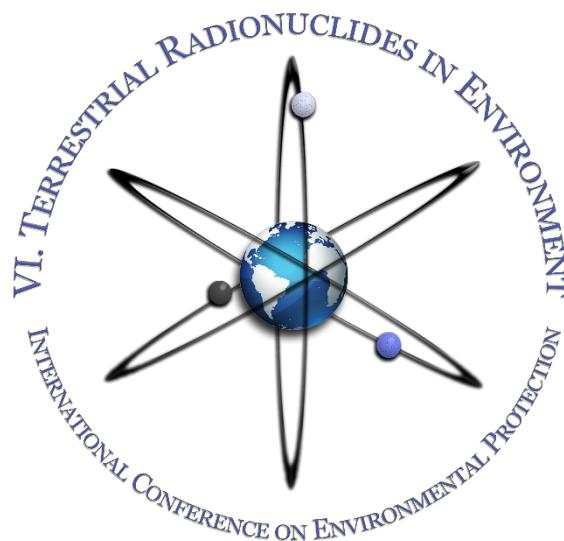


# VI. TERRESTRIAL RADIOISOTOPES IN ENVIRONMENT

International Conference on Environmental Protection



VESZPRÉM

2018

Social Organization for Radioecological Cleanliness

VI. TERRESTRIAL RADIOISOTOPES IN ENVIRONMENT  
International Conference on Environmental Protection

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## **VI. TERRESTRIAL RADIOISOTOPES IN ENVIRONMENT**

International Conference on Environmental Protection

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ISBN 978-615-00-2168-3

DOI 10.18428/TREICEP-2018

Published by the Social Organization for Radioecological Cleanliness

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## Variance and sensitivity analysis of food-chain models

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Artificial radionuclides are produced by nuclear explosions and nuclear facilities, if released into the environment, may reach the human body through several transfer pathways is considered as one of the important routes through which radionuclides can enter the human body via food-chain. The sensitivity analysis of input parameters for food-chain models [1-2] was performed as a function of deposition coefficient and transfer factors for the long-lived radionuclides (Sr-90, Cs-137). The input parameters were used from different databases [2-3]. The influence of input parameters for short and long-term contaminations of the foodstuffs after a deposition was also investigated. Their sensitivity indices were quantified as partial rank correlation coefficient. In our calculations, we were the most conservative approach to the realistic (best estimate) modeling conditions, which in practice meant that the calculation results declined significantly, up to several orders of magnitude. PRCCs were strongly dependent on the contamination period of foodstuffs as well as the deposition time of radionuclides. The results of this study may be serve as a useful information for improving the reliability of predictive results and saving a major effort in the collection of relevant data by entifying the main contributor of input parameters to model results.

[1] Generic models for use in assessing the impact of discharges of radioactive substances to the environment – Safety Report Series 19., IAEA, Vienna, 2001.

[2] Generic models and parameters for assessing the environmental transfer of radionuclides from routine releases – exposure of critical groups – Safety Series 57., Vienna, 1982.

[3] Handbook of Parameter Values for the Prediction of Radionuclide Transfer in Terrestrial and Freshwater - Technical Reports Series 472., IAEA, Vienna, 2010.

## Airborne gamma-ray spectrometry for investigating radon vertical profile

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<sup>222</sup>Rn gas has always been recognized as a sizable source of systematic uncertainty for the estimation of terrestrial <sup>238</sup>U concentration by means of Airborne Gamma-Ray Spectroscopy (AGRS) measurements. <sup>238</sup>U ground abundance is conventionally retrieved by monitoring the 1765 keV Energy Window (BEW) associated to the decay of <sup>214</sup>Bi, a daughter isotope that occurs after <sup>222</sup>Rn in the <sup>238</sup>U decay series. This prevents to distinguish the gamma signal generated by <sup>214</sup>Bi in the ground from the one emitted by <sup>214</sup>Bi attached to airborne aerosols and produced after the decay of <sup>222</sup>Rn exhaled into the atmosphere. A deep interest exists in understating the <sup>222</sup>Rn distribution as it has implications in tracing air vertical mixing processes, studying the dynamics of the atmospheric boundary layer and investigating health impacts of human exposure to low-level ionizing radiation.

We present the results of a dedicated off-shore AGRS campaign which led to the acquisition of 14688 1-second radiometric measurements performed in the (70 – 3000) m altitude range with a 16L NaI(Tl) detector. Experimental data were tested against a theoretical model describing the overall count rate recorded in the BEW (nBEW) as a superposition of a constant component due to the radioactivity of the aircraft plus a height dependent contribution due to cosmic radiation and atmospheric <sup>222</sup>Rn. The altitude profile of the <sup>222</sup>Rn component of the nBEW outlines the combination of a detector field of view effect, reflecting the 1765 keV photon mean free path in air (~ 175 m), and of the vertical distribution of <sup>222</sup>Rn itself. The latter has been modeled as a single air layer extending up to a cutoff altitude  $s$  and having uniform <sup>222</sup>Rn concentration  $aRn$  located at the bottom of a radon-free layer. Thanks to the large flight altitude range covered during the data taking and to the adoption of a refined  $\chi^2$  based statistical analysis we obtained not only a conclusive evidence of AGRS <sup>222</sup>Rn detection but also a <sup>222</sup>Rn concentration  $aRn = (0.96 \pm 0.07)$  Bq/m<sup>3</sup> and an atmospheric layer depth  $s = (1318 \pm 22)$  m fully compatible with literature data.

## **Natural radioactivity mapping via gamma-ray spectroscopy: Integrating different techniques and multivariate information**

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Among naturally occurring terrestrial radioisotopes, K and some radioisotopes in the U and Th decay chains emit  $\gamma$ -rays having energy of the order of MeV and can be easily detected. Gamma-ray surveys performed in laboratory, in field setting and from aircraft allow for monitoring the environmental radioactivity as well as provide valuable insights on geological mapping, structural geology and soil surveying. Natural radioactivity mapping provides the opportunity to integrate  $\gamma$ -ray spectroscopy data acquired with different measurement methods and field of views of the investigated area. In this framework the elaboration of a unique cartographic product with an appropriate descriptive legend cannot be pursued without critically dealing with some delicate issues. We dealt the problematic aspects regarding the statistical distributions of analyzed datasets, the heterogeneous experimental uncertainties and the spatial resolution of the measurements. Adopting a rigorous study of the data distributions and appropriated geostatistical interpolators, we integrated the results coming from the statistical treatment of the rock analysis and the spatial interpolation of the airborne measurements to which are associated distinct frequency distributions. Taking on the challenge to treat heterogeneous input uncertainties data in the spatial interpolation, we took into account the degree of the confidence associated to different gamma-ray techniques during the estimation process. Multivariate spatial estimations could enhance the estimation of radioelement distribution taking advantage of the correlation existing between the under-sampled  $\gamma$ -ray measurements and the continuous distributions of geological formations. Adopting this approach on the basis of a sophisticated geostatistical interpolator, we obtained radiometric maps tackling the complex aspects related to the performing of spatial interpolation of quantitative data using the qualitative information of the geological map as constraint. The described methods were validated through several surveys that cover approximately 50000 km<sup>2</sup> of the Italian territory: specific cases of studies will be presented and discussed.

## **Interaction of uranium(VI) with in-situ generated magnetite nanoparticles**

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Fate of uranium ions in an iron containing media is relevant for remediation of wastewaters resulted at the front-end of the nuclear fuel cycle and for geological nuclear waste disposal. Different processes such as direct immobilization of the uranyl ion ( $\text{UO}_2^{2+}$ ) on the magnetite surface as well as chemical, electrochemical, or biological reduction of U(VI) to U(IV)/ U(V) have been extensively investigated within the last decades. During our recent studies on the production of  $\text{AnO}_2$ -nanoparticles by hydrothermal decomposition of the associated oxalates, we have screened the applications of the method towards other relevant metallic elements. Thus, we found that iron(II) oxalate decomposes to magnetite ( $\text{Fe}_2\text{IIFeIII}\text{O}_4$ ) under certain reaction conditions. This leads to the ease of the reductive incorporation of uranium(VI) into magnetite based on the partial oxidation of iron(II). Indeed, in the presence of excess of uranyl ions in aqueous solution (pH= 7), the final reaction products are nanometric-sized  $\text{Fe}_3\text{O}_4$  and  $\text{UO}_{2+x}$ /  $\text{U}_4\text{O}_9$  (with quantitative precipitation of uranium). For smaller initial amounts of U(VI), the XRD indicates the formation of the  $\text{Fe}_3\text{O}_4$  as a sole reaction product, while the solution became uranium-free. That suggests the quantitative incorporation of uranium in the magnetite structure. Transmission electron microscopy (TEM) and X-ray absorption spectroscopy (XAS) analysis are presently ongoing in order to understand the mechanism of the uranium reduction/ immobilization during the decomposition of iron(II) oxalate under hot compressed water.

## **Soil-gas and indoor radon measurements in the Eastern sector of Mt. Vulsini volcanic district (northern Latium, Central Italy)**

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The knowledge of spatial distribution of Radon concentrations in soils is important in order to define Radon Prone Areas (RPAs), because geogenic radon is, along with some peculiar types of building materials, the major potential source of indoor radon accumulation. The present work was aimed at entifying RPAs in a selected volcanic area of northern Latium, in order to prove local Administrations of a useful tool for land-use planning and effective strategies aimed at health risk reduction. The studied area is located about 80 km NW of Rome, in the eastern border of the Quaternary Vulsini volcanic district, characterized by extensional tectonics, high heat flow and widespread endogenic gas emissions. Past volcanic activity mainly produced pyroclastic products and minor lavas flows. Soil-gas surveys were carried out in order to measure concentration of endogenic gases (radon and CO<sub>2</sub>) exhaling from deeper environments. Data were elaborated using the kriging algorithm that led to geo-referred maps of radon anomalies, mainly localized in the western sector, highlighting a sharp correlation with outcropping volcanic formations and faults. Long-term indoor radon activity was measured with charcoal canisters, and values show a mean value of 468 Bq/m<sup>3</sup>, far above the national Italian mean (77 Bq/m<sup>3</sup>). About 50% of the measured dwellings show radon indoor values above 300 Bq/m<sup>3</sup> (threshold value recommended according to the 2013/59/Euratom Directive). In some places, short-term radon concentration was also measured continuously using ionization chamber that highlighted radon fluctuation due to variations of meteorological parameters. Results show that the investigated area is generally characterized by a high geogenic radon potential; building materials of older houses, mainly constituted with local volcanic tuffs, and present life habits appear to be also important features affecting indoor radon accumulation. Being the studied area sparsely populated, the elaborated maps may facilitate local Administrations in appropriate urban planning policies, also focusing on the issue of public health management.



## **Preparation of a radon-in-water proficiency test: sampling, transport and sample handling conserations**

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This work focuses on the preparation of the next proficiency test on radon-in-water measurements organized by the European Commission-Joint Research Centre. As a pre-selection step candate natural waters were collected and analysed with the aim of using them as proficiency test materials. For the material characterization standard measurement methods based on gamma-ray spectrometry, emanometry and liqu scintillation counting were tested and verified. Since the proficiency test material would not be produced at the PT organizer`s site, the influence of sampling, transport and sample handling on radon loss was tested and the conditions were described.

## **A new approach to monitor the soil moisture tritium content in the unsaturated zone of the Paks Nuclear Power Plant**

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The existing monitoring network of the Paks Nuclear Power Plant has been designed solely to monitor the saturated geological media. The three-phase zone, which extends from the surface to approximately 8-meter-depth, has been given less attention. In order to understand the transport processes dominating in the unsaturated zone better, a new approach is applied. We established three groups of wells within the controlled area of the power plant. Arrangement of each well group is syringe-like, consisting of 5 wells filtering different depths. The individual wells have two 20-20 cm long screens at their bottoms. Sampling of soil moisture was carried out using silica bags, isolating the screens of each well by inflatable sacs. Two months averaged samples were collected for 10 months using this new system. The tritium content of water samples desorbed from silica gel traps was determined by LSC method. According to the results tritium loaded parts of unsaturated zone of the controlled area are well separated. Tritium activity concentration values in two well groups fluctuated above natural values (2 and 70 Bq/l) and an upward migration of the contamination from the saturated zone can be observed in the soil column. Concentrations measured in the third well group fluctuated between 0 and 2 Bq/l, so this well group carries background information about the area.

## **Temporal variations of radon concentration: human and natural factors**

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The dynamic of indoor radon concentration in residential and public buildings is mainly determined by two factors – weather conditions (external temperature, speed and direction of the wind, etc.) and the mode of operation of the room (ventilation, airtightness of windows and other ways of air leakage into the building, heating system operation, etc.). To assess the temporal variations of radon concentration, a series of long-term (from 3 to 6 months) measurements of radon concentration in apartments and offices were performed during the warm and cold seasons. The measurements were carried out using the AlphaGUARD radon monitors with measurement interval 1 hour. The surveyed rooms were kept in the ordinary mode of operation. The total number of surveyed rooms was: apartments – 20, offices – 6. It was observed that within the season radon concentrations changes up to one order of value. In a number of cases, the distribution of radon concentration is bimodal, representing different modes of the room use: the period of active functioning and the stationary mode without any expressed activity of its inhabitants. In many cases, significant diurnal variations in radon concentration values were observed due to changes in the operation mode of the room use: human activities or absence (sleeping). The difference between the average levels of radon concentration at daytime and night hours can be 20–30 %. The diurnal dynamics of the coefficient of variation of radon concentration also correlated with human activity in the room. The patterns observed in apartments have been confirmed by measurements results in offices analyzed separately on working days and weekends. For the majority of the urban buildings diffusion mechanism of radon entry was experimentally proved. Under dominating diffusion mechanism of radon entry, the radon entry rate independent on the temperature difference between indoor and outdoor atmosphere. At the same time, under the constant effect leakage area of the room, the natural ventilation increased in cold season due to increasing of stack effect. For all apartments, with high influence of their inhabitants on the room operation regime, a statistically significant excess of the mean values of radon concentration in the cold period over the mean values for the warm period was observed. When air exchange conditions in the room in general determined by natural processes opposite situation is expected for the dominant diffusion mechanism of radon entry. Thus, the "temperature effect" can influence not only the radon entry rate into building but also the behavior of its inhabitants. In our opinion, the season dynamic of radon concentration in the room is not only the physical process associated with the temperature difference between the outdoor air and the building envelope but also is the result of the living and operation habits.

