Environmental factors influencing the spatial distribution of geogenic radon sources (uranium, radium) in a granitic area

Theses of doctoral dissertation

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I. Introduction and objectives

In average, the population receive a total effective dose of 2.4 mSv y^{-1} from natural sources, from which terrestrial ionizing radiation is responsible for it of approximately 84 % (2.02 mSv y^{-1}) (UNSCEAR, 2008). The main contributors to the total effective dose are radon (1.15 mSv y^{-1}) and gamma radiation (0.48 mSv y^{-1}). The exposure to radon represents the second cause of lung cancer after smoking (UNSCEAR, 2000). Therefore, national and supranational institutions have established normative to minimize the risk. In the European Union, the European Commission proposed in the newest Euratom Basic Safety Standards (BSS) (Council Directive 2013/59/Euratom, 2013) to establish reference level for indoor radon concentration and to develop national radon action plan. That includes to assess relevant parameters for indoor radon and provides scientifically based maps of potential natural radioactivity hazard.

This research main aims to study natural radioactivity (gamma and radon) in a selected area and understand their relationship with local geology and properties of the soil and its rock debris. This research focuses on the detailed study of terrestrial natural radioactivity through the independent evaluation of 1) ambient gamma dose equivalent rate and 2) geogenic radon potential (GRP) referring to the potential risk of geogenic radon to the human health.

The evaluation of the ambient gamma dose equivalent rate focuses on the relationship with the underlying geology (rock types, forms, fractures, topography) through the application of digital spatial pattern analysis.

The evaluation of GRP focuses on two main aspects: 1) determination of the effect of soil properties on soil gas radon concentration and on soil gas permeability to obtain the controlling factors, and 2) test the usability of theoretical and empirical predictive models by comparison with the field measured corresponding values.

II. Sampling and methods

The study area is located in the western side of Velence Hills, which mass is made up by the outcropping Velence Granite Formation that was formed in the Variscan orogeny in the Carboniferous (280-300 Ma) (Horváth et al., 2004).

Evaluation of ambient gamma dose equivalent rate

The 19.8 km^2 evaluated area covers the full extent the granitic superficial outcrop. Ambient gamma dose equivalent rate was measured in situ by FH 40 G-L10 energy filtered proportional counter tube instrument (Thermo Fisher Scientific Inc.). Field measurements were carried out at ground level at 300 sites along a 250 m x 250 m regular grid. Field measured ambient gamma dose equivalent rates were interpolated using the triangular irregular network (TIN) method (Davis, 2011), and smoothed to suppress high frequency noise and enhance large scale spatial pattern. A systematic digital image processing methodology is applied to the interpolated ambient gamma dose equivalent rate map according to method of Evans (1972) as extended by Jordan et al. (2005) and Jordan (2007).

By the simple ‘higher than’ algorithms local maxima (peaks) and local minima were calculated (Jordan, 2007; Takahashi et al., 1995) and they revealed anomalous ambient gamma dose equivalent rates (Garbrecht and Martz, 1995). A dike density map was calculated by using total length of all dikes, regardless of their origin in order to highlight possible spatial relationship between ambient gamma dose equivalent rates and dike density.

Local variability of ambient gamma dose equivalent rates was measured by two different methods. Relief was calculated by using the range divided by the median value of the ambient gamma dose equivalent rates. Variability index was calculated by taking the square root of the absolute value of the squared differences between the maximum and minimum of ambient gamma dose equivalent rate. Relief and variability index maps were overlain by all type of dikes to see whether high dike density corresponds to high local variability of the ambient gamma dose equivalent rates (Beltrán et al., 2018).

The gradient calculations by Prewitt-operator were performed to identify the largest change of the gamma dose rates (‘slope’) and its direction (‘aspect’) at each grid point (Gonzalez and Woods, 1993). Uniform aspect with high gradient magnitudes along linear features may indicate geological influence on the ambient gamma dose equivalent rates distribution. Profile curvature is the second derivative of ambient gamma dose equivalent rates indicating sudden change in gradient magnitude and identifies inflection lines between convex (negative curvature values) and concave (positive curvature values) areas.

Ambient gamma dose rate lineaments are displayed as sharp linear edges on the interpolated surface and show sudden changes in the ambient gamma dose equivalent rates. Lineament density gives information on the local variance of ambient gamma dose equivalent rates and it was calculated by the total length of ambient gamma dose equivalent rate lineaments, similar to the dike density calculation. The resulting maps of digital image processing analysis were compared to geological maps with special emphasis on dikes and faults using GIS overlay. Spatial modelling was performed with Surfer 10, ILWIS 3.8 and ArcGIS 10 applications (Beltrán et al., 2018).

