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RADON EMANATION OF SOILS FROM DIFFERENT LITHOLOGICAL UNITS

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Abstract: In soil samples collected from 58 points over 7 different lithological units most often appearing in Slovenia, at which radon in indoor and outdoor air and in soil gas had been measured in previous surveys, emanation fraction of ^{222}Rn has been determined. The number of samples for a unit differed and was the highest for carbonates which cover more than two thirds of the country. Emanation fraction differed significantly from unit to unit, as well as from sample to sample within the same unit. The following ranges and averages were obtained: 0.015–0.079 and 0.035±0.016 for alluvial and glacial deposits, 0.014–0.306 and 0.089±0.085 for clastic sediments containing clay, 0.027–0.084 and 0.056±0.040 for coarse clastic sediments, 0.016–0.114 and 0.053±0.037 for flysch, 0.010–0.547 and 0.186±0.178 for carbonates, 0.016–0.041 and 0.029±0.018 for metamorphic rocks, and 0.262–0.418 and 0.340±0.110 for sea and lake deposits. Correlation of the emanation fraction with the radium concentration in soil and radon concentration in soil gas showed correlation coefficients of 0.56 and 0.26, respectively.

Keywords: radon, radium, soil samples, emanation fraction, lithology

1. INTRODUCTION

During their creation by α-transformation of ^{226}Ra (half-life, $t_{1/2}$ = 1600 years) in the earth's crust, radon (^{222}Rn) atoms receive about 80 keV recoil energy which enables to those being close enough to the grain surface, to emanate from the grain into void space, from where they start to migrate through the medium by diffusion and to longer distances by convection/advection, and eventually exhale into the atmosphere and enter the buildings (Etiope & Martinelli, 2002). The emanation fraction (f_{em}), also called emanation coefficient or emanation power, is ratio of the number of radon atoms emanated and number of radon atoms created in the grain. Emanation is the first bottleneck in radon transport from its origin to living and working environment, and as such, one of the key parameters in modelling radon levels in soil gas, as well as in outdoor and indoor air (Abumurad & Al-Tamimi, 2001; Bossew, 2003; Iskandar et al., 2005; Savović et al., 2012). The emanation fraction has been comprehensively reviewed by Nazaroff et al., 1988, Nazaroff (1992) and recently by Sakoda et al. (2011). Its values range from 0.05 to 0.70 (UNSCEAR, 2000), with representative values of 0.03 for minerals, 0.13 for rocks, 0.20 for soils, 0.17 for mill tailings and 0.03 for fly ash (Sakoda et al., 2011).

In Slovenia, radon emanation fraction of soil has only been roughly estimated at several sites (Brajnik et al., 1992). In order to remedy the lack of this information, soil samples were collected from soils over different lithological units and emanation fraction was determined. In the paper, measurements are described, and results of the analysis are presented and commented on.

2. MATERIALS AND METHODS

Soil samples were collected from a depth of 80 cm at 70 points (Fig. 1). Our measurement points were classified into 7 units, based on the lithological classification. Each sample was obtained as a typical soil at each of the following lithological units: A – alluvial and glacial deposits represent mainly unconsolidated clastic sediments (soil type: Calcaric Cambisol and Renzic Leptosol), B1 – clastic sediments containing clay (soil type: Planosol), B2 – coarse clastic sediments (soil type: Dystric Leptosol, Distric and Eutric Cambisol), B3 – flysch (soil type: Eutric and Calcaric Cambisol), C – carbonates (soil type: Chromic Cambisol, Rendzic Leptosol), D – metamorphic rocks (soil type: Dystric Cambisol), E – sea and lake sediments (soil type: Gleysol). The number of samples for a unit differs and was highest for carbonates which cover more than two thirds of the country.