## **Determination of non-exchangeable organically bound tritium concentration in reference material of pine needles (NIST 1575a)**

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Non-Exchangeable Organically Bound Tritium (Nx-OBT) in a biological reference material: NIST 1575a Pine Needles was determined by two different analytical methods which are radiometry and noble gas mass spectrometry. In radiometry, the sample was combusted to obtain OBT as water and tritium activity in the water was measured by liquid scintillation counting. In mass spectrometry, the sample was stored in a glass vessel and tritogenic <sup>3</sup>He was measured by noble gas mass spectrometer. Before the analysis by each method, bottled reference material powder around 100 g was immersed in tritium free water of approximately 200 mL at room temperature during overnight. The immersed sample was dried using vacuum drying method. This treatment was repeated three times to remove exchangeable OBT completely. After the measurement, the data were decay corrected to reference date (1 March 2002). Nx-OBT concentration ranged from 1.08 to 1.45 Bq/L-combustion water (CW) (n=4) with mean value ( $\pm$ S.D.) of  $1.25\pm 0.15$  Bq/L-CW in radiometry and ranged from 1.12 to 1.35 Bq/L-CW (n=3) with mean value of  $1.22\pm 0.25$  Bq/L-CW in mass spectrometry. For the Pine Needles reference material, there is no proposed values about Nx-OBT, but our results showed a good agreement with the data determined by the different method. Thus, it is useful material for quality control of Nx-OBT measurement.

## Statistical regularities of terrestrial radioisotopes distribution in mountainous regions of Transcarpathia: global and local factors

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Earth crust, soils, mud, rocks, etc. are formed by chemical elements with the stable and radioactive nuclei, which are products of decay of the natural series  $^{238}\text{U}$ ,  $^{232}\text{Th}$ ,  $^{235}\text{U}$ ,  $^{237}\text{Np}$ . The content and ratio of the formed trace elements and radioisotopes determine the geochemical and radiation conditions of biota living in this territory. This pattern is also maintained for soils formed on the sites of the previous location of rocky rocks, because under the influence of winds, rains, the transfer of matter from the soil horizons takes place. Soils of low-lying mountains areas are formed in result of an accumulation of sediment deposits from higher horizons due to water passing through them and processes of the mutual mixing of their chemical and isotopic components. This report presents the results of summarizing and statistical processing of the results of the radioecological monitoring of Transcarpathia areas for the data of the research on the sediments of mountains rivers and soils of the protected areas. The distance between the points of sampling was within 200-400 m; the samples were taken on the surface, from a depth of 20 and 50 cm. The protected areas were at a distance of 100-300 km from each other. The composition and ratio of terrestrial radioisotopes both of natural and artificial origin were studied, statistical regularities of their accumulation and distribution under the influence of geochemical, meteorological and technogenic factors were systematized. The basis of the research methodology is the constancy of the correlations of gamma-active nuclei (GAN) of the natural series of  $^{238}\text{U}$  and  $^{232}\text{Th}$ ,  $^{40}\text{K}$ , their isotopic abundances in the objects of the environment, and their variability under the influence of spatial (the mountains area altitude, depth of soil layers, the riverbed location) and seasonal factors. The method of correlation and factor analysis was used to establish the patterns of distribution of GAN in the sediments from mountain rivers, for various soil horizons depth, on mountain slopes and rges. Comparison of data of radioecological and geochemical monitoring of the content of heavy metals and their GAN in environmental samples also done. It is shown that the actual state of the background radioactivity for a given territory is the result of both the global factors caused by the isotopic abundances content of GAN as products of the decay of natural radioactive rows and the intensity of numerous external factors. The latter has a different effect on the degree of statistical clusterization of sampling points for the high- and lowland areas of Transcarpathia. Mountains are suitable the model objects for studying the nature of changes in the natural activity of mountains and nearby territories under the influence of external factors, the formation of isolated mountain areas with a special isotopic and microelement composition of their soils. The results of such studies may be useful for other European mountain systems and in case of radiation mapping of areas.

## **Neutron Activation Analysis in determining radionuclides in environmental, geological and biological samples: an overview**

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The radioactivity of the  $^{235,238}\text{U}$  and  $^{232}\text{Th}$  isotope decay chains for environmental samples can usually be assumed to be in equilibrium due to their age. Similarly, one can assume that the isotopic mass proportions are equal to natural isotopic abundance. Current methods used to ascertain activity in these decay chains involve alpha particle spectrometry, ICP-MS or passive gamma-ray spectrometry, all of which can be laborious and time consuming. In this research, we have used thermal and epithermal neutron activation analysis (NAA) of small sample sizes of various geological materials in order to ascertain these activities. By using NAA, we aim to obviate cumbersome sample preparation, the need for large samples and extended counting time. In addition to the decay chains of uranium and thorium,  $^{40}\text{K}$  was also determined using epithermal neutron activation analysis to determine total potassium content and then subtracting out its isotopic contribution. Neutron Activation Analysis (NAA) is well known to determine many elements in a wide range of samples. However, NAA can determine uranium, potassium and thorium using the  $^{238}\text{U}(n,\gamma)^{239}\text{U}$ ,  $^{41}\text{K}(n,\gamma)^{42}\text{K}$  and  $^{232}\text{Th}(n,\gamma)^{233}\text{Th}\rightarrow^{233}\text{Pa}$  reactions. Once these concentrations are converted to activities one can then determine  $^{234}\text{U}$  and  $^{235}\text{U}$  assuming equilibrium and knowing the fixed ratio of  $^{238}\text{U}/^{235}\text{U}$ . As well  $^{234}\text{U}$  also in the  $^{238}\text{U}$  decay chain can be determined. We have also shown how the soil-to-plant transfer factor(s), TF(s) and plant-to-animal transfer coefficient(s), TC(s) two of the most important parameters widely used to estimate the internal radiation dose from radionuclides through food and water ingestion has been used by the determination of total strontium and cesium using NAA.

## **Radon as an indicator of atmospheric stability over the Ljubljana Basin in the cold season**

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Diurnal variations in radon activity concentrations in the lower atmosphere are a direct measure of atmosphere's air mixing capacity. Recently, use of radon as an indicator of lower atmosphere vertical mixing has been steadily increasing. Vertical mixing is expressed by the level of stability of the atmosphere within several categories that closely corresponds with urban pollutant concentrations.

In the present study, radon in air was continuously (ones an hour) monitored, using an AlphaGuard radon monitor (Saphymo, Germany) in diffusion mode. The device was set up in a weather instrument shelter, 1.5 m above the ground at the Ljubljana-Bežigrad meteorological station of the Slovenian Environment Agency.

The aim of the study was the testing of the recently-developed 'radon-based stability' classification with our radon data. The complex landscape of the Ljubljana Basin, characterised by the lake and river sediments (with enhanced radon exhalation rate), with generally calm winds, exhibits frequent persistent and strong nocturnal cold-air pool. Thus, discerned 'radon-based stability' classifications are needed for the 'warm' and 'cold' periods of the year.

By using the deviations of radon activity concentrations by more than  $\pm\sigma$  from the winter season mean value, a 'five-category radon-based stability' classification was built up for the cold season in the Ljubljana Basin by slightly modifying the 'radon-based stability' classification.

The 'five-category radon-based stability' was then compared with the commonly used 'Pasquill-Gifford (P-G) stability' classification (categories: extremely unstable, moderate unstable, weakly unstable, neutral, weakly stable, moderate stable, strongly stable) and with the previous 'radon-based stability' classification (categories: near neutral, weakly stable, moderate stable, stable), admitting that the strongly stable category in the 'P-G' and stable category in the 'radon-based stability' is less or not selective for the most stable conditions (persistent/nocturnal cold-air pool) as strongly stable category of 'five-category of radon-based stability' (categories: weakly unstable, neutral, weakly stable, moderate stable, strongly stable). This means that the other stability classifications may underestimate pollutant concentrations in the strongly stable atmosphere in the Ljubljana Basin in wintertime. Lastly, it has been demonstrated that radon observations at a single height are applicable for monitoring atmospheric stability during cold season in the sites with low wind conditions and situated far from coastal influence, when advective effects are minimal.

## **Determination and distribution of $^{210}\text{Po}$ in tobacco plants at during various growth stages from Manisa, Turkey**

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The main alpha-emitter in tobacco is  $^{210}\text{Po}$  ( $t_{1/2}=138$  d). It is also the most radiotoxic nucle to human beings. Tobacco plants take around a couple of months to get the right size for harvesting. In the study, the soil and tobacco plant samples were collected monthly between transplant to harvest (April, May, June and July 2016). The average  $^{210}\text{Po}$  activity concentrations were determined in the different parts of tobacco plants (leaves, mdle part and root) and soil samples.  $^{210}\text{Po}$  was determined by alpha spectrometry using PIPS detectors after radiochemical separation and spontaneous deposition of polonium on a copper disc. Transfer factors of  $^{210}\text{Po}$  from soil to tobacco plants were determined. The results of  $^{210}\text{Po}$  activity concentrations of this study were in the range of values measured elsewhere in the world.



## **Metrological tools to support environmental analysis of safeguards samples**

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The JRC Directorate G for Nuclear Safety and Security has a long history of supporting nuclear safeguards by producing certified reference materials and organising inter-laboratory comparisons in compliance with ISO 17034 and ISO 17043, respectively. In this paper, some aspects of the latest Nuclear Signatures Inter-laboratory Measurement Evaluation Program NUSIMEP-8 will be discussed in the view of the performance of participating laboratories measuring Pu amount ratios with the help of Naji plots. Furthermore, the recent certification of the IRMM-042a <sup>244</sup>Pu spike reference material will be presented as potential isotopic reference material for environmental sample analysis.

## **Natural radionuclides ( $^{226}\text{Ra}$ and $^{210}\text{Po}$ ) in groundwater and tap water in İzmir, Turkey**

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In the study,  $^{210}\text{Po}$  and  $^{226}\text{Ra}$  activity concentrations of 9 groundwater samples and 30 tap water samples were determined.  $^{210}\text{Po}$  was determined by alpha spectrometry using PIPS detectors after radiochemical separation and spontaneous deposition of polonium on a copper disc.  $^{226}\text{Ra}$  measurements in water samples were performed by using collector chamber method. The method is based on the measurement of the alpha particles emitted by the radon daughters. A copper plate with a diameter of 4.5 cm, a collector chamber having a volume of 2.47 L and an applied voltage of 600 V are optimal conditions for the collector chamber method. The natural activity concentrations of  $^{210}\text{Po}$  and  $^{226}\text{Ra}$  are within the range recommended by World Health Organization. All values for each type of water, as well as for each population group, were well below the recommended reference dose level (RDL) of 0.1 mSv from one year's consumption of drinking water according to the European Commission recommendations of 1998.

## **Soil radioactivity at Lake Chagan**

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On January 15 of 1965, an artificial lake in Kazakhstan was created by The Chagan nuclear test. The aim of our work has been to determine the physical processes that took place during the explosion that modified the composition of the soils near the lake. The research has been carried out examining three samples of the terrain with gamma spectroscopy. We have successfully identified several radioactive isotopes, determined their activities and offer a hypothesis for the cause of their presence in the soil. The results have been compared to other similar measurements carried out in New Mexico and Semipalatinsk nuclear test sites.

## **Determination of natural radionuclides for the samples very rich in thorium in Mandena deposit, South Madagascar**

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The Mandena deposit occurs as black sand placer on the East-South of Madagascar. The deposit contributes about 2/3 of the total heavy mineral resources of this country which is estimated about 20 million tones in total of ilmenite, rutile and zircon minerals. The average concentration of the ilmenite, monazite, zircon and other minerals are 66.72%, 2.3%, 2.8% and 28.18% respectively. The laboratorial measured activity concentration of <sup>238</sup>U, <sup>226</sup>Ra and <sup>232</sup>Th of sand samples in this deposit range from 2060 Bq/kg to 4220 Bq/kg, 1500 Bq/kg to 2880 Bq/kg and 11,000 Bq/kg to 24,400 Bq/kg respectively. The absorbed dose rate ranges from 7350 to 16,000 nGy/h, the dose rates are from 120 to 270 times higher than the average terrestrial radiation background of 60 nGy/h, the effective dose rate for the human being living in this area could be up to 140 mSv/year with principal contribution from <sup>232</sup>Th activity concentration.

