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RADON IN SOIL GAS AND CONSTRUCTED GEOGENIC RADON POTENTIAL IN CROATIA

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ABSTRACT
Radon concentrations in soil gas in Croatia were measured from 2005-2013 with the AlphaGUARD and from 2013- with RM-2 measuring systems, while soil permeability was measured with the Radon-JOK device since 2015. Measurements of both parameters were performed at 412 locations, mostly in the vicinity of schools and kindergartens and the obtained average value of radon in soil gas was 48 kBq m^{-3} with standard deviation of 52 kBq m^{-3}. It is important to emphasize that there are areas with radon concentrations up to 550 kBq m^{3}.
Geogenic radon potential (GRP) has been constructed by "Neznal approach" and a new subclassification with 7 classes (1- very low, 2 – low, 3 – medium lower, 4 – medium higher, 5 – high, 6 – very high, 7 – extremely high) is being introduced. The obtained average GRP of 28 ± 29 ("Neznal equation") or 3.30 ± 0.96 (classes) classifies the soil of Croatia into soil of medium GRP.

1. INTRODUCTION

Radon (^{222}Rn) is continuously generated in rocks and soil as a product of radioactive decay of radium (^{226}Ra) in the natural decay series of uranium (^{238}U). A very small fraction of radon emanates from its place of origin into the air-filled pore spaces of rocks. Driven by some transport mechanisms, such as advection (due to pressure differences), convection (temperature differences) or diffusion (concentration differences), it moves toward the surface and exhalates to the atmosphere or enters into the buildings above. The resulting indoor radon concentration in buildings is a function of natural ("what the Earth delivers") and anthropogenic factors (construction types: state and age of building, building materials used, presence of basement or not, and living habits of the residents: ventilation rate, type of heating and air-conditioning etc.) and therefore it is temporally variable and unique for each particular house or building.
From the radon mapping point of view, it is wise to use that kind of parameter which is more or less constant over a long period of time. Such parameter should take into consideration as much physical processes as possible which contributes to conditions for potential indoor radon. The most used input quantities are: radon in soil gas (c, kBq/m^{-3}) at certain depth which is the result of production (radioactive decay of radium) and emanation from minerals while soil permeability (k, m^2) describes an advective transport process which usually dominates over the others in most cases. Recently, these two variables were combined and define so called geogenic radon potential (GRP) which is widely used as a measure of geogenic hazard of "what Earth delivers" in term of radon [1-3]. Although, there are several very different definitions and meanings of GRP in literature [4-7], we are using the Czech approach, the parameter called "Neznal's" GRP which has been developed from radon index (RI) of building site [8]. That GRP is defined in the following way:

$$GRP = \frac{c}{-log_{10}k\text{-}10}. \text{ GRP}=\frac{c}{-\log_{10}k-10} \quad (1)$$

In the Czech Republic, three classes of GRP are used and they are correlated to radon index: low (GRP < 10), medium (10 ≤ GRP < 35) and high (35 ≤ GRP).

The main goal of this paper is to present GRP maps based on radon in soil gas and soil permeability measurements in Croatia. The difference between continuous (by using equation (1)) and discrete (classes) GRP approach will be investigated and the subclassification of GRPs will be introduced and justified due to very different and diverse geological structures as well as climatological conditions in Croatia.

2. MATERIAL AND METHODS

Radon concentrations in soil gas in Croatia were measured from 2005-2013 with the AlphaGUARD (Manufacturer: Genitron Instruments, Germany) and from 2013- with the RM-2 (Manufacturer Radon v.o.s., Czech Republic) measuring systems, while soil permeability was measured with the Radon-JOK device (Manufacturer: Radon v.o.s., Czech Republic) from 2015. Measurements of both parameters were performed at 412 locations, mostly in the vicinity of schools and kindergartens. Radon in soil gas measurements were generally performed immediately after the soil permeability measurements at the same soil probe and at the depth of 0.8 m (with 5 cm of sampling geometry). Up to 2013, radon was measured by the AlphaGUARD measuring system according to sampling protocol from manufacturer (AlphaGUARD manual). In both types of radon detectors for soil gas measurements, thoron was excluded from the results because measurements were performed 15 minutes after gas sampling.

