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STUDY

Radon prediction in Hungary: a short overview

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This paper is a short overview of radon prediction applied in Hungary until now. After an introduction about the radon problem, the brief history of radon measurements in Hungary is summarized. Then the methods used for radon prediction of unmeasured areas in Hungary are described.

Highlights:
- Overview of the methods used for radon prediction in Hungary
- Prediction based on only indoor radon values
- Spatial analysis applied for radon prediction
- Radon prediction by environmental co-variables

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1. Introduction

Radon and radioactive radiation are ubiquitous in our nature. Terrestrial radiation is responsible of approximately 84% of the total effective dose of 2.4 mSv y^{-1}, at which humans are exposed as worldwide average (UNSCEAR, 2008). Radon (^{222}Rn and ^{220}Rn), the only gaseous intermediate radioactive product of the uranium and thorium decay chain, is the main contributor (1.25 mSv y^{-1}) to the population annual average effective dose (UNSCEAR 2008). In Hungary, these values are a little bit higher because of the elevated radon concentration compared to the world average, total effective dose is 3 mSv y^{-1} (Marx, 1999). In Hungary, the absorbed dose rates inside dwellings varies in a wide range of 11 to 236 nGy h^{-1} with an average of 95 nGy h^{-1}, whereas the absorbed dose rates outdoors varies in the range of 15 to 130 nGy h^{-1} with an average of 61 nGy h^{-1} (Nikl, Sztanyik 1988, Nikl 1996, UNSCEAR 2008). According to UNSCEAR (2008), Hungary, Malaysia, China, India, Albania, Portugal, Australia, Italy, Spain, Sweden and Iran present the highest values of absorbed dose rates inside dwellings in the range of 95–115 nGy h^{-1} compared to the world population-weighted average of 84 nGy h^{-1}, which reflects the wide use of stone or masonry materials in buildings (UNSCEAR, 2008). In this sense, areas in Hungary with expected elevated indoor radon concentration due to the contribution building material or the local geology has been evaluated. For instance, Szabó et al. (2014b) estimated that 7% of the 53 adobe dwellings, studied in the great Hungarian plain, exceeds in 10 mSv y^{-1} the world average. In school buildings, containing coal slag insulation in the cities of Transdanubia (e.g., Ajka, Tatabánya and Veszprém), were studied by Somlai et al. (1997) who found that the external dose rate in Tatabánya, in classrooms closed for 24 h, were in the range of 500–900 nGy h^{-1}. Thus, the attention should be paid to existing radiation situations in such large quantities of natural radioactive isotopes whose health consequences are no longer negligible and, therefore, require actions to protect public health.

The main source of radon in the air of dwellings, workplaces, underground places, caves, is geogenic radon, which derives from the soil and rock (77%) (UNSCEAR 2000). Since there are the most likely two reasons for such large quantities of natural radioactive isotopes: 1) during a very long period, which can be measured on the Earth his-

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torical scale, natural enrichment in soils and rocks (e.g., loess, granite, NORM [Naturally Occurring Radioactive Materials]), and 2) artificial enrichment as a result of human activities, mainly mining and industrial processing (building material using artifacts as raw materials: e.g., slag, fly ash, TENORM [Technologically Enhanced Naturally Occurring Radioactive Materials]). The second main source of radon in indoor air is the building material (18%), other sources (e.g., tap water) have smaller than 5% role (UNSCEAR 2000).

It is important to clarify that radon itself, as a non-reactive noble gas, does not denote hazard for human health directly. However, its danger relies on the fact that it can migrate through the soil, rock and building material and reach the ambient air, and making possible the contact of its progenies with the human tissues by inhalation (Nazaroff 1992). The short-lived decay products of radon can deposit on the respiratory tract cells and may cause damage due to the chemical reactivity and energy associated to their decay process, increasing in this way the risk of lung cancer (Nazaroff 1992). Therefore, the World Health Organization (WHO) recommends a 100 Bq m^{-3} reference level for radon activity concentration to minimize the health hazard due to indoor radon exposure. However, when this limit is reached, the concentration should not exceed 300 Bq m^{-3} (WHO 2009). Epidemiological studies and evidences show that even at relatively low exposure (100 Bq m^{-3}), a significantly higher risk of lung cancer is still present (WHO 2009).

