Final report for OTKA-PD 115810
Principal investigator: Dr. Szabó Katalin Zsuzsanna
Duration: 36 months (2015-09-01 - 2018-08-31)

Title: Numerical modeling and spatial mapping of natural radioactivity for environmental risk assessment based on the geochemical and physical properties of the potential source rocks and soils

The general objective of this project was development, testing, calibration and reliability analyses of numerical modeling and spatial mapping methods of natural radioactivity (uranium, thorium, radon). A further scientific objective was the use of geochemical and physical properties of selected radioactivity source rocks and soils such as uranium content, mineralogical composition and soil porosity as independent input parameters into predictive natural radioactivity models.

In the first year of the project (September 2015 – August 2016) preparation for prediction and reliability analysis for natural radioactivity risk assessment (field and laboratory work) was started. In line with the work plan in the first three months equipment and software acquisition, tests of the instruments and equipments and literature review were performed. Also, search for known studies, results and data in The State Geological, Geophysical and Mining Data Store of the Mining and Geological Survey of Hungary (MBFH) were performed. In the Spring field surveys were conducted in the possible study areas. An outmost importance in the project was attached to the direct knowledge transfer to young researchers. Thus students were take part in the project from the beginning. PhD students from Szent István University and from Eötvös Loránd University were participated in the field surveys and the field works. For the highest level of research, I was collaborating during the 3 years with experts of the different research fields and I discussed with them the arised questionable parts. Csaba Szabó (ELTE), Győző Jordán (SZIE and GEM-RG), Zsolt Benkó (ATOMKI, MTA) and László Kupi (SRK Exploration Services) geologists were so kind and joined us in the field trips. During these preliminary field trips we 1) checked the accessible sites in the potential study areas, 2) identified the type of dikes, veins, altered zones, 3) determined the weathered and fresh rock types 3) collected rock samples, 4) checked the thickness of the soil, since for the measurements we need at least 80 cm soil, 5) made gamma dose equivalent rate measurements since it gives quick results, 6) assigned possible measurement areas, 7) made preliminary soil gas radon and soil air permeability measurements.

In the first year in accordance with the objectives of the project two IF papers and one book-chapter were published by analyzing our data which already existed. It was described in the Research Plan that radon prone areas with a high probability of elevated indoor radon concentrations must be identified for the protection of human health. This health hazard is quantified by the ‘geogenic radon potential’ (GRP). In addition, the main hypothesis was that the radon potential of a given rock formation can be assessed based on the physical and geochemical properties of the rock and the overlying soil types e.g. soil air permeability, porosity, arithmetic mean particle diameter, bulk density. Thus **in the first IF paper (Pásztor et al., 2016)** we investigated a new approach (regression kriging) for mapping geogenic radon potential (GRP), which has several advantages such as application of environmental co-variables (used in spatially continuous form), refined spatial resolution and inherent accuracy assessment. Our results demonstrated that not only pure geological information should be relied on GRP mapping, but further environmental variables (related to climate, land use, soil and topography) also play important roles. **In the second IF paper,** submitted on February 2016, analyzing ambient gamma dose equivalent rate by digital image processing and spatial analysis methods we confirmed that the main spatial features identified in the gamma dose rate map are influenced by not only the underlying geological conditions but also the surface sediment distribution defined by morphological conditions in the study area **(Szabó et al., 2017)**. **In the book chapter** we summarized the methods and applications for environmental geochemistry modeling including advanced spatial analyses at the regional scale: radon risk assessment **(Jordan and Szabó, 2016)**.

In Szabó et al. (2017) we applied 3.2 km average sampling distance (142 measurement sites for a 7200 km^2 area, Pest County) and we decided to check if the successful spatial analysis methods work in case of higher sampling resolution. Also during the field trips we realized differences in the gamma dose rate. Thus a 250 m average sampling distance was applied in the selected study area, the western part of the Velence Hills granitic area (20 km^2). Granite is a well-known rock types which have high radioactive element content compared to most of the other rock types. Field ambient gamma dose equivalent rate measurements were done with FH detector in the first year. 300 measurement sites were investigated (averaging from 3-6 measurements per each sites). My PhD student, Silvana Beltrán with Stipendium Hungaricum Scholarship, whose co-supervisor I am, was involved into the project (one of the student employment in the project) and took part in the field trips and laboratory analyses. Also, Ábel Szabó, PhD student (second student employment in the project) took part in the field works. Apart from the training of university students, I find it important to orient the young people towards the scientific field. Thus two high school students, who spent their summer practice at the SZIE under my supervision, were took part in these field measurements. Since in the second year (September 2016 – August 2017) the planning and also the intensive field soil radon and permeability measurements and soil sampling were performed, we made the evaluation of gamma dose rate measurements parallel. Thus results of this study were submitted only in the third year of the project, in March 2018. This study has shown that digital spatial analysis methods, including digital image processing techniques, are efficient in revealing spatial pattern in gamma dose rates and in identifying the relationship between the spatial pattern and the underlying geological setting at high resolution local scale. It is concluded that these methods provide useful means for the recognition and characterization of spatial pattern in field measured ambient gamma dose equivalent rate at the local scale, too. Results of this study were published **in the third IF paper (Beltran et al., 2018)**.

