Journal of Environmental Radioactivity 164 (2016) 245–252

Contents lists available at ScienceDirect

Journal of Environmental Radioactivity

journal homepage: www.elsevier.com/locate/jenvrad

Radon in soil gas in Kosovo

Dafina Kikaj [a, *], Zvonka Jeran [b], Meleq Bahtijari [c], Peter Stegnar [a]

[a] Jožef Stefan International Postgraduate School, Jamova 39, 1000, Ljubljana, Slovenia
[b] Jožef Stefan Institute, Jamova 39, 1000, Ljubljana, Slovenia
[c] University "Hasan Prishtina", George Bush NA, 10 000, Prishtina, Faculty of Medicine, Prishtina, Kosovo

ARTICLE INFO

Article history:
Received 5 May 2016
Received in revised form
26 July 2016
Accepted 31 July 2016

Keywords:
^{222}Rn
Soil gas
^{226}Ra
CR-39 detectors
Lithology
Seasonal variation

ABSTRACT

An assessment of the radiological situation due to exposure to radon and gamma emitting radionuclides was conducted in southern Kosovo.

This study deals with sources of radon in soil gas. A long-term study of radon concentrations in the soil gas was carried out using the SSNTDs (CR-39) at 21 different locations in the Sharr-Korabi zone. The detectors were exposed for an extended period of time, including at least three seasonal periods in a year and the sampling locations were chosen with respect to lithology. In order to determine the concentration of the natural radioactive elements ^{238}U and ^{226}Ra, as a precursor of ^{222}Rn, soil samples were collected from each measuring point from a depth of 0.8 m, and measured by gamma spectrometry.

The levels (Bq kg^{-1}) of naturally occurring radionuclides and levels (kBq m^{-3}) of radon in soil gas obtained at a depth 0.8 m of soil were: 21–53 for ^{226}Ra, 22–160 for ^{238}U and 0.295–32 for ^{222}Rn. With respect to lithology, the highest value for ^{238}U and ^{226}Ra were found in limestone and the highest value for ^{222}Rn was found in metamorphic rocks. In addition, the results showed seasonal variations of the measured soil gas radon concentrations with maximum concentration in the spring months.

1. Introduction

Radon is a radioactive noble gas having the three naturally-occurring radioactive isotopes ^{219}Rn (Actinon), ^{220}Rn (Thoron) and ^{222}Rn (Radon). The parent of these series can be found in all natural materials, so all three radon isotopes are released into the air from the surfaces of rocks, soils and buildings materials. Their respective half-life's ($\tau_{1/2}$) are 3.96s, 55.6 s and 3.82 d. The abundance of actinon in the gases from geological sources is limited due to its short half-life, which means that most of it will decay during slow exhalation processes from rocks and soils, and it is virtually always produced in much smaller amounts than is ^{222}Rn, since the natural ^{235}U/^{238}U ratio of these ultimate progenitors is 0.00719. Hence, ^{219}Rn is largely ignored. ^{220}Rn too is short lived relative to ^{222}Rn and, consequently, moves a much smaller distance from its source than does ^{222}Rn.

Radon emanates and diffuses from rocks/soils and tends to concentrate in enclosed spaces, such as underground mines and houses. In these confined environments, ^{222}Rn accumulates leading to a potential hazard. Radon has been confirmed as the second major cause of lung cancer, soon after cigarette smoking (Darby et al., 2005; WHO, 2009) and is also the main source (approximately 55%) of internal radiation exposure to human life (ICRP, 1993). Worldwide average annual effective dose from natural sources is estimated to be 2.4 mSv, of which approx. 1.0 mSv is due to radon exposure (UNSCEAR, 2000). This knowledge has attracted considerable attention by the national and international authorities that have conducted nationwide radon surveys. In recent decades, most European countries have, therefore, adopted a number of regulations and made large efforts to identify radon-prone areas, e.g. in Austria (Friedmann, 2005), U.K (Green et al., 2002), Germany (Kemski et al., 2001), Czech Republic (Mikšová and Barnet., 2002) and Slovenia (Žvab et al., 2008). In Kosovo, the first radon measurements were conducted in the course of uranium prospecting in the period 1983–1989 (Jakupi et al., 1990) and monitoring of indoor radon in dwellings started ten years later. Indoor radon concentrations in the range of 20–450 Bq m^{-3} were found, with the exception of several houses built of stone in the uranium ore deposit area where values exceeded 1000 Bq m^{-3} (Jakupi et al., 1997). In order to make the radon map more complete, in 2002 indoor radon was measured in elementary and high schools in several towns (Bahtijari et al., 2007, 2006, 2008a; Nafezi et al., 2014) and in a krast cave (Bahtijari et al., 2008b).