1.1. Normative

In Europe, on December 2013 the European Commission proposed the latest Euratom Basic Safety Standards (BSS) (Council Directive 2013/59/Euratom) laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation and repealing the former directives. This includes, among others, establishing reference level for indoor radon concentration with a maximum of 300 Bq m^{-3} and developing national radon action "for addressing long-term risks from radon exposures in dwellings, buildings with public access and workplaces for any source of radon ingress, whether from soil, building materials or water" (Council Directive 2013/59/Euratom). The national radon action plan includes assessing relevant parameters and providing scientifically based maps of potential areas "where the radon concentration (as an annual average) in a significant number of buildings is expected to exceed the relevant national reference level" (Council Directive 2013/59/Euratom). The Directive was implemented in the Govt. decree 487/2015. (XII. & 30.) in Hungary. The Hungarian radon action plan for 2018–2023 was prepared and accepted by the Government in the first half of 2019 (Emberi Erőforrások Minisztériuma 2018, available in Hungarian). In the framework of the Hungarian national radon action plan, a national representative radon survey is planned. This survey would contain indoor radon, soil gas radon and radon exhalation measurements, also gamma dose rate measurements should be considered (Emberi Erőforrások Minisztériuma 2018).

1.2. European Atlas of natural radiation

Each European country performed different radon mapping and predicting procedures (Dubois 2005). Complementing the national efforts to identify and document the radon prone areas at a harmonized level, the Joint Research Center of the European Union (JRC) is leading and permanently developing the European Atlas of natural radiation project since 2006 (Bossew et al. 2013, 2015, Cinelli et al. 2018, De Cort et al. 2011). This project encompasses the European maps of annual cosmic-ray dose, indoor radon, concentration of uranium, thorium and potassium in soil and in bedrock, terrestrial gamma dose rate, soil permeability and geogenic radon. The atlas displays the geographical distribution of the radon related parameters on a reference grid of 10 km × 10 km defined with 32 participant countries (until August 2017). Digital version of the atlas is already available (Cinelli et al. 2018).There are areas on each map that have not yet been measured, hence further radon measurements and prediction for health risk assessment are needed.

2. Brief history of radon measurements in Hungary

Radon has been extensively studied in Hungary since 1980 by different institutions, universities and laboratories and by their collaborations. Surveys were performed chronologically (literary references without completeness, seeking the latest and most important):

1) by the Laboratory of National Public Health Center National Research Directorate for Radiobiology and Radiohygiene (under different names over time) between 1980 and 2019: measurements at ~2000 sites in dwellings, workplaces, public places (kindergartens, schools etc.) and mines; building materials and waters were investigated, and the biological effects of radon, as well (Déri et al. 2003, Gundy et al. 1995, Hámori et al. 2004, Juhász et al. 2002, Lázár et al. 2003, Nikl 1996, Nikl, Köteles 2000, Homoki et al. 2017, Sándor et al. 2003),
2) by the Hungarian Academy of Sciences, Institute for Nuclear Research (ATOMKI) between 1980 and 2019: several hundreds of radon measurements in indoors, soil gas, thermal baths, caves and wine cellars; building materials and waters were also investigated (Csige 2008, Csige et al. 2013, 2018a,b, Hakl et al. 1996, 1997, Hunyadi et al. 1991, Sóki, Csige 2016, 2017; Sóki et al. 2018, Szabó et al. 2014b),
3) by the Institute of Radiochemistry and Radioecology, University of Pannonia between 1992 and 2019: several thousands of radon measurements in dwell-

ings, workplaces and public places (kindergartens, schools, hospitals etc.), thermal baths, caves and building materials, and waters were also investigated (Csordás et al. 2018, Fábián et al. 2017, Kávási et al. 2006, 2010, Kovács et al. 2017, Müllerová et al. 2018, Németh et al. 2005a, Sas et al. 2015, Shahrokhi et al. 2016, Somlai et al. 1997, 2004, 2006, 2007),

4) by the RAD Laboratory between 1994 and 2006: ~20.000 measurements in dwellings, workplaces, public places (kindergartens, schools, libraries, churches) were studied (Hámori et al. 2006a,b, Minda et al. 2009, Tóth, Hámory 2005, Tóth, Selmeczi 1994),
5) by the MecsekÉrc Ltd. (later as Mecsek-Öko Ltd.) in the region of the Hungarian closed uranium mine (Mecsek Mts.) performed many indoor, soil gas, water radon measurements and made the recultivation as well in the area (Juhász et al. 2002, Németh et al. 2005b, Várhegyi et al. 2004, 2009),
6) by the Geodetic and Geophysical Institute, Hungarian Academy of Sciences the relationship between rock deformation and radon concentration was investigated together with temperature and barometric pressure effects for many years (Mentes, Eper-Pápai 2015, Mentes 2018),
7) by different research groups of Eötvös Loránd University: Lithosphere Fluid Research Group studied mainly geology and building material and also a radon remediation method (Nagy et al. 2012, Szabó Z. K. et al. 2013, 2014, Szabó Zs. et al. 2013, 2014, Völgyesi et al. 2014); Department of Atomic Physics studied especially waters (Horváth et al. 2015) and Ra distribution in rock (Freiler et al. 2015), Department of Physical and Applied Geology studied the underground waters and hydrogeological aspects (Erőss et al. 2012).