Results of the first year were presented as presentation at the International Workshop on the European Atlas of Natural Radiation (IWEANR 2015), November 9-13, 2015, European Commission JRC, Verbania (Italy).

(Note: the project could start with almost three months' delay in November 2015).

In the second year of the project (September 2016 – August 2017) detailed planning for field campaigns and also the intensive field measurements and sampling for soil and rock were performed. The difference to the plan is that one study site was selected instead of the planned three. Considering the tasks coming up meantime and the unexpected level of costs, two far sites were excluded. But this change gave the possibility to investigate the study site much more in detail and gave more established results. We selected a 0.8 km^2 meadow on the granitic area which has enough soil thickness for the measurements and it is easily accessible (VH01, Fig. 1). The underlying geological formation is re-deposited slope debris above the granite formation. It is at the western part of the Velence Hills near to Pákozd settlement. In this second year field measurements works were conducted in this study area. According to the planned supervised systematic soil sampling design 90 soil gas radon and soil permeability measurements were carried out at 30 sites at 80-100 cm depth (three at each sites) according to the standard accepted and suggested by the European radon community (Fig. 2). The 30 measurement sites were selected randomly in the study area. Soil gas radon activity concentration was measured by AlphaGUARD, soil air permeability was measured by Radon-JOK equipment (Fig. 3). Simultaneously sample collection for radioactivity, geochemical and soil physical measurements was done. Altogether 62 disturbed and 108 undisturbed soil samples were collected from two depths (30-40 and 80-120 cm) according to the planned sampling strategy (Fig. 4). We took samples from 80-120 cm which is the depth for soil radon and permeability measurements and also from an upper layer, 30-40 cm depth, to see the similarity or difference between the two layers. Perhaps the upper layer is sufficient for obtaining information and do not need to excavate deeper which is more time consuming. Soil samples were taken from 100 cm×80 cm soil sections at three sites and from drillings at the remaining 27 sites. Disturbed samples were taken by Edelman auger. Three soil sections were used for calibration and reliability analyses of soil sampling. Standard procedure in soil science that undisturbed samples are taken from the wall of a section by metallic cylinder (ring). Considering the costs of the field trips and the fact that we had to perform the field soil radon and permeability measurements in the same time with the soil sampling we planned not to perform the time-consuming soil sections at each 30 sites. Thus in three soil sections we tried a “top” undisturbed soil sampling and compared to the conventional method. With this “top” method we get the sample vertically from above the soil and not from the wall horizontally. This “top” method give the opportunity to get the sample from a drill hole from 80-120 cm without excavating a section. The comparison gave reliable results thus we used this method for the remaining 27 sites. Meteorological parameters were also recorded for each measurement just like the ambient gamma dose equivalent rate. Again for training and knowledge transfer for youngsters and to encourage them to learn further three high school students, who spent their summer practice at the SZIE under my supervision, were took part in these field and laboratory measurements. At the end of this year statistical evaluation of field measurements were done and the laboratory analyses were started (Fig. 3). In the laboratory the bulk density, water content, calculated porosity were determined.

Results of the second year were presented as presentation at the

- XIII. Environmental Science Conference of the Carpathian Basin, April 5-8, 2017, Cluj-Napoca (Romania),

… as a poster at the

- 8th Conference on Protection against Radon at Home and at Work and 13th International Workshop on the Geological Aspects of Radon Risk Mapping (Prague, Czech Republic, September 12-16, 2016).
- EGU General Assembly 2017, April 23-28, 2017, Vienna (Austria).

In the third year (September 2017 – August 2018) the laboratory analyses were accomplished such as further soil physical measurements: particle size distribution, pH, organic matter, clay mineral and carbonate content and the radioactivity lab measurements of soil and rock samples: isotope content (U-238, U-235, Ra-226, Th-232, K-40) by gamma-spectroscopy and ICP-MS, radon emanation coefficient and radon exhalation (Fig. 3). To carry out a large number of laboratory analyses, it was necessary to employ a non-researcher for 5 months. This change was requested on 16 October 2017 and it was approved.

