RADIONUCLIDES DISTRIBUTION AND ASSOCIATED ECOLOGICAL RISK IN THE ENVIRONMENT OF ARMENIAN MOUNTAINS

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Abstract

The paper summarizes the results of radioecological monitoring in the mountainous regions of Armenia in order to identify the distribution of radionuclides (natural Ra-226, Th-232 and artificial Cs-137) and the associated environmental risks in the mountainous environment of Armenia. Soil samples from the surface of eight mountain ridges and massifs were collected along altitude (900-3200 m above sea level). Gamma spectrometry has been used to identify radionuclides and measure activity concentration. A significant correlation was found between Ra-226 and Th-232, no statistically significant correlation was observed between Cs-137 and natural radionuclides. The results of the Kruskal-Wallis test reveal the height-dependent pattern of the studied radionuclides in the soil: the concentration of Cs-137 activity increases with height, the variances of the average values of the Ra-226 and Th-232 activity concentrations do not change in absolute height. An assessment of the ecological risk to non-human biota using the ERICA Tool identified risk factors (RQs) and limiting reference organisms in the mountain environment.

Keywords: Cs-137, NORM, mountains, Erica Tool, Armenia

РАСПРЕДЕЛЕНИЕ РАДИОНУКЛИДОВ И ЭКОЛОГИЧЕСКИЙ РИСК В ГОРНОЙ СРЕДЕ АРМЕНИИ

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Аннотация

В данной статье обобщены результаты радиоэкологического мониторинга в горной среде Армении с целью выявления распределения радионуклидов (естественный Ra-226, Th-232 и техногенный Cs-137) и связанных с ними экологических рисков в горной среде Армении. Образцы почв с поверхности восьми горных хребтов и массивов отбирались по высоте (900-3200 м над уровнем моря). Для идентификации радионуклидов и измерения удельных активностей использовалась гамма-спектрометрия. Между Ra-226 и Th-232 была обнаружена значимая корреляция, а между Cs-137 и естественными радионуклидами статистически значимой корреляции не наблюдалось. Результаты теста Крускала-Уоллиса выявляют закономерности распределения исследуемых радионуклидов в почвах: активность Cs-137 увеличивается с высотой, а дисперсии средних значений активности Ra-

226 и Th-232 не изменяются по абсолютной высоте. Оценка экологического риска с помощью ERICA Tool выявила коэффициенты риска (RQ) и ограничивающие референтные организмы в горной среде.

Introduction

The interest in environmental radioactivity research has grown after the historical nuclear accidents, incidents, and nuclear weapon tests in the 20th century. The radionuclide composition of the environment has several components: naturally occurring radioactive materials (NORM) in the terrestrial environment, airborne radionuclides originating by cosmic rays in the earth's atmosphere, artificial radionuclides from global radioactive fallout. U-Ra and Th radioactive series' radionuclides, as well as K-40 are the main gamma dose forming natural radionuclides and are widespread in the environmental compartments [1–4]. H-3, Be-7 and C-14 are main secondary cosmogenic radionuclides that originate from the interaction of cosmic rays and atmospheric gases [4]. From global fallout radionuclides, artificial Cs-137 has the most focus in environmental issues due to its long half-life (30.2 y). Given the diversity of radioactive sources, the prevalence of radionuclides, there are still many areas of interest for assessing radioactive levels. Mountainous areas are such an example. Compared with adjacent valleys, mountains characterize by higher precipitations and may serve as barriers for atmospheric pollutants [5–7]. Moreover, the local geological features determine the distribution of NORM in soils. Mountain areas, especially the highest terrains, are not for permanent inhabitation and are mainly used as a temporary pasture. However, it is important to assess the risk to other organisms, especially when the mechanism for assessing the risk associated with radionuclides is not yet complete for non-human biota.

Taking into account the above, this study is aimed at determining the activity concentrations of natural Ra-226, Th-232 and artificial Cs-137, as well as assessing the ecological risk in accordance with the integrated ERICA approach in the mountainous environment of Armenia.

Materials and Methods

The highest mountains of Armenia, including eight mountain ridges and massifs, were selected for the study: Aragats Mountain, Syunik Highland, Pambak Ridge, Geghama Ridge, Vardenis Ridge, Bazum Ridge, Meghri Ridge, Khustup-Katar Ridge (*Fig. 1*). Armenia (total area 29.743 km^2) is located at the south of the great mountain range of the Caucasus and northeast of the Armenian Highlands (latitudes $38^050'$ and $41^018'$, longitudes $43^027'$ and $46^037'$). The territory of Armenia is mostly mountainous; the average height above sea level is 1830 m, with the lowest point of 379 m (Araks river canyon) and the highest of 4090 m (Aragats Mountain).