Slovenia lies on the junction region between the Alps and the Dinarides which incorporates the Eastern Alps, Southern Alps, Dinarides, Panonian basin and the Adriatic-Apulia foreland (Placer, 2008). The western and south-western parts of Slovenia are characterised by NW-SE trending dextral strike-slip faults (Fig. 1). The Adriatic-Apulian foreland comprises the larger part of Istria in the south-western corner of Slovenia consisting of rocks of the Adriatic segment of the Adriatic-Dinaric Mesozoic carbonate platform, and the flysch rocks resulting from its degradation (Placer, 2008). The entire southern part of Slovenia belongs to the Dinarides, characterised by the thrust and nappe structure, consisting mainly of carbonate rocks and sediments resulting from disintegration of the Adriatic-Dinaric carbonate platform: Upper Cretaceous carbonate turbidites, Cretaceous-Palaeocene and Eocene flysch (Placer, 2008). The Southern Alps are palaeogeographically a part of Dinarides. Mesozoic rocks of the Slovenian basin and Upper Triassic rocks of the Julian carbonate platform are exposed within them (Placer, 2008). The Eastern Alps are a geologic-orographic term comprising the complex of Precambrian and Old Palaeozoic high and low grade metamorphic rocks and of Permian and Mesozoic sedimentary rocks north of the Periadriatic fault (Placer, 2008). The Pannonian basin, in the north-eastern part of Slovenia, consists of individual depressions that originated during Palaeogene and Neogene. They are filled with sediments of the Paratethys deposited on subsided continuations of the Eastern and Southern Alps and Dinarides (Placer, 2008). In addition to the major lithological units described above, alluvial and glacial deposits, extending along major valleys, have to be considered. In the Ljubljana basin, situated in the central part of Slovenia and in the Krško basin in south-east, sea and lake sediments prevail.

All samples were analysed for ^{226}Ra (Gregorič et al., 2012) and 58 were taken for emanation measurements. These were first dried to constant weight in the oven at 105°C for 24 hours, milled in an agate mortar, and sieved. 25 g of the 660 μm mesh fraction were filled into a 50 cm^3 glass ampoule, weighed, air-tight sealed and left for a month to reach the secular equilibrium between ^{226}Ra and ^{222}Rn. Emanation fraction was determined by a direct measurement of radon amount emanated from the sample (Somlai et al., 2008). For that purpose, the ampoule was broken in a 181 cm^3 metal chamber and radon so released was transferred into a Lucas type scintillation cell, using nitrogen gas as its carrier. Measurement started after three hours, necessary to reach secular equilibrium between ^{222}Rn and its short-lived decay products, i. e., ^{218}Po (α, 3.11 min), ^{214}Pb (β/γ, 26.8 min), ^{214}Bi (β/γ, 19.9 min) and ^{214}Po (α, 164 μs) (Nero, 1988). Signals from the scintillation cell were analysed using two different instruments: (i) an EMI photomultiplier connected to a NP 420 single channel amplitude analyser (hereafter called NP method), and (ii) a GammaTech NDI detector (hereafter called NDI method) (Abbady et al., 2004).

3. RESULTS AND DISCUSSION

Values of emanation fraction (f_{em}) obtained with both NP and NDI methods do not differ significantly (Fig. 2), and they both roughly fit lognormal distribution (Fig. 3). Hereafter only the NDI results will be interpreted. They vary from 0.010 to 0.547, the range being similar to those reported for soil samples by de Martino et al., (1998) and found in the review by Sakoda et al. (2011), but considerably higher than those reported by Baixeras et al., (2001), Abumurad & Al-Tamimi (2001), and Bossew (2003).

Distribution of f_{em} with respect to the lithological units is shown by the box & whiskers plot in figure 4 and is summarised in table 1. The highest values, with the widest range, was observed in samples over carbonates covering more than two thirds of the south-west part of the country, which is crossed by a number of tectonic faults (Fig. 1). The highest average value was found in soil samples over the sea and lake deposits (E) and the lowest, over metamorphic rocks (D). The former were composed of fine grain sediment, mostly clay, containing a high amount of organic matter with iron on which radium prefers to adsorb (Miklyaev & Petrova, 2011), and the latter, of larger compact particles with low porosity. The highest emanation fractions are ascribed to radium atoms adsorbed on the surface of nano particles (Miklyaev & Petrova, 2011). Differences within a lithological unit (e. g., carbonates) are even bigger than between units. Comparison of our values with those from other studies is not straightforward because of different lithological classification used by different authors.