In fact, soil permeability was calculated from Darcy's law according to measurement of time needed to withdraw certain amount of soil air by means of negative pressure established by Radon-JOK measurement set-up (Radon-JOK manual).

Geogenic radon potential for each measuring location was calculated by using equation (1) and taking into account the maximum measured value of radon in soil gas and the average soil permeability value. The obtained values were classified into classes according to Table 1.

Table 1. Subclassification of geogenic radon potential (GRP)

GRP – "Czech" approach	Class	Radon index	GRP – "Croatian" subdivision	Class	Description
GRP < 10	1	Low	GRP < 5	1	Very low
			5 ≤ GRP < 10	2	Low
10 ≤ GRP < 35	2	Medium	10 ≤ GRP < 22.5	3	Medium lower
			22.5 ≤ GRP < 35	4	Medium
GRP ≥ 35	3	High	35 ≤ GRP < 60	5	High
			60 ≤ GRP < 125	6	Very high
			GRP ≥ 125	7	Extremely

Radon values as well as GRPs were presented in the form of maps based on internationally suggested 10 x 10 km square grid which has been used for the European indoor radon map as well as for the future European geogenic radon map [1, 9].

3. RESULTS AND DISCUSSION

The obtained results of radon in soil gas measurements in Croatia as well as calculated (by Equation (1)) and constructed (according to classification described in Table 1) geogenic radon potential are summarized in Table 2. The arithmetic mean of all measurements of radon in soil gas is 48 $kBqm^{-3}$ with standard deviation of 52 $kBqm^{-3}$. while the average GRP with its standard deviation is 28 ± 29. According to the used subclassification, it belongs to higher medium GRP. Normal probability plots of logarithmic values for both parameters are presented in Figures 1A and 1B. The applied statistical tests for normality (Lilliefors, Kolmogorov-Smirnov, Shapiro-Wilk) show that both parameters do not obey log-normal distribution, which was expected, since the measuring locations were not randomly chosen. The measurements of radon in soil gas and soil permeability were mostly performed in the vicinity of schools and kindergartens as a part of targeted surveys of indoor radon.

Table 2. The basic statistical analysis of radon in soil gas measurements and calculated and constructed geogenic radon potential (GRP) for Croatia (AM – arithmetic mean, SD – standard deviation, GM – geometric mean, GSD – geometric standard deviation)

	Radon in soil gas [kBq m^{-3}]	**GRP**	**GRP-classes**
Number of measurements	874	795	
Number of locations	785	412	
AM	**47.6**	**28.4**	**3.30**
SD	52.1	28.7	0.96
Median	34.7	15.9	3
GM	33.0	16.8	3.0
GSD	2.4	2.6	1.6
Min - Max	1.0 - 566.0	0.3 - 470.4	1 - 7

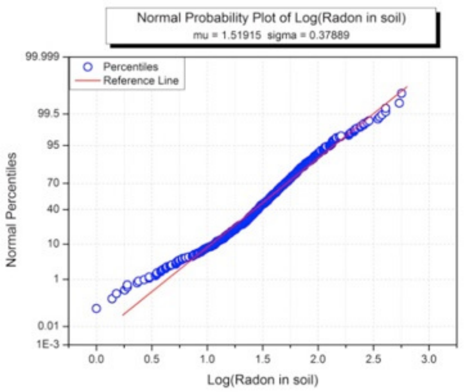

Figure 1A. Normal probability plot of logarithmic values of radon in soil gas in Croatia

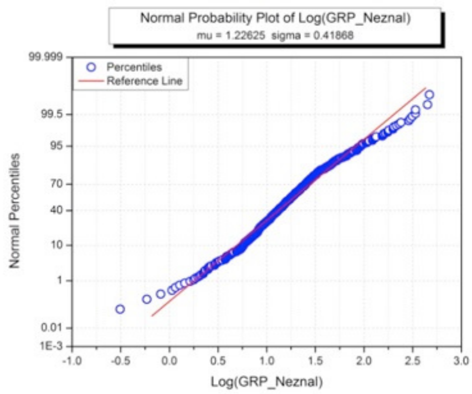

Figure 1B. Normal probability plot of logarithmic values of GRPs in Croatia

The strong influence of water content in soil on radon concentration in soil gas is presented in Figure 2. In two consecutive years (2014 and 2015), the measurements of radon in soil gas were performed in the vicinity of schools and kindergartens in

