncertainty budget in the calculation of the assigned activity value.

Uncertainty component	u (%) ($k=1$)		
	^{226}Ra	^{232}Th	^{40}K
Sample and background counting statistics	1.60	1.01	6.80
Standard reference material and background counting statistics	0.70	0.50	0.60
Activity of the reference material	0.30	1.38	1.43
Heterogeneity of the sample material	1.42	1.87	2.74
Density difference between the sample material and the standard reference materials	1.0	1.0	2.0
Sample weight[a]	0.03	0.03	0.03
Half-life[a]	< 0.01	< 0.01	< 0.01
Dead time correction[a]	0.01	0.01	0.01
Combined standard uncertainty	2.48	2.76	7.76
Expanded uncertainty ($k=2$)	4.96	5.52	15.52

[a] Negligible contributions in the combined standard uncertainty calculation.

Please cite this article as: Şahin, N.K., et al., A review of the nationwide proficiency test on natural radioactivity measurements by gamma spectrometry. Appl. Radiat. Isotopes (2015), http://dx.doi.org/10.1016/j.apradiso.2015.12.053