The measured emanation fractions were related to the ^{226}Ra activity concentration (C_{Ra}) previously determined in soil samples from the same lithological units (Gregorič et al., 2012), whose distribution with respect to the lithological units is shown by the box & whiskers plot in figure 5a and is summarised in table 2. Here, the highest average value was obtained in soils over carbonates (83.5Bqkg^{-1}) and the lowest, in soils over flysch (32.8Bqkg^{-1}). The overall average of all samples of 62.7Bqkg^{-1} is markedly higher than the UNSCEAR world average of 30Bqkg^{-1} (UNSCEAR, 2000). Because the lithological units with the highest f_{em} and C_{Ra} average values do not coincide, an excellent correlation between the two

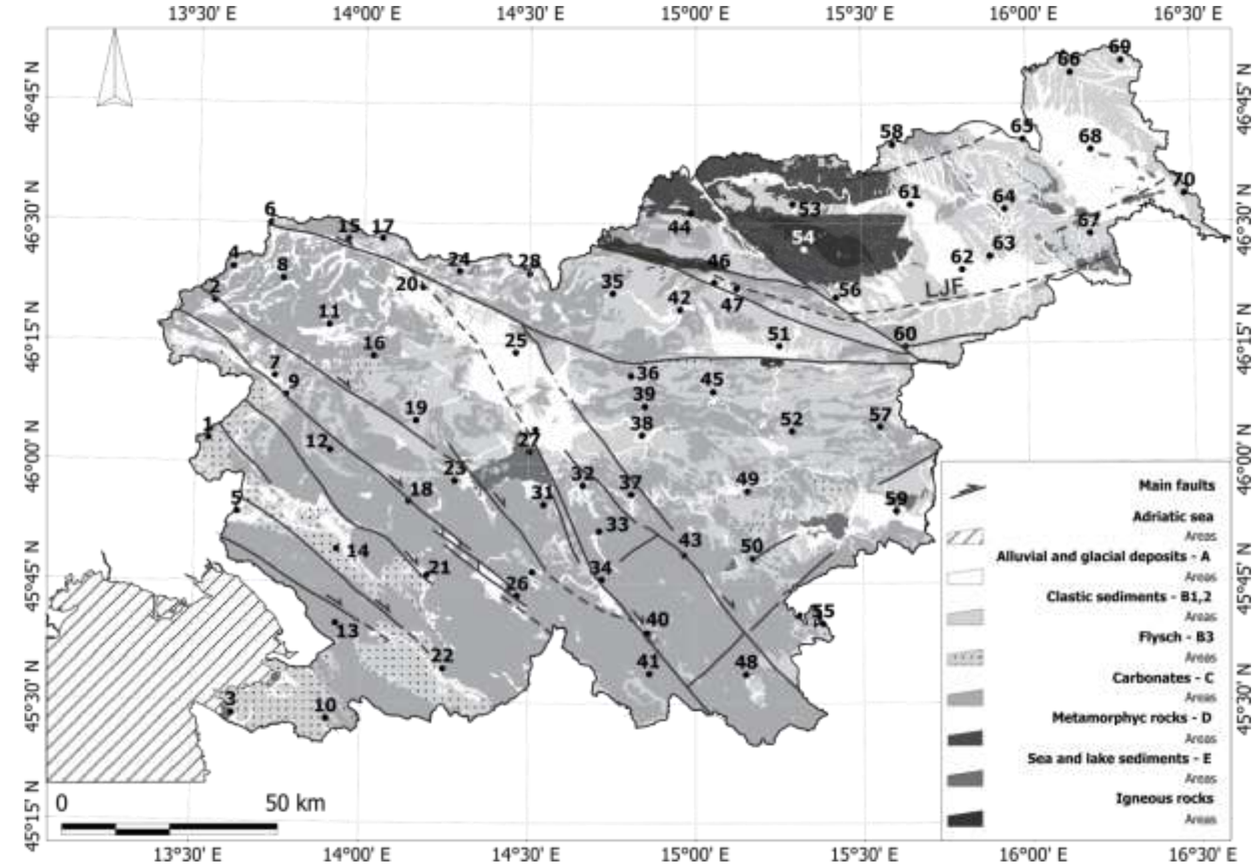

Figure 1. Map of Slovenia (surface area 20 thousand km^2), showing soil sampling points, the following lithological units: A – alluvial and glacial deposits, B1 – clastic sediments containing clay, B2 – coarse clastic sediments, B3 – flysch, C – carbonates, D – metamorphic rocks, E – sea and lake deposits, and the main tectonic faults.

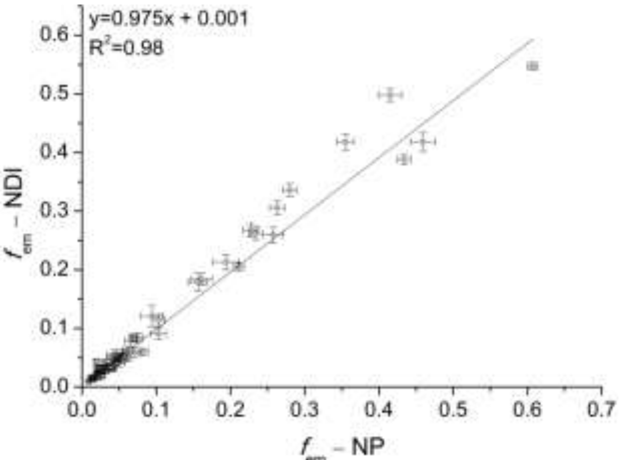

Figure 2. Comparison of radon emanation fractions (f_{em}) in Slovenian soil samples, obtained with the NP and NDI methods.

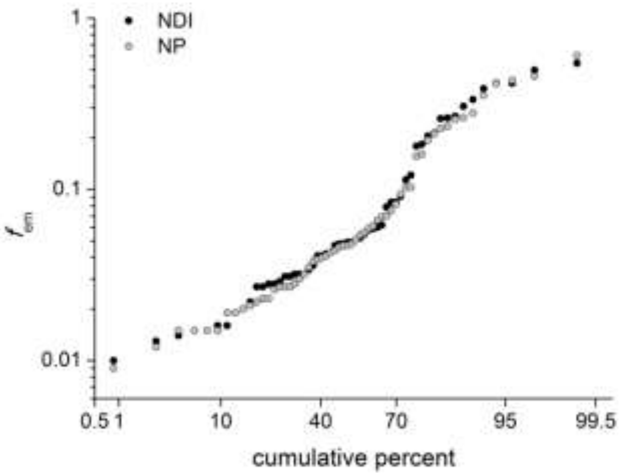

Figure 3. Log-normal plot of radon emanation fractions (f_{em}) in Slovenian soil samples, as obtained with NP and NDI methods.