Požega-Slavonia county and repeated a year later. In 2014, there were severe rainfalls with lots of floods in the area of interest a week before measuring campaign and a year later (in 2015), there were normal weather conditions (the cumulative precipitation for September 2014 and 2015 for Požega weather measuring station are presented as the right part of Figure 2). Radon in soil gas were approximately twice higher during the period with significant water content in soil than during the period with normal conditions (normal quantity of precipitation for that area).

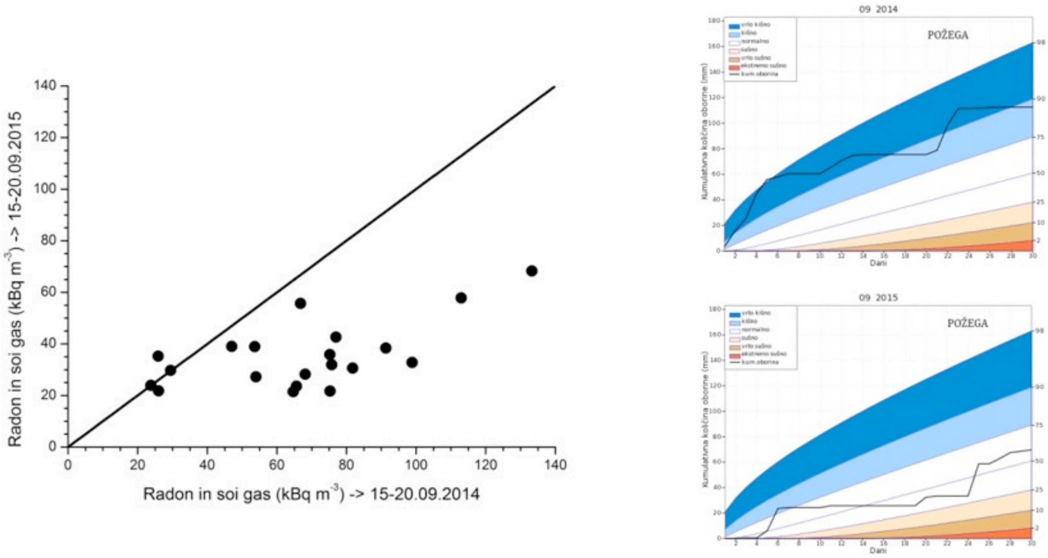

Figure 2. Comparison of radon in soil gas concentrations measured in Požega-Slavonia county at the same locations in two consecutive years. In 2014, the measurements were performed a week after severe rainfalls (upper right) and in 2015 during normal weather conditions (lower right). The line (y(x) = x) on the graphical presentation of empirical results suggests no difference between measured values.

In order to examine temporal variations of local GRP associated to meteorological and climate variations, the measurements of radon in soil gas and soil permeability were undertaken several times a week during the whole year (from April 2015 until April 2016). The obtained results of calculated GRP are presented in Figure 3. There were 82 measurements and the average value and its standard deviation of adjoint GRP class was 3.76 ± 0.58. In 93% of cases, the associated GRPs belonged to medium GRP class. Two values, at the end of the presented measuring cycle, were significantly different from the other results. These anomalies were associated with the preparation for construction work by heavy machinery nearby the measurement point. This was, at the end, the reason to stop measurements at that measuring site.