## **An empirical function of energy and distance for the gamma-ray efficiency of a HPGe detector**

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One of the important tasks of ecology investigations is the reliable gamma-spectrometric measurements of highly active samples (for example, the Fukushima nuclear accident). To solve this problem, it is necessary to increase the distance between the sample and the end cup of the detector. This makes it possible to reduce the dead time of measurements and, accordingly, to improve the accuracy of the results. We carried out the experimental studies of the efficiency of the HPGe- detector ("ORTEC", 150 cm<sup>3</sup>) at 7 different distances from the source of radiation to the end cup of the detector (5 ÷ 24.5 cm) for the energy range 59.5 ÷ 1408.0 keV (for 18 gamma lines). The detector efficiency was determined using 8 standard point sources of gamma radiation, namely Na-22, Co-57, Co-60, Cd-109, Ba-133, Cs-137, Eu-151, Am-241. The spectrometric standard sources were produced by the Metrology Institute (St. Petersburg, Russia) and Eckert & Ziegler Isotope Products GmbH (California, USA). A formula is proposed for describing the dependence of the efficiency on the distance (5 ÷ 25 cm) for the mentioned energy interval.  $\log_e(E) = a_0(d) + a_1(d)(\log E/E_0) + a_2(d)[(\log E/E_0)]^2 + a_3(d)[(\log E/E_0)]^3 - a_4/E^{a_5}$ , where  $e$  – efficiency,  $E$  - gamma-rays energy (keV),  $d$  - distance from sources to the end cap of the detector (cm),  $E_0 = 1$  keV,  $a_0(d)$ ,  $a_1(d)$ ,  $a_2(d)$ ,  $a_3(d)$ ,  $a_4$ ,  $a_5$  – parameters. The formula has been successfully tested on the existing experimental data on the energy-efficiency dependences for a wide range of distances between the sample and the surface of the detector.

## Natural radioactivity of evolving biogeochemical precipitate in thermal spring caves of Gellért Hill area, Budapest

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The high radioactivity of the springs of Gellért Hill area (Buda Thermal Karst, Hungary) is known since the early 1900's. The thermal springs of the Gellért, Rudas and Rác Spas are famous about their medicinal radon content. Despite the several hypotheses for the possible sources of <sup>222</sup>Rn, neither could be proved by measurements. It became clear, that the high <sup>222</sup>Rn concentration cannot be explained directly by the <sup>226</sup>Ra activity of the waters. The maximum values of <sup>226</sup>Ra concentrations do not overlap with the highest <sup>222</sup>Rn activities. Reddish brown iron hydroxides and different forms of carbonates (e.g. calcite rafts, carbonate mud) are the most abundant precipitates in recent spring caves of the Gellért Hill area. In the iron hydroxide precipitates bacterial species were found, therefore, these were handled as biofilm. Biofilms are known to adsorb radionuclides in hot springs in Japan, causing high radioactivity. In the Gellért Hill area, the radioactivity of biofilms in different spring caves was measured at several locations (Nagy Spring of Rác Spa, Török Spring and Diana Hygieia Spring of Rudas Spa, Ósforrás of Gellért Spa) to survey the distribution of radioactivity in biofilms. Furthermore, detailed investigation was made in Török Spring, where radioactivity was found to be the highest beside Diana Hygieia Spring. Mineral, biological precipitates and the host rock were measured by  $\gamma$ -spectrometry to screen the distribution of <sup>226</sup>Ra. Based on the findings an in situ experiment was carried out for a year in Török Spring to study the evolution of biofilm and the adsorbed <sup>226</sup>Ra activity in time. According to the results the iron hydroxide biofilm adsorbs <sup>226</sup>Ra in the highest concentration. It could be proved by the experiment, that the adsorption takes place parallel to the formation of biofilm. The highest <sup>222</sup>Rn activity of the waters does not overlap with the highest adsorbed <sup>226</sup>Ra activities, because of the characteristics of the spring caves. These were investigated by air measurements. The research was supported by the NK 101356 research grant.

## **Estimation of radon potential in towns around Saldanha Bay by modelling granite geology**

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The geology of an area can be used as a predictor for radon potential. Granite rock normally contains high concentrations of uranium and subsequent elevated emanation of radon gas. The geology in and around Saldanha Bay, South Africa, is dominated by granite rock outcrops and was consequently investigated for radon exhalation. Radon results from granite rich environments in India were employed to extract a relationship between radon gas and uranium concentrations. Uranium concentrations were then measured on a large granite hill in the Saldanha Bay area and the relationship between radon gas and uranium were used to model radon emanation for this outcrop. The results were then extrapolated to similar areas around the bay that are inhabited. Radon exhalation rates larger than 0.35 Bq/m<sup>2</sup>h were predicted. Estimated indoor radon concentrations in excess of 400 Bq/m<sup>3</sup> were finally predicted for certain neighbourhoods in the town of Saldanha.

## Contribution to the origin of the P-Mn-U-Be-HREE-enrichment in phosphatite, near Bükkszentkereszt, NE Hungary

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The U-Be-REE-bearing phosphate bodies are associated with Mn-oxe ore, found in strongly deformed phosphatite layers occurring in the microbrecciated crack zones of the weathered metarhyolite tuff of the Triassic Bagolyhegy Metarhyolite Formation. Rejuvenation of the volcanism (owing to the tectonics) brought submarine, hydrothermal exhalations and/or heated flus originating from alpine regional epimetamorphism. The microbrecciated zones and their surroundings were first silicified, and then phosphatised, resulting in the formation of Mn-ore with U-Be-REE contents; this was due to metasomatic replacements. The mineral composition in cream-colored bands are mostly quartz and some fluorapatite, while the dark bands are significantly enriched in fluorapatite with some quartz and a strongly variable Mn-oxe content in massive veins. Concerning main and trace element compositions the maximum concentrations are: MnO-44.6 wt. %; P<sub>2</sub>O<sub>5</sub>-35.3 wt. %; Fe<sub>2</sub>O<sub>3</sub>-3.1 wt. %; U-998 ppm; Be-1550 ppm; ΣREE-500 ppm; As and Zn-1400 ppm. The apatite contains the U-Be-HREE in inhomogenous dispersion, but the Be is enriched mostly in Mn-oxes. Recent study was made by high resolution optical microscopy, FTIR-ATR, and 1800 Raman spectra along a profile were elaborated. Observations show thin, Mn-oxe and Fe-oxe-hroxide bands, stromatolite-like, filamentous and cocco-like microstructures, which contain ferrihydrite and pyrite, and also variable embedded organic compounds. These structures can be interpreted as series of Fe-rich biomats. According to hypothesis the Bükkszentkereszt occurrence is a mineralized microbially mediated deposit, and the recent results support this scenario based on the mineralized microbially produced structures, their micromineralogy as they are built up by ferrihydrite, goethite (pyrite), variable manganese-oxes (cryptomelane, pyrolusite, manganite, birnessite, manjiorite), braunite, and rhodochrosite, apatite and embedded variable organic compounds. Also the P, Mn, U, Be, (As, Zn) are bioessential elements. As a summary, the rhyolite-hosted manganese ore mineralization bounded U-Be-phosphate enrichment can be proposed as a fossil mineralized microbially mediated deposit. Supported by NRDIO-OTKA 125060.



## **Cesium and $^{137}\text{Cs}$ distribution of urban geochemical samples in Salgótarján city, Hungary**

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Cesium is a rare and highly mobile and reactive alkali metal occurring in the environment mostly from erosion and weathering of rocks. Therefore, mining and milling of certain ore minerals can produce cesium into environment, particularly into air, water and soil. Cesium-137 an artificial radionuclide, which generates from atomic bomb tests and nuclear accidents (e.g., Chernobyl). This work focuses mostly on the spatial distribution of (total) cesium and  $^{137}\text{Cs}$  in attic dusts and urban soils collected in Salgótarján city. We also compare amount of cesium and  $^{137}\text{Cs}$  between the two environmental sample types, which can be used for origin determination of cesium within the study area. City of Salgótarján was divided into 38 1x1 km<sup>2</sup> cells from where 40 attic dust and 36 urban soil samples were collected. Sampling sites of attic dusts are divided into four groups: kindergardens (3 samples), family houses (30 samples), churches (6 samples) and blockhouses (2 samples). Attic dust samples were collected away from the entrance of the attics and at the highest possible point to minimize disturbing effects of recent activities. Urban soils were collected at playgrounds (11 samples), kindergardens (9 samples), parks (6 samples), along transport routes and train track (6 samples) and other open communal areas (4 samples) to aim human receptor. The total cesium concentration of the samples was analyzed by ICP-MS, whereas the  $^{137}\text{Cs}$  is measured by well-type HPGe detector for attic dusts and coaxial HPGe detector for soil samples. We found that the total cesium concentration of attic dust (1.93-3.32 g/t) was higher than in the soil samples (0.87-1.93 g/t). Two attic dust outliers (3.15 and 3.32 g/t, respectively) were observed in family houses built in the early 1920s. In addition, geochemical background sample from a forest soil, located 6 km from the city, shows higher total concentration (2.14 ppm) than urban soils. The spatial distribution of total cesium and  $^{137}\text{Cs}$  of attic dust and soil samples in Salgótarján city is used to explain geochemical and atmospheric origin and to distinguish natural and anthropogenic processes.

## Natural radioactivity and radon emanation coefficient in soil in Ninh Thuan Province, Vietnam

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The natural radioactivity (<sup>238</sup>U, <sup>226</sup>Ra, <sup>232</sup>Th and <sup>40</sup>K) and radon emanation coefficient of 85 soil samples were studied by gamma ray spectrometry and RAD7 detector. The average <sup>238</sup>U, <sup>232</sup>Th, <sup>226</sup>Ra, <sup>40</sup>K are 40.41±6.26 Bq.kg<sup>-1</sup>, 96.00±6.81 Bq.kg<sup>-1</sup>, 70.62±3.37 Bq.kg<sup>-1</sup>, 1127.16±93.64 Bq.kg<sup>-1</sup>, respectively and emanation coefficient was 0.32±0.02. Radionuclides <sup>238</sup>U and <sup>226</sup>Ra were found in disequilibrium with ratio of specific activities (<sup>238</sup>U/<sup>226</sup>Ra) from 0.2 to 0.9 for most of the samples. Radium equivalent for some of samples are higher than the allowed maximum value of 370 Bq.kg<sup>-1</sup>. The emanation coefficient does not strongly depend on the uranium and radium contents. For all of five classes of particle size, the emanation coefficient increases with moisture content and reaches a constant value with a different moisture content. The constant values for less than 0.1 μm, (0.1 to 0.2) μm, (0.2 to 0.3) μm, (0.3 to 0.5) μm (0.5 to 0.8) μm of grain sizes was 0.59, 0.46, 0.35, 0.25 and 0.21, respectively with moisture contents are 16%, 14%, 10%, 6% and 4% in the order above. There was a trend for the radon emanation coefficient to drastically increase with the decrease and increase in particle size and moisture content, respectively. The radon emanation coefficient is almost unchanged with grain size for the large particle sizes.

## **High indoor radon concentration observed in Okinawa, southwestern part of Japan**

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Short-term and annual measurements of indoor radon concentration were conducted from 2006 in dwellings in Yomitan-son, a village of Okinawa prefecture located in subtropical region of Japan, in which the highest annual average of indoor radon concentration (220 Bq m<sup>-3</sup>) has been observed by a nationwide survey at the end of 20th century. Also in a private house in Yomitan-son, anomalously high radon concentrations (400-1000 Bq m<sup>-3</sup>) with an active radon monitor (AlphaGUARD) were reported in 2009. In this study, to estimate the cause of high radon concentration, measurements of indoor radon were made with passive monitor (RADUET) and analyses for the natural radionuclides in soils were performed with a Ge semiconductor detector. The seasonal variation that the indoor radon concentration in winter is obviously higher than that in summer was observed in several dwellings by the annual measurement. The highest concentration, 211 Bq m<sup>-3</sup>, was observed by the short-term measurement during winter season in a private house that is different one in the nationwide survey. The results for the soil analyses suggested that the radon source is the red soils distributed widely over Okinawa.

## **Geogenic radon potential assessment based on geochemical and physical soil properties in a granitic area, Hungary**

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Terrestrial radiation is the most important source of natural radioactivity we are exposed to. Hence, it has a high importance to define the role of physical and chemical soil properties on geogenic radon, especially in areas nearby settlements. It is known that high terrestrial natural radiation levels are associated with acidic igneous rocks such as granite due to its formational process. It leads to elevated concentration of uranium, radium and thorium in certain silicate and phosphate minerals. This study was performed in the western se of Velence Hills in Hungary. The goal of the study was to use and test the usability of the we range of the soil geochemical and physical properties as independent input parameters into predictive natural radioactivity models. In order to avo the influence of the structural features such as dykes and tectonic faults, a homogeneous re-deposited slope deposits formation area was selected, surrounded by Velence granite formation. To check the theoretical models, field measured soil gas radon activity concentration and soil gas permeability was compared to the model calculations. These models use the laboratory measured radium content, radon emanation coefficient and soil physical properties, (e.g. dry bulk density, porosity, arithmetic mean particle diameter and water content of soil samples). Effects of additional geochemical properties, such as clay mineral, organic matter and carbonate content, were also studied. Measurements and soil sampling were carried out in three replicates at 30 randomly selected sites on a 6640 m<sup>2</sup> areas. Magnitude of the soil gas radon concentration at the area was in the range of 13.4 to 137 kBq m<sup>-3</sup> with an average of 31.7 kBq m<sup>-3</sup> and a median of 27 kBq m<sup>-3</sup>. Geogenic radon potential (GRP) was also calculated for each measured site from soil gas radon activity concentration and soil gas permeability. Therefore, the relationship between all of the studied geochemical and physical soil parameters and GRP will be also discussed.

## **An extensive indoor radon survey – challenges, experience and first results**

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According to the new EU BSS all member states shall establish a national action plan addressing long-term risks from radon exposure and have to identify areas where the radon concentration in a significant number of buildings is expected to exceed the relevant national reference level. Identifying such areas will be fundamental for establishing legislation and future strategies. For this purpose an extensive indoor radon measurement campaign was designed in Austria with planned radon measurements in about 35.000 dwellings. The measurements are carried out in the houses of selected members of the voluntary fire brigades for 6 months. The measurement locations are selected based on a regular 2x2 km grid, taking geology into account. Because of resources and administrative issues, the campaign is carried out over several years and is done province by province. Until now, measurements in 3 provinces are finished, in 4 provinces the measurements are under way. In the contribution the design of the survey as well as the experience and challenges with the planning and conduction of such an extensive survey will be presented. Possible impacts for the success of a survey (return rate, quality of results) will be evaluated. In addition the first results and the processing of the data are discussed - addressing effects as representativeness of the measurements, influences of different building characteristics, differences in the provinces (housing stock, geology etc.) and their impact on the results and the modelling. The discussions will be based on the results and experiences of the surveys in the already measured provinces.