Soil gas infiltration is recognised as the most important source of

* Corresponding author.
E-mail address: dafinakikaj90@gmail.com (D. Kikaj).

http://dx.doi.org/10.1016/j.jenvrad.2016.07.037
0265-931X/

indoor radon (Brown et al., 1993; Durrani and Ilić, 1997) and this has given rise to a number of studies and legal measures worldwide.

The radon activity concentration in gas at a point in soil is a result of its production in mineral grains by the radioactive transformation of ^{226}Ra (a member of the ^{238}U natural radioactive chain), its emanation into the void space between the grains, its transport by diffusion and advection/convection and eventually its exhalation from the soil into boreholes to reach the detectors (Etiope and Martinelli, 2002; Nazaroff and Nero, 1988). Emanation depends mainly on the distribution of radium in the grains (Greeman and Rose, 1996; Sakoda et al., 2010b), kind of mineral (Abumurad and Al-Tamimi, 2001; Jobbágy et al., 2009; Kardos et al., 2015; Sakoda et al., 2010c), grain size (Breitner et al., 2010; Greeman and Rose, 1996; Sakoda et al., 2010a), content of clay and organic matter (Greeman and Rose, 1996), moisture content in grains (Bossew, 2003; Sas et al., 2012), temperature of the grains (Iskandar et al., 2004; Lee et al., 2012) and the weather modification of grains (Schumann and Gundersen, 1996; Washington and Rose, 1990). Radon transport and exhalation are dependent mainly on soil permeability, moisture, and temperature and pressure gradients (Åkerblom and Wilson, 1982; Ershaidat et al., 2008; Etiope and Martinelli, 2002; Nazaroff and Nero, 1988; Papachristodoulou et al., 2007; Schery and Petschek, 1983; Singh and Virk, 1996; Tanner, 1980).

As a part of its projected European Atlas of Natural Radiation (EANR), the Joint Research Centre (JRC) of the European Commission, in cooperation with research institutions and radioprotection authorities all over Europe, is currently developing a map of the Geogenic Radon Potential (GRP).

The geogenic radon potential (GRP) is identified using variables that include geology data (geological units, lineaments, special geological features) and (radiometry) point data like soil gas radon concentration, geochemical concentrations of U, Ra, Th in soils, geophysical data (eU (*equivalent uranium*) by airborne gamma), external dose rate, soil permeability and outdoor radon concentration (De Cort et al., 2011). One suggested approach to quantify the geogenic radon potential for the geogenic radon map of Europe is developed by (Neznal et al., 2004) based on the soil gas radon concentration and soil gas permeability, followed by Kemski et al. (2001). Another suggested approach for the geogenic radon map has been reported in (Appleton and Miles, 2010; Barnet et al., 2005, 2008; Cinelli et al., 2011) based on the soil gas radon concentration, geology and indoor radon concentration.

The present paper is focused on the results obtained from a pilot study of soil gas radon measurements and its parents ^{238}U and ^{226}Ra in soil measurements carried out in the Sharr-Korabi zone of Kosovo. The results of the analysis are presented and discussed below. The variation of radon availability from the ground is multifactorial and, therefore, generalisations were not easy to obtain.