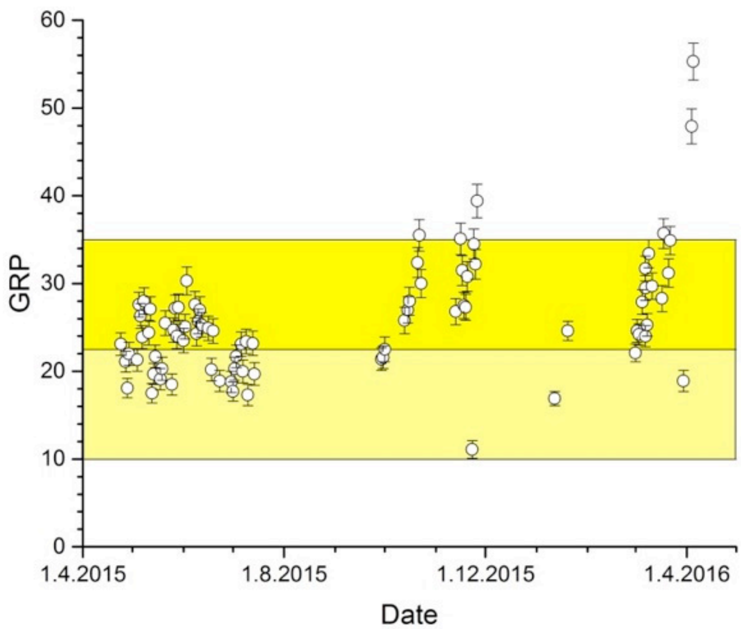

Figure 3. Time series of GRP values calculated from measurements of radon in soil gas and soil permeability at one measuring point in the courtyard of Department of Physics in Osijek. Yellow areas represent the medium (lower and higher) GRP.

Geogenic radon potential maps of two selected counties (Istria and Osijek-Baranja) using grid approach were created and presented in Figure 4. These two counties have a very different geological structure. Istria consists of a limestone plateau and dolomites formed in Late Cretaceous, much of which lacks water owing to its karst topography while carbonate rocks, especially flysch are significantly formed during Paleogene period. Osijek-Baranja county consists geologically mainly of sandstones and soft clay while sediments are of gravels, loess and silts. All these sediments are the product of mostly continental environments and alluvial fans. Differences in geological structure of these two areas inevitable have, as a consequence, very different geogenic radon potential as it can be seen in Figure 4.