## **The impact of geographic factors on the distribution of natural terrestrial radionuclides on Baviaansberg in the Saldanha Bay military area of South Africa**

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The most significant naturally occurring radionuclides are uranium ( $^{238}\text{U}$ ), thorium ( $^{232}\text{Th}$ ) and potassium ( $^{40}\text{K}$ ). These nuclides are of primordial origin and they all constantly emit gamma radiation that is readily detectable by means of radiation detectors. Naturally occurring radionuclide concentrations were measured in a relatively undisturbed site named Baviaansberg and radioelement maps were constructed. These radioelement maps were used to establish the influence of local geographic factors on the distribution patterns of the natural nuclides. The Baviaansberg outcrop is situated in the Saldanha Bay Military Area on the West Coast of South Africa and was formed by underlying granite bedrock. Radioelement maps were used to assess the relationship between the distribution patterns of the natural nuclides and geology, topography, soil type, hydrology, wind regime, and human activities. Radiation measurements were executed by means of in situ measurements using a NaI(Tl) scintillation detector that was connected to a tablet computer. Radioelement maps were created once the nuclide concentrations over the total area were extracted. Qualitative analysis was performed on the radioelement maps by comparing it to the slope rasters, slope profiles, flow accumulation rasters, wind data, Google Earth images, human activities and soil type maps. Results indicated relationships between the distribution of natural nuclides and geology, topography, soil type, hydrology, and wind.

## **Determination of the activity and average annual dose of absorbed actines in drinking water from Warsaw**

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The specific activity of <sup>210</sup>Po, <sup>234</sup>U and <sup>238</sup>U was determined for samples of drinking water from Warsaw metropolitan area. The most important source of drinking water in Warsaw is tap water coming from surface and sub-surface intakes. Other source is water from deeper intakes, which is mainly from the level of Oligocene sands. In the area of Warsaw are about 200 such intakes, half of which are in continuous use and are available to the residents of the capital. Annual monitoring was done for selected surface and deep intakes. For other intakes the samples were collected once during study period. Measurements of uranium and polonium isotopes activity in collected water samples were performed using an alpha spectrometer (ORTEC DUO). Spatial and temporal changes of activity was recognised. The mean values of <sup>210</sup>Po, <sup>234</sup>U and <sup>238</sup>U in surface intakes were 0.13, 3.91 and 2.61 (mBq dm<sup>-3</sup>) and for deep water intakes these were 0.60, 0.24 and 0.32 (mBq dm<sup>-3</sup>), respectively. The annual dose absorbed as a result of the consumption of drinking water by a statistical inhabitant of Warsaw was calculated on the basis of measurements of specific activity of polonium and uranium. The calculations showed a difference in the value of the annual dose absorbed depending on the source of drinking water supply. It was calculated that the annual absorbed dose from water is only a negligible part (about 0.01%) of the dose taken from all sources, the value of which is estimated at 2.8 mSv year<sup>-1</sup>. The source of the water consumed has no significant effect on the amount of radiation absorbed.

## Determination of the thoron emanation coefficient using a powder sandwich technique

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Thoron ( $^{220}\text{Rn}$ ) is a natural radioactive gas, tasteless, odourless, colourless, undetectable without proper equipment. This gas is carcinogenic, just like radon ( $^{222}\text{Rn}$ ) but due to the short half-life (55.6s) and a small amount in the environment, its share in the absorbed radiation dose is often neglected. However, in areas rich in thorium ( $^{232}\text{Th}$ ), the radiation dose from the thoron can be much larger and quite significant. The problem is to measure the concentration of the thoron due to its short decay time as well as the fact that it is alpha-emitting as radon. An even greater challenge is to determine the emanation coefficient for the thoron. The method used in this experiment was developed by S.D. Kanse based on the work of D.J Greeman and adapted to the equipment used in Laboratory of Radiometric Expertise IFJ PAN. In the technique used to determine the thoron emanation coefficient, a closed loop system is used in which thoron is pushed out by means of a flow system from the sample and measured by a AlphaGuard DF2000 detector that is adapted to determine concentration of this gas. A sample of the material is placed between 2 filters in the geometry of the sandwich. This arrangement ensures that the thickness of the powder sample is significantly less than the length of the thoron diffusion, thus avoiding significant loss of the thoron due to intergranular absorption and facilitates the complete removal of this gas escaping from the powder. Using this technique, it is important to determine the concentration of  $^{226}\text{Ra}$  and the  $^{232}\text{Th}$ , since for the AlphaGuard detector, the ratio between thoron and radon should not exceed 5:1 for proper determination of the thoron concentration. Measurements of  $^{226}\text{Ra}$  and  $^{232}\text{Th}$  activity were carried out using gamma spectroscopy (HPGe detector) and were  $^{226}\text{Ra}$ :  $34\,795 \pm 204$  Bq/kg (according to the  $^{214}\text{Bi}$  isotope) and  $^{232}\text{Th}$ :  $93\,180 \pm 889$  Bq/kg (according to the  $^{228}\text{Ac}$  isotope). The emanation factor calculated on the basis of the results obtained from the experiment was 0,146, it is comparable to the result obtained by means of another calculation method, which was 0,149. It was examined how the type of filter and humidity affects the obtained results. The air flow rate in the system does not affect the emanation factor.



## **Interlaboratory comparison of H-3 and C-14 activity determination in water**

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The RadiÖko Ltd. radioanalytical laboratory and Laboratory for low-level radioactivities of the Ruđer Bošković Institute (RBI) performed an interlaboratory comparison for the determination of H-3 and C-14 activities in water samples. Three samples were measured for H-3, two for C-14 activity. Each laboratory applied their own sample preparation techniques for H-3 determination and the activities were measured by LSC Quantulus 1220. At the RBI the low H-3 activity sample A was measured by both direct and electrolytic enrichment method. For the determination of the radiocarbon activity the Zagreb laboratory used AMS technique, whilst RadiÖko used an own developed sample oxizer followed by LSC measurement using a Quantulus LSC. It is interesting to note that all three H-3 results of the RadiÖko group are lower than those obtained at RBI. The results of the two radiocarbon measurements reveal that in the case of sample D the measurement results of the laboratories lie close to each other. Sample E had too high activity to be determined by AMS technique, this way it is not possible to compare the results. In all the successfully conducted cases it is apparent that the differences are well below the 10% satisfactory range.

## **An approach to compare personal, at home and at workplaces radon exposures**

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Population's exposure to radon gas is estimated, in general, using data from surveys carried out at home or/and at workplaces, mostly in radon affected areas, considering fixed exposure times. This approach brings several uncertainties. A study has been organized thanks to 48 volunteers: 35 participants were from Russia, the others from Austria, Belgium, Germany, Hungary, Italy, Japan, Republic of Serbia, Slovenia, Spain and Sweden. Each participant received 5 CR-39 track detectors and was asked to record hours spent at home and at workplace daily. Three sets of radon data were obtained: dwellings, workplaces and personal radon concentrations. For the group of volunteers involved in the study (active working population) the average proportion of time spent at home is 0.55, significantly less than the recommended value (0.7–0.8). The comparison of radon in dwellings with "personal" radon concentration was proved. The ratio Cp/Cd is well described by lognormal distribution with average of 1.05, GM of 0.90 and GSD of 1.70. The multiplicative uncertainty due to radon exposure in other places than dwelling was estimated as  $\sigma=0.55$  (CI 0.43–0.74). A 50% contribution of occupational exposure or higher was observed in 25% of participants. Even if radon measurements performed simultaneously in dwellings and workplaces, the accuracy of the calculated exposure compared with the measured personal exposure remains low. It can be explained both by an unidentified exposure to radon outside home and workplace and by the uncertainty due to daily radon variations in rooms.

## **Hungarian Radon Action Plan**

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A big change could be observed in the field of recommendations for radiation protection in the last 10 years. Much larger attention was put to the exposition from natural sources. This approach was adopted by the European Union and the Council reviewed the European Basic Safety Standard (EU BSS). The new EU BSS (2013/59/EURATOM Directive) was published at the end of 2013, which has to be transposed to the national legislation until February of 2018. According to the directive, all Member States shall establish a national Radon Action Plan (RAP). The directive regulates the minimum content of the RAP. Its main goals shall be the evaluation of the exposition of general public from the natural isotopes (especially from radon gas), to inform the public about the estimated level of its health risk and set up and apply practices to minimize their radiation burden. In Hungary, the National Public Health Center initiated collaboration between the radon experts and decision makers to support the preparation of the RAP in 2015. After the assessment of the actual situation, it was obvious it is needed to make a new national radon survey, to set up a national data base, to prepare a communication campaign, to adopt radon prevention and remediation technics and to develop the legislation framework. At first of January in 2016 steps into force the 487/2015. Govt. decree setting up reference level (300 Bq m<sup>-3</sup>) for indoor radon concentration both at workplaces and in homes. The RAP was finalised during the Autumn in 2017. At the end of November was published the 1862/2017. Govt. decree regulating the tasks, deadlines and responsible ministries defined in the first Hungarian RAP.

## **Radon metrology as part of measurement quality assurance in Germany - Developments in the radon calibration laboratory**

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Since the implementation of the EU Basic safety standards (BSS) into German law the Federal Office for Radiation Protection BfS is responsible for the quality assurance of radon measurements for work places and dwellings in Germany. One essential part of this responsibility is to assure that measuring institutions are able to prove the traceability of the radon activity concentration to national metrological standards and to be prepared for future requirements and questions, e.g., measurements of potential alpha energy concentration (PAEC) or thoron. For this purpose the BfS Radon Calibration Laboratory offers calibration services accredited according to ISO 17025 as well as scientific support, further development of measuring methods and international collaborations in the field of radon metrology. The talk will focus on developments and changes for the work of the Radon Calibration Laboratory due to the new German Radiation Protection Act and the new situation of radon metrology in Germany. Radon gas, radon decay product and thoron calibrations will be addressed.

## Natural radioactive isotopes in soil of Kharkov region

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Dynamics of migration of radioactive isotopes of the soil of the Kharkiv area (Ukraine) is studied. A migratory series of radioactive elements (Ra > U > Th) corresponds to the humid climate. High mobility of radium in relation to uranium and thorium is found out. The high correlation between <sup>40</sup>K versus <sup>232</sup>Th allows asserting, that thorium and potassium are a part of parent minerals because of the low mobility of thorium. Measurement of the age of soil is obtained. The registration method  $\alpha$ -radiations from radionuclides uranium family in the assumption the equilibrium of <sup>226</sup>Ra - (<sup>214</sup>Pb - <sup>214</sup>Bi) and calculation of <sup>238</sup>U content through the contribution <sup>235</sup>U in 186 keV  $\alpha$  line gives the value of age about 15 thousand years.

## **The concentration of $^{234}\text{U}$ , $^{238}\text{U}$ , $^{226}\text{Ra}$ and $^{228}\text{Ra}$ in the water samples in Kim Boi, Hoa Binh and drinking water in Vietnam**

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Uranium and radium isotopes ( $^{234}\text{U}$ ,  $^{238}\text{U}$ ,  $^{226}\text{Ra}$ ,  $^{228}\text{Ra}$ ) were measured in 17 water samples (6 samples of groundwater, 3 samples of surface water and 8 samples of drinking water in Vietnam). The groundwater and surface water samples were taken in Kim Boi district, Hoa Binh province where contains sandstone, gravel, and limestone. The radioactivity of these samples are quite low, varied from 0.19 to 8.95 mBq/L, from 1.60 to 18.63 mBq/L, from 3.00 to 29.20 mBq/L and from 4.30 to 12.80 mBq/L for  $^{238}\text{U}$ ,  $^{234}\text{U}$ ,  $^{226}\text{Ra}$ , and  $^{228}\text{Ra}$  respectively. Meanwhile, the drinking water samples are famous brands on the market and the radioactivity of drinking water samples distributed in a we range, for example,  $^{226}\text{Ra}$  fluctuated between the lower limit of detection to 202.10 mBq/L. The samples that have the highest concentration of uranium and radium isotopes were taken at the highest depth or from the confined aquifer. The result shows the evence for the correlation between the concentration of radium isotopes and the sampling depth.

## **Evaluation of radon suppression in low background gamma-ray spectroscopy: Monte Carlo simulation approach**

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This study proposes the approach to remove the contribution of  $^{222}\text{Rn}$  from the measured gamma-ray spectra, which achieved from a HPGe spectrometry. Several scenarios of flushing the central detector volume with nitrogen gas to actively remove the radon isotopes were simulated to evaluate the effect of Radon on the low-background gamma-ray spectra. To quantify the gamma background a set-up was prepared, the MCNPX code was adopted to evaluate the scenarios will be applied to the gamma-ray spectroscopy system consisting of a coaxial ORTEC HPGe detector (model number - GMX40-76) and the energy of events up to 3 MeV at the NAA lab (KAERI, Republic of Korea).  $^{222}\text{Rn}$  in the air around the detector contributes significantly to the remaining background and attention will be pa to the radon concentration, especially inse the sample chamber. The optimal configuration based on the simulations presented that a factor 4 improvement on the daughter decay peaks of  $^{222}\text{Rn}$  with a ~30% reduction again in the integrated rate between 100-2700 keV to  $0.751\pm 0.001$ , and translating to improved sensitivity of the detector for the  $^{238}\text{U}$  lines. The results also show a slight reduction in the 238 keV energy peak, more than expected from purging radon purely. This could be as a result of thoron reduction inse the main detector volume, which could explain why only the 238 keV peak was reduced and not the 338 and 911 keV peaks. Therefore, it can be concluded that the HPGe spectroscopy system at the NAA lab is fully operational and capable of making reliable measurements of sample activity with competitive sensitivity. It is now ready for use in material screening and selection for the environmental experiment.

