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Radioactivity of natural and artificial building materials – a comparative study

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ABSTRACT

Building materials and their additives contain radioactive isotopes, which can increase both external and internal radioactive exposures of humans. In this study Hungarian natural (adobe) and artificial (brick, concrete, coal slag, coal slag concrete and gas silicate) building materials were examined. We qualified 40 samples based on their radium equivalent, activity concentration, external hazard and internal hazard indices and the determined threshold values of these parameters. Absorbed dose rate and annual effective dose for inhabitants living in buildings made of these building materials were also evaluated. The calculations are based on ^{226}Ra, ^{232}Th and ^{40}K activity concentrations determined by gamma-ray spectrometry. Measured radionuclide concentrations and hence, calculated indices and doses of artificial building materials show a rather disparate distribution compared to adobes. The studied coal slag samples among the artificial building materials have elevated ^{226}Ra content. Natural, i.e. adobe and also brick samples contain higher amount of ^{40}K compared to other artificial building materials. Correlation coefficients among radionuclide concentrations are consistent with the values in the literature and connected to the natural geochemical behavior of U, Th and K elements. Seven samples (coal slag and coal slag concrete) exceed any of the threshold values of the calculated hazard indices, however only three of them are considered to be risky to use according to the fact that the building material was used in bulk amount or in restricted usage. It is shown, that using different indices can lead to different conclusions; hence we recommend considering more of the indices at the same time when building materials are studied. Additionally, adding two times their statistical uncertainties to their values before comparing to thresholds should be considered for providing a more conservative qualification. We have defined radon hazard portion to point to the limitations of the internal hazard considerations based on only measured ^{226}Ra activity concentrations without direct radon measurements. Our data are compared to those obtained in other countries and they provide a good basis to expand the database of radioactivity of building materials and gives information about the safety and situation of the building material industry in this central region of Europe.

1. Introduction

Since people in developed countries spend about 80% of their time indoors, knowledge of natural radioactivity levels in building materials is important (e.g. Kovler, 2009). The radionuclides ^{226}Ra, ^{232}Th and ^{40}K, originating in the Earth's crust, are present as naturally occurring radioactive materials (NORM) in building materials or in additives of building materials (e.g. Abo-Elmagd et al., 2010; O'Brian and Cooper, 1998), sometimes in technically enhanced amount (TENORM). These can cause both external and internal radiation exposure to inhabitants. External exposure arises due to gamma radiation, internal exposure arises due to inhalation of radon and thoron and their progenies, which are exhaled from the building materials and accumulated in indoor air (e.g. Abo-Elmagd et al., 2006).

This paper presents the results of a comparative study of radioactivity in two types of Hungarian building materials, i.e. natural and artificial ones. The materials were classified into these groups based on the presence of any induced heat treatment during the technologic preparation process of any raw materials or additives. Heat treatment can influence the radon exhalation properties via internal structure changes in the material (Sas et al.,

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2012). The natural type is adobe, which is made of soil (clay and sand), water and organic material. One can still find many adobe dwellings in Hungary. The NORM content of adobe is expected to be close to the Hungarian soil averages, i.e. 33 Bq kg^{-1} for ^{226}Ra, 28 Bq kg^{-1} for ^{232}Th and 370 Bq kg^{-1} for ^{40}K (UNSCEAR, 2000). However, local variations need to be considered. The artificial type is a grouping of many other types of building materials that occur in Hungary. This group is of interest mainly because of a widespread use of coal slag (bottom ash) and fly ash as additives during the 50s and 60s (Guler et al., 1995; Karangelos et al., 2004; Somlai et al., 1997, 2008). Coal slag was also used up till the 80s as an under-floor filling and insulating material (Somlai et al., 1998). Thus the radioactivity of the coal (from which the coal slag and fly ash is derived) can become a significant hazard for inhabitants of those buildings. In Hungary, the ^{226}Ra activity concentration in coal is highly variable, exceeding up to ten or twenty times the world average value (Bódizs et al., 1993). After burning, the remaining slag or ash can be enriched in radionuclides (Németh et al., 2000; Somlai et al., 2006). Hence, when these are used for building construction, an elevated gamma-dose rate is expected (Somlai et al., 1996).

In this paper, activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, and radium equivalent, activity concentration, external and internal hazard indices for building material qualification furthermore absorbed and annual effective dose rates are determined. Our main aims are to compare and qualify the different types of building materials used in Hungary and to provide data to the database of radioactivity of building materials, as well as place the samples into an international scale. We also compare the results of the different applied indices to each other and define a new value, radon hazard portion ($H_{R\%}$) based on the difference between the results of two hazard calculations for pointing to the limitations of internal hazard considerations frequently used without necessary explanation.

2. Samples

Two categories of samples: natural and artificial building materials were examined in this study. If none of the raw materials in the building material was artificially heated up above 30–40 °C (maximum summer temperature in Hungary), it belongs to the natural group. If any of the additives or the whole material has gone through a heat treatment during the production, it belongs to the artificial group. It is critical to distinguish these two groups because the heat treatment causes internal structure changes (Sas et al., 2012) and hence can influence radon and thoron exhalation, which is approximated in some hazard evaluations. Moreover, it can lead to enrichment of radionuclides (TENORM).

