IS HIGH INDOOR RADON CONCENTRATION CORRELATED WITH SPECIFIC ACTIVITY OF RADIUM IN NEARBY SOIL? A STUDY IN KOSOVO AND METOHIJA.

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Acknowledgements

The present work was supported by the Ministry of Education, Science and Technological Development of the Republic of Serbia, under the Project No. III41028 and the Project No. 171021.

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Abstract

This paper presents indoor radon concentrations and specific activities of natural radionuclides measured in soils of Kosovo and Metohija. The measurements of radon concentration were performed during two consecutive six-month periods in two rooms of 63 houses using CR-39 detectors. The annual radon concentration ranged from 30 to 810 Bq m^{-3} with the average value of 128 Bq m^{-3}. Almost 15% of houses had radon concentration higher than 200 Bq m^{-3}. The difference between radon concentrations measured in the two six-month periods was analyzed, showing, as expected, a slightly higher radon concentration in the "winter period" than in the "summer period". The variation between different rooms of the same houses was also analyzed, showing that 20% of the dwellings had a significantly higher radon concentration (> 100 Bq m^{-3}) in one room compared to the other (the coefficient of variation ranged up to 96%). The specific activities of natural radionuclides in the nearby soil were determined by gamma spectrometry. The estimated average value (and standard deviation) of ^{226}Ra, ^{232}Th and ^{40}K specific activities were 32 (13), 35 (16), 582 (159) Bq kg^{-1}, respectively. The correlation between indoor ^{222}Rn and ^{226}Ra content in soil was investigated. Only a weak correlation was found (Spearman's rho=0.220) indicating that other factors might affect diffusion and accumulation of radon indoors, as confirmed also by the high variability between the rooms of the same houses.

Keywords: indoor radon; radionuclides; correlations; seasonal variation; lung cancer risk; effective dose

1. INTRODUCTION

The only gaseous product of disintegration in ^{238}U series is radioactive noble gas radon, ^{222}Rn which is produced by the decay of ^{226}Ra. In general, the main sources of radon are soil and rocks that are rich in uranium and radium or they are located near ore deposits. Due to the relatively long half-life (3.824 days), radon can be found far from its sources (Duranni and Ilic, 1997). Fissures in the soil under the buildings, as well as cracks in building construction enable efficient transport and entry to the indoor atmosphere by diffusion and advection that leads to the radon buildup and contamination of living space. The contribution of radium from soil to indoor radon concentration depends on the coefficient of emanation and permeability of the ground, as well as on thickness and tightness of the building structure. Many studies have been conducted aiming to investigate the correlation between indoor radon concentration and radium (or uranium) content in nearby soil. Some of them found either no correlation, or a weak correlation (Singh et al., 2005; Celik et al., 2008a; Vinay Kumar Reddy et al., 2012; Bossew et al., 2013; Gulan et al., 2013; Forkapic et al., 2016), while others indicated a strong association between these two variables (Singh et al., 2002; Mehra et al., 2006; Kucukomeroglu et al., 2009; Abd El-Zahed, 2013; Friedmann et al., 2016; Farid, 2016). Some of these studies were local in character while others included big areas and large number of samples (Nero et al. 1994; Bossew et al., 2013). The earlier indoor radon studies conducted in Kosovo and Metohija were either limited on 3-month measurements (Milic et al., 2010, 2011), or carried out in specific areas (Zunic et al., 2001, 2010). These studies contained no data on soil radioactivity.

The current study was a continuation of a survey started in December 2009, which included a lower number of measurements (Gulan et al., 2013a). This work describes a radioecological survey of Kosovo and Metohija; the study included annual radon measurements in 63 houses and measurements of radionuclide concentrations in soil near the houses. Bearing in mind the possible adverse effect of radon on health, relative risk for lung cancer and relative contributions to effective dose from radon and terrestrial radionuclides were estimated. However, the main purpose of the study was to investigate the correlation between natural radionuclides in soil and indoor radon concentration. An effort has been done for better understanding, implication and correlation with geology, in order to investigate a possibility of further predicting of indoor radon in some indicative cases. Besides, the study intended to determine a possible difference between radon concentrations measured in different rooms of the same buildings, as well as to investigate the seasonal variations of indoor radon levels.