## **A fully automated gamma-ray spectrometer for NORMs characterization**

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The large production of naturally-occurring radioactive material (NORM) represent a severe radiological hazard for health and for this reason has to be subject to regulation. We designed, built-up and tested an ad-hoc laboratory instrument, the MCA\_Rad, dedicated to the quantification of the specific activity of <sup>40</sup>K and several radioisotope of <sup>238</sup>U and <sup>232</sup>Th decay chains. MCA\_Rad is a high-resolution gamma-ray spectrometer system made of two 60% relative efficiency coaxial p-type HPGe detectors, with an energy resolution of ~1.9 keV at 1332.5 keV (<sup>60</sup>Co). The detectors are lead and copper shielded for reaching a low background, positioned facing each other and cooled at approximately -190 °C by electromechanical stirling cryocoolers. The instrument is equipped with an automatic sample changer, which allows managing independently the measurement of 24 samples without any human intervention. The material is contained in a small cylindrical polycarbonate box of 180 cm<sup>3</sup> of useful volume and labeled by a barcode that uniquely entifies the sample. A dedicated software has been developed in order to automatically manage the acquisition and the spectral analysis: sum of the spectra, background subtraction, energy calibration and abundances estimation. The fully energy calibration of the detectors of the MCA\_Rad was performed using certified reference materials of natural origin. The chemical composition and density were conserved in order to account for different photon attenuation within the source material itself. In our approach the correction factor for self-absorption effect was determined via Monte Carlo simulation. The overall uncertainty about the absolute efficiency of the MCA\_Rad system is estimated to be <5 %. The MCA\_Rad allows for checking the secular equilibrium among the U and Th decay chains segments. This makes this instrument particularly suitable for the characterization of the NORMs which are, respect to a typical rock or soil sample, more subject to several chemical and physical processes that disturb the secular equilibrium. Moreover the overall uncertainty about the absolute efficiency of estimated to be <5 % and the full automation of the experimental setup, represent adding values for the accurate and of the systematic characterization of a large number of samples and in turn for the regulation of the NORMs.



## **In situ experimental study of natural radioactivity of microbiological and chemical precipitates in a flowing thermal water**

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The high <sup>226</sup>Ra adsorbing capacity of biofilms causing high radioactivity of thermal springs is known in Japan, in Switzerland and in the Buda Thermal Karst, Hungary. This naturally revealed ability of biofilms can be a useful tool in bioremediation. However, the time factor, ie. the development of radioactivity of biofilms in the function of time, in controlled circumstances is challenging to study in natural environments. Therefore, a 12-week-long in situ experiment was carried out in the tunnel of Gellért Hill, Buda Thermal Karst, Hungary. Thermal water was pumped with constant volume discharge into a 120-m-long trapezoid canal. Glass slides were put into the canal at certain distances from the outflow to study the formation and evolution of the precipitates. The physicochemical parameters of the water (temperature, specific electric conductivity, pH, dissolved oxygen content) and activity of radionuclides (<sup>226</sup>Ra, <sup>238+234</sup>U, <sup>222</sup>Rn) in the water were measured three times, at the beginning, at half-time and at the end of the experiment. The detailed analysis of the evolved precipitates (study by stereo microscope, transmitted light microscope, scanning electron microscope and X-ray powder diffraction), together with the measurement of the radioactivity (by  $\gamma$ -spectroscopy) was made twice, at half-time and at the end of the experiment. At the first ~10 m from the thermal water outflow, intense red coloured, amorphous iron-oxyhydroxide precipitate formed. Further away light red, then yellowish grey calcite with rhombohedral and dendrite calcite crystals evolved. The red precipitate turned out to be biofilm. Relatively high <sup>226</sup>Ra activity could be measured in it compared to the carbonates. However, it turned out that calcite also has elevated radioactivity. This adsorbed <sup>226</sup>Ra supply the <sup>222</sup>Rn activity of the water, which continuously decreases along the flow path due to degassing. The research was supported by the European Union and the State of Hungary, co-financed by the European Regional Development Fund in the project of GINOP-2.3.2.-15-2016-00009 'ICER'.

## **From the European indoor radon concentration map to a European indoor radon dose map**

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The European Indoor Radon Concentration Map, part of the European Atlas of Natural Radiation [1], displays the geographical distribution of long-term mean indoor radon (Rn; <sup>222</sup>Rn) concentration in ground floor living rooms. This definition has been chosen after some discussions when the project was initiated in 2006, because it was thought that sufficient and representative data from as many European countries as possible were available only for ground floor. Individual data are aggregated into 10 km×10 km cells by participant countries and cell statistics (number of data, arithmetic mean and standard deviation, AM and SD of ln-transformed data, median, minimum, maximum) communicated to the JRC. There the results are collected, checked for errors and mapped. A map which displays doses resulting from indoor Rn would be at least equally relevant. The main challenge consists in the fact that most people, in particular in cities, do not live in ground floor dwellings; as indoor Rn concentration usually decreases with increasing floor level, a dose map based solely on ground floor data would lead to overestimation of dose. In this study we investigate the possibility of a model-based regionalized dose estimation since a representative European indoor Rn data is still not available. The logic is planned as follows. (1) We assume that in average, the frequency of dwellings in higher floor levels is higher in cities than in the countryside. As proxy to decide whether one particular cell of the indoor Rn database (we do not have the individual data for reasons of data protection) belongs to a city or to countryside, we use population density in that cell, which is available Europe wide. (2) We establish a model of frequency of floor levels, in dependence of population density, from regional datasets which are population representative (3) We apply "floor correction" to each cell in accordance to the floor distribution derived from the model which relates it to population density. Also the correction is derived from empirical data. Thus we get a floor-corrected version of the indoor Rn map. (4) Finally, we use recent dose conversion factors to estimate doses. The procedure must be tested for consistency and plausibility by common statistical methods such as data partition and bootstrap. At this first experimental stage, we do not attempt to produce an authoritative European indoor Rn dose map. After all, it may be that the correction models underlie themselves a regional trend, possibly due to climatic or cultural factors. This can only be decided by applying the procedure to different regional datasets.

[1] <https://remon.jrc.ec.europa.eu/About/Atlas-of-Natural-Radiation>

## **Research testing of a new IAQ real time monitoring system (radon, CO<sub>2</sub>, CO, VOC, T, P, rH)**

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Indoor air quality (IAQ) is the general concern of the present times, both for the general public and for researchers. The European Council Directive 2013/59/EURATOM (EU-BSS), which is currently implemented in EU member states, has led to a significant progress in the development of the metrological instruments across Europe in the field of radon. Multiple IAQ systems are available on the market but only a handful proving the level of radioactive pollutants of the indoor environment. The “Constantin Cosma” Radon Laboratory (LiRaCC) from Babeş-Bolyai University has developed in the frame of SMART\_RAD\_EN project an IAQ monitoring system equipped with Radon, CO<sub>2</sub>, CO, VOC, T, P, rH sensors, which can actively monitor indoor air quality and, if necessary, activate a ventilation system. Due to the active logging of data, real time remediation can be applied. An internal calibration program to assess the effectiveness, precision, accuracy and stability of the device was started. All 100 prototypes build at the LiRaCC are currently being calibrated in the laboratory and tested in different indoor environments. The traceability valuation and the quality assurance of the prototype system will be carried out within accredited European laboratories. In this paper are being presented the results of testing and comparisons between prototype units and reference equipment in order to evaluate the quality of radon measurements.

Acknowledgement: The research is supported by the project P\_37\_229, Contract No. 22/01.09.2016, with the title „Smart Systems for Public Safety through Control and Mitigation of Residential Radon linked with Energy Efficiency Optimization of Buildings in Romanian Major Urban Agglomerations SMART-RAD-EN” of the POC Program.

## **Measurement of terrestrial radiation level in a neotectonic fault system in Trinidad**

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The relations between geological fault systems and increased terrestrial radiation level was investigated by many researchers in the world, and still is being investigated due to the complex nature of the work and unavailability of suitable locations that can prove continuity in data collection for possible earthquake forecasting. Present study area, which is located in the Central Range Fault (CRF) system in the island of Trinidad, can prove solution to that problem as this strike-slip fault system is reported to be active in present time (during Holocene, most current geological epoch) with a movement of geodetic rate of 9-15 mm/y. If this rate is typical of last several thousand years, then for a lapsed period of >550 years (time since no large major earthquake occurred in this fault system) and the accumulated strain energy equivalent of 9.4 m slip can produce earthquakes of  $M > 7$ . Moreover, the selected fault system is accompanied by mud volcanoes, which can amplify the level of radiation activity by slight but significant anomalies as described by some researchers from other areas. Furthermore, being one of the first environmental radiation study in the Caribbean Islands, this study is significant. With the aim to achieve a continuous monitoring of radiological data, objectives of this preliminary investigation are as follows: (a) to investigate the possible anomalies in radiation level in the fault system, (b) to correlate the relation between the radiation level and the controlling physical properties of soil and rock, and finally (c) to locate the most suitable geological section within this neotectonic fault system for the recording of least biased radiation level for possible earthquake prediction. The measurement points are selected based on a regular 2x2 km, taking accessibility, terrain and vegetation into account. As a pilot study, radiation measurements were carried out with the help of a portable Geiger Muller counter in 50 locations along the 25km long section of CRF between Naved Dam and Pointe-a-Pierre. Radiation measurements were taken both at one meter level above the surface and at nearly one meter below the ground. Results from this pilot study showed positive anomalies in terrestrial radiation in the fault system (2-3 times higher than the average level found in other parts of the island) and reaffirmed the recent tectonic activities. Authors also found that radiation values increase at subsurface levels (nearly 2 times higher than the surficial values). In this contribution, the first set of results of these evaluations will be presented and discussed, as well as the relations of these values with the controlling factors will explained.

## **Spatial and temporal variations of radon gas in dry carbon dioxide spas**

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Upwelling carbon dioxide in post volcanic activity areas are often used as therapeutic agent in the form of dry carbon dioxide spas to cure patients suffering from vasoconstriction and some other diseases. Radon gas is also present in the gas that is seeping out from the ground. This radon gas may be used as a natural tracer to follow the sources and transport of carrier carbon dioxide. It also poses some risk, especially for the staff of such spas, associated with the radiation exposure from the inhaled short lived daughters of the gas. Therefore, it is both useful and necessary to monitor the spatial and temporal variations of radon gas in such carbon dioxide spas. In this work we have used active and passive radon monitors to obtain Rn-222 activity concentration data with high spatial and temporal resolutions. We have also monitored the weather with automated weather stations. Experimental data showed significant spatial and temporal variations at locations relevant for patients and staff. We have done model calculations to describe the transport of carbon dioxide and radon gas in the pools and in the connected rooms of carbon dioxide spas. Based on these model calculations we have designed optimized ventilation system in and around the pools which ensures high carbon dioxide concentration for treatment at lower body parts but guarantees low carbon dioxide and radon gas concentrations at inspiration levels.

## Indoor radon mapping in Romania from large-scale to small-scale

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Systematic data for indoor radon measurements in Romania were obtained since 2006, through research projects implemented by Babeş-Bolyai University. The main purpose of the present paper is to display the current results of the annual average indoor radon concentration in Romania, based on approximately 6000 radon measurements performed by using nuclear track detectors CR-39, as an update to the preliminary Romanian Indoor Radon Map. In 2018 the large scale mapping (10x10 km cells) was updated with 1000 new data on a small-scale (1x1 km cells) for five major agglomerations with a high density of population and settlements (Bucharest, Cluj-Napoca, Iaşi, Sibiu and Timișoara). The measurements were completed within SMART\_RAD\_EN European Project. The qualitative evaluation of the indoor air quality, the variability of the measured data between all the surveyed regions, the correlation with the local geology, as well as the relationship between radon-related quantities was investigated and presented. The preliminary results reveal that in about 11% of the investigated houses the radon concentration exceed the recommended level in Romania, of 300 Bq m<sup>-3</sup>. The results will contribute to the further implementation of the European Council Directive 2013/59/EURATOM (EU-BSS) in Romania.

Acknowledgement: The research is supported by the project P\_37\_229, Contract No. 22/01.09.2016, with the title „Smart Systems for Public Safety through Control and Mitigation of Residential Radon linked with Energy Efficiency Optimization of Buildings in Romanian Major Urban Agglomerations SMART-RAD-EN” of the POC Programme.

## **Radionuclides in groundwater flow systems – case studies from drinking water supply systems in Hungary**

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Uranium, radium and radon are usually found in groundwater. Using these radionuclides is a novel approach to characterize flow of groundwater flow systems and understand mixing processes. Particularly, in regional discharge areas, where different order flow systems convey waters with different temperature, composition and redox-state to the discharge zone. Radium and uranium are redox-sensitive parameters, which causes fractionation along groundwater flow paths. Discharging waters of regional flow systems are characterized by elevated total dissolved solid content (TDS), temperature, reducing conditions, and therefore with negligible uranium content. Whereas local flow systems have lower TDS and temperature and represent oxidizing environments, and therefore their radium content is low. Due to the short transit time, radon may appear in local systems' discharge, where its source is the soil zone. However, our studies revealed the importance of FeOOH precipitates as local radon sources throughout the adsorption of radium, which is transported by the waters of regional flow systems. These precipitates can either form by direct oxidizing of waters at the discharge zone, or by mixing of waters with different redox state. Therefore elevated radon content often occurs in regional discharge areas as well. Since 2016 the measurement of radioactivity of springs and wells is compulsory in groundwater monitoring in those cases, when they are used for drinking water supply. In our study we present case studies from Hungary, where the understanding of groundwater flow systems and the hydrogeological environment helped to explain the occurrence of radionuclides in these systems. This study was supported by the ÚNKP-17-4 New National Excellence Program of the Ministry of Human Capacities.