According to the Research Plan two transfer functions applied and tested in this research, one theoretical model for soil gas radon activity concentration (1) and one empirical model for soil air permeability (2).

1) $C_\infty = C_{Ra}\, \varepsilon\, \rho\, p/(1-p)$

where C_∞ *= equilibrium concentration of* ^{222}Rn *in soil air, in kBq* m^{-3}*,* C_{Ra} *= Ra concentration in dry soil (Bq* kg^{-1}*),* ε *= emanation factor (dimensionless),* ρ *= dry bulk density (kg* m^{-3}*), p= porosity (dimensionless).*

2) $k = (p/500)^2\, d_4/3\, \exp(-12w_4)$

where p = total porosity, d = arithmetic mean particle diameter (m), excluding particles larger than mesh 4 (= 4.75 mm), w = volume fraction of water saturation (i.e. water content).

The following key questions are answered in this project:

1) How the model predicted value compares to the exact field measurement value? How the input parameters affect the output radon potential? How sensitive is the calculated radon potential to the variations in the input parameters?

2) How the geochemical composition of the studied rock and soil types such as mineralogical composition, uranium and radium speciation and mobility, or the organic matter, clay mineral and carbonate content of the soil, affect the soil gas radon concentration?

The model predicted values were not the same than the field measured values. They were not even the same in magnitude, however for soil radon they were significantly correlated (r=0.6, p=0,001). For soil permeability they were not the same and they are not correlated. We tried some procedures to find out why is the difference and we tried to get better results. We applied other parameters in the models such as effective porosity instead of total porosity or different water content definitions. We measured again the radon emanation and exhalation with another method in an other lab than previously. We searched for other models in the literature, we compared and tested them (Fig. 5). Also we found that thorium content and thoron emanation of the soil and rock is higher than expected which has an impact on the measured radon value. Also, we found statistically significant correlation between the measured radon and the thorium content of the soil (r=0.6, p=0,001). Which confirmed the thoron interference in the soil radon measurements. Another fact is that only the soil properties are considered in the models and not the rocks properties. This caused that we had to modify the measured radon values according to the thoron contribution since the model does not contain thoron part. For this we had to made new measurements with RAD7 in the field in summer of the third year to have the ratio of the radon and thoron in the study area. Next we included rock properties (Ra-226 content, emanation). After these procedures we obtained much better results, with the same magnitude. But some analyses are still in progress. Sincet he study area is a little bit downhill at the upper part the rock is much more closer to the surface and cause higher radon concentrations. This means that in case of a shallow soil layer the rock must alsobe taken into account in the models. For answering the other questions we correlated the soil gas radon with the other parameters and we found that it is higher in case of higher clay content and reversely lower in case of higher carbonate content. Soil gas radon and calculated gegenic radon potential is decreasing with the higher amount of sand and increasing with the higher amount of silt and clay, since the radioactive element bearing heavy minerals can be find below the sand fraction. But some analyses are still in progress.

At the conferences where we presented the results we got a lot of positive feedback and ideas and we had useful discussions. Based on these we had to make more analyses which take more time. Thus **the fourth IF paper** is under writing, we have a lot of new results we have to include into the paper and we will submit it until June 2019 and report about it to the NKFIH.

In cooperation with Colleagues from the Institute for Soil Sciences and Agricultural Chemistry, Centre for Agricultural Research, Hungarian Academy of Sciences (former TAKI) we planned to do a similar analysis than previously (Pásztor et al., 2016) since at this study area we have much more measured parameters and we can improve our previous modeling and spatial mapping. Accordingly, we made a one-day field trip together in July 2018 with the leadership of László Pásztor, to obtain high resolution soil information from the study area which can then be used for analyses combined with the existed parameters measured by us. We obtained information for the 90 points e.g. elevation, aspect, slope, profile curvature, ridge top flatness, valley bottom flatness, relative slope deposition, SAGA wetness index, terrain ruggedness index. Also field energy selective gamma measurements and XRF measurements were performed at the 30 sites and at some important points such as rock outcrops, nearby fluorite mine, nearby dikes and the nearby Juliska spring to obtain total uranium, thorium, potassium content and element composition. We took also aerial photos from the study area and aerial gamma dose rate measurements in the framework of the Safecast measurements. We are also working on to complete this study and submit the fifth IF paper of the project. However, it is expected that this paper will be published one year after the end of the OTKA, but I will report about it to the NKFIH.

The third IF paper was published meanwhile in the middle of the third year based on the gamma dose rate study (300 measurements) (Beltran et al., 2018).

