NATURAL RADIOACTIVITY OF TERRESTRIAL ORIGIN IN ALBANIAN SOIL SAMPLES

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Abstract

This study aims to discuss the first data on the concentration of natural radioactivity in soil gathered within the framework of the didactic activities at the Physics Department. Top soil samples are randomly collected from near fifty different locations. The average activity concentrations measured by using high-resolution gamma ray spectroscopy method are found to be respectively 394 ± 121 Bq kg^{-1}, 22 ± 8 Bq kg^{-1}, 30 ± 10 Bq kg^{-1}, 27 ± 11 Bq kg^{-1} and 25 ± 10 Bq kg^{-1} for the ^{40}K, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th. The existence of secular equilibrium in ^{232}Th and ^{238}U decay chains is confirmed through the activity ratios of ^{228}Ra/^{228}Th and ^{226}Ra/^{210}Pb respectively. The top soil activity concentration of lead is found to be significantly affected by the unsupported lead, which seems to be an indicator of high soil radon emanation processes. The average absorbed dose rate and the corresponding annual effective dose rate in outdoor air are found to be 42 ± 13 nGy h^{-1} and 0.05 ± 0.02 mSv y^{-1} respectively. These values are comparable with the world-wide levels reported in literature. These results can be used in the future in order to build the natural radioactivity map of the Republic of Albania.

Key words: Natural radioactivity, terrestrial radioactivity, annual dose rate, radiological hazard, gamma spectrometry.

Introduction

The environmental radioactivity and especially the naturally occurring radionuclides are generally found in various concentrations due differences in the geological composition of the earth's crust. The most significant source of natural exposure for humans arises from ^{40}K and ^{232}Th and ^{238}U decay chain progenies (UNSCEAR 2008). To our knowledge, no systematic surveys are conducted to study the distribution of the levels of the natural activity concentration across the territory of the Republic of Albania. However, in few studies the concentration of ^{137}Cs due to radioactive fallout is reported to vary from <0.3 Bq kg^{-1} to 90.0 Bq kg^{-1} (Xhixha,. *et al.* 2019a). In Berat and Fier regions average concentration of natural radionuclides, of 20 ± 5 Bq kg^{-1}, 25 ± 10 Bq kg^{-1}, 25 ± 9 Bq kg^{-1} and 326 ± 83 Bq kg^{-1}, respectively for ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K (Xhixha *et. al.* 2015), are reported.

This work aims to present and discuss the first data on natural radioactivity concentration across country as a starting point for the realization of the natural

radioactivity map of soils in Albania. For each soil sample, the activity concentration of ^{210}Pb, ^{226}Ra, ^{228}Ra, ^{228}Th and ^{40}K is measured by using a High Purity Germanium (HPGe) detector. The equilibrium in the ^{238}U and ^{232}Th decay chains is checked by the ^{210}Pb/^{226}Ra and ^{228}Ra/^{228}Th ratios, respectively. These data are used to evaluate the radiological hazard for the population.

Materials and methods

Sampling and sample preparation

Soil samples (49 samples), up to a depth of 5-10 cm, are collected randomly within the territory of Albania. The coordinates of each location are recorded using a GPS device and a panoramic photo is generally taken for documentation. Soil samples are first hand cleaned of any impurities (vegetation or stones) and then dried for at least 24 h at a constant temperature of 105°C. Samples are homogenized to a grain size of less than 2 mm and transferred into Marinelli beakers with a volume of 500 cm^3. The containers are sealed for radon gas and left undisturbed for four weeks in order to establish a radioactive equilibrium in the ^{226}Ra decay chain.

Gamma-ray spectrometry measurement

Soil samples are measured using a low background HPGe detector of relative efficiency of 40%. The HPGe detector has an energy resolution of 1.8 keV for 1.33 MeV gamma line of ^{60}Co. The absolute efficiency is calibrated in the energy range from 30 keV to 2000 keV using LabSOCS (Laboratory Sourceless Calibration Software). The uncertainties in the determination of the absolute efficiency vary from 10% at low energies to 4% at high energies (Shyti, 2019). The calibration procedure is then cross-validated through the participation in the wide open proficiency tests (IAEA-TEL-2018-03). The intercomparison showed generally a relative bias of less than 10% for most radionuclides, except for ^{210}Pb where the relative bias is found to be 50% (however this is reasonable due to the sensitivity of the correction for self-absorption).

Each soil sample is measured for 86400 seconds. The activity concentration of ^{226}Ra (^{238}U decay chain) is determined through its short-lived progenies ^{214}Pb and ^{214}Bi. In the ^{232}Th decay chain the activity concentration of ^{228}Ra is determined through the direct progeny ^{228}Ac, while ^{228}Th is determined through its short-lived progenies ^{212}Pb and ^{208}Tl (gamma energies are stated in (Xhixha, *et al.* 2016). In the case of the determination of the activity concentrations of ^{210}Pb (^{238}U decay chain) and ^{40}K their single gamma emissions are used.