## Contribution of thoron to the inhalation dose in Angolan adobe houses

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As known, radon has always been considered to be the main contributor to the inhalation dose indoors. Thoron in contrast has always been ignored due to its short half-life (55s). It is true that there is not enough time for thoron to accumulate indoors if it comes from the soil. However, for buildings made of porous building materials like mud and adobe houses, thoron plays a significant role in the inhalation dose. In Hungary for example, a study made in adobe houses showed a contribution of thoron of at least 30% to the total inhalation dose. In Central-Chinese Loess Plateau, the contribution of thoron was determined to be up to 50% in earthen houses. In Germany thoron progeny concentrations contributed to about 4 mSv/y in same kind of houses. In Angola, 45 houses have been monitored within this study for radon and thoron with etched track passive detectors for a duration of one year divided into two seasons (rainy and dry) in 3 different areas, Cabinda, Huambo and Menongue at the North, Central and South parts of the country, respectively. The average concentrations in Bq/m<sup>3</sup> at the respective mentioned localities were: 29±8, 99±43 and 57±18 for radon and 153±96, 248±147 and 106±74 for thoron. Annual inhalation doses in mSv/y corresponded to 0.7, 2.5 and 1.4 from radon and 1.7, 2.8 and 1.2 from thoron. The inhalation doses of both radon isotopes are higher in the central part of the country. In this study the contribution of thoron to the inhalation dose is up to 52%. All the values apart from radon in Cabinda, are considerably high if compared to the world average, which is 1.15 mSv/y.



## **Investigation of the fingerprint of climate changes in Apa Roşie peat bog (central Romania) by using $^{210}\text{Pb}$ dating method (preliminary results)**

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The Apa Roşie (Veresvíz) peat bog is a protected area in the north-eastern part of Covasna county, in the Nemere Mountains, along the Apa Roşie (Veresvíz) stream at 990-1030 m altitude. It is a natural area (deciduous forests, coniferous forests, mixed forests, transition forests, bushes, hedges, meadows, peatlands and marshlands) framed in the alpine bioregion of the southern slope of the Nemere Mountains (group of the Carpathians' Moldo-Transylvanian region, belonging to the Oriental chain of the Carpathians). The region was chosen as the subject of our research as it contains all the necessary characteristics of our study: isolation, habitats in good conservation status can be found here, forests, grasslands and marshy areas are in good condition. In peat formation systems, small changes in external factors may cause large alterations in the functioning of the ecosystem, making these areas sensitive indicators of past as well as recent climatic changes. Bogs are sensitive to climate condition changes. The changes of temperature and precipitations lead to slower or faster peat mass accumulation. Longer and warmer growing seasons could also be playing a key role in the recently observed increase in the mass of peat bog. The object of our research is how these longer and warmer periods stimulate the accumulation rate of peat bog production.

The grow rate of the peat was determined by measuring the mass accumulation of the organic matter per year. The age of each investigated layer was determined by using the  $^{210}\text{Pb}$  dating method. The activity concentration  $^{210}\text{Pb}$  was measured by gamma spectrometry using a Well –type high-purity germanium (HPGe) detector. Additionally some of the samples was measured by alpha spectrometry (through  $^{210}\text{Po}$ ) for valation the gamma spectrometric measurements.

## **Production of glass-ceramics from NORM material**

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Along with the gradually increasing of the quantity of the coal fly ash as residue containing NORM (Naturally Occurring Radioactive Material) appropriate management and treatment have become an urgent environmental protection problem. Glass-ceramics materials are prepared by controlled crystallization of glass. The physical, chemical and radiological properties make the coal fly ash an adequate potential material for production of glass-ceramics. The aim of the study was to investigate the possibility of production of glass-ceramics from fly ash obtained from the thermal power plant REK Bitola in R. of Macedonia. The crystallization mechanism was investigated by differential thermal analysis (DTA), X-ray diffraction (XRD) and scanning electron microscopy (SEM). The glass transition temperature ( $T_g$ ) and peak temperature ( $T_p$ ) obtained by DTA were 580°C and 1020°C, respectively. The crystallization tendency of the parent glass estimated by Hruby coefficient obtained by DTA curve was 2.3. X-ray diffraction patterns show that the major crystalline phases were calcium aluminum silicate (anorthite) and hematite. SEM analysis confirmed that the crystals were homogeneously dispersed within the parent glass. Physical and mechanical properties of the glass-ceramics such as density, porosity, linear shrinkage, bending strength and E-modulus have been reported and discussed.

## Using total gamma radiation to assess soil water content

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The soil water content is a fundamental parameter linking the interaction between the land surface and the atmosphere. Despite its relevance for both scientific (e.g. climate) and practical (e.g. agriculture) applications, soil moisture is difficult to assess, particularly at small spatial scales and short temporal resolutions. Gamma radiation emitted by terrestrial radioisotopes (K, U, Th) is largely determined by the water content of the soil, which on one hand attenuates the propagation of gamma rays in the subsurface, and on the other influences the mobility of radon gas and its subsequent exhalation to the atmosphere. Therefore gamma radiation is a potentially useful indicator of soil water content. However, in order to be able to use low-cost and simple total counting measurements of gamma radiation at ground level as a proxy of soil moisture, all the other influencing factors need to be assessed, including space (cosmic rays), atmosphere (aerosols, precipitation) and soil (radon gas) contributions. Disentangling the multiple interacting factors is a challenging task that requires both high-resolution and continuous measurements of gamma radiation as well as high-resolution and co-located measurements of soil and meteorological parameters. The gamma radiation monitoring campaign at the Eastern North Atlantic (ENA) facility in the Graciosa island (Azores) was set-up in order to take advantage of the detailed meteorological and atmospheric information available from ENA, a permanent site of the Atmospheric Radiation Measurement (ARM) program. Gamma radiation is measured at the ENA facility every 15-minutes since May 2015 using a NaI(Tl) scintillator counting all gamma rays in the range from 475 KeV to 3 MeV. The resulting time series of total gamma counts displays a complex temporal pattern conveying the multiple interacting influences. However, after removal of the direct effect of precipitation, the temporal variability of gamma radiation reflects to a large extent the water content of the soil, including very fast (sub-daily) changes in soil moisture conditions.

## **Evaluation of the methodology for estimation of radon hazard for public places (case study: Sardinia, Italy)**

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Indoor Air Quality (IAQ) in public and residential buildings has become a highly important environmental issue for municipalities, especially in large, densely populated urban areas. The inhalation of indoor radon ( $^{222}\text{Rn}$ ) and its progenies accounts for half of the annual dose of ionizing radiation. Accumulation of radon gas into confined spaces may increase the risk of lung cancer, depending also on the number of residents and their exposure time. On the average, people spend most of their time in confined spaces; considering these facts and the higher vulnerability of children, elderly and patients, radon monitoring in schools, retirement homes and hospitals should be the first priority. This article discusses the most common approaches suitable for radon monitoring and radon assessment risk in public buildings. Based on literature data, previous experiences, comparison of different measurement methodologies and available instruments, a comprehensive program for the evaluation of radon risk in public buildings is presented. The results of the first step of this research will be used as an action plan for real-time radon measurements and radon risk analysis in the main cities of Sardinia. The geogenic factors controlling Rn proneness will be also investigated and selection of the city for doing measurements in public buildings will be based on the geogenic map of radon potential which is an ongoing project as a part of a three year Ph.D. research programme.

## **Main results of the second radon-in-field international intercomparison for passive measurement devices**

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During last decades, many radon intercomparison exercises have been organized in order to assure the quality of radon measurements. Most of them took place in "radon chambers", in standard conditions of temperature, humidity and radon concentration. Nowadays, many radon laboratories have expressed interest in testing their monitoring systems during in-field exercises, with exposures taking place in less controlled and much more variable real conditions, and for this reason very similar to the ones in which devices are normally exposed. The first radon in-field international intercomparison for passive measurement devices was held in Italy during 2014 with the aim of evaluating the performances of passive radon devices exposed in "extreme" environmental conditions (very high radon level, huge fluctuations of the radon concentration values, very high humidity levels, etc.).

The second edition was organized in order to focus on two different issues: the effect of long time sampling at low and medium radon levels and the effect of the simultaneous presence of radon and thoron.

This last intercomparison was attended by 48 laboratories (from 12 European countries and 3 non-European countries), with an increment of about 4% respect to the first intercomparison, and with the exposure of 66 sets of devices. Passive devices were mainly SSNTD (76%) and electrets (20%).

Three different exposures were carried out from November 2016 to April 2017: the first two exposures were performed at ARPA laboratory in Ivrea (Piemonte, Italy) and lasted 5 months, the third exposure was performed for about 10 days in a cellar of a private dwelling in Quittengo (Piemonte, Italy).

In order to gather information about laboratories procedures in term of managing data and to analyze the reliability of passive devices, the participants were requested to prove results both in terms of radon exposure and of arithmetic mean of radon activity concentration.

In the present paper, the main results are showed and discussed. However, looking at overall radon concentration results, no particular effect of long time sampling (fading) has been observed. Conversely, the simultaneous presence of radon and thoron could affect accuracy, in particular in case of electrets, although also the simultaneous interference of humidity cannot be neglected.

## **Preliminary study of cesium immobilization in geopolymer matrix**

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Geopolymers are modern synthetic materials which have many beneficial properties, they have excellent mechanical properties, are fire and heat resistant, shrink slightly, mold in shape based on the above mentioned behaviour of geopolymers, they could be applicable as conditioning materials e.g. hazardous waste, radioactive materials. However their long-term applicability needs to be investigated.

The aim of the study is to investigate the mobility of different radioactive isotopes, (Cs-137, Co-60) in embedding agents. Different geopolymer matrixes (metakaolin, kaolin, bentonite, zeolite, fly ash) were tested. Leach test was carried out according to ASTM C1308 - 08(2017) standard, the activity concentration of Cs-137 isotopes was measured using semi-conductor HpGe gamma-spectrometry. According to our preliminary results those matrixes have given the most sufficient immobilization effect in which bentonite, cement, fly ash and 8M NaOH were used, approx. 10% of cesium was released.

## **The effect of concrete composition on its neutron activation properties**

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The European Spallation Source (ESS) will be the world's most powerful pulsed neutron source. A vast amount of concrete will be used for radiation shielding purposes in the construction of this research facility. Since high neutron fluxes will be applied as analytical tools, it is an important step of safety planning to determine the neutron activation properties of concretes.

MCNP and CINDER'90 simulation codes were used to estimate the activation products of concretes and the dose contributions of these activation products. Long-lived radioisotopes are important in terms of radioactive waste management, while short-lived radioisotopes can have higher contributions to the radiation level around the instrument particularly in maintenance periods.

Two different concretes were examined: a reference concrete and the so called PE-B4C-concrete, developed at the ESS, which is the mixture of the reference concrete, polyethylene and B4C (these components enhance the neutron shielding properties of the concrete). Beside the nominal concrete composition simulations were carried out with compositions calculated based on our previously accomplished PGAA, XRF and NAA measurement results.

The experiments proved a deeper understanding on the significance of certain trace elements and a comparison of the activation properties of the 2 concrete samples.

## Natural occurring radon measurement method using fiber optics

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It is extremely important to be able to measure and monitor radon concentrations due to its relevance as a natural environmental tracer. Several measurement methods are currently employed for radon monitoring in liquid and gas samples. In gas samples, as for the indoor concentration assessment, most systems use passive solid-state nuclear track detectors. This system lacks temporal resolution as it gives an integrated value of radon concentration for the detection time of exposure. In liquids, samples are normally sent to a laboratory where a liquid scintillation counter is used to determine radon concentration. This technique is very time-consuming, expensive and with no temporal resolution. Other techniques currently used in liquid samples rely on expensive pumping systems (e.g. RAD7), presenting some temporal resolution. Gamma-scintillation detectors avoid the need of pumping systems and are well suited for continuous monitoring, presenting high sensitivity as well as being more cost effective. In this work, a new scintillation detector based on optical fiber is proposed. The system, composed by scintillating optical fiber, a photomultiplier and a micro-computer, is robust, autonomous and cost effective. The detection of Uranium-enriched rock samples placed in an enhanced confined mode (ECM) system was used as proof-of-concept. The proposed system successfully detected the presence of radon, as well as radon concentration variations. A commercial solid state nuclear track detector (CR-39) was used in order to access the average value of radon concentration inside of the detector encasing.