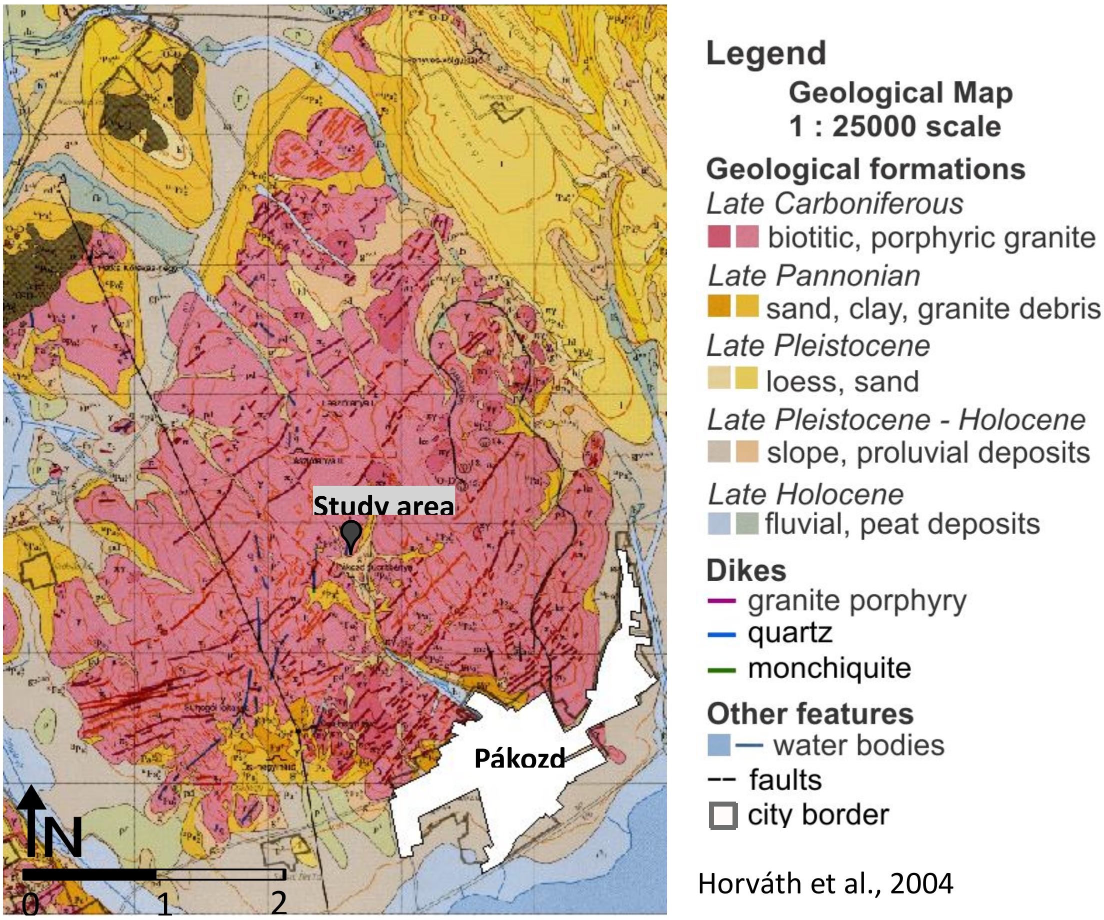
Results of the third year were presented as a presentation at the

- 2nd International Workshop on the European Atlas of Natural Radiation (IWEANR 2017), November 6-9, 2017, JRC, European Commission, Verbania (Italy),
- 6th International Geo-hazards Research Symposium 9th Dresden Symposium, IGRS Conference 2018, March 4-9, 2018, Dresden (Germany),
- XIV. Environmental Scienitfic Conference of the Carpathian Basin, April 5-7, 2018, Gödöllő (Hungary),
- VI. Terrestrial Radioisotopes in Environment International Conference on Environmental Protection, May 22-25, 2018, Veszprém (Hungary)
- 9th Assembly of Petrology and Geochemistry, 6-8 September 6-8, 2018, Szentkút (Hungary)
- 2 lectures during our visit connected to my bilateral TÉT_16-1-2016-0087 Project, 7 June 2018, INESC TEC, Porto, Portugal