Evaluation of the radiological hazards

The human exposure due to the external gamma radiation related to the presence of natural radionuclides in soil is calculated according to UNSCEAR (2008):

$$E\ (mSv\ y^{-1}) = 0.7 \cdot 8760 \cdot [0.462 \cdot C_{Ra\text{-}226} + 0.604 \cdot C_{Th\text{-}232} + 0.0417 \cdot C_{K\text{-}40}] \cdot 10^{-6}$$

where E (mSv y^{-1}) is the annual effective dose rate in air at one meter height above ground. The measured activity concentrations C_{Ra-226}, C_{Th-232} and C_{K-40} respectively, for ^{226}Ra, ^{232}Th and ^{40}K are given in Bq kg^{-1}. The constants 0.462, 0.604 and 0.0417 are the dose rate conversion coefficients expressed in nGy h^{-1} per Bq kg^{-1}. The constant 0.7 is a conversion factor that takes into account the effect of radiation to humans and is expressed in nSv nGy^{-1} (effective to absorbed dose ratio).

Results and discussions

The statistical data for the activity concentrations of ^{40}K, ^{226}Ra, ^{210}Pb, ^{228}Ra, ^{228}Th and ^{137}Cs are summarized in Table 1. The skewness and kurtosis indicate symmetric but spread distribution of the activity concentration values of ^{40}K and ^{210}Pb, while for ^{226}Ra, ^{228}Ra, ^{228}Th, and ^{137}Cs the distributions are positively skewed and very peaked respect to a normal distribution. However, the averages and the standard deviations are calculated for all radionuclides as representative values for the comparison. The average values of the activity concentration of ^{40}K, ^{226}Ra, ^{210}Pb, ^{228}Ra and ^{228}Th in the soil samples are found to be respectively 394 ± 121 Bq kg^{-1}, 22 ± 8 Bq kg^{-1}, 30 ± 10 Bq kg^{-1}, 27 ± 11 Bq kg^{-1} and 25 ± 10 Bq kg^{-1}. Similar values are found in the Balkan region (Xhixha, *et al.* 2019b and references within).

Table1. Statistical data on the activity concentration (Bq kg^{-1}) of radionuclides in soil samples.

		^{238}U decay series		**^{232}Th decay series**	
	^{40}K (Bq kg^{-1})	**^{226}Ra (Bq kg^{-1})**	**^{210}Pb (Bq kg^{-1})**	**^{228}Ra (Bq kg^{-1})**	**^{228}Th (Bq kg^{-1})**
range	111-632	7-54	7-57	5-69	5-65
average	**394**	**22**	**30**	**27**	**25**
median	406	20	29	26	24
st. dev.	121	8	10	11	10
skewness	-0.4	1.8	0.3	1.0	1.1
kurtosis	0.1	5.0	0.2	3.3	3.5

The ratio of ^{228}Ra/^{228}Th is found to be close to unity with a good correlation (C_{Th-228} = [0.93±0.10] ·C_{Ra-228}), confirming the existence of secular equilibrium in the decay chain. On the other hand, the ratio of ^{226}Ra/^{210}Pb indicates a very poor correlation (C_{Pb-210} = [0.63±0.16] C_{Ra-226} + [15.85±3.73]), probably due to the high uncertainty in the measurement of ^{210}Pb and due to unsupported lead (Figure 1). Indeed, several studies have

demonstrated that the contribution of unsupported lead is relevant (He and Walling 1997). Therefore, a soil core at depth up to 50 cm is sampled every 10 cm in order to check the evidence of the contribution of unsupported ^{210}Pb (Figure 2). Data shows that the contribution of unsupported ^{210}Pb in top soil is relevant, while at depths below 20 cm the activity concentration of ^{226}Ra and ^{210}Pb are comparable within the uncertainty. These results seem to indicate the existence of secular equilibrium in the uranium decay series for the investigated soil samples.