## **Radon measurements for the planned thermal bath complex in Chunqing (Ba'nán) region, China**

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The health effect of radon is a well-known phenomenon in the whole world. The increasing lung cancer risk in case of the miners is a harmful effect, but in a lot of cases the healing effect of this radioactive gas is used. There are several caves with high radon activity concentration in the world where the treatment of patients with respiratory problems can be carried out. Besides the respiratory problems radon content are used with very good results in healing of inflammatory rheumatological diseases, mainly spondylitis ankylopoetica. In Europe – especially in Hungary – there are some bathes with high radon concentration in the water which are used for the rheumatological treatment. Based on the Hungarian experiments the aim of this study is to investigate the possibility of the radon thermal bath in China. The venue of the planned thermal bath complex in Chunqing (Ba'nán) region of China. They examined the measurable radon content in the Hot cave's air, in the water of the lake in the cave, in the spring near the cave, and in the spring that can be found on the other side of the mountain. The cave is a hot cave, with a high percent of humidity (95-99%), with a temperature of 30-37,8 °C. The air of the cave contains radon, however measurements in different parts of the cave have shown quite different results, values between 629 Bq/m<sup>3</sup> and 6290 Bq/m<sup>3</sup> were measured. The measurements were carried out by AlphaGUARD PRO radon monitor. Worthwhile radon activity was low (0.9 Bq/L) in the water of the cave's lake. The water of the well near the cave contains calcium magnesium sulphate, it is a mineral water with high rates of minerals, with a temperature of 45 °C. It contains small amount of radon (1.5 Bq/L). Due to its consistence it is useful for preserving health, and can be used for wellness, recreation, and preventional purposes. Since its radon content is minimal (vanishing), it is suitable for bathing for masses. The water of the spring on the other side of the mountain is mineral water containing calcium magnesium sulphate, has a high mineral rate, and has radon content. Its radon content is 62,8 Bq/dm<sup>3</sup>. radon baths in Europe have a much higher radon concentration, however in Hungary, in the city of Eger, there is effective healing in progress with similar radon concentration in the Turkish Bath, and in the thermal Bath of the Markoth Ferenc Hospital.

## **Metrological aspects of the second radon-in-field international intercomparison for passive measurement devices: dwellings and workplaces**

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The “Second radon-in-field international intercomparison for passive measurement devices”, held in Italy in 2016-2017, was focused to specific issues affecting the quality of radon concentration measurements in real situations. Each participants sent at least 3 sets of passive devices to use in three different exposures. Passive devices were exposed in two workplaces (office and storage room, respectively) for 5 months at typical indoor radon activity concentrations, in order to evaluate the effect of fading. The third exposure was shorter but in a “complex situation”, such as a cellar characterized by the simultaneous presence of radon and thoron, with the aim to evaluate both the thoron exposure itself and if the response of radon passive devices is affected by thoron presence. In the present paper a synthesis of the metrological aspects of the intercomparison is given. Indeed particular attention was pa to metrological characterization of the 26 between radon and radon/thoron monitors, based on different measurement principles, with particular attention to background estimation, calibration and their response to radon and radon/thoron atmosphere. The active monitors used in these intercomparison exercises were calibrated by ENEA-INMRI (National Metrological Institute of Ionizing Radiation) and in order to give the radon reference values in field exposures several steps were performed: the experimental details will be given.

## **Background and counting efficiencies variation of two liquid scintillation spectrometers for low level tritium measurement**

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Liquid scintillation counting (LSC) is the most commonly used technique for measuring low-energy beta emitters, namely for tritium. ISO 9698/2010, International Standard specifies the conditions for the determination of tritium activity concentration in samples of environmental water or of tritiated water using liquid scintillation counting. This paper presents the results of a comparative study conducted with four Tritiated standard solutions, using two ultra-low level liquid scintillation spectrometers Quantulus 1220. We investigated the variation of three measurement parameters: the counting efficiency, the figure of merit and the background counting rates. Measurements were performed using two ultra-low level liquid scintillation spectrometers Quantulus 1220, one in service from 1998 manufactured by Wallac (Q1) and the other from PerkinElmer in service starting with 2011 (Q2). In order to compare the performance of the studied spectrometers during the period December 2014 – December 2017 (more than 150 observations), the background, counting efficiency, figure of merit and detection limit were determined for each of them. Even if the mean value of the background for Q2 is higher than the mean value for Q1, the detection limit of Q2 is lower than that of Q1 due to better counting efficiency. The mean value of maximum figure of merit recorded during studied period was higher for Q2 than Q1, also. In fact, figure of merit incorporates the spectrometers ability to measure low radioactivity level of the weak beta emitters, like tritium, and it is of major importance in establishing an optimum window of spectrum interpretation. The defined region of the spectrum has special characteristics, the best counting efficiency at the low level of background. The differences of studied parameters between the two ultra-low level liquid scintillation spectrometers are due to aging of the photomultipliers of Q1, after 20 years operation, but the two spectrometers have a detection limit lower than 5 TU (approx. 0.6 Bq/l) suitable for tritium environmental monitoring.

**Acknowledgments:** This paper was prepared in connection with the work done for project PN 18 12 03 04, part of Core Program ICSI 4E supported by the Romanian Ministry of Research and Innovation, and monitoring programme of Tritium Removal Facility - PESTD.

## **Study of tritium level and individual precipitation of the warm months in Rm. Valcea, Romania**

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Precipitation is the main mechanism for removing H-3 from the atmosphere. Moreover it is the vehicle for the downward transport of H-3 within the troposphere. Romania reports now by Rm. Valcea location isotopes in precipitation, but the records period start with 1999 for tritium and 2011, for stable isotopes. Regarding the tritium level in precipitation at Rm. Valcea location there was identified a particularity for the monthly tritium average and monthly precipitations average of long term period of observations: the months with maximum concentration are also months with maximum quantity of precipitations. In order to investigate this particularity there was collected and measured individual precipitation of the warm months during the period 2008-2017. Here we present the results of tritium concentration in individual precipitation with the aim of prove a relation between tritium concentration with the amount of the precipitation. Important differences were recorded for the monthly amounts of precipitation, but no correlation were established between the amount of individual precipitation in warm months and measured tritium concentration. Peaks of tritium concentration were measured in individual precipitations demonstrating the influence of continental nuclear activity, but tritium concentration in our area is still low, an early average of 10 TU being registered. Acknowledgements: This paper was prepared in connection with the work done for project PN 18 12 03 04, part of Core Program ICSI 4E supported by the Romanian Ministry of Research and Innovation, and monitoring programme of Tritium Removal facility PESTD.

## **Radiocesium contamination in riverine areas around the Fukushima Dai-ichi nuclear power plant**

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The radiocesium (<sup>137</sup>Cs, T<sub>1/2</sub>=30.17 y) was released into the atmosphere and Pacific Ocean from the accident of Fukushima dai-ichi nuclear power plant (FDNPP). The dry and wet deposition of radiocesium caused a large area contamination in northwest of FDNPP. To minimize the health risk of the accident, intensive clean-up efforts were successfully accomplished in contaminated urban areas. However, the remediation of forest areas is much more difficult thus radiocesium will remain there for a long period of time. The radiocesium migration can be affected by many soil parameters such as, grain size, adsorption properties of soil phase, mineralogy, organic matters and ion concentration. Monitoring of different environmental matrices is very important to understand the fate of radiocesium dispersion in forest and riverine areas. In this study, we collected 20 river bed sediments (RBS) and 20 flood basin sediment samples (FBS) from five forest area river systems e.g., Ukedo River, Takase River, Maeda River, Kuma River and Tomioka River within 20 kilometres from the FDNPP between 30 July and 2 August 2017. Radiocesium activity concentration was measured in all with HPGe gamma spectroscopy. The organic matter content of the samples was determined by weight loss on ignition (LOI) method. In case of six samples, particle size distribution was determined using dry and wet sieved with mesh sized in the range from 45 to 2000 µm. Major oxides concentrations also were measured using an X-ray fluorescence (XRF) spectrometer. The range of radiocesium concentration in RBS is between 119 and 25,503 Bq/kg, in FBS is 106 and 135,557 Bq/kg. The results show correlation with radiocesium contamination observed in governmental monitoring program the (MEXT,2011). Analysing the radiocesium concentration in different particle size fractions, we observed around 70% of the radiocesium attached to fine sand, silt and clay particles. The concentrations of major oxides and organic matter of the samples show dependence on geological locations.

## **Building material radon emanation and exhalation rate: potential information and critical aspects of an advanced data collection**

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The role of building materials as radon source indoors is an issue increasing in research activities in the last years. This is mainly due to some important reasons. In the last decades international organizations (ICRP, WHO, etc) recommended a decrease of the radon reference levels on the basis of epidemiological evidences. Moreover, the increasing recycling of NORM residues as components of new building materials determines a radiation protection concern not only for gamma exposure but also for radon. That's why the EU BSS indicates that the building material radon contribution shall be taken into account in the National Radon Plans. Consequently, the authors have started a collection of radon emanation and exhalation rate data which looks particularly interesting and useful in the light of previous considerations. A first database containing data of about 1450 samples was published in the beginning of 2018. This data set contains informations collected to 2015. In this work a collection updated to 2018 is presented and discussed. Notwithstanding the huge amount of data and relevant potential information, the database analysis shows some critical aspects, such as the use of different units to express the exhalation rate due to different measurement approaches, different measurement techniques, lack of information of density and thickness of measured samples, etc. All these aspects do not let to compare reliably collected data. From all these remarks the need of research activities aimed to develop a standard on exhalation rate measurements based on reliable and user-friendly methods emerges.

## Study of the short-term changes of radon concentration in the soil air

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In the soil air,  $^{222}\text{Rn}$  (radon) activity concentration usually reaches several  $\text{kBq}\cdot\text{m}^{-3}$ , and is not stable. While radon activity concentration depends mainly on the soil type and on the content of  $^{226}\text{Ra}$  in the soil, it also depends on atmospheric pressure, soil humidity, precipitation and temperature. Naturally,  $^{222}\text{Rn}$  activity concentration in the soil air is the result of simultaneous influences of several factors in real natural conditions. Therefore it is difficult to search for the dependence of soil  $^{222}\text{Rn}$  on individual factors in isolation. However, the study of the short-term changes in radon activity concentration with one of the factors changed, while others remain relatively constant, is one of the possible approaches to evaluate these influences. An example of such an approach can be the evaluation of the relation between the daily variations of radon activity concentration and the regular daily oscillations of atmospheric pressure.  $^{222}\text{Rn}$  activity concentration in the soil air has been continuously monitored by a scintillation cell of Lucas type with a volume of 125 ml. The air was sampled from a depth of 0.8 m and dried in the refrigerator before entering the Lucas cell. The monitor allows measuring the radon activity concentration in the soil air on the level of around  $10\text{ kBq}\cdot\text{m}^{-3}$ , with a relative error of 1.5 % during a one hour long measurement interval. The measurements show that the changes in  $^{222}\text{Rn}$  activity concentration during the day are not very significant. The deviations of  $^{222}\text{Rn}$  activity concentrations during a day from its average daily value were only on the level of (1 – 5) % for individual months. The diurnal time series of  $^{222}\text{Rn}$  have not the same shapes for all months. The measurement analyses show that the regular short-term changes in  $^{222}\text{Rn}$  activity concentration in the soil air could be connected to the changes in atmospheric pressure. A more significant linear correlation between the deviation of the  $^{222}\text{Rn}$  activity concentration during a day from its average daily value and the relative change in the atmospheric pressure was obtained mostly for the summer months.

**Investigation of the spatial distribution relation between outdoor absorbed gamma dose levels and cation exchange capacity (CEC) of soil samples from Arıklı Uranium Mineralisation Region, Çanakkale, Turkey**

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Radioactive material exploration sites are a current concern due to the possible radioactive material contamination risks. In 1959, radioactive anomalies were found in Arıklı village in Turkey and its surroundings by airborne gamma measurement. After orientation studies, uranium explorations was continued by drillings but the exploration studies were cancelled due to the lack of economic reserves. The present research helps to understand spatial distribution of outdoor absorbed gamma dose levels by taking into conseration the relationship with cation exchange capacity (CEC) measurements as a geochemical control parameter, in a catchment including the abandoned Arıklı uranium mineralisation site. The study area was split into grs and 156 outdoor gamma measurements were taken at the gr nodes using ESP-2 Na(I) probed Eberline gamma detector. In addition, surface soil samples were collected from the same nodes. For the CEC measurements, the barium chlorite method was used by ICP-OES. Homogeneity test of univariate distributions, and bivariate scatter plots were used for statistical analysis. Strong correlations were obtained especially at the alluvial accumulating flat bottoms of valleys.



## **The determination of $^{90}\text{Sr}$ transfer factors in the environment using stable strontium as a surrogate and neutron activation analysis**

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In the last decades many food-chain models have been employed to predict the transfer processes (soil-to-plant transfer factor(s) [TF], plant-to-animal transfer coefficient(s) [TC] and concentration ratio [CR]) that are likely to be important for radiological assessment arising from nuclear accidents and former atmospheric weapons testing. Due to the relatively high uranium fission cross-sections Cs-137 and Sr-90 have become ubiquitous in the environment and have been exhaustively studied. The determination of Cs-137 requires large volumes of samples with long counting times of its characteristic 661.7 keV gamma-ray. Since Sr-90 exhibits pure beta decay its determination requires digestion and radiochemical separation with possible interferences. Very recently, we have successfully shown how Cs-137 transfer factors can be determined by the evaluation of stable cesium using neutron activation analysis. In this work we report on using epithermal neutron activation analysis in conjunction with Compton suppression to determine strontium using the  $^{86}\text{Sr}(n,\gamma)^{87\text{m}}\text{Sr}$  reaction and its 2.5 hr half-life. This technique is non-destructive requiring no digestion with sample sizes between 0.3-0.4 g. Typically about 8-10 samples can be analysed in one working day. The liquid comparator, samples and reference materials were irradiated with an epithermal neutron flux  $2.2 \times 10^{11} \text{ n.cm}^{-2}\text{s}^{-1}$  for 10 minutes, and a decay of 30-45 minutes and a one hour counting time using the 1.1 MW TRIGA Mark II research reactor at the Nuclear Engineering Teaching Laboratory. Results for stable strontium as surrogate for Sr-90 will be presented.