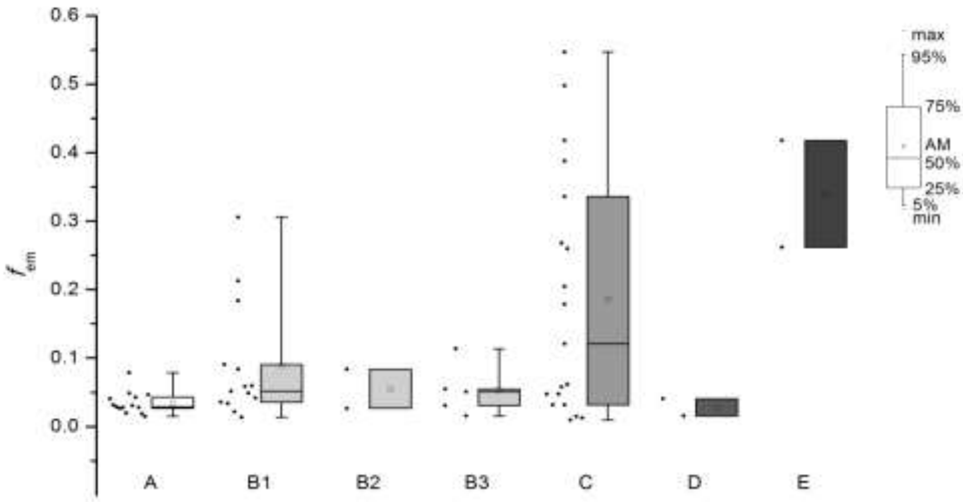

Figure 4. Box and whiskers diagram of distribution of radon emanation fraction (f_{em}) with respect to the lithological units in Slovenia: A – alluvial and glacial deposits, B1 – clastic sediments containing clay, B2 – coarse clastic sediments, B3 – flysch, C – carbonates, D – metamorphic rocks, E – sea and lake deposits.

Table 1. Values of minimum (min), maximum (max), median, arithmetic mean (AM) and standard deviation (SD) of emanation fraction of ^{222}Rn (f_{em}), for the following lithological units: A – alluvial and glacial deposits, B1 – clastic sediments containing clay, B2 – coarse clastic sediments, B3 – flysch, C – carbonates, D – metamorphic rocks, E – sea and lake sediments.

	Lithological units	No. samples	min	max	median	AM	SD
f_{em}	A	14	0.015	0.079	0.030	0.035	0.016
	B1	14	0.014	0.306	0.056	0.089	0.085
	B2	2	0.027	0.084	0.056	0.056	0.040
	B3	5	0.016	0.114	0.051	0.053	0.037
	C	19	0.010	0.547	0.121	0.186	0.178
	D	2	0.016	0.041	0.029	0.029	0.018
	E	2	0.262	0.418	0.340	0.340	0.110
	total	**58**	**0.010**	**0.547**	**0.097**	**0.112**	**0.069**

Table 2. Values of minimum (min), maximum (max), median, arithmetic mean (AM) and standard deviation (SD) of ^{226}Ra activity concentration in soil and ^{222}Rn activity concentration in soil gas, for for the lithological units: A – alluvial and glacial deposits, B1 – clastic sediments containing clay, B2 – coarse clastic sediments, B3 – flysch, C – carbonates, D – metamorphic rocks, E – sea and lake sediments.

Radionuclide	Lithological units	No. samples	min	max	median	AM	SD
^{226}Ra (Bq kg^{-1})	A	15	23.4	102.1	51.1	55.0	23.1
	B1	18	20.1	169.9	50.6	55.8	32.0
	B2	3	41.9	48.5	46.4	45.6	3.4
	B3	6	21.7	42.1	31.5	32.8	7.5
	C	24	11.5	269.3	63.3	83.5	61.8
	D	2	34.4	42.6	38.5	38.5	5.8
	E	2	70.0	74.7	72.4	72.4	3.3
	total	**70**	**11.5**	**269.3**	**48.3**	**62.7**	**43.9**
^{222}Rn (kBq m^{-3})	A	15	2.9	97.0	27.7	37.1	33.2
	B1	18	2.4	93.8	25.2	32.4	28.6
	B2	3	23.0	73.9	30.7	42.5	27.4
	B3	6	4.2	9.7	7.8	7.5	2.1
	C	24	2.5	211.4	50.3	61.9	55.6
	D	2	1.1	14.6	7.9	7.9	9.5
	E	2	17.2	34.6	25.9	25.9	12.3
	total	**70**	**1.1**	**211.4**	**25.1**	**30.7**	**24.1**

quantities was neither expected nor observed, as evidenced in figure 6. The correlation coefficient (R=0.57) is similar to that observed by Greeman & Rose (1996) and Thomas et al., (2011), but markedly

