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Radioactivity studies in soils from Northwestern Greece

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Abstract Natural radioactivity is examined in soil samples collected from Northwestern Epirus, specifically in Pogoni and Zagori areas. The focus was to determine experimentally the specific activity for naturally occurring isotopes (40 K, U and Th series) and the anthropogenic 137 Cs, in the samples collected from a generally unexplored area of Greece. The measurements were carried out with the use of two High-Purity Germanium (HPGe) detectors, calibrated with two different bulk geometry samples. Based on the measurements of specific activities, the absorbed dosage and index of external hazard, H_{ex} have been estimated through models of UNSCEAR, contributing to the level of environmental sustainability of the area.

Keywords Soil radioactivity, Chernobyl, HPGe, Dosimetry, Gamma-ray spectroscopy

INTRODUCTION

Radioactivity is the spontaneous disintegration of atomic nuclei, extensively omnipresent in the natural environment. Radioactivity studies in soils can provide insights on fallouts (e.g. Chernobyl), long-term impact of nuclear testing, while they can be exploited to monitor industrial applications (e.g. mining or land remediation). We studied the generally unexplored mountainous areas of Pogoni and Zagori in NW Greece, known to have been impacted by the Chernobyl fallout [1,2]. This work is focused on measuring the activity of U-series, Th-series, ⁴⁰K, and ¹³⁷Cs and calculating the dose rate from these isotopes. Results for most of the locations studied in this work are presented for the first time. Two different generation High-Purity Germanium detectors were used, in order to study their relative responses in radioactivity experiments.

EXPERIMENTAL DETAILS

Ten (10) soil samples across an undisturbed area of $\approx 150 \text{ km}^2$ were collected from the remote area of Pogoni and Zagori in NW Greece (see Fig. 1), following IAEA's standards and removing any excess waste and organic matter. The samples were dried overnight in a special oven at 60°C, cleaned from large objects (e.g. gravel) and organic matter, sifted using a sieve with 2-mm holes and sealed in 200 ml PVC containers (Figs. 2A-B). This sample geometry is the standard one used in the Environmental Radioactivity Laboratory at NKUA, where the gamma spectrometers are calibrated with reference materials of the same bulk geometry [3,4].

All soil samples were measured in two γ -spectroscopy stations (Figs. 3A-B), called TIGER and GEROS [5]. The main characteristics of the detectors are summarized in Table 1. For the bulk samples, the duration of each measurement was 80'000 s (live time). The collected spectra were calibrated and analyzed using the software suite SPECTRW [6] and bulk sample activities were deduced using IAEA and CEA reference materials of similar matrices with the soil samples.

Both spectrometers were energy-calibrated in advance with a standard point ¹⁵²Eu source [7]. The equation used to fit the data from the point-source and deduce the absolute energy efficiency of each detector is:

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 $e_{absolute} = \frac{AE^b}{1000C + E^d} , \qquad (1)$ where *E* is the energy and *A*, *b*, *C*, *d* are free parameters to be determined after a best fit.



Figure 1. Area of sample collection



Figure 2. (a) Soil samples during weighing



Figure 3. (a) The spectroscopy station "TIGER"



Figure 2. (b) Soil samples during sieving



Figure 3. (b) The spectroscopy station "GEROS"

	TIGER	GEROS	
Manufacturer	MIRION, 2021	Tennelec, c.1985	
Туре	p, coaxial	p, coaxial	
Ge crystal Radius (mm)	30.3	26.8	
Ge crystal Length (mm)	63.3	53.4	
Bias (V)	+3500	+1300	
FWHM at 1332 keV (keV)	1.8	2.06	
Relative Efficiency	40%	22%	
Shielding	Lead + copper, rigid	Lead, makeshift	
Cooling medium	LN2	LN2	
Detector's window material	Aluminum	Aluminum	

