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High variability of indoor radon concentrations in uraniferous bedrock areas in the Balkan region



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HIGHLIGHTS

• The excessive radon values are explained from geological point of view.

• The excessive radon values must not be rejected, which is often performed.

• Rejection of excessive concentrations cause serious errors in the dose estimation.

• Method to estimate the annual mean concentration when some seasons are missing.

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ABSTRACT

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Keywords: Indoor radon Indoor radon variations Uraniferous bedrock areas Geological structure Radon/groundwater migration In this work the strong influence of geological factors on the variability of indoor radon is found in two of three geologically very different regions of South-Eastern Europe. A method to estimate the annual mean concentration when one seasonal measurement is missing is proposed. Large differences of radon concentrations in different rooms of the same house and significant difference in radon concentrations in one season comparing it to the others are noted in certain cases. Geological factors that can lead to such behavior are discussed.

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1. Introduction

In the reports of the United Nations Scientific Committee on Exposure to Atomic Radiation (UNSCEAR, 2000) it has been estimated that excluding doses from radiotherapy and nuclear accidents, the global average annual effective dose to a member of the public is about 3.0 mSv. The largest contribution of 52% to the annual effective dose is due to the exposure to radon and its progenies. Recent re-evaluation of Rn (we put Rn in short for ²²²Rn) dosimetry suggests that the contribution of Rn might be even much higher (Harrison and Marsh, 2012). This important exposure to radon is mainly due to radon in indoor environments

http://dx.doi.org/10.1016/j.apradiso.2014.08.018 0969-8043/© 2014 Elsevier Ltd. All rights reserved. such as homes, schools and workplaces. At the same time, the radon concentration is affected by spatial and temporal variability at different scales, depending on geogenic and meteorological factors (Porstendörfer et al., 1994). Overall, radon is perhaps the most variable contributor to the total dose.

Indoor radon concentration depends on natural and on anthropogenic factors. The latter include construction style of houses, position of a dwelling within a house, building materials, living habits etc. The consequence of this convolution of different influences is that the exactly same building, built on the same geological ground can show very different indoor radon concentrations due to different living habits of inhabitants (Gruber et al., 2013). The geogenic potential for high indoor radon concentrations depends on many factors, which includes the ²²⁶Ra content of the soil underneath buildings and the soil permeability. Therefore, indoor radon has a tendency to be correlated with local geology (Appleton and Miles, 2010; Kemski et al., 2009;

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Scheib et al., 2009). Accordingly besides mapping indoor radon concentrations, which may serve as proxy to exposure, a mapping of the geogenic radon potential (RP) can be very useful in the identification of radon prone areas, i.e. regions in which for geogenic reasons elevated indoor radon concentrations can be expected, depending on anthropogenic factors.

On an European level the project of mapping Rn has been underway for several years. Chronologically, the geogenic RP map, still in its initial phase, was preceded by a European map of indoor radon concentrations, which is not yet completed in all European countries. Since the RP depends on the radionuclide content and the permeability of the soil, it is not influenced by anthropogenic factors. and thus it is considered more reliable for the identification of radon prone areas (Gruber et al., 2013). For instance, in the Czech Republic, radon characterization of the ground of every new building site is required by law; in France priority areas are defined where radon surveys are obligatory for several types of public buildings (lelsch et al., 2010). Sometimes airborne gamma spectrometry referring to an equivalent uranium content in soil is used construct RP maps (Appleton et al., 2011; Smethurst et al., 2008). In order to map RP and to investigate the geological influence on the indoor radon concentration, indoor radon mapping can be used in conjunction with geological boundaries (Appleton et al., 2011).

The RP can vary significantly between different and within the same geological unit. Together with the variability of the anthropogenic factors this results in high variability of indoor radon concentration, so that only relatively little of it can be explained by the bedrock and superficial geology. Variability explained by geology ranges from a few percent (Kemski et al., 2009; Bossew et al., 2008) up to 25% (Appleton and Miles, 2010). In this paper, only the relation between indoor radon and the bedrock of the building site is investigated. Other radiometry, such as measurement of the radionuclide content in soil or airborne gamma measurement, was not performed in this investigation.

Some authors identified geology as a key factor in increased indoor radon concentration levels (Zhu et al., 2001; Gillmore et al., 2005; Miles and Appleton, 2005). Moreover, some authors suggest that the correlation between bedrock and radon concentration can be used for prediction of the radon risk, especially for regions where few measurements are available (Kemski et al., 2009, Sundal et al., 2004).