3. Radon prediction

Radon prediction is an important tool for radon risk management by the identification of areas with elevated radon concentration. Besides this, prediction of radon is a complex task because several influencing factors should be considered and many input parameters can be used. In general, two types of radon prediction exist regarding the used input parameters. Both have advantages and disadvantages.

The first type of radon prediction uses indoor radon values since the main contributor of the effective dose it is, especially radon progeny, and these data are often measured Europe-wide. However, this type of prediction has a disadvantage that indoor radon values vary from house to house because of the building characteristics (e.g., building materials, foundation type, building types) and living habits (e.g., ventilation frequency by the occupant). Different building characteristics and living habits can cause large difference in indoor radon data in houses next to each other. This fact gives the limitation of prediction radon spatially. Despite of restriction, this type of radon prediction, spatial extension of the indoor radon, is the most widespread on the world since carrying out the measurement of indoor radon is relatively easy, simply and cheap. Only a passive track detector system (detectors and evaluation) and a distributing system (e.g., placing the detectors by post) is required.

The second type of radon prediction is by predicting the potential radon using other environmental parameters as well, which can be a determining factor for indoor radon. These parameters can be, for instance, geological and soil parameters, and other environmental radioactivity parameters. This prediction provides information of the potential risk of the area and not about the indoor radon itself. Its main advantage is that this technique provides the possibility to characterize areas for radon risk, where indoor radon measurements are not available (Gruber et al. 2013, Bossew et al. 2015). The predicted radon potential can be a continuous variable, but usually it is category variable like low, medium and high. However, each category can be associated with an indoor radon value probability.

4. Methods applied for radon prediction in Hungary

4.1. Radon prediction based on indoor radon concentration

Indoor radon level were measured by several researchers, however, only in few studies were applied prediction for indoor radon of unmeasured places/dwellings (Hámori et al. 2004, 2006a,b, Minda et al. 2009). Hámori et al. (2004) estimated the number of homes expected to be above 400 Bq m^{-3} with a lognormal distribution based on the radon level over 15,000 ground floor homes. With appropriate statistical procedures (lognormal fitting, χ^2 test, homogeneity grouping), they estimated indoor radon level for 92% of the Hungarian population and flats. By considering the stratum system in a lognormal radon distribution, Hámori et al. (2006a) determined the estimated percentage of dwellings above 150 and 600 Bq m^{-3} at different regions of the country. Using similar approach, Hámori et al. (2006b) determined the estimated percentage of dwellings above 200 and 400 Bq m^{-3} in Hungary and in upper floors, and first floors of houses in cities, towns and villages. Following the international practice of indoor radon surveys Minda et al. (2009) proposed to use the indoor radon index (IRI), which shows the ratio of houses above 200 Bq m^{-3} based on the lognormal distribution of 6154 one-storied, no-basement houses indoor radon data. In these works 21 areas were defined in Hungary, based on different geological background and calculated the IRI for these areas (Minda et al. 2009).

4.2. Radon prediction based on soil gas radon concentration and soil gas permeability

In Hungary, Szabó K. Z. et al. (2014a) applied firstly the method of radon potential (RP), for the central part of Hungary, which was developed by Neznal et al. (2004). The quantification of radon potential (RP) is based on the mathematical expression proposed by Neznal et al. (2004), which relates to the radon potential with the equilibrium concentration of ^{222}Rn in soil air and the soil gas permeability. Soil gas radon concentration and soil gas permeability were measured at 192 points over 41 geological formations (Szabó K. Z. et al. 2014a). Their results show that the study area in central Hungary can be characterized by low and medium radon potential. High risk occurs only locally corresponding to the slope sediments in the hilly areas and the lowest values of radon potential was found in drift sand, fluvioeolic sand and fluvial sand and loess formations (Szabó K. Z. et al. 2014a).

