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^{226}Ra activity distribution of rocks in the Sopron Mts. (West-Hungary)

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Abstract Radon gas is the largest natural source of human exposure to ionizing radiation and most of that exposure occurs in indoor air. Bedrock geology is an important factor in radon hazard evaluation of an area. The presence of rock types usually rich in uranium can be considered an indication of a potential radon hazard. In this study the average ^{226}Ra activity concentration of the main rock types (orthogneiss, micaschist, leucophyllite) in the Sopron Mountains was measured by gamma-spectroscopy, to reveal the uranium rich areas. This work is focusing on the distribution of ^{226}Ra among the different rock types of the Sopron Mountains with similar geological origin. The effect of different retrograde processes such as mylonitisation, fluid migration and argillitic–limonitic alteration on ^{226}Ra activity concentration was investigated. A few anomalies occurred in these metamorphic rocks. One explanation of the high uranium concentration is the high radioactive level of the rocks before the metamorphosis, but we demonstrated the significance of the above mentioned secondary processes as well. At Nándormagaslat quarry the presence of radium anomaly we found in the limonitic alteration of weathered gneiss (range: 131–726 Bq kg^{-1}) in fractures explains the high air concentration nearby in houses (96–2,051 Bq m^{-3}) and in a corresponding tunnel (maximum 600 kBq m^{-3}).

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Introduction

The health effect of radon (^{222}Rn) is well documented; this radioactive gas is also considered to be the leading cause of lung cancer second only to smoking [1]. Radon gas is the largest natural source of human exposure to ionizing radiation and most of that exposure occurs at homes [2]. The rocks and soil beneath dwellings are the principal sources of indoor radon. The amount of radium in the rocks and soil determines the amount of radon generated there [3, 4]. Mapping of radium concentration in rocks and soils helps to determine the risk areas and adds a new aspect to radon mapping encouraged by Radioactivity environmental monitoring (REM) group at the Institute for Transuranium Elements (ITU), Joint Research Centre (JRC) [4, 5]. According to the Council Directive 2013/59/EURATOM of 5 December 2013, laying down basic safety standards for protection against the dangers arising from exposure to ionizing radiation it is written that: “Member States shall identify areas where the radon concentration (as annual average) in a significant number of buildings is expected to exceed the relevant national reference level” [6, 7]. We can also contribute to this database with our work.

Sopron Mountains is an interesting place in terms of radioactivity. It composed of medium-grade crystalline schists, different types of orthogneiss and leucophyllite which may have high radium concentration according to the literature [8]. Gundersen et al. showed that 1/3 of the U.S. is characterized by high radon potential and a possible reason is the occurrence of metamorphic rocks and granite with high uranium content and high grade of deformation/

mylonitisation [8]. In some areas of the Sopron Mountains mylonitic rocks of granitic origin occur. Also high concentrations have been already detected in the area. One of the highest indoor air radon concentration was measured in the Sopron Mountains in Sopronbánfalva Geodynamic Observatory of the Geodetic and Geophysical Research Institute of the Hungarian Academy of Sciences where 600 kBq m^{-3} as maximum radon concentration can be measured at summer [9].

Mapping of radium activity concentration can provide significant information on pollution sources of radionuclide as it was described by Karadeniz [10].

The aim of this work is to show the distribution of ^{226}Ra activity concentration as the main source of air radon concentration in dwellings, among geological formations and main rock types around the Sopron Mountains. By locating areas with high radium activity concentration in the base rock, we can determine areas, where a risk of higher indoor radon concentration is to be expected. On the other hand, the exclusion areas can be defined, where no preventive measures have to be considered.

Sampling site

The Sopron area is the easternmost part of the Eastern Alps (Fig. 1). Several rock types including orthogneiss, micaschist, leucophyllite and amphibolite were described here. [11–13]. Three main geological formations were chosen to our examination: the Óbrennberg Micaschist-, Vöröshíd Micaschist- and Sopronbánfalva Gneiss Formations.