2. MATERIALS AND METHODS

2.1. Study area

Kosovo is located in Southeastern Europe, between latitude 41°51' to 43°15' N and longitude 20°01' to 21°48' E; it occupies an area of 10.887 km^2 with an average altitude of about 800 m, but with extreme altitude changes of relief and morphology (Gavrilov et al., 2017). The province is mostly mountainous, the lowest parts are located at an altitude of 297 m and the highest point is at an altitude of 2656 m. Tectonic Kosovo basin and Metohija valley stand out in the varied relief. The mountain rim of basin is made up of Palaeozoic schist, Mesozoic limestone, volcanic and metamorphic rocks with layers of marl, sandstone and coal deposits (Dimitrijevic, 1997). The diversity of geological structures and materials of Kosovo and Metohija are conditioned by various periods in their creation from Cambrian (Paleozoic era) to the Holocene in Quaternary. It is a unique space of characteristic tectonic and magmatic events, which includes vertical and horizontal faulting. Two faults stretch toward North-Northwest and South-Southeast along the narrow and long tectonic zone of Senonian flysch deposits (Geological Atlas of Serbia, 2002).

2.2. Radon measurements

Measurements of indoor radon activity concentrations were conducted in 34 mostly rural settlements of 10 municipalities of Kosovo and Metohija (Fig. 1). Sampling locations were selected based on demographic structure: the study involved settlements with high population density. One year measurements (from December 2010 to 2011) were performed in 63 houses during two seasons: winter-spring (I period) and summer-autumn season (II period) with semi-annual exchange of detectors. The detectors were placed in two rooms (a living room and a bedroom) at a distance of about 30 cm from walls (to reduce the contribution of thoron) and on height of 1.5–2 m from the room's floor. Complete measurements were carried out in 54 houses. In 4 houses, measurements were carried out for a single one-year period. In other 5 houses measurements were performed in one six-month period or/and in one room only.

Houses were chosen randomly, regardless of the year of construction. Generally, houses in Kosovo are detached buildings, mainly single-storey, with or without concrete foundation slabs and with an average age of about 30 years. Usually they are built on sandy soil or clay. All the houses under investigation had doors and window frames made of wood, unable to provide tight sealing. Radon concentration was measured by a passive device based on CR-39 detector (model TASTRAK). It consists of a dome-shaped diffusion chamber, made of conductive plastic with 4.5-cm diameter and 2-cm height, and a detector (CR-39 with the area of 2.5 × 2.5 cm^2 and 1 mm thickness) placed on the bottom of the diffusion chamber.

Figure 1. Map of Kosovo and Metohija

(grey- surveyed municipalities, brackets- no. of measurements)

After the exposure detectors were chemically etched in a thermal bath with a solution of 6.25 M NaOH for 1 h at 98 °C. In order to stop the etching, detectors were washed with hot distilled water, kept for 30 minutes in a 2% aqueous solution of acetic acid with continuous mixing, and then placed in a drying chamber with a fan (Carpentieri et al., 2011). Etching, tracks counting and evaluation of radon concentration was done by the Italian National Institute of Health. A calibration of diffusion chamber was performed by exposing in a chamber with high radon concentration; values of calibration exposure ranged from 300 to 65000 kBq h m^{-3}. The calibration coefficient for exposures up to 4000 kBq h m^{-3} (corresponding to 926 Bq m^{-3} for six months) was 1.92±0.13 [tracks cm^{-2} (kBq m^{-3} h$)^{-1}$].

The measurement uncertainty was evaluated taking into account both calibration component and variability component. The uncertainty component due to the calibration is largely due to the uncertainty of the reference radon concentration (5%, k=1) since the number of detectors exposed in the chamber during the calibration was chosen to minimize the variability component (10 detectors). The variability component of the measurement uncertainty was evaluated both in radon chamber (by the CV of groups of the 10 detectors exposed at different levels) and in field by means of the variability of paired devices (Carpentieri et al., 2011, Bochicchio et al, 2014). The total uncertainty of the single measurement varies between 6% and 8% depending on the radon concentration.