RESULTS AND DISCUSSION

Specific activities

After measuring each sample in both spectroscopy stations, photopeak analysis in the spectra resulted in specific activities (Bq/kg) for ⁴⁰K and ¹³⁷Cs, as well as Th-series (daughters: ²⁰⁸Tl, ²¹²Pb, ²²⁸Ac) and U-series (daughters: ²¹⁴Pb, ²¹⁴Bi), presented in Table 2 and Figs. 4A-D (results from "TIGER" are shown exclusively). In Table 2, the ratios of ⁴⁰K over ¹³⁷Cs and ²³²Th over ²³⁸U are of great importance, the former for a comparison between natural and manmade radioactivity and whether ¹³⁷Cs can be adsorbed [8] and the latter is used in the study of Earth's crust and mantle [9].

	Minimum	Maximum	Mean	Standard Dev.
40 K	142	271	200	40
^{137}Cs	1	26	6.7	8.4
²³² Th series	3	13	7.1	3.3
²³⁸ U series	9	25	17.8	4.7
⁴⁰ K over ¹³⁷ Cs	7	310	93.6	93.7
²³² Th over ²³⁸ U	1.5	4.1	2.9	0.8

 Table 2. Specific activities (in Bq/kg) and relevant ratios



Figure 4. (a) Specific Activity of ^{238}U



Figure 4. (b) Specific Activity of ²³²Th



Figure 4. (c) Ratio of specific activities of ⁴⁰K over ¹³⁷Cs, in logarithmic scale



Figure 4. (d) Ratio of specific activities of ²³²Th over ²³⁸U

Dosage and Hex

In this section the dose in the areas of interest is examined. The absorbed dose rate, D, as well as the index of external hazard, H_{ex} for natural and manmade radioactivity are extracted using UNSCEAR models [10]. These parameters are of importance for determining the sustainability of sampling site in terms of radioactivity concentrations. Note that typical values for the index of external risk for soil samples are in the range from 0.1 to 1. The results from the presently reported measurements are summarized in the table below (see Table 3).

	Minimum	Maximum	Mean	σ
Natural Dose Rate (nGy/h)	13 (5)	29 (11)	21.4	4.9
Natural H _{ex}	0.11 (11)	0.25 (10)	0.19	0.04
Manmade Dose Rate (nGy/h)	0.11 (10)	3.2 (17)	0.83	1.04
Manmade H _{ex}	0.001 (10)	0.028 (6)	0.0073	0.0091

Table 3. Natural and manmade dosage rate and H_{ex} . Errors are shown in parenthesis.

Comparison of the two detectors

From the comparison of the detector performance, the aged detector, GEROS, is found to fall short when compared to the much newer TIGER, especially in terms of overall absolute efficiency and energy resolution (FWHM). The latter is prominent at the spectrum areas where multiple photopeaks are present. A typical representation can be seen in the image below (see Fig. 5) where, at the same energy

range, the two photopeaks from TIGER's spectrometer coincide in GEROS' and appear as one due to lower resolution.



Figure 5. *The 352 keV line of*²¹⁴*Pb (range 350-360 keV) in both spectrometers. The resolution is significantly better in TIGER (left) than GEROS (right).*

CONCLUSIONS

The purpose of this work was to study and evaluate the radioactivity levels, as well as the sustainability of the remote areas of Pogoni and Zagori in NW Greece, using two different HPGe detectors. Through γ -ray spectroscopy, a total of ten soil samples were analyzed and the values of specific activity for ⁴⁰K, ²⁰⁸Tl, ²¹²Pb, ²¹⁴Pb, ²¹⁴Bi, ²²⁸Ac and ¹³⁷Cs were deduced. It can be concluded that the levels of natural radioactivity are comparable to other geographically similar regions in Greece [11-14]. Anthropogenic radioactivity (¹³⁷Cs) is found at background level, in agreement with [2], which showed in 1989 to be \approx 27 Bq/kg in nearby regions. Furthermore, no risk from external dose hazard was found when calculating the dosage and H_{ex}. A comparison between the two detectors showed that the aging GEROS shows worse operational status compared to the newer one, TIGER.

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