… as a poster at the

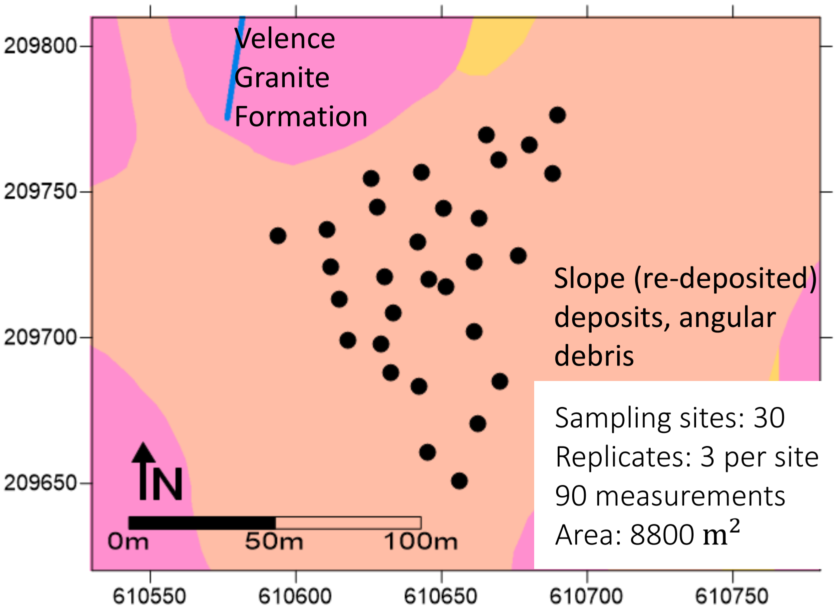
- 9th International Conference on High Level Environmental Radiation areas - For Undestanding Chronic Low-Dose-Rate Radiation Exposure Health Effects and Social Impacts (ICHLERA 2018) September 24-27, 2018, Aomori, Japan
- 5th Educational Symposium on Radiation and Health by Young Scientists, September 29-30, 2018, Sapporo, Japan.

During these 3 years the results of the OTKA project were presented in 9 presentations and 4 posters at 12 different conferences and meetings.



Horváth et al., 2004

Figure 1.



Sampling sites: 30
Replicates: 3 per site
90 measurements
Area: 8800 m^2

Figure 2.