Length and frequency distribution of ambient gamma dose equivalent rate lineaments were shown in rose diagrams and compared to those of faults/fractures and dikes to understand their correlation (Beltrán et al., 2018).

Evaluation of Geogenic Radon Potential (GRP)

For the evaluation of GRP, 30 sampling points selected by the simple random sampling method in a total area of 0.8 km^2 located in the center of the exposed granite mass, in a slope sediment formation that belongs the Upper Pleistocene-Holocene transitional period. This superficial formation contains contracted slope sediments, angular debris, clays and sands. To determine the GRP of the study area by field measurements, soil gas permeability was determined by Radon-JOK equipment and soil gas radon concentration was measured with the active detectors AlphaGUARD and RAD7 connected to a soil probe from depths around 0.8 m after Neznal et al., (2004). At each sampling site, disturbed and undisturbed soil samples were collected to determine physical and geochemical properties of soil. Bulk density and water content have been determined by gravimetry, drying the samples at 110 °C at the at the Lithosphere Fluid Research Lab of the Eötvös Loránd University (ELTE). Soil effective porosity was determined from the previous analyses. Grain size analysis of soil were performed by two methods: dry sieving at the Lithosphere Research Lab (LRG), ELTE combined with sedimentation at Department of Soil Science and Agricultural Chemistry, Szent István University and laser diffraction by Horiba Partica 950-V2 LA Analyzer at the Laser Diffraction Particle Size Distribution Analyzer Laboratory of the Research and Instrument Core Facility of Faculty of Sciences, ELTE. The clay fraction was considered as representative clay mineral content based on its positive correlation with the elevated concentration of aluminum and rubidium in the bulk sample as characteristic components of clay mineral. Soil organic content was determined by ignition at 110 °C and carbonate content by treatment with

1N KCl solution and measured with a Scheibler calciminer and soil pH in the Department of Soil Science and Agricultural Chemistry, Szent István University. The chemical composition of soil samples has been analyzed by Inductively Coupled Plasma Mass Spectrometry (ICP-MS) by Bureau Veritas Commodities Canada Ltd. Canada. The activity concentration of the gamma emitters: ^{238}U, ^{232}Th, ^{226}Ra, ^{40}K, ^{232}Th, was analyzed by gamma spectrometry with ORTEC GMX40-76 HPGe semiconductor detector at the Institute for Radiochemistry and Radioecology, University of Pannonia. Radon exhalation rate and radon emanation were measured following the method published by Sas et al. (2015) in the same institute.

To test the usability of soil gas permeability, the empirical model developed by Rogers and Nielson (1991) was evaluated. Regarding to the soil gas radon concentration, theoretical models developed by Porstendorfer (1994) and by Várhegyi et al. (2013) were evaluated.

III. Thesis points of the doctoral study

1) I determined a high ambient gamma dose equivalent rate anomaly, spatially coincides with the high dike density area. Also, anomalous high local variability areas of the gamma dose rate coinciding with the area of high dike density characteristic for the southern part of the study area (Beltrán et al., 2018).

2) I identified that the gradient (slope, aspect, curvature), which represents the sudden changes of gamma dose equivalent rates in the study area coincide with the corresponding prevailing orientations of the underlying granitic dikes (SW-NE) and fractures (NW-SE) (Beltrán et al., 2018).

3) I determined that in the measured area, at pH < 8, the governing geochemical process is preferential adsorption of radium on organic material and clay at low concentration of calcium present as mainly carbonate mineral. In that process the controlling factors are the organic material, carbonate and clay content on the soil. At pH > 8, as the carbonate content increases, this process vanishes because calcium ions (generally, in higher concentration than radium at this pH) competes with radium ions for adsorption sites in organic material. Thus, the controlling factor in these conditions is the clay mineral content (Beltrán Torres et al., 2019b).

4) I proved that the empirical equation developed by Rogers and Nielson (1991) for the determination of soil gas permeability cannot be used for such a small area (< 1 km^2) with reduced range of soil gas permeability applied in this study. Therefore, I modified the model by changing the exponent of the particle diameter (from 4/3 to 8/5). Thus, the median soil gas permeability, obtained by the modified equation, is in excellent agreement with the field measured median value (Beltrán Torres et al., 2019a).

5) I determined that the two theoretical models, proposed by Porstendorfer (1994) and Várhegyi et al. (2013), for soil gas radon concentration underestimate the measured values. Based on the multiple linear regression analysis, I included the carbonate, organic material and clay content of the soil into the models. Thus, I obtain higher correlation and the same order magnitude of the modeled and the field measured values (Beltrán Torres et al., 2019a).

V. References

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VI. Publications related to doctoral study

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