Overall, 130 soil samples from upper horizon (0-5 cm) were collected along altitude (900-3200) in every 200 m a.s.l. Soil sampling procedure, laboratory preparation were carried out according to [8–10] and are described in more detail in [5].
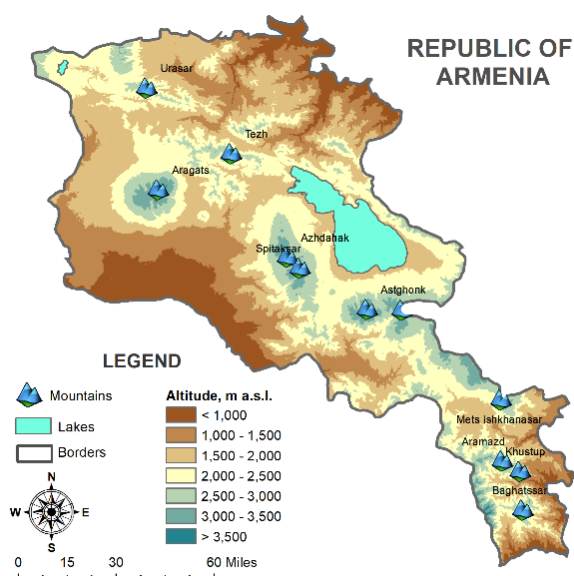
Fig. 1. The highest mountain ridges and massifs of Armenia

Gamma spectrometry (CANBERRA) was applied for identification and specific activity measurements of radionuclides. System consists of high purity germanium coaxial detector (HPGe) with energy resolution 1.8 keV FWHM for the Co-60 gamma-ray energy line at 1332 keV, which is coupled to a DSA-1000 multichannel analyzer. Soil sample spectrum acquisition and analysis were performed using Genie2K software. The efficiency calibration was performed using LabSOCS [11]. For energy calibration of the gamma spectrum, gamma lines of Eu-155, Na-22 and Co-60 were used as reference ones. Background measurements were implemented every week using clean and empty Marinelli beakers. Gamma spectrum acquisition time for each sample was 20000 sec. Cs-137 was determined directly from the 661.5 keV energy line. Ra-226 and Th-232 were measured using the daughter product gamma lines.

For QC/QA IAEA-447 reference material was used and an inter-laboratory comparison program with the National Nuclear Forensic laboratory was conducted. Statistical analysis was performed by means of IBM SPSS Statistics 20 and RStudio software. For mapping, ArcMap 10.3 program was used and ecological risk assessment was carried out using ERICA Tool (Version 1.3) program.

Results and Discussion
The descriptive statistics of investigated radionuclides is presented in (

Table 1). Cs-137 was found in all soil samples. For Ra-226 and Th-232, only one sample contains minimum detectable levels (<MDA). Cs-137 is more abundant at higher altitudes (from 2000 m). The greatest activity was observed in the area of Mount Aragats at an altitude of 3000 m. The activity concentrations of Ra-226 and Th-232 are within the ranges of UNSCEAR values for the soils of Armenia. Ra-226 and Th-232 did not exhibit altitudinal dependence. Moreover, significant correlation was found between Ra-226 and Th-232 radionuclides (Spearman's rho, 0.748 at 0.01 level, 2-tailed). No correlation was observed between Cs-137 and other radionuclides.

Table 1. Descriptive statistics of investigated radionuclides.

Parameter	Cs-137	Ra-226	Th-232
Mean	46.7	17.1	32.9
Median	31.6	15.4	29.3
Std. Deviation	52.8	9.7	17.6
Variance	2792.6	94.8	310.9
Minimum	0.96	2.49	2.89
Maximum	445.7	70.1	97.6