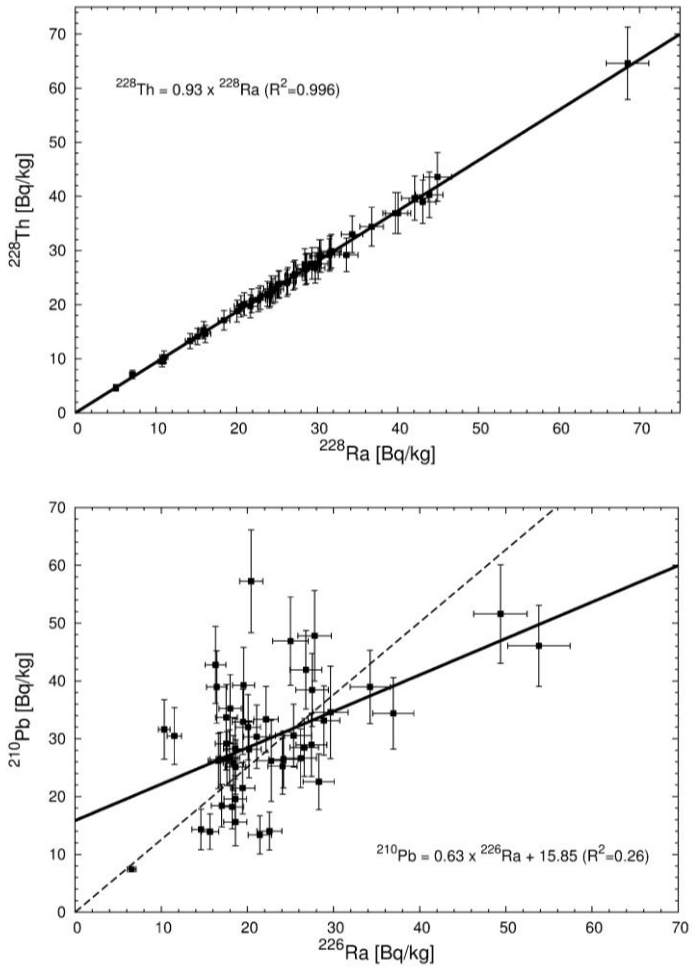

Figure 1. Correlations between ^{226}Ra/^{210}Pb and ^{228}Ra/^{228}Th.

The calculated values of gamma absorbed dose rate are found to vary from 12 to 82 nGy h^{-1} with an average of 42 ± 13 nGy h^{-1}. Considering an outdoor occupancy factor of 20%, this corresponds to an annual effective dose ranging from 0.01 to 0.10 mSv y^{-1} with an average of 0.05 ± 0.02 mSv y^{-1}. These results are comparable with the world average outdoor absorbed

gamma dose rate of 58 nGy h^{-1}, which corresponds to an average effective dose rate of 0.07 mSv y^{-1} (UNSCEAR 2008).

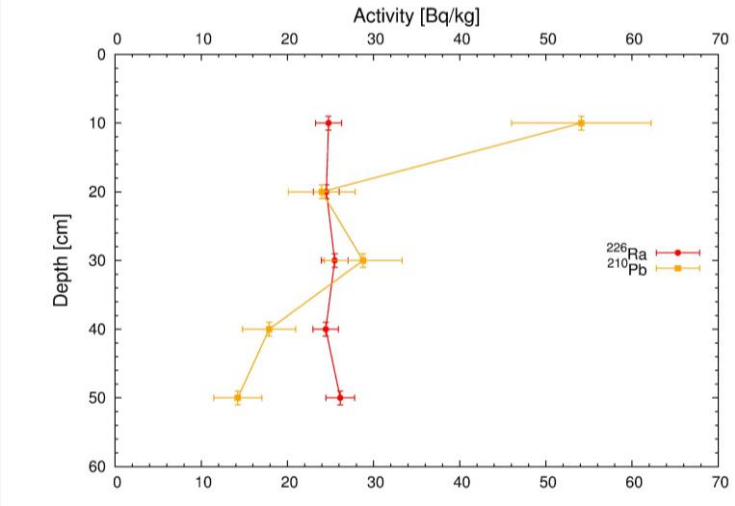

Figure 2. Activity concentration of ^{226}Ra and ^{210}Pb in the soil core sample at different depths.

Conclusions

Based on these preliminary results we conclude that:

The activity concentration values of ^{40}K, ^{226}Ra (^{238}U) and ^{232}Th are generally found to be comparable with activity concentrations found in literature regarding the Balkan region.

The existence of secular equilibrium in thorium decay chain is confirmed by the activity concentration ratios of ^{228}Th/^{228}Ra, which is found to be close to unity (0.93±0.1).

The activity concentration ratios of ^{226}Ra/^{210}Pb indicate a relevant contribution due to unsupported lead. This conclusion is supported by the analysis of soil core at depth up to 50 cm, showing that below 20 cm the activity concentrations of ^{226}Ra and ^{210}Pb are comparable within the uncertainty and indicating the existence of secular equilibrium in the uranium decay chain.

The calculated average value of the annual effective dose rate is found to be 0.05 ± 0.02 mSv y^{-1}, which is comparable with the worldwide average values of 0.07 mSv y^{-1}.

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