The metamorphic history of the Sopron area is quite complex, at least one pre-Alpine and an Alpine metamorphic stages were recorded. During the formation of the Sopron Mountains micaschist, the largest rock mass was the first to form from sedimentary rocks, then granitic rocks intruded into the micaschist and metamorphosed together during the Alpine orogeny. During the retrograde metamorphism the micaschist and orthogneiss (or metagranite) underwent several retrograde processes. These processes involved fluid migration and fluid–rock interactions, which locally may have increased U and Ra activity concentration in gneiss rocks. Fluid migration was facilitated by deformation during which cracks and fractures were opened in the rocks and mylonitic or cataclastic gneisses were created by plastic or rigid deformation from orthogneiss. Another fluid facilitated process when leucophyllite was created during Mg-metasomatism and the third one is an argillitic-limonitic alteration with different degree of phosphate mineralisation as it was described by Török [14–16].

Materials and methods

134 rock samples were collected around the Sopron Mountains. Samples are originating from surface sampling

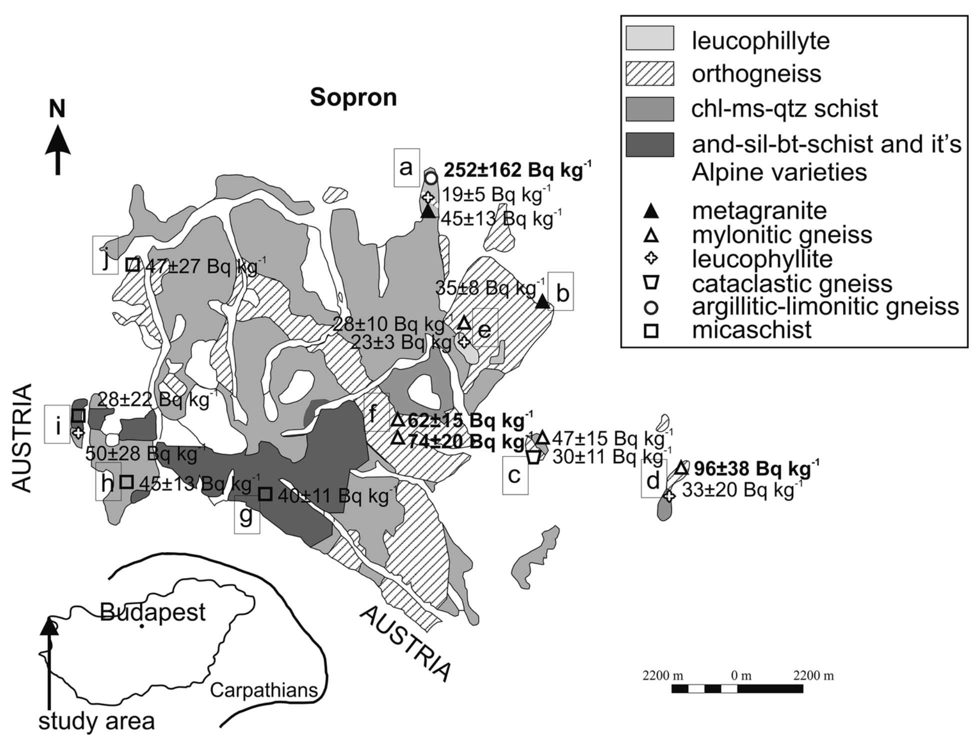

Fig. 1 Distribution of ^{226}Ra among geological formations and main rock types around the Sopron Mountains. *First number* indicates the average of the values and the *second* is the empirical scattering of the values of the several samples at a point. The anomalous values were demonstrated by *bold letters*. (*a*) Nándormagaslat quarry; (*b*) Várisi quarry; (*c*) Róka ház; (*d*) Harka quarry; (*e*) Vas hegy quarry; (*f*) Csalóka Spring & Road to Csalóka Spring; (*g*) Szarvas hegy; (*h*) Kovácsárok; (*i*) Kőbércorom;(*j*) Vörös híd quarry

of metagranite, orthogneiss, leucophyllite and micaschist rocks. Ten sampling sites were chosen to describe the specific ^{226}Ra activity concentration of the different dominant rock types of the area. The sites represent all the known rock types and varieties from the area. In most cases we could sample the rocks in outcrops from fresh rock. However, where no outcrops of the host rock was found within ca. 10 m around the given area we collected the rock samples from debris.