4.3. Radon prediction using environmental co-variables

Regression kriging was applied by Pásztor et al. (2016) for the interpolation of radon potential using spatially exhaustive auxiliary data on soil, geology, topography, land use and climate. Categorical and numerical parameters were used as environmental co-variables and its significance (within 95–99.9% confidence interval) was calculated from the step-wise regression method. An important contribution of this study is the determination of the influence of the environmental parameters in the radon potential. The authors made a 90% interval estimate for the areal extension of the three RP risk categories (low, medium and high) (Pásztor et al. 2016).

4.4. Radon flux prediction using gamma dose rate as a proxy

Szegvary et al. (2007) used gamma dose rate measurements to predict radon flux in Hungary, Switzerland, Germany and Finland. The radon flux was measured using AlphaGUARD and accumulation chamber and the gamma dose rate was measured by an autonomous gamma probe at 1 m above ground, soil moisture and precipitation was also measured. Linear correlation was found between gamma dose rate and radon flux despite the variability of the data. The radon prediction is based on a linear correlation between radon flux and gamma dose rate. The predicted means of each country are within the error proving the effectivity of the approach. Szegvary et al. (2007) found that an increase of moisture causes a decrease in both variables as follows: when the soil moisture increases the gas diffusivity is reduced as well as the radon flux, and the increasing moisture enhances the electron shielding, therefore the gamma dose rate decreases.

4.5. Geogenic radon potential assessment by empirical and theoretical models

Beltrán Torres et al. (2019a) tested the usability of empirical and theoretical models to predict soil gas radon concentration and soil gas permeability for the geogenic radon potential assessment in a granitic area. The authors proved that the evaluated models can be applied with modifications based on physical and geochemical properties of the soil. This study pointed out the importance of geochemical processes that constrains the soil gas radon concentration such as the preferential adsorption of radium on organic material and clay minerals at low carbonate content (Beltrán Torres et al. 2019b).

5. Future perspectives of radon prediction in Hungary

In Hungary, especially areas, where elevated radon concentration is expected, are well documented, but still there are areas (e.g., south western and eastern Hungary) where basically there is no information on radon. Predictions and new measurements help to enhance our knowledge on these regions. The predictive models have some remarkable advantages, which allow the estimation of radon potential in areas where there are no field measurements available and the parameters used in the model are generally available in databases. It should also be noted for sake of the completeness that several methodologies of radon prediction (e.g., using different predictors such as gamma dose rate, geology, airborne radiometry, soil geochemical parameters, topography, expected lung cancer incidence, or applying different approach such as statistical univariate/multivariate) have been successfully applied in different countries (e.g., Germany, Spain, United Kingdom, France, Italy, Norway, Canada, Sweden, Ireland, United States, Belgium). Radon potential can be predicted based on gamma dose rate and geological information (García-Talavera et al. 2013, Quindós et al. 2008), airborne gamma dose rate (Smethurst et al. 2016, Wattananikorn et al. 2008) and the combination of several environmental parameters (e.g., geology, gamma dose rate, indoor radon etc.) (Appleton et al. 2008, Bossew et al. 2008, Branion-Calles et al. 2015, Cinelli et al. 2018, Elio et al. 2017, 2018, Ferreira et al. 2018, Ielsch et al. 2010, Schumann 1993, Zhu et al., 2001). In Hungary expert and specialist have gained already successful experience, based on detailed studies on natural radioactivity (gamma, radon) in soil, water, outdoor, underground and indoor spaces as listed above in Section 2. An important complementary information is that there are extensive studies about local factors controlling natural radioactivity that constitutes a solid base for the formulation or application of predictive models. For instance, Pásztor et al. (2016) pointed out that not only geology but also soil parameters are also important factors on geogenic radon potential. Furthermore, based on hydrogeological measurements, Erőss et al. (2012) proposed the geogenic uranium, radium and radon as natural tracers to study groundwater flow system. Mentes (2018) described a complex relationship between strain, temperature, barometric pressure and radon concentration in the gneiss rock at the Sopronbánfalva Geodynamic Observatory. On the same region, Freiler et al. (2015) clearly demonstrated that even within a homogeneous geological formation, geochemical processes (e.g., fluid migration, alteration) can cause changes in the distribution of radioactive isotopes. Beltrán Torres et al. (2019a,b) pointed out that the geogenic radon potential is constrained by geochemical processes involving such variables as carbonate and clay mineral content and pH. Based on the Hungarian experiences in radon prediction summarized in this paper and the radon mapping program of the European Atlas of natural radiation, it is obvious that in order to deliver a most proper survey/assessment for the country, not only the indoor radon should be considered, but also the variables associated to the geogenic radon potential.

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