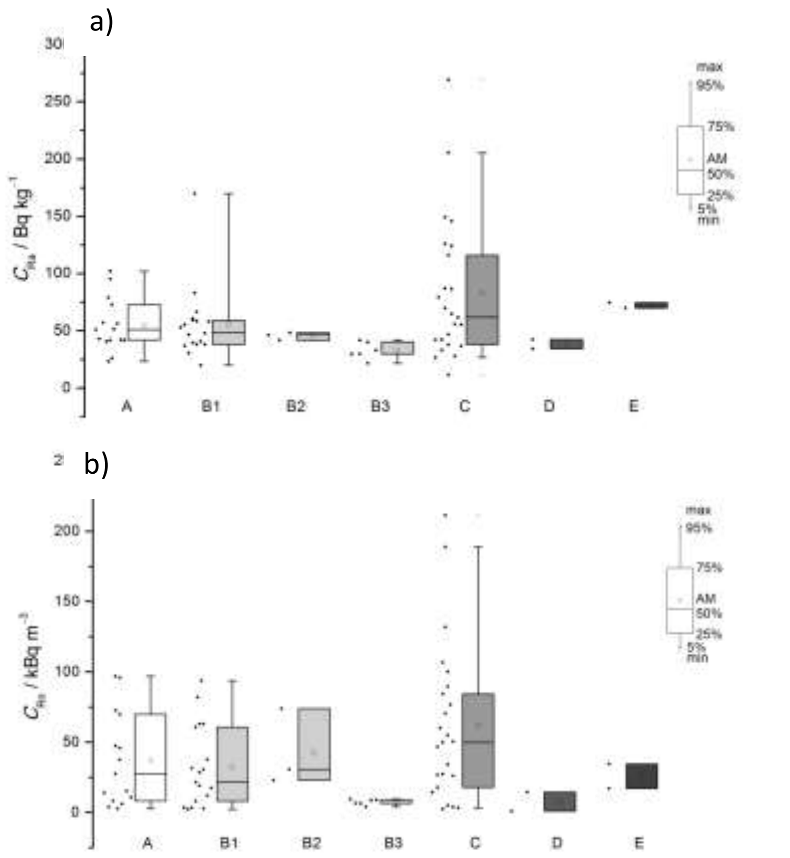

Figure 5. Box and whiskers diagram of activity concentration distribution with respect to the lithological units in Slovenia: A – alluvial and glacial deposits, B1 – clastic sediments containing clay, B2 – coarse clastic sediments, B3 – flysch, C – carbonates, D – metamorphic rocks, E – sea and lake deposits; of a) ^{226}Ra in soil and b) ^{222}Rn in soil gas.

lower than that by Baykara et al. (2005). Closer look at this relationship would reveal that soil samples over alluvial and glacial deposits show even a negative correlation, and emanation fraction of soils over flysch sediments and coarse grained clastic sediments does not depend on the ^{226}Ra content. Thus, the relatively high correlation coefficient is mostly contributed from samples over carbonates and clastic sediments containing clay, whose number is by far the largest.

It is reasonable to expect that higher emanation fraction would be reflected in higher radon concentration in soil gas (C_{Rn}). This was checked with C_{Rn} values, obtained at the same points with solid state nuclear track detectors in a previous study (Vaupotič et al., 2008). Their distribution with respect to the lithological units is shown by the box & whiskers plot in figure 5b and is summarised in table 2.

As with C_{Ra}, the highest average value of C_{Rn} was obtained in soil samples over carbonates (61.9Bqm^{-3}) and the lowest over flysch (7.5Bqm^{-3}). Actually, figure 7 shows a positive correlation of the $C_{Rn} - f_{em}$ relationship, but with a very low correlation coefficient (R=0.26). This is not disappointing, because the emanation is only the initial step in radon travel from its origin in the grain to the detector exposed in the borehole. This travel is influenced by hydro-meteorological conditions (e. g., humidity, temperature, barometric pressure, wind) and soil characteristics (e. g., grain size distribution, porosity, fraction of pores filled with water) and therefore a clear role of emanation fraction is hardly to be elucidated.