During sample preparation the rock samples were cut to have nearly cylindrical shape in the laboratory. These are about 4–5 cm in height and their diameters are about 6 cm.

The concentration of ^{226}Ra isotope in rock samples were determined by a GC1520-7500SL coaxial type HPGe detector (energy resolution 1.8 keV at 1 MeV, detection limit (DL) 0.5 Bq kg^{-1}). The cylindrical rock samples (mass between 70 and 270 g) were placed to the top of the semiconductor detector. The isotope concentrations were determined using the standard method of evaluating the 186 keV (^{226}Ra) peak of the spectra [17]. The duration of each measurement was 24 h. The radioactive equilibrium between ^{238}U and ^{226}Ra was assumed and was checked using a low statistics 1,001 keV peak intensity. In the total count rate of the 186 keV peak was split to two parts, 58.3 % of ^{226}Ra and 41.7 % of ^{235}U at a radioactive equilibrium [18]. The efficiency of the detection was calculated by Monte-Carlo simulation, which was taking into account the self-absorption, too. The uncertainties of each measurement were maximum 4 %.

Results and discussion

The average ^{226}Ra activity concentration and the empirical scattering of values of each rock types at each site (metagranite, mylonitic gneiss, cataclastic gneiss, leucophyllite, micaschist and clayey limonitic gneiss) are shown on Fig. 1.

In general, the activity concentrations of *metagranite*s are relatively low (average: 33 ± 3 Bq kg^{-1}, range: 6–45 Bq kg^{-1}). Metagranites are those rocks, where the original granitic texture was preserved and no visible signs of the above listed retrograde processes were observed. However we could detect anomalously high average activity concentrations of 96 Bq kg^{-1} (range: 50–146 Bq kg^{-1}) in a metagranite-gneiss quarry near the village of Harka (point d. on Fig. 1). Low values can be compared to the average radium activity concentration of the Hungarian soils, which is 33 Bq kg^{-1} (range: 14–76 Bq kg^{-1}) [2]. It shows that the distribution of radium have not been homogenous even in the original intruding granite.

Mylonitic gneisses were sampled at four sites (c. Róka ház; e. Vas hegy quarry; f. Csalóka Spring & Road to Csalóka Spring on Fig. 1.). The average radium activity concentration was 53 Bq kg^{-1}. The two highest values, both at Csalóka Spring, were 62 and 74 Bq kg^{-1} these are two times higher than the average of metagranite at Várisi quarry on Fig. 1. These values indicate that the chemical composition and activity of the rock may change during deformation process [8].

At the Nándormagaslat gneiss quarry (a. on Fig. 1) we found a thin layer of weathered gneiss with *argillitic–limonitic alteration* where the average radium activity concentration was as high as 252 Bq kg^{-1} (range: 131–726 Bq kg^{-1}). This was the highest measured radium activity concentration in the Sopron Mountains so far. Our results indicate that the weathering and the argillitic–limonitic alteration changes both the activity and the chemistry of the rock. This layer with high radium activity concentration cuts the tunnel of the Geodynamic Observatory, where extreme radon concentrations were measured [9]; presumably this layer is the source. To check the presence of the anomaly in the above mentioned layer we have taken samples from along this layer at every 0.5 m, horizontally. Furthermore, samples were taken from the fresh gneiss rocks, too, vertically about 1 m apart from the anomalous layer, and measured one order of magnitude lower values compared to the layer.

Anomalously high indoor radon concentrations were measured (96–2,051 Bq m^{-1} in 24 houses) in houses of Sopronbánfalva village that is in the vicinity of this site [19]. According to the new Council Directive of the European Union (2013/59/Euratom) "The reference levels for the annual average activity concentration in air shall not be higher, than 300 Bq m^{-3}" [20]. Sampling and measuring the radium activity concentration of the soil around the quarry and the nearby houses with high indoor radon concentration would help to limit the risk area where measurements should be taken to avoid damages in human health. These measurements also indicate the importance of radium activity mapping and may reveal the significant relationship between the radium activity and indoor radon concentration.

Micaschists (andalusite-sillimanite-bitote schist and chlorite-muscovite-quartz schist on Fig. 1) have the radium activity concentration of 46 ± 4 Bq kg^{-1}, which is somewhat higher than that of the orthogneisses, but not anomalous.