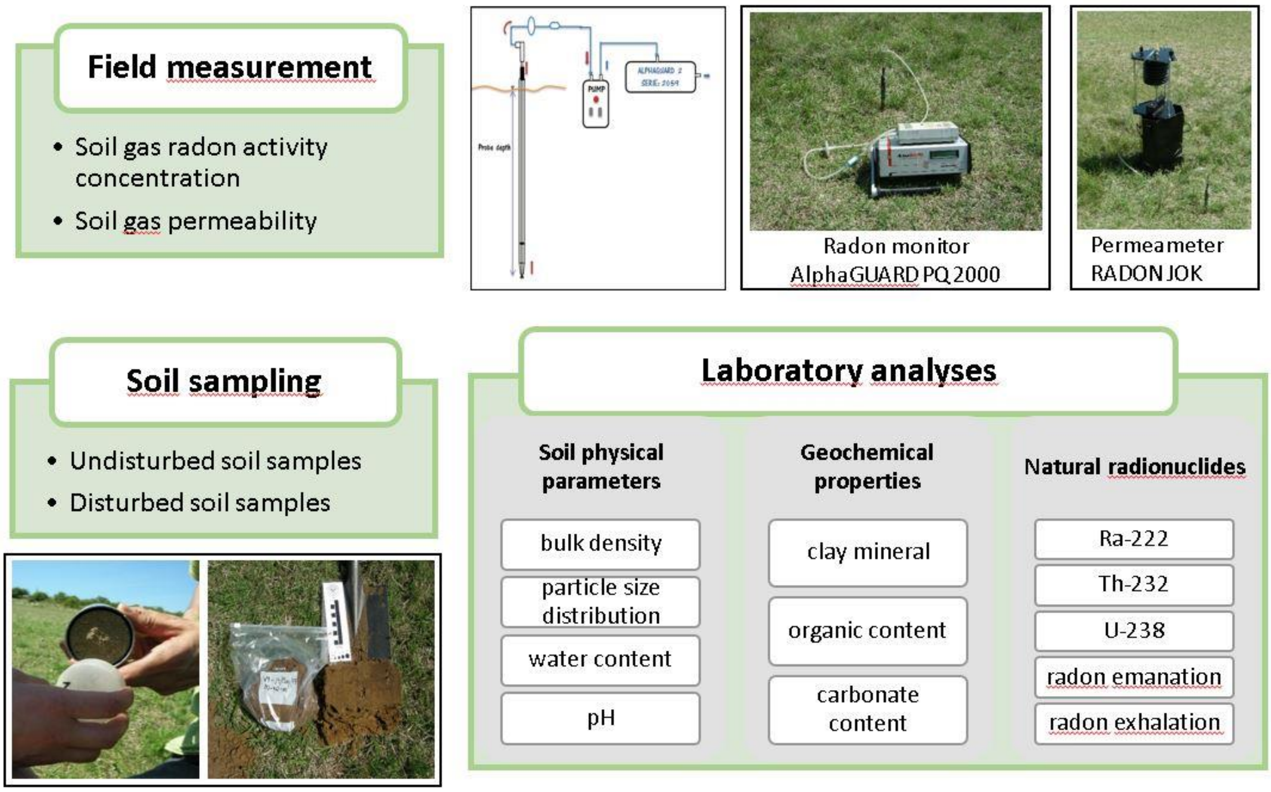

Field measurement
• Soil gas radon activity concentration
• Soil gas permeability
Probe depth
PUMP
ALPHAGUARD 2
SERIE: 2059
Radon monitor
AlphaGUARD PQ 2000
Permeameter
RADON JOK
Soil sampling
• Undisturbed soil samples
• Disturbed soil samples
Laboratory analyses
Soil physical parameters
bulk density
particle size distribution
water content
pH
Geochemical properties
clay mineral
organic content
carbonate content
Natural radionuclides
Ra-222
Th-232
U-238
radon emanation
radon exhalation

Figure 3.

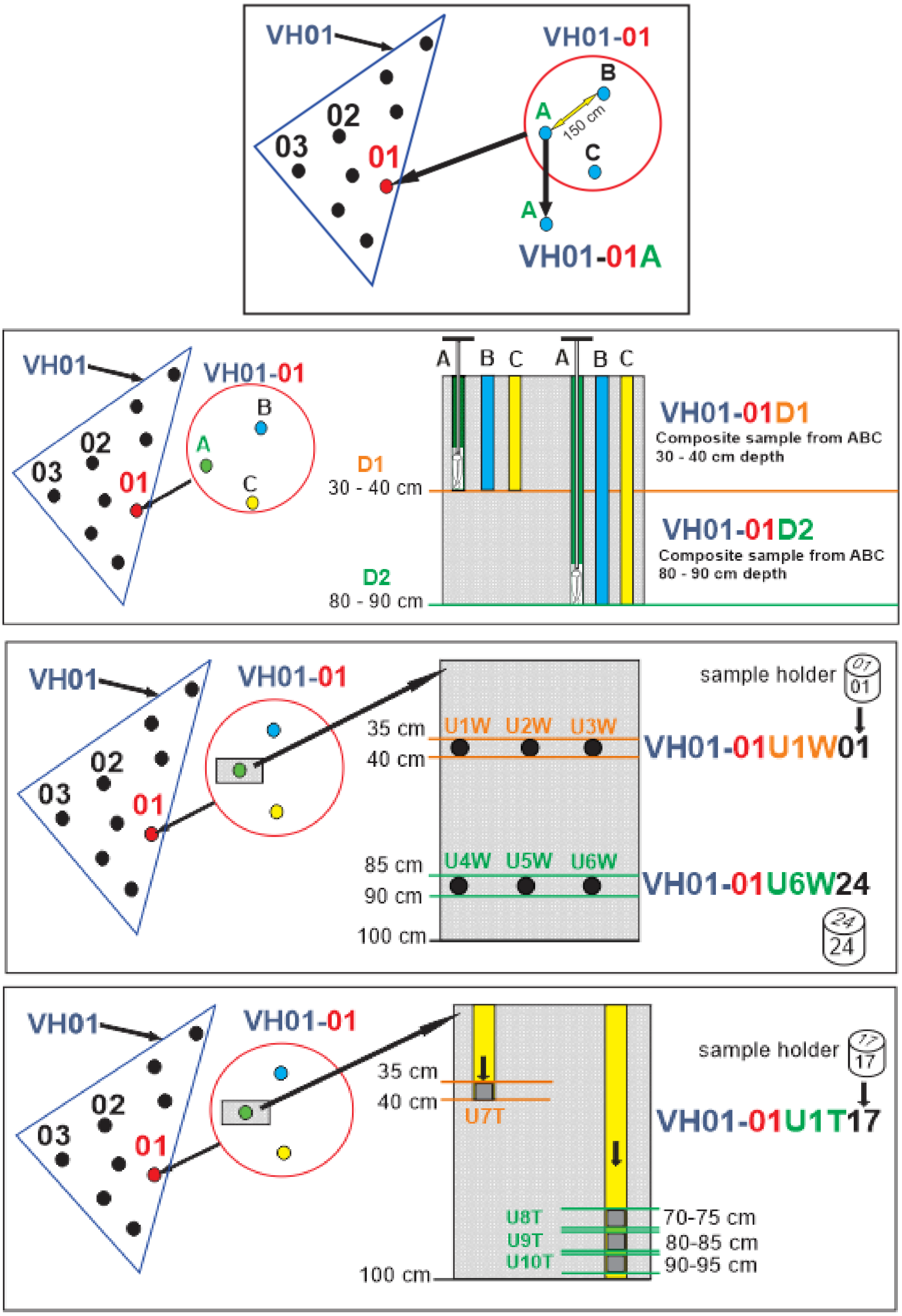
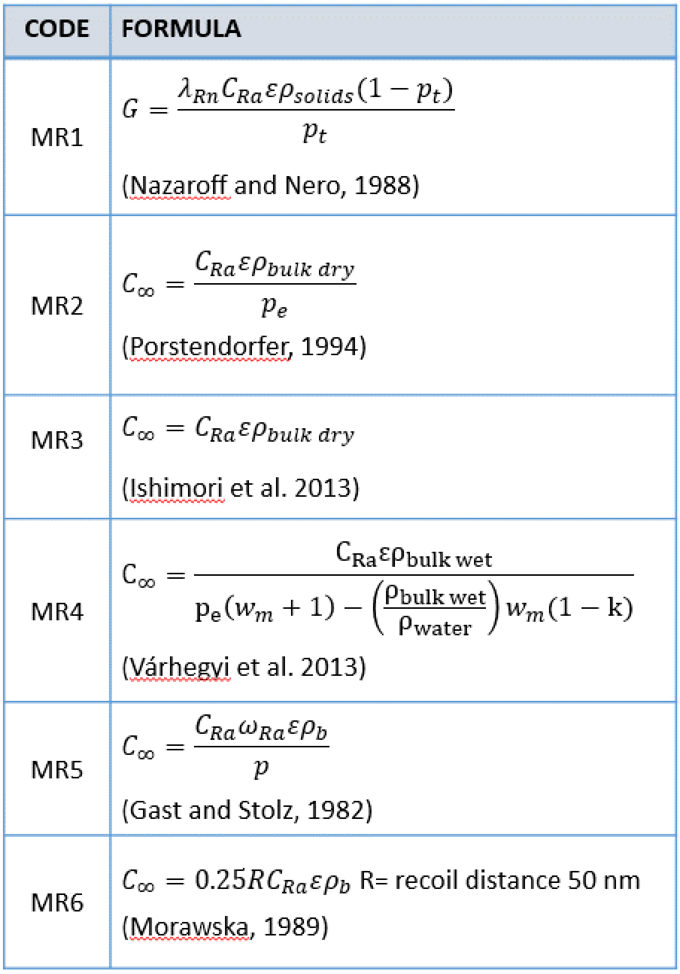