2.3. Determination of gamma activity concentrations of radionuclides in soil

Surface samples of undisturbed soil (0–5 cm) were collected near the houses where radon was measured (at the distance of 2–3 m) during April 2011. The sampling was performed according to the recommendations of International Atomic Energy Agency (IAEA, 1989). After removing stones and roots, samples were air-dried and homogenized using a 2 mm sieve. They were hermetically sealed in 450 ml Marinelli beakers and left for more than 4 weeks in order to achieve a secular equilibrium between radium and its progeny.

Gamma-spectrometry measurements were performed using coaxial HPGe detector (model GEM30-70, ORTEC) with a relative efficiency of 32% and energy resolution (FWHM) of 1.85 keV at 1.33 MeV (^{60}Co). The detector was calibrated using a mixture of gamma-emitting radionuclides (MBSS 2) provided by Czech Metrological Institute. The detector was shielded in lead of 10 cm thicknesses. Each measurement lasted for 10800 s. Specific activity of ^{226}Ra was determined based on the gamma-ray lines corresponding to ^{214}Pb (351.9 keV) and ^{214}Bi (609.3 keV). Specific activity of ^{232}Th was obtained by the gamma-ray lines of ^{228}Ac (911.1 keV and 968.9 keV) and ^{208}Tl (583.0 keV and 860.6 keV). The gamma-ray line at 1460.7 keV was used for estimating specific activity of ^{40}K.

3 RESULTS AND DISCUSSION

3.1. Indoor radon concentrations

Table 1 presents descriptive statistics of the measurement results. The mean annual ^{222}Rn concentration ranged from 30 to 810 Bq m^{-3}, with an average value of 128 Bq m^{-3}. The same table also presents the results of estimating relative risk for lung cancer based on the assumption that the relative risk increases by 16% per each 100 Bq m^{-3} increase of radon concentration (Darby et al. 2004). The difference between indoor radon concentrations measured during the two periods under consideration was analyzed. Generally, the concentrations were lower during the summer-autumn season. According to Table 1, the difference in mean values for these periods is evident and rather expected. In addition to seasonal meteorological variations and poor ventilation, a reason of such difference is also the fact that most of the population used solid fuels for room heating in the cold period of the year.

Table 1. Descriptive statistics of radon and radionuclides activity concentrations

	Indoor ^{222}Rn concentration (Bq m^{-3})				Radioactivity in soil (Bq kg^{-1})		
	I period	II period	Annual	RRLC[*]	^{226}Ra	^{232}Th	^{40}K
Minimum	28	21	30	1.05	9	7.2	242
Maximum	881	740	810	2.30	91	103	1061
Median	106	71	96	1.15	30	32	573
Average	139	115	128	1.20	32	35	582
SD	136	110	118		13	16	159
GM	104	85	97	1.16	30	32	561
GSD[*]	2.1	2.1	2.0		1.6	1.9	1.4

SD=Standard Deviation; GM=Geometric Mean; GSD=Geometric Standard Deviation; RRLC=Relative Risk for Lung Cancer

[*] dimensionless

Table 2 presents average annual radon concentration measured in each municipality under the study as well as the corresponding number of houses with increased radon levels. Almost 50% of all houses had annual radon concentration higher than 100 Bq m^{-3}. Due to the low number of cases, one has to expect rather large uncertainty of estimated percentages, as can be seen from the large confidence intervals. The lowest average annual radon concentration (30 Bq m^{-3}) was measured in Leposavic municipality, on the first floor of a house, as well as in Pristina municipality (33 Bq m^{-3}) on a ground floor. The highest average annual radon concentration (810 Bq m^{-3}) was measured in a ground floor house (with poor concrete slab) in Zvecan municipality. Relatively high concentration of radon for measuring spot on the first floor (271 Bq m^{-3}) was found in another house in close vicinity. There is an "anomaly" of radon at these locations. A detailed comparison with specific activities of radionuclides in nearby soil (hereinafter) indicates that soil is an important source of indoor radon at these locations. Besides, the use of local stone as a building material might also pose a significant contribution to these high indoor radon levels.