Leucophyllites—formed from orthogneiss and micaschist via Mg-metasomatism—cover essentially the same range as their parent rocks, despite the strong chemical and mineralogical changes during metasomatism that was demonstrated by ICP analysis [15].

Results of our measurements are summarized in Table 1.

Conclusions

In this study the distribution of ^{226}Ra activity concentration of different gneiss types and micaschists were measured around the Sopron Mountains in detail. Ten geologically different areas were chosen to the investigations. In case of metagranite and micaschist quarries we measured the ^{226}Ra activity concentrations of the original rock types where no retrograde processes occurred. The results of these rocks are in the range of 19–50 Bq kg^{-1} except for the metagranite quarry near Harka. Rocks from this quarry together with other three locations (Nándormagaslat quarry, Csalóka Spring, Road to Csalóka Spring) belong to the anomalous points with average activities between 62 and 252 Bq kg^{-1}. In case of the other three sites retrograde processes such as mylonitisation, fluid migration, argillitic–limonitic alteration occurred. All anomalous areas belong to the Sopronbánfalva Gneiss Formation. We concluded that anomaly can occur where (1) the distribution of radium was initially high in the original granitic rocks (2) presumably created by retrograde processes which caused chemical change in the rocks. The degree of changes in radium activity concentration depends on the mobilisation of U and Ra from the minerals and on their precipitation process along the cracks and fractures of the rock during the processes.

We showed that around the Csalóka Spring plastic deformation process resulted in a raised radium concentration of the gneiss rock up to 62, 75 Bq kg^{-1} from the initial value that is considered to be represented by the 35–45 Bq kg^{-1} of the metagranite of Váris and Nándormagaslat quarries.

The most significant anomaly that we have found occurred in weathered gneiss and limonitic alteration in fractures enhanced the radium concentration up to as high as 726 Bq kg^{-1} around Nándormagaslat quarry. This radium anomaly caused a detectable effect in the air of the nearby houses to the quarry. These results demonstrate that even within a homogeneous geological formation geological processes can cause changes in the distribution of

Table 1 List of sampling sites and rock types with the average ^{226}Ra activity concentration, standard deviation (SD) that represents the local spatial variability around the site, range of measured data, number of measured samples and the prevailing retrograde metamorphic or subsurface process

Area	Sampling site	Rock type	Average ^{226}Ra activity concentration (Bq kg^{-1})	SD	Range (Bq kg^{-1})	Nr. of samples	Retrograde process
a	Nándormagaslat quarry	Metagranite	45	13	27–57	5	–
a	Nándormagaslat quarry	Argillitic-limonitic gneiss	252	162	131–726	13	Argillitic limonitic alteration
a	Nándormagaslat quarry	Leucophyllite	19	5	15–27	5	Mg-metasomatism
b	Várisi-quarry	Metagranite	35	7.6	24–48	15	–
c	Róka ház	Mylonitic gneiss	47	15	24–89	18	Plastic deformation
c	Róka ház	Cataclastic gneiss	30	11	6–45	9	Rigid deformation
d	Harka quarry	Metagranite	83	46	19–147	6	–
d	Harka quarry	Leucophyllite	33	20	18–48	2	Mg-metasomatism
e	Vas hegy quarry	Mylonitic gneiss	28	10	17–39	3	Plastic deformation
e	Vas hegy quarry	Leucophyllite	23	3	21–25	2	Mg-metasomatism
f	Csalóka Spring	Mylonitic gneiss	62	15	34–82	12	Plastic deformation
f	Road to the Csalóka	Mylonitic gneiss	74	20	24–100	17	Plastic deformation
g	Szarvas-hegy	Micaschist	40	11	16–65	12	–
h	Kovácsárok	Micaschist	45	13	33–70	6	–
i	Kőbércorom	Micaschist	28	22	17–54	3	–
i	Kőbércorom	Leucophyllite	50	28	19–74	3	Mg-metasomatism
j	Vörös híd quarry	Micaschist	47	27	18–70	3	–

The letters in the first column refer to the location of the sites, as indicated in Fig. 1

radioactive isotopes. This fact should be taken into account during mapping of high activity areas and locating radon prone areas.

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