VH01
03
02
01
VH01-01
B
A
150 cm
C
A
VH01-01A
VH01
02
03
01
VH01-01
B
A
C
D1
30 - 40 cm
D2
80 - 90 cm
A
B
C
A
B
C
VH01-01D1
Composite sample from ABC
30 - 40 cm depth
VH01-01D2
Composite sample from ABC
80 - 90 cm depth
VH01
VH01-01
02
03
01
35 cm
40 cm
U1W U2W U3W
85 cm
90 cm
U4W U5W U6W
100 cm
sample holder
01
01
VH01-01U1W01
VH01-01U6W24
24
24
VH01
VH01-01
02
03
01
35 cm
40 cm
U7T
U8T
U9T
U10T
100 cm
70-75 cm
80-85 cm
90-95 cm
sample holder
17
17
VH01-01U1T17

Figure 4.



CODE	FORMULA
MR1	$G = \frac{\lambda_{Rn} C_{Ra} \varepsilon \rho_{solids} (1 - p_t)}{p_t}$ (Nazaroff and Nero, 1988)
MR2	$C_\infty = \frac{C_{Ra} \varepsilon \rho_{bulk\ dry}}{p_e}$ (Porstendorfer, 1994)
MR3	$C_\infty = C_{Ra} \varepsilon \rho_{bulk\ dry}$ (Ishimori et al. 2013)
MR4	$C_\infty = \frac{C_{Ra} \varepsilon \rho_{bulk\ wet}}{p_e (w_m + 1) - \left(\frac{\rho_{bulk\ wet}}{\rho_{water}}\right) w_m (1 - k)}$ (Várhegyi et al. 2013)
MR5	$C_\infty = \frac{C_{Ra} \omega_{Ra} \varepsilon \rho_b}{p}$ (Gast and Stolz, 1982)
MR6	$C_\infty = 0.25 R C_{Ra} \varepsilon \rho_b$ R= recoil distance 50 nm (Morawska, 1989)