Three distinct areas of Hungary were selected for collecting adobe (AD) samples belonging to the natural group of building materials. These samples are made of the local soil. The studied areas are Békés County (BC, SE-Hungary), E-Mecsek Mts. (EM, S-Hungary) and Sajó and Hernád Rivers Valleys (SH, NE-Hungary) (Fig. 1.), where alluvial sediments of Körös and Berettyó Rivers, loess and redeposited loess are prevailing, respectively (Gyalog, 2005). Seven samples from Békés County (from Gyomaendrőd, Gyula, Sarkad, Újiráz, Vésztő), six from E-Mecsek Mts. (from Bátaapáti, Feked, Mórágy, Véménd) and seven from Sajó and Hernád Rivers Valleys (from Alsódobsza, Hernádnémeti, Sajóhídvég, Sajókeresztúr, Sóstófalva, Újcsanálos) (Fig. 1) were studied in detail. In an earlier study (Simó, 2008) adobe samples were collected from the Central Hungarian region (from Kunszentmiklós, Tass) (Fig. 1 in italics) giving the opportunity for a comparison with our results.

The artificial building material samples consist of two brick (B) samples, two concrete (of block of flats) (CBF) samples, six coal slag (CS) samples, three coal slag concrete (CSC) samples and seven gas silicate (GS) samples, six of which contain fly ash as additive. These were each collected from one of eight locations (from Budapest, Dunaharaszti, Gödöllő, Kecskemét, Kiskunhalas, Kistarcsa, Sülysáp, Százhalombatta) in the Central Hungarian region (CHR) (Fig. 1.).

Altogether, 20 adobe samples and 20 artificial samples are studied in this paper. All of the adobe samples and 14 of the artificial samples are used in bulk amount, whereas the other six artificial samples were in restricted usage (i.e. the six coal slag samples were used for under-floor filling). Note that the artificial building material sampling focused on finding hazardous materials, in contrast to the adobe samples which were collected randomly from typical areas. This difference in sampling strategy probably slightly influences the results obtained.

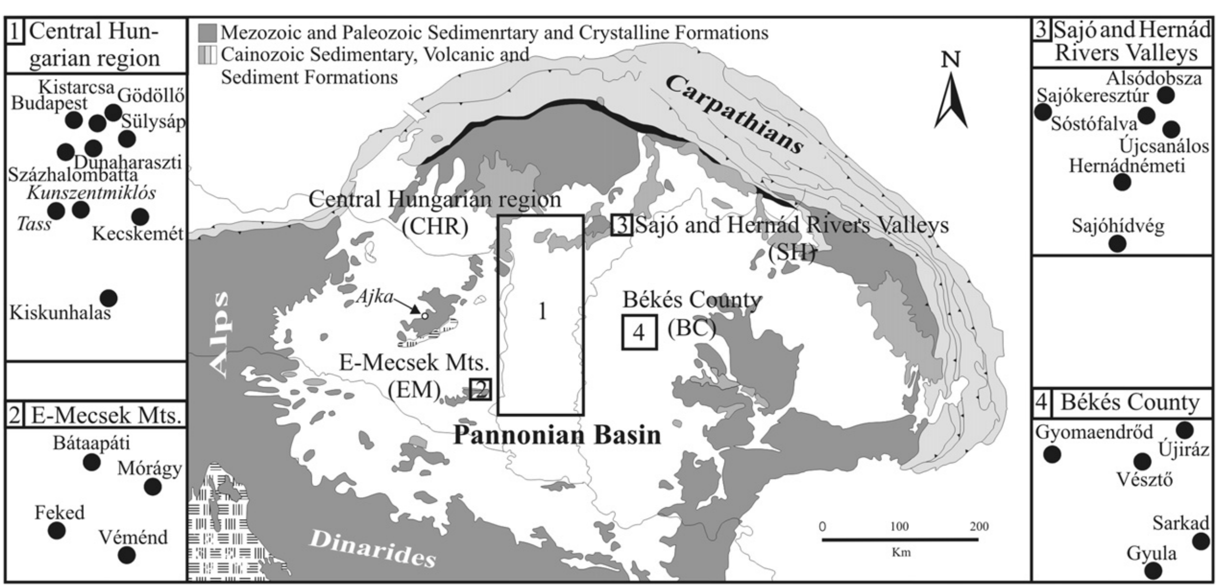

Fig. 1. Schematic geological map of Hungary and its adjacent area indicating localities of natural samples, i.e. adobes (Békés County, E-Mecsek Mts., Sajó and Hernád rivers Valley) and artificial building materials (Central Hungarian region). Three localities are indicated by italics: Kunszentmiklós and Tass (Simó, 2008) and Ajka (Somlai et al., 1996).