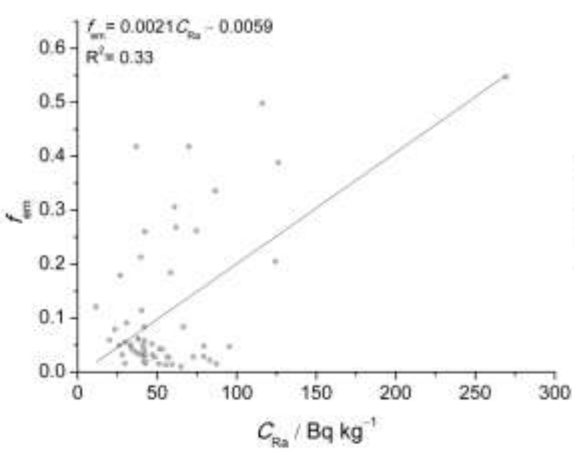

Figure 6. Relationships between radon emanation fraction (f_{em}) and ^{226}Ra content (C_{Ra}) in the same soil samples (Gregorič et al., 2012) in which emanation was measured.

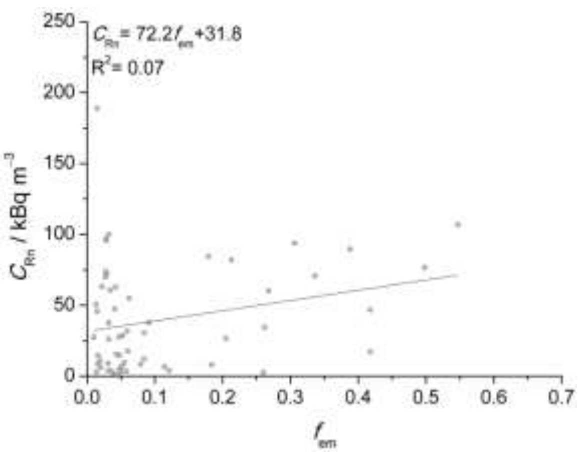

Figure 7. Relationships between radon emanation fraction (f_{em}) and radon concentration in soil gas (C_{Rn}) obtained at the same points where soil samples were taken for emanation analyses (Vaupotič et al., 2008).

4. CONCLUSION

Emanation fraction varied substantially (from 0.010 to 0.547) from lithological unit to lithological unit, as well as within a lithological unit. The highest average value was found in soils over sea and lake deposits (0.340) and the lowest, over metamorphic rocks (0.029). A positive correlation was observed between emanation fraction and both radium content in soil (R=0.57) and radon concentration in soil gas (R=0.26).

In using radon as a tracer in studying local and global transport of air masses, not always the measured values of radon exhalation rate or concentrations in outdoor air or soil gas are considered, but rather (because of lack of measurements) they are calculated using models based on the literature averages of radium content and emanation fraction. According to our results, a great uncertainty is inherent in applying this approach.

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REFERENCES

Abbady, A., Abbady, A.G.E. & Michel, R. 2004. *Indoor radon measurement with the Lucas cell technique*. Applied Radiation and Isotopes, 61, 1469–1475.

Abumurad, K.M. & Al-Tamimi, M., 2001. *Emanation power of radon and its concentration in soil and rock*. Radiation Measurement, 34, 423–426.

Baixeras, C., Erlandsson, B., Font, L.l. & Jönsson G., 2001. *Radon emanation from soil samples*. Radiation Measurements, 34, 441–443.

Baykara, O., Doğru, M., İnceöz, M. & Aksoy, E., 2005. *Measurements of radon emanation from soil samples in triple-junction of North and East Anatolian active faults systems in Turkey*. Radiation Measurements, 39, 209–212.

Bossew, P., 2003. *The radon emanation power of building materials, soils and rocks*. Applied Radiation and Isotopes, 59, 389–392.