The radon potential depends on the geological features such as lithological variations and geochemistry and accordingly these features are of high importance in radon mapping (Kemski et al., 2005; Shi et al., 2006; Sundal et al., 2004). Moreover, a large-scale variability could be the consequence of geological features like different kind of faults (Ciotoli et al., 1998; King et al., 1996). The soil constitutes an interface cover between the solid geology (basement rocks) and the atmosphere and may also influence the vertical radon migration, according to its permeability. The water saturation of the shallow part of the soil can be responsible for seasonal variation of soil-gas radon concentration (King and Minissale, 1994).

In Serbia a strong dependence between bedrock and indoor radon concentration is already noticed in Niška Banja (Žunić et al., 2007a; Zunić et al., 2007b). The strict proof was not obtained, however all the houses with high indoor radon concentration are built on a travertine, which is permeable and holds a high content of ²²⁶Ra. High correlation between soil gas radon concentration, gamma dose rate and ²²⁶Ra content (which spatially corresponds to travertine bedrock) in soil were found.

In this paper, the correlation between indoor radon concentration and the geological and geochemical environment (i.e., rocks, soil and water) is investigated. Indoor radon measurements were performed in time series of different durations and sometimes longer than one year. A method to estimate average annual radon concentration is presented in the case of an interrupted time series. A method of non-linear regression is proposed to estimate the annual mean concentration when one seasonal measurement is missing.

The goal of this study consists of a qualitatively investigation of the relationship between the indoor radon concentration and the geological background, in order to outline geogenic radon prone areas. Variation of an order of magnitude of indoor radon concentration measured in several houses in one season is explained from a geological point of view.

2. Material and methods

In Serbia, which is a part of the Balkan region, there are uranium deposits of different types, therefore radioactive geological anomalies are mainly caused by enhanced uranium and thorium concentrations in rock and soil. Several hundred anomalous zones have been identified. mostly due to elevated uranium or thorium concentrations or both (Jankovic, 1990). Since radon is a member of the uranium radioactive decay series, geogenic conditions for enhanced exposure of population to radon in closed spaces exist in Serbia (Zunic et al., 2007b). The reasons for occurrence of high indoor radon levels are many and complex: they include geological factors, building characteristics, usage patterns, etc. (Komatina, 2004). Geological studies in Serbia have clearly identified regions, which have high levels of uraniferous material, but little or no investigations on the exposure of humans to natural radiations in these areas have heretofore taken place. However, results of radon measurements in dwellings in the Balkan region show a close dependence between radon distribution and geological features (Komatina, 2004; Komatina-Petrovic, 2011). In the period from 1997-1999, a survey of natural radiation sources was performed (Zunic et al., 2001) in three investigated areas, two of which are rural ones: Gornja Stubla, situated in the southwest part of Kosovo, in Kalna a former uranium mine in the Eastern part of Serbia, and in the Montenegrin coast of the Adriatic sea.

2.1. Geological structure of the study areas and migration of groundwater and radon

2.1.1. Kalna

Kalna is a former uranium mining district which was exploited from 1948 to 1966, when the mine was closed. It is located in the Stara Planina mountain region in the eastern part of Serbia, where the discovered uranium mineralization is mostly limited to the western slopes of its central part, draining towards the river Trgoviski Timok through its tributaries, the Crnovrska, the Inovska and Gabrovnicka rivers.

The most important mineralization and uranium ore deposits occur between 300 and 1000 m above sea level, limited to the granite massive of Janja. The hydrothermal lentice-like layers of deposits and the appearance of uranium are connected to a number of time-activated cleavage faults. Deposits of Gabrovnica are clay-like, crumbling, near-surface cleavage zones. Generally, the mean uranium content in the granite ranges between 6 ppm and 20 ppm.

In the area of Stara Planina which is a significant metal-genetic zone, there are four uranium deposits which were temporarily exploited (Jankovic, 1965). The most significant uranium deposit near Kalna is concentrated in the granite massive of Janja (Fig. 1). Hydrothermal veins and saline deposits with the presence of uranium are linked to active brittle zones. The deposit of Mezdreja belongs to the same genetic type.

The deposits of Gabrovnica are tied to clay and porous surface layers of brittle zones. The granite massive of Janja ("Janja–Inovo–Gabrovnica" – see Fig. 1) is elongated in the NW–SE direction. It is

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