Kruskal-Wallis test (one-way analysis of variance) was applied to see the differences of means of radionuclides in different altitudes. The results show that only for Cs-137 the null hypothesis is rejected, which means that variances of means of Cs-137 are different at selected altitudes (p-value = 3.117e-06). Null hypothesis is accepted Ra-226 and Th-232, which does not show a similar distribution, their averages are almost the same at different heights (p-value = 0.4, 0.5, respectively). This is also confirmed by boxplots (*Fig. 2*) and visible in distribution maps (*Fig. 3*).
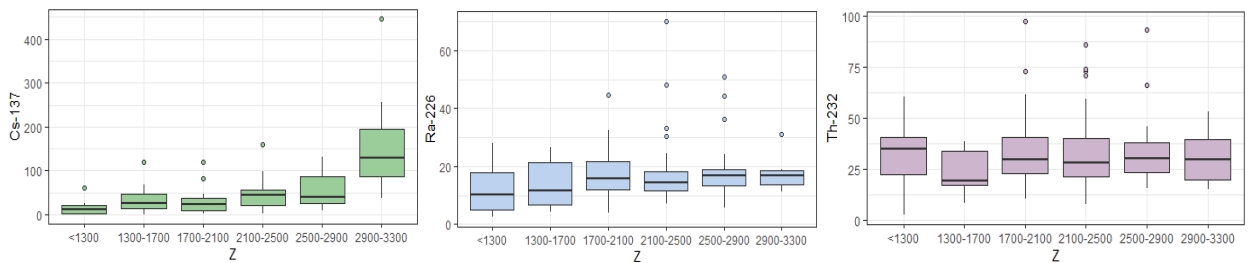

Fig. 2. The distribution of investigated radionuclides in different altitudes

The average and highest activity concentrations of radionuclides in soil were used to assess ecological risk for non-human biota with Tier 1 Approach in ERICA Tool. With average values the Risk Quotient (RQ) is 1.03, which is below 10 µGy/h reference dose rate. Using the maximal activities, the RQ was found to be 4.5, exceeding the reference dose rate. The limiting reference organisms are lichens and mosses and the dose mainly comes from Ra-226.
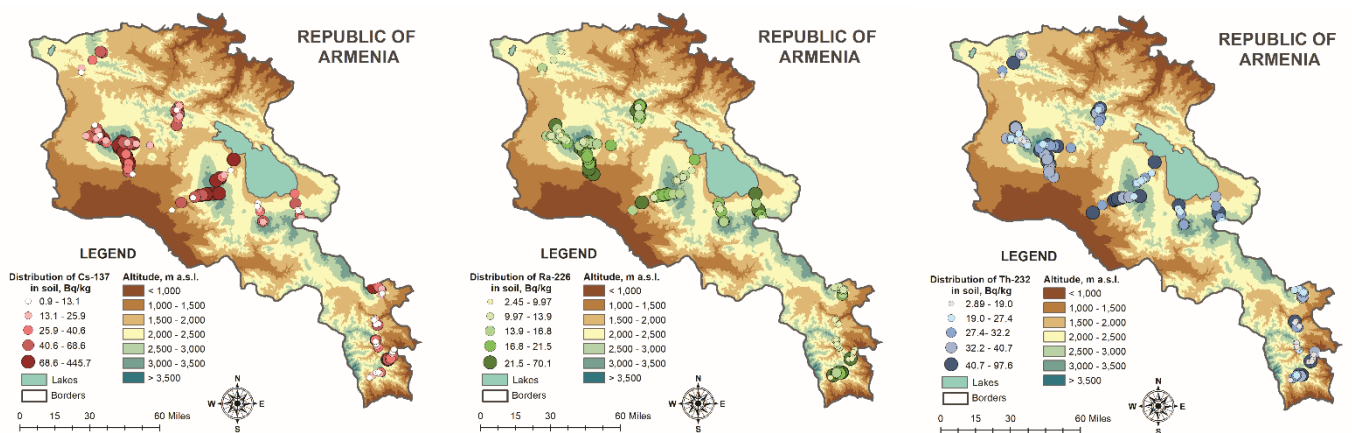

Fig. 3. Distribution of Cs-137, Ra-226, Th-232 activity concentrations in soils of mountainous environment of Armenia

Conclusions

The study brings the following conclusions: Mountainous environments are in special focus in studying environmental radioactivity issues. The distribution of artificial Cs-137 in the mountain areas of Armenia has altitudinal-dependence patterns, emphasizing the cross-border transfer of global pollutant Cs-137. The activity concentrations of natural Ra-226 and Th-232 radionuclides are within the ranges of UNSCEAR average for the soils of Armenia. Ecological risk assessment shows that from the highest values of investigated radionuclides (in particular Ra-226) lichens and mosses are exposed. The study plays an important role in further evaluation of the background and baseline activities of radionuclides in Armenia and cross-border transfer in the region.

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