Table 2. Houses with elevated annual radon concentration

Municipality (no. of measurements)	Average Rn concentration in the Municipality (Bq m^{-3})	Number of dwellings with annual radon concentration		
		100–200 Bq m^{-3}	200–300 Bq m^{-3}	>300 Bq m^{-3}
Pristina (9)	81	2	--	--
Obilic (6)	101	3	--	--
Lipljan (6)	158	2	1	1
Strpce (4)	66	1	--	--
Gnjilane (1)	72	--	--	--
Pec (5)	152	2	1	--
Leposavic (10)	71	3	--	--
Zvecan (12)	191	3	3	1
Vucitrn (2)	245	--	--	1
Kosovska Mitrovica (8)	127	6	1	--
Total number		22	6	3
Total percentage [%]		35 (24 – 47)*	10 (24 – 47)*	5 (2 – 13)*

*95% confidence interval of the proportion (Wilson approximation)

In most of the houses, the average annual radon concentrations were approximately equal in both rooms. However, there were about 20% of locations (12 houses) where a significantly higher radon concentration (> 100 Bq m^{-3}) was measured in one room compared to the other (in both periods). Fig. 2 presents coefficient of variation (CV) and difference in absolute value between average radon concentrations in the two monitored rooms for all annual measurements. For above mentioned 12 houses, CV varied between 27% and 96% – quite higher than the estimated measurement variability (Carpentieri et al., 2011; Bochicchio et al, 2014), and difference in absolute value varied from 111 to 630 Bq m^{-3}.

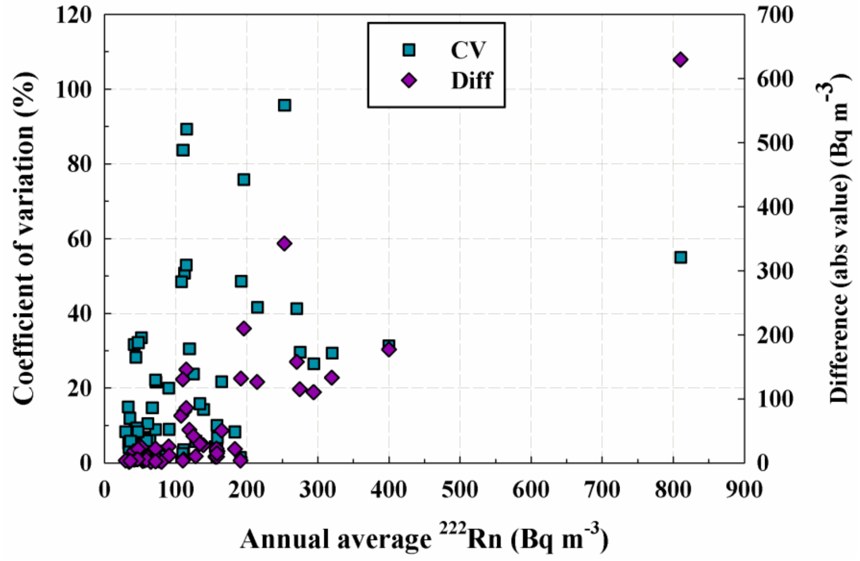

Figure 2. Coefficient of variation, CV (left y-axis) and difference in absolute value (right y-axis) as a function of average radon concentrations in the two measured rooms

The reasons for this might be found in different floor structures that influence radon diffusion from soil, as well as in other possible factors such as building materials, room ventilation or living habits of inhabitants. Besides, some additional sources of radon might exist in some of these places. Eleven of twelve houses are ground floor, and one is a house with cellar. In the case of close contact with ground, it can be expected that radon sources located in one room or a part of a house cause an uneven radon inflow from soil. Another remark refers to selection of room type; lower concentrations were measured in living rooms compared to the bedrooms in both periods. That could be because inhabitants spend most of their daytime hours in living rooms with frequent opening windows and doors. Bedrooms are usually used only for sleeping, not for daily duties, so most of them have poor ventilation that allows radon accumulation indoors.