The present EIA (Environmental Impact Assessment) of Kosovo is inadequate due to insufficient funds to assess every aspect of this issue. Therefore, the data from this study will form a basis for establishing appropriate environmental radiation safety criteria for the regulatory body in Kosovo. Furthermore, the main aim is the identification of radon-prone areas, as suggested by the International Commission for Radiological Protection (ICRP), the EU Council Directive 96/29/EURATOM (1996) and the new EURATOM Directive on Basic Safety Standards published in January (EC, 2014).

2. Materials and methods

2.1. Geology of Sharr-Korabi zone

Kosovo is located in the southeastern part of Europe and extends into the central part of the Balkan Peninsula. It is stretched between the northern latitudes of 41° 50′ 58″ and 43° 15′ 42″, and the eastern longitudes of 20° 48’ 02” and occupies a key setting in the geological structure of the Central and Western parts of the Balkans (Aubouin and Ndojaj, 1964). The area of Kosovo is characterised by a variety of geological formations. Among these are rocks ranging from old crystalline Proterozoic to the youngest Quaternary age comprising sedimentary and magmatic rock types together with rather less frequent metamorphic rocks.

The following main units have been distinguished:

Dardania massif (Dardanides, DM, the Kosovarian part of the Serbo-Macedonian massif), Vardar zone (VZ) consisting of Internal Vardar subzone (IVZ), Central Vardar subzone (CVZ), and External Vardar subzone (EVZ), and Dinarides-Hellenides, subdivided into the Drin-Ivanicki element (DIE), East Bosnian – Durmitor zone (EBDZ), Ophiolite belt (OB) and Sharr-Korabi zone (SKZ).

Sharr-Korabi zone is related to the Pelagonian zone in Hellenides. This zone consists of the oldest formations. With reference to the Korabi zone, the available data are scarce, but in analogy with the neighbouring country Albania, we made a description of the area. Sharr- Korabi zone is composed of two sub-zones: Sharri subzone and Korintiku subzone. Sharri Mountain is stretched in the southern and southwestern of Kosovo, Sharri subzone thrusts on to the Koritniku subzone.

The upper Palaeozoic consist of greenschist, marble, chert, recrystallised cherty limestone, ignimbrite, etc. (at the Sharri subzone). The overlying Mesozoic rocks consist of Middle –Upper Triassic limestone, dolomite, and chert. The Middle – Upper Jurassic comprises volcanic-sedimentary unit is overlain by weakly metamorphosed shale, schists, sandstones and reddish conglomerates of inferred Permo-Triassic age (i.e. ‘verucano’ facies) are of a high Ti and calc-alkaline type. The sedimentary succession of Late Jurassic–Early Cretaceous age within the Korabi zone is regionally overlain by ophiolite. In Kosovo, ophiolite formation is spread mainly in the Vardar zone and in the Miredita-Gjakova ophiolite belt (Elezaj and Kodra, 2008). The ophiolite belt consists of a mélange, traditionally known as the Dinaride oliostrome (Robertson et al., 2009).

Sharr-Korabi zone has strong tectonic deformations (Elezaj, 2009).

The most important geologic characteristics influencing the radon concentration in soil gas are the litho-stratigraphical and the tectonic fractures. For this study, our measurement points were determined from the geological and tectonic maps of Kosovo on a scale of 1: 200,000 (ICMM, 2006; Knobloch et al., 2006) and classified into 3 units, based on the lithological classification:

- Metamorphic rocks (phyllite, quartz schist, sericite schist, metasandstone). Surficial material in this type of lithology is shallow brown soil with gneiss, marbles, quartzite, etc.
- Limestone (limestone with chert, dolomitic limestone, cherty limestone). Terra rossa (red soil) is heavy and clay-rich soil, strongly reddish (Fe-oxides) developed on limestone.
- Lake and river sediments (Mélange with olistoliths). Surficial material over lake and river sediments is sand, silt and clay, sandy gravel and mud/silt.

2.2. Sample collection and preparation

To quantify the spatial and long-term temporal variability of radon concentration in the soil gas, it was monitored at 21 different locations (house's and school's backyards) in the Sharr-Korabi zone. In the soil over the metamorphic rocks, 6 sampling points, on limestone 11 and on lake and river sediments 4 sampling points