Brajnik, D., Miklavžič, U. & Tomšič, J., 1992. *Map of Natural Radioactivity in Slovenia and its Correlation to the Emanation of Radon*. Radiation Protection Dosimetry, 45, 273–276.

de Martino, S., Sabbarese, C. & Monetti, G., 1998. *Radon emanation and exhalation rates from soils measured with and electrostatic collector*. Applied Radiation and Isotopes, 49, 407–413.

Etiope, G. & Marinelli, G., 2002. *Migration of carrier and trace gases in the geosphere: an overview*. Physics of Earth and Planetary Interiors, 129, 185–204.

Greeman, D.J. & Rose, A.W., 1996. *Factors controlling the emanation of radon and thoron in soils of the eastern U.S.A.* Chemical Geology, 129, 1–14.

Iskandar, D., Iida, T., Yamazawa, H., Moriizumi, J., Koarashi, J., Yamasoto, K., Yamasaki, K., Shimo, M., Tsujimoto, T., Ishikawa, S., Fukuda, M. & Kojima, H., 2005. *The transport mechanism of ^{222}Rn in soil at Tateishi as an anomaly spot in Japan*. Applied Radiation and Isotopes, 63, 401–408.

Gregorič, A., Kobal, I., Kovács, T., Szeiler, G., Fábián, F., Kardos, R. & Vaupotič, J., 2012. *Natural radioactivity of Slovenian soils*. In: Barnet, I., Neznal, M. & Pacherová, P.: Proceeding of the 11th International Workshop on the Geological Aspects of Radon Risk Mapping. Czech Geological Survey, Prague. pp. 116–125.

Miklyaev, P. S. & Petrova, T. B., 2011. *Studies of radon emanation from clays*. Water Resources, 38, 868–875.

Nazaroff, W.W., Moed, B.A. & Sextro, R.G., 1988. *Soil as a source of indoor radon: generation, migration, and entry*. In: Nazaroff, W.W. & Nero, Jr., A.V.: Radon and Its Decay Products in Indoor Air. John Wiley & Sons, New York. pp. 57−112.

Nazaroff, W.W., 1992. *Radon transport from soil to air*. Reviews of Geophysics, 30, 137–160.

Nero, Jr., A.V., 1988. *Radon and its decay products in indoor air: an overview*. In: Nazaroff, W.W. & Nero, Jr., A.V.: Radon and Its Decay Products in Indoor Air. John Wiley & Sons, New York. pp. 1−53.

Placer, L., 2008. *Principles of the tectonic subdivision of Slovenia*. Geologija, 51, 205–217.

Sakoda, A., Ishimori, Y. & Yamaoka, K., 2011. *A comprehensive review of radon emanation measurements for mineral, rock, soil, mill tailing and fly ash*. Applied Radiation and Isotopes, 69, 1422–1435.

Savović, S., Djordjevich, A. & Ristić, G., 2012. *Numerical solution of the transport equation describing the radon transport from subsurface soil to buildings*. Radiation Protection Dosimetry, 150, 213–216.

Somlai, J., Jobbágy, V., Somlai, K., Kovács, J., Németh, Cs. & Kovács, T., 2008**.** *Connection between radon emanation and some structural properties of coal-slag as building material*. Radiation Measurements, 43, 72–76.

Thomas, P.A., Anderson, D.W., Zhang, W.H., Baweja, A.S. & Tracy, B.L., 2011. *Radon emanation in Saskatchewan soils*. Health Physics, 100, 565–573.

UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation)., 2000. *Sources and effects of ionizing radiation*, Vol I: Sources. UNSCEAR 2000 Report to the General Assembly, with Scientific Annexes.

Vaupotič, J., Žvab, P., Gregorič, A., Kobal, I., Kocman, D., Kotnik, J. & Križman, M., 2008. *Radon mapping in Slovenia based on its levels in soil gas*. In: Book of Abstracts of the 33rd International Geological Congress, August 2008, Oslo, Norway, 6–14.

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