3.2. Specific activities of radionuclides in soil

Specific activities of natural radionuclides are presented in Table 1. The values obtained for ^{226}Ra and ^{232}Th vary by an order of magnitude. The worldwide average values for soil are given as follows (UNSCEAR 2008, Annex B, sec. II, par. 77, p. 233): ^{226}Ra (32 Bq kg^{-1}), ^{232}Th(45 Bq kg^{-1}) and ^{40}K (412 Bq kg^{-1}).. The mean value of measured ^{226}Ra (32 Bq kg^{-1}) is equal to the current worldwide value, while the mean value for ^{232}Th (35 Bq kg^{-1}) is lower than the corresponding worldwide average. There is also a great variation in the values of specific activities of ^{40}K, with mean value of 582 Bq kg^{-1}, which is higher than the worldwide average. Frequency distribution of ^{226}Ra is presented in Fig. 3.

A wide range of values indicates uneven distribution. There are several locations in the vicinity of Zvecan municipality with higher values of specific activities of ^{226}Ra and ^{232}Th compared to the northern parts (Leposavic municipality). Elevated levels of natural radionuclides in the soil of Zvecan vicinity could be the consequence of the ore deposits (the Trepča complex, formerly one of the most important mining areas in Europe, is located here). Extensive mining activities, production of mineral fertilizers and other industrial activities have created several flotation tailings, landfills, metallurgical slag and intermediate products. Data published in recent years report about the levels of diffuse contamination of the environment with heavy metals of mining and surrounding area (Di Lella et al., 2004; Borgna et al., 2009; Nannoni et al., 2011; Gulan et al., 2013b).These technological activities could also alter the level of natural radiation of the environment (Murty and Karunakara, 2008).

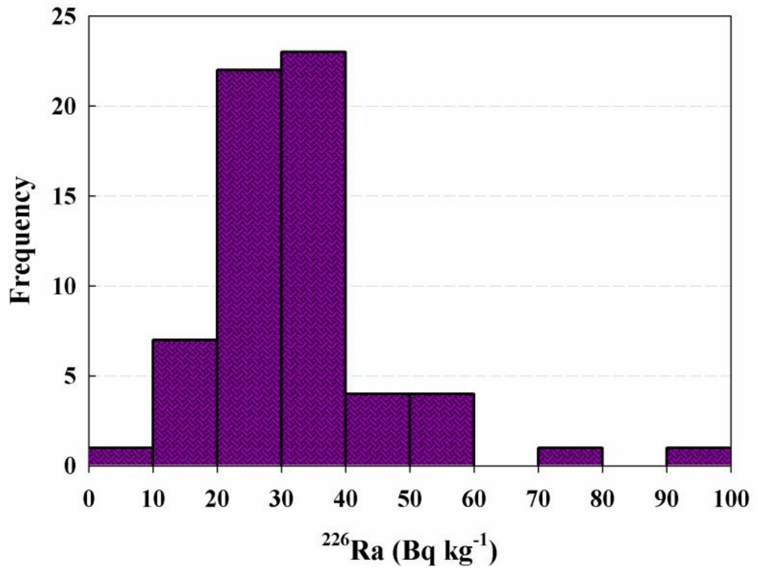

Figure 3. Frequency distribution of ^{226}Ra

This area (Zvecan, Kosovska Mitrovica and Vucitrn municipalities) is also particular in geological terms. High concentrations of radionuclides in soil could be associated with existence of deep fault and seismogenic fault zones (Geological Atlas of Serbia, 2002). Faults were produced by the volcanic activity, typical for this area. These are extremely permeable structures that enable mobility of radionuclides and their decay products to the surface.

In addition, some houses with elevated indoor radon concentrations were identified. It was found that higher values of specific activities of radionuclides in soil at certain locations in Zvecan, Kosovska Mitrovica and Vucitrn municipalities (Table 2) are mainly responsible for high indoor radon concentrations, which is a direct confirmation of radon geogenic origin. On the other hand, a detailed analysis of houses in the municipalities of Lipljan and Pec indicate that high indoor radon levels were not just the consequence of high content of radium in soil, but they were also affected by other factors such as soil porosity, building styles and lifestyles.