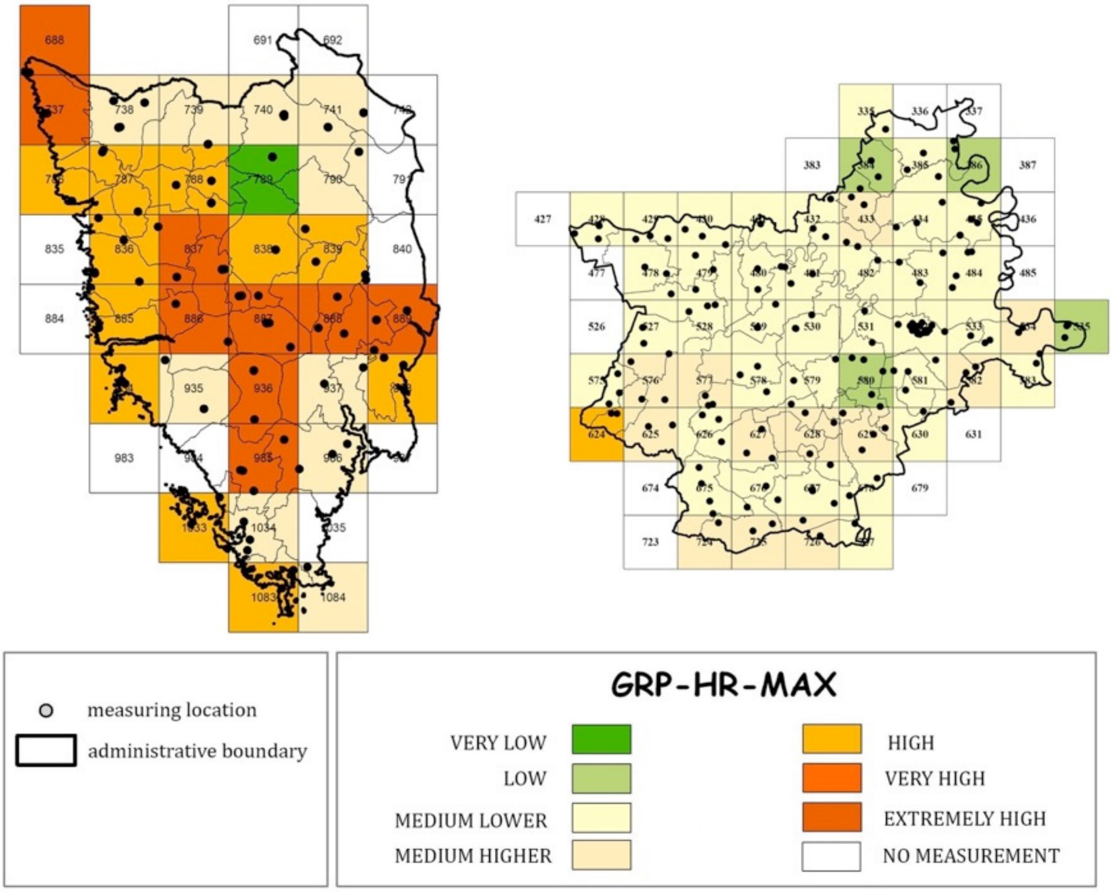

Figure 4. Geogenic radon potential maps of Istria (left) and Osijek-Baranja (right) counties

In more than 2/3 of the Istrian peninsula there are high (or even very high) GRP while in Osijek-Baranja county medium GRP prevails. This kind of maps could be useful for identification [10] as well as definition of so called radon-prone areas which have been under special attention in EU BSS [11]. The next phase of research is to connect GRP and indoor radon maps and to find some kind of functional or geostatistical relationship between those spatial variables as well as to introduce new ones (other terrestrial radionuclide maps, climate maps, geology maps, etc.).

4. CONCLUSION

Radon in soil gas and soil permeability were measured in more than 400 locations in Croatia. The geogenic radon potential for each location was calculated and the obtained results show that the average GRP in Croatia is in medium class. Although, there are areas with very high (or even extremely high) GRP. It was shown that radon in soil gas concentration itself is highly sensitive on water content in soil. The variations of GRP throughout a year are much smaller than radon in soil gas and it is wise to use this quantity to describe the potential hazard of radon. GRP maps of several Croatian counties were created and can be used as a tool for identification or even for definition of radon-prone areas. Nevertheless, it should be pointed out that no map can fully reflect the true reality of variations of radon potential due to heterogeneity of soil under the surface. Moreover, such maps can provide an useful information about soil as the main radon source in buildings but they cannot be used for direct extrapolation and estimation on indoor radon concentrations inside the buildings at some areas of interest.

5. ACKNOWLEDGEMENT

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RADON U TLU I IZRAČUNATI GEOGENSKI RADONSKI POTENCIJAL U HRVATSKOJ

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APSTRAKT
Koncentracije radona u tlu u Republici Hrvatskoj su mjerene AlphaGUARD mjernim sustavom (Genitron Instruments) od 2005. do 2013., a potom se mjere RM-2 mjernim uređajem (Radon v.o.s., Češka Republika). Premeabilnost tla mjeri se od 2015. godine Radon-JOK uređajem (Radon v.o.s., Češka Republika). Mjerenja se uglavnom obavljaju u blizini škola i vrtića. Dosad je izvršeno 412 mjerenja oba parametra. Prosječna koncentracija radona u tlu iznosi 48 kBq m^{-3} s pripadnom standardnom devijacijom od 52 kBq m^{-3}. No, postoje i područja u kojima je mjerena koncentracija radona u tlu i do 550 kBq m^{-3}.
Geogenski radonski potencijal (GRP) je izračunat tzv. češkim pristupom iz empirijske jednadžbe, a uvedena je i nova subklasifikacija kojom se kontinuirane vrijednosti GRP-a svrstavaju u klase: (1- vrlo nizak, 2 – nizak, 3 – niži srednji, 4 – viši srednji, 5 – visok, 6 – vrlo visok, 7 – izrazito visok). Izračunate vrijednosti GRP-a iznose 28 ± 29 (empirijska kontinuirana vrijednost) odnosno 3,30 ± 0,96 (klase) čime se tlo u Republici Hrvatskoj, u prosjeku, klasificira u srednji geogenski radonski potencijal.