Figure. 5

Several equipments used during the 3-year OTKA project were provided by different Institutes and Ltd's who I collaborate with. I would like to thank 1) to Csaba Szabó, Lithosphere Fluid Research Lab, ELTE for the FH instruments, meteorological station, RAD7 and for providing the availability of his laboratory and opportunity to discuss every kind of question connected to the research project, 2) to Ákos Horváth, Department of Atomic Physics, ELTE for the FH and RAD7 instruments, 3) to Mátyás Gáti, Mining Support Ltd. for the AlphaGUARDs, 4) to Zsolt Homoki and Géza Sáfrány, OSSKI for the Radon-JOK, 5) to Erika Michéli, Department of Soil Science and Agrochemistry, SZIE for the soil sampling equipments, 6) to Tibor Kovács, RRI University of Pannonia and Social Organization for Radioecological Cleanliness for the analyses on soil and rock samples.

This Postdoctoral OTKA project had not got only research part but also my salary, thus gave me a job, too. It gave me the opportunity to stay in the scientific research sector. **I would like to highlight**, that this PD OTKA enabled me to conduct many steps of professional scientific career parallel with the scientific research covered by this Fund. Thanks to this Postdoctoral Fund I could continue the cooperation with my former colleagues and I could make new collaborations, both in Hungary and abroad. These Colleagues, all of them, helped me a lot to see how to become an expert and also become a valuable member of society.

I could be a supervisor or mentor of students such as Msc, PhD and high school students as well. My PhD student was directly involved into this postdoctoral OTKA project.

It allowed me to remain a member of 4 and be a member of 2 new Scientific Associations.

It enabled me to become a member of the Hungarian Academy of Sciences and a member of the ELTE Doctoral School of Environmental Sciences.

During these 3 years I was a session chair and member of scientific committees at 5 international conferences and meetings, i.e. International Workshop on the European Atlas of Natural Radiation (IWEANR) JRC European Commission in 2015 and in 2017; 8th Conference of Protection against Radon at Home and at Work and 13th International Workshop on the Geological Aspects of Radon Risk Mapping in 2016; V. and VI. Terrestrial Radioisotopes in Environment International Conference on Environmental Protection in 2016 and in 2018.

I was a co-convener of a session (Geoscience applications of environmental radioactivity) at the EGU General Assembly in 2017 and in 2018 in collaboration with Susana Barbosa convener from INESC TEC, Portugal and other excellent researchers.

I was an invited speaker at The RAMARO International Workshop, Radon map (residential, geogenic, water) for Centre, West and Northwest Regions from Romania) Babeș-Bolyai University, Faculty of Environmental Science and Engineering, and at the 21st ELTE Hungarian Summer School of the Carpathian Basin in 2016.

It enabled me to take part in all of the Expert Meetings on Hungarian Radon Action Plan and become a member of the Work Group supporting the implementation of the Hungarian Radon Action Plan coordinated by the EMMI.

This Postdoctoral Fund let me to apply and successfully conduct other projects during these 3 years, i.e. bilateral TÉT project (TÉT_16-1-2016-0087), National Talent Program Fund Project (NTP-MTTD-15-0040) and four ERASMUS Mobility Grants.

During these 3 years, I was asked to review TDK, BSc, MSc and PhD theses and papers for 7 different journals.

Szabó Katalin

Szabó Katalin Zsuzsanna
principal investigator

Budapest, 2018. december 21.

Irodalomjegyzék

Beltrán Torres, S., Petrik, A., **Szabó, K.Z.,** Jordan, G., Yao, J., Szabó., C. (2018) Spatial relationship between the field-measured ambient gamma dose equivalent rate and geological conditions in a granitic area, Velence Hills, Hungary: An application of digital spatial analysis methods, Journal of Environmental Radioactivity 192, 267-278. **IF: 2.263, independent citations: 0**

Jordan G., **Szabó, K.Z.** (2016) Environmental geochemistry modeling: Methods and applications. In: Gruiz K., Meggyes T., Fenyvesi, É. (eds.): Engineering tools for environmental risk management. Vol. 3. Site assessment and monitoring. ISBN 9781138001565. p. 380.

Pásztor, L., **Szabó, K.Z.**, Szatmári, G., Laborczi, A., Horváth, Á. (2016) Mapping geogenic radon potential by regression kriging. Science of the Total Environment 544, 883–891. **IF: 4.9, independent citations: 6**

Szabó, K.Z., Jordán, G., Petrik, A., Horváth, Á., Szabó, C. (2017) Spatial analysis of ambient gamma dose equivalent rate data by means of digital image processing techniques, Journal of Environmental Radioactivity 166, 309-320. **IF: 2.31, independent citations: 5**