3.3. Dose estimation

3.3.1. *Effective dose due to indoor radon*

The annual effective dose E (mSv y^{-1}) due to exposure to radon and its progeny was calculated using the following formula:

$$E = C \cdot F \cdot t \cdot DCF \tag{1}$$

where C (Bq m^{-3}) is the annual average radon concentration; F= 0.4 is the assumed equilibrium factor between radon and progeny (ICRP 65, 1993; ICRP 115, 2010); t = 7000 h is the time spent indoors during one year; DCF = 9 nSv (Bq h $m^{-3})^{-1}$ is dose conversion factor for radon and its short lived progeny, adopted by UNSCEAR (UNSCEAR, 2008). Accordingly, the average value of annual effective dose E (mSv y^{-1}) due to exposure to radon and its progeny equals 3.2 mSv. It ranges from 0.8 to 20 mSv. Estimated effective dose is lower than the worldwide average of 1.15 mSv y^{-1} for radon inhalation (UNSCEAR 2008, Annex B, sec. II, par. 97, p. 236) only at 8 of 63 locations.

ICRP (ICRP 115, 2010) has recently adopted a dose conversion factor higher than the one adopted by UNSCEAR. According to ICRP recommendations (DCF = 12 nSv (Bq h $m^{-3})^{-1}$), annual effective doses ranged from 1.1 to 26.7 mSv, with an average value of 4.5 mSv.

3.3.2 Effective dose due to radionuclides in soil

The annual effective dose due to external exposure to radionuclides in soil D_E (mSv y^{-1}) was calculated using the conversion coefficient of 0.7 Sv Gy^{-1} for:

$$D_E = 0.7 \cdot t \cdot p \cdot \dot{D} \tag{2}$$

where t is the annual exposure time (8760 h) and p is the assumed outdoor occupancy factor of 0.2 for time spent outdoors. The external gamma dose rate in air at 1 m above ground level, $\dot{D}$ (nGy h^{-1}) was calculated according to the following formula (UNSCEAR 2008, Annex B, sec. II, par. 81, p. 234):

$$\dot{D} = 0.462 \cdot A_{Ra} + 0.604 \cdot A_{Th} + 0.0417 \cdot A_K \tag{3}$$

where A_{Ra}, A_{Th} and A_{K} are the values of specific activities of ^{226}Ra, ^{232}Th and ^{40}K (Bq kg^{-1}). The average value of the dose rate in air due to natural radionuclides in soil is 60 nGy h^{-1}. The average value of annual effective dose, 74 µSv (range 25–177 µSv), is slightly higher than the annual average value of 66 µSv for external exposure to natural terrestrial sources of radiation (UNSCEAR 2008, Annex B, sec. II, par. 81, p. 234).

3.4 Correlation analyses

Spearman correlation analysis has been performed using SPSS 20 (IBM, US) software. The results are presented in Table 3. A strong positive correlation exists between pairs of natural radionuclides in soil. Strong correlation between ^{226}Ra and ^{232}Th (Spearman's rho=0.847) can be explained by the assumption that ^{238}U (the progenitor of ^{226}Ra) and ^{232}Th have similar behavior during their transport and they commonly occur together in nature (Chandrasekaran et al., 2015). There was also a strong correlation between ^{226}Ra and ^{40}K (rho=0.657), as well as between ^{232}Th and ^{40}K (rho=0.669).

Table 3. Spearman correlation coefficients (Spearman's rho)

	^{222}Rn	**^{226}Ra**	**^{232}Th**	**^{40}K**
^{222}Rn	1	0.220	0.181	0.082
^{226}Ra		1	0.847**	0.657**
^{232}Th			1	0.669**
^{40}K				1

**Correlation is significant at the 0.01 level

On the other hand, a weak correlation between ^{222}Rn and ^{226}Ra has been found (95% confidence interval of rho ranged from -0.036 to 0.443). Fig. 4 illustrates the correlation between mean annual radon concentration (averaged over the two periods and the two rooms of each house) and radium content in nearby soil. Geogenic factors, like local variability of geogenic radon potential, as well as permeability of soil (including soil moisture, density, type), could be the main reason for a weak correlation between indoor radon and radium in soil. Besides, anthropogenic factors (type of building, room ventilation etc.) could also be the cause of such result. It is obvious from Table 3 that indoor radon was also not correlated with ^{232}Th or ^{40}K in soil, however, such a result was rather expected.

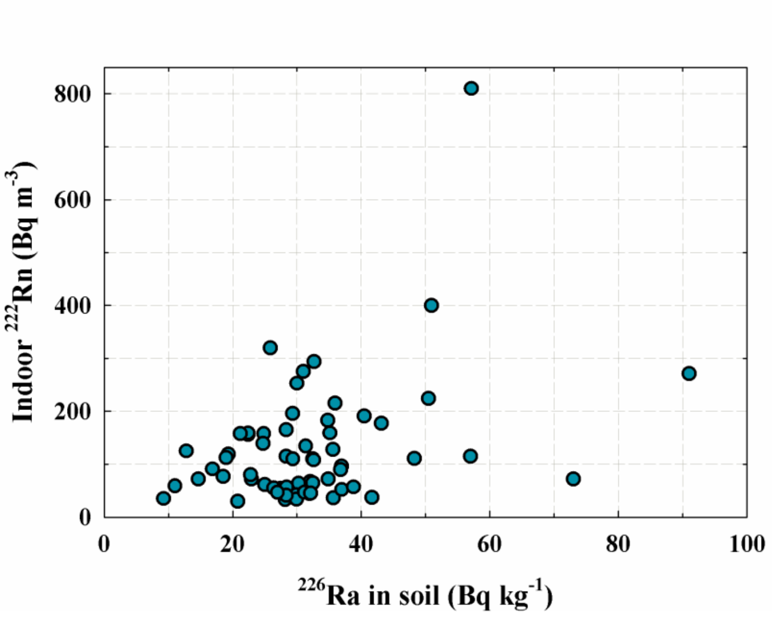

Figure 4. Correlation between mean annual ^{222}Rn concentrations and specific activities of ^{226}Ra in the nearby soil

Similar studies have been conducted in other countries obtaining quite different results. A survey of indoor radon and radium in the soil near 97 dwellings was conducted in Trabzon (Turkey); Pearson coefficient of determination between ^{226}Ra and ^{222}Rn was R^2=0.682 (Kurnaz et al., 2011). Another survey in Giresun (Turkey) involved 71 houses and determination of radionuclides content in the nearby soil (0–15 cm layer); it was concluded that there is a positive correlation (R^2=0.54). Radon concentrations were significantly higher in houses built on soil with a higher content of radium. However, there are no general rules that apply for all houses, because in addition to geology, some local variability factors, as well as factors of building types and usage influence the indoor radon concentration (Celik et al., 2008). In a study conducted in Norway (Sundal et al., 2004) correlations between indoor radon and geology (radium content and permeability of soil in the vicinity of the building) indicate that the geological data are useful in the identification of radon-prone areas, although they could not provide the assessment of the level of radon in buildings. A study conducted in the central part of India also found no direct relationship between radon in rooms with the content of uranium, thorium and potassium content in soil (Kher et al., 2008). In addition to geology, anthropogenic factors like building properties (building material, nature of the surface of walls and floors, ventilation, type of windows, etc.) and lifestyles of inhabitants significantly affect indoor radon concentration.

4. CONCLUSION

Bearing in mind that radon is the largest source of radiation in non-accidental situations and a major contaminant of indoor space, this study enabled the identification of locations with a large range of indoor radon concentration levels useful to evaluate correlation with radium content in nearby soil. A weak correlation (Spearman's rho=0.220) between indoor radon concentrations and radium content in nearby soil was found. Soil is assumed to be a dominant source of indoor radon in all locations under the study. However, geogenic causality could not be proven: in addition to geology, indoor radon level is strongly influenced by many other variable factors such as climatic conditions, design and construction of building, household habits, etc. This conclusion is supported by the fact that the study revealed a relatively large number of locations where average annual radon concentration varied significantly between different rooms (at the same floor) of the same houses. Accordingly, it would be rather difficult or even impossible to predict indoor radon levels from Ra in soil, due to the large number of factors involved.

Conflict of Interest: The authors declare that they have no conflict of interest.

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