## **Investigation of NORM in Turkish bauxite residue (red mud) using neutron activation analysis for the determination U-234,235,238, Th-232 and K-40 and their leaching characteristics**

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The measurement of natural radioactivity in the environment from naturally occurring radioactive material (NORM) is of significant importance for evaluating the radiological impact of non-nuclear industrial activities. The production of alumina ( $\text{Al}_2\text{O}_3$ ) from bauxite ores using the Bayer process produces a waste material which is called red mud. The residue is mainly composed of silica, iron, aluminum, calcium, and titanium oxides in different proportions dependent on the country of origin of the bauxite ore. Industries dealing with raw materials containing various concentrations of naturally occurring radionuclides often produce large amounts of waste. Due to its high alkalinity and the significant amount of radioactive elements and rare-earth elements (REEs) contents, red mud should be evaluated as an environmental problem. On the other hand, these waste materials may potentially be used as an industrial by-product. When re-using these residues, it is of importance to evaluate the radioactive and toxic elements, chemical composition, and the leaching features of these materials. In this study, the activity concentration of  $^{234,235,238}\text{U}$ ,  $^{232}\text{Th}$  and  $^{40}\text{K}$  in bauxite residue was determined by neutron activation analysis (NAA) and Compton suppression methods. SRM 1632d and 1633c from the National Institute of Standards and Technology (NIST) and the liquid standards of uranium, thorium and potassium from Inorganic Ventures were used to calibrate the measurement system. We will show how rapidly these NORM isotopes can be evaluated in red mud with minimal sample sizes that does not include long counting times with the usual self-attenuation of gamma rays in bulk samples.

## Search for Fukushima NPP accident traces in soil samples based on $^{239+240}\text{Pu}$ analysis

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The impact of the actines after Fukushima NPP accident on the environment is still unclear. There are a few reports on contamination level of Pu isotopes in the soil to understand about global fall out, weapon grade Pu and Chernobyl nuclear accident. The activity concentration of  $^{239+240}\text{Pu}$  in our soil samples from Fukushima exclusion zone (Namiemachi area) ranged between (13-360) mBq/kg, whereas  $^{238}\text{Pu}$  was detectable in a few samples. This study suggests especially global fallout as a main source of Pu isotopes in analyzed samples. Activity ratio of  $^{238}\text{Pu}/^{239+240}\text{Pu}$  about 0.03 mainly referred as global fallout ratio. We are allowed to estimate that level of the  $^{238}\text{Pu}$  concentration, which should be closed to the detection limit for alpha spectrometry. The activity concentration of  $^{239+240}\text{Pu}$  indicated contribution of the other source than global fallout for a few samples with activity ratio  $^{238}\text{Pu}/^{239+240}\text{Pu}$  higher than 0.3. However, additional  $^{241}\text{Am}$  analysis may be able to clarify and update our results.

## **Effects of leaching test results on internal dose assessment on the example of phosphate fertilizer**

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A large variety of leaching tests and soil-water distribution coefficient determination methods are commonly used to assess the environmental availability of heavy metals and radionuclides. Interpretation and comparison of these results can be challenging and have implications for several assumptions made during the internal dose assessment process. Furthermore, the differences between leaching tests can mean an order of magnitude or higher difference in the resulting distribution ratio and affect the following modelling or internal dose assessment severely. Two leaching procedures were applied and compared the MSZ 21470-50 (Hungarian standard) and the Tessier five-step sequential extraction method on eight commercial available phosphate fertilizer samples. A conservative scenario, taking into consideration the Tessier V. fraction from the results, which is commonly regarded as not available under natural conditions, would present an upper estimate for the increase of the internal radiation dose. This would have a significant effect (between -11 and +100%) on the results of the dose assessment, however the dose increment by the common rate of fertilizer use would be less than 0,45% compared to the those calculated for the unaltered soil.

## **A new application of mass spectrometry instruments, $^{90}\text{Sr}$ analysis**

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Five isotopes of strontium element exist in the environment, four of them ( $^{84}\text{Sr}$ ,  $^{86}\text{Sr}$ ,  $^{87}\text{Sr}$ ,  $^{88}\text{Sr}$ ) are stable and have natural origin while  $^{90}\text{Sr}$  is a radioactive isotope ( $T_{1/2}=28.8$  y) and originates from nuclear weapon tests and nuclear accidents. With regard to dose estimation, the  $^{90}\text{Sr}$  monitoring in environmental samples is essential because it easily permeates into human skeleton causing long-time radiation exposure. After the Fukushima accident, the drawbacks of the radiometric methods for  $^{90}\text{Sr}$  determination were revealed. Because of the necessity of rapid  $^{90}\text{Sr}$  analysis, attempts have been made to develop new  $^{90}\text{Sr}$  measurement methods using mass spectrometry instruments, such as accelerator mass spectrometry (AMS); resonant laser ionization mass spectrometry (RIMS); inductively coupled plasma mass spectrometer with dynamic reaction cell (ICP-DRC-MS); triple quadrupole inductively coupled plasma mass spectrometer (ICP-QQQ-MS) and thermal ionization mass spectrometer (TIMS). For each technique, the principles used for  $^{90}\text{Sr}$  analysis, and the merits along with demerits will be discussed.

## **Progress report of a new research project on dose assessment for residents of Namie Town, Fukushima Prefecture –Focused on radon research**

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Namie Town, Fukushima Prefecture decided that the evacuees began to return to their houses from March 2017. Accordingly, we established new research project on the dose estimation for the residents of Namie Town. A fact that is often ignored by the general public is that people were exposed to natural radiation sources such as radon, cosmic-rays and terrestrial gamma-rays before the FDNPP, and that such exposure continues on a daily basis. According to the report on the nationwide indoor radon survey, indoor radon concentration  $>100 \text{ Bq m}^{-3}$ , which corresponds to 3 mSv, as an annual effective dose was found in a house in Fukushima Prefecture. Information about natural radiation doses should be the baseline for the public when considering the effects of artificial radiation. It is well known that the radon concentration has been observed daily and seasonal variations due to the human behavior and meteorological parameter. In this study, a passive-type radon monitor will be installed to estimate an internal dose taken into account of variation of radon concentration. This work was supported by Research on the Health Effects of Radiation organized by Ministry of the Environment, Japan.

## **Overview of a new research project on dose assessment for residents of Namie Town, Fukushima Prefecture**

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Namie Town, Fukushima Prefecture allowed the evacuees to return to their houses from March 2017. Accordingly, we established new research project on the dose estimation for the residents of Namie Town. Indoor and outdoor radon concentrations are measured using a radon-thoron discriminative monitor (RADUET) to estimate internal doses by its inhalation. Indoor thoron progeny concentration is also measured using a passive type thoron progeny monitor. Furthermore, airborne radiocesium concentrations at five points are analyzed by HPGe detector to estimate internal doses. To estimate an internal dose by radiocesium in domestic water, we are collecting river water and sediment at five points. On the other hand, a car-borne survey along the main road was carried out using a 3-in NaI(Tl) scintillation spectrometer to know the dose rate distribution in Namie Town. Ambient dose rates are measured using a 3-in NaI(Tl) scintillation spectrometer at the fixed measurement points in each 1 km to make a dose rate distribution map. The gamma-ray pulse height distributions are analyzed using a response matrix method to evaluate the dose rates for natural and artificial components. Individual external doses for the residents are estimated using an integral pocket dosimeter. We will introduce about overview of our new research project for residents of Namie Town. This work was supported by Research on the Health Effects of Radiation organized by Ministry of the Environment, Japan.

## Sintering of NORM based ceramics

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Red mud as naturally occurring radioactive material (NORM) was used in the clay matrix in order to examine its behaviour on the sintering of the composites consisted of clay and 10, 20 and 30wt.% red mud.

The origin of illite clay and red mud employed in this investigation were from R. Macedonia and Hungary, respectively. The both raw materials were characterized from chemical, mineralogical and physical aspect. DTA/TG (heating rate of 100°C/min, temperature up to 10500°C, air atmosphere) was used to follow the thermal behaviour (dehydration, dihydroxylation and phase transformation) of clay, red mud and composites.

Consolidation of the compacts was realized by pressing ( $P = 70\text{MPa}$ ) and sintering from RT to 10500°C. Expansion/shrinkage of clay, red mud and composites during polythermal sintering of the pressed compacts were followed by dilatometer (heating rate of 100°C/min, air atmosphere). By introducing the red mud in the clay matrix, it is influenced on the lowering the shrinkage and beginning of sintering. The lowest beginning of the sintering ( $T = 8500^\circ\text{C}$ ) showed the composites composed of 30wt% red mud and 70wt% clay.

The XRD patterns of the sintered composites showed that the dominant mineralogical phases are: quartz, cristobalite, hematite and plagioclase. The microstructure observation realized by scanning electron microscopy (SEM) showed homogeneous distribution of red mud in the clay matrix.



## **The role of LSC methods in implementation of the Euratom Directive 51/2013 for the monitoring of radioactivity of water supplies in Poland**

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In October 2013, the European Commission issued the Directive Euratom/51/2013, related to the monitoring of radioactivity of drinking water supplies. In Poland this Directive has been introduced into the national law system in November 2015 as the Ordinance of the Ministry of Health. In the implementation of the Ordinance into practice the main role play methods based on LS spectrometry. This robust technique can be applied for monitoring of the majority of the radionuclides, required by the Ordinance, it means tritium, radon and concentration of radium isotopes in water. In this paper the description of the Polish approach to the Euroatom Directive implementation is given as well as brief description of the LSC methods, used in the monitoring system of water supplies, proved by Silesian Centre for Environmental Radioactivity.

## **Monitoring of radioactivity of water supplies in Poland – first two years of the implementation of the Euratom/51/2013 Directive**

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In November 2015 the Ordinance of the Ministry of Health related to the monitoring of radioactivity of drinking water supplies has been issued. It is an implementation of the European Commission Euratom/51/2013. Actually the year 2016 is the first year of the implementation of the Ordinance into practice. Very important for the introduction of the Ordinance is application of LS spectrometry methods for the monitoring of the radionuclides, present in water. Polish Ordinance requires monitoring of tritium, radon and radium isotopes in water. In the Silesian Centre for Environmental Radioactivity, Central Mining Institute, measurement methods for all mentioned above radionuclides are based on LSC. In the paper results of the first year of the monitoring of radioactivity in drinking water supplies are presented. Almost 2000 water samples have been analysed within 2016, in most of the samples results of monitoring were below the detection limits of particular methods. No elevated concentrations of tritium have been found, only in several cases the elevated concentration of radon were found, exceeding 100 Bq/l. In case of radium isotopes, their activities are used to estimate the total indicative dose. For the several water supplies, the estimated annual dose for users exceeded reference value 0,1 mSv/year. The preliminary investigations will continue till the end of the year 2017. At that time it will be possible to have to final pattern of distribution of radioactivity in drinking water supplies in Poland.

## **Application of LSC for measurements of concentrations of radon and thoron decay products in air**

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Liqu scintillation counting (LSC) is a measuring technique, broadly applied in environmental monitoring of radionuclides. One of the possible applications of LSC is the measurement of radon and thoron decay products. But this method is suitable only for grab sampling. The LSC method can be used for calibration of portable radon decay products monitors as the LSC method has the advantage to be an absolute one. A comparison of the measurements of has been performed in the radon/thoron chamber of the Chinese National Institute of Metrology in Beijing. The method has been previously developed and tested in the Silesian Centre for Environmental Radioactivity with application of Triathler and Quantulus LS spectrometers. Measurements in the Chinese National Centre of Metrology have been performed with use of TriCarb 2770 and Hex L-300 LS spectrometers. The results show a good compliance of the method's transition to any of LS spectrometers without any additional calibration. For long term measurements a different technique can be applied – monitors of potential alpha energy concentration (PAEC) with thermo luminescent detectors (TLD). The readout of TL detector shows directly potential alpha energy, with no dependence on equilibrium factor etc. This technique, which had been used only for radon decay products measurements, was modified to allow simultaneous measurements of radon and thoron PAEC. The TLD monitors have been checked in the radon/thoron chamber of NIM in Beijing, too.

## **Application of zeolite for radium removal from water – results of RATZEO project**

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In the formation waters, occurring in coal mines in the Upper Silesian Coal Basin (USCB), Poland different pollutants are present which may cause damages to the natural environment, among them radium isotopes. In several collieries the radium removal from mine water was necessary to mitigate negative results of radium release with mine effluents. The most efficient method of radium removal is based on application of barium chlore, implemented in full technical scale in two of Polish collieries. The treatment stations were unique, the first underground installations for the removal of radium isotopes from saline waters in non-uranium mines. The removal efficiency, exceeding 95% of the initial activity, was achieved. Another possibility of removal radium isotopes from salty waters is an application of zeolites. In this paper results of laboratory experiments are presented, describing results of mine water treatment with use of zeolite and barium chlore solution. During the period of exploitation of underground treatment stations for radium removal some drawbacks of the application of barium chlore have been found. The most important ones were as follows: the need for continuous feeding of the powdered chemical and the maintenance of installation. Therefore investigations were focused on such methods, which would allow automation of the process, or would be a passive ones, with no need of continuous maintenance. It has been found, that two possible methods were – application of zeolite for radium removal from water in form of a passive barrier or application of barium chlore as a solution, which would give the opportunity to make the process of feeding fully automatic. The national research project RATZEO was awarded and executed in the period 2014-2017. Therefore the prototype of the treatment installation was constructed and tested within whole year in the active coal mine. The main goal of the investigations described in the paper was to check the capability of zeolites to remove natural radionuclides from mine waters. Additionally, one of the important goals of the project was to compare the possibility of application of both methods in active coal mine in underground galleries.

## **Preliminary results on study of the behavior of Pb-210, Cs-137 and Po-210 isotopes in the sediments column under different chemical conditions**

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The sediment deposits can be considered a data base of the changes in environmental processes and pollution in recent past. The most used radionuclide for dating the sediment layers for established the date of different events are Pb-210, Ra-226, Po-210 and Cs-137 (in some cases Am-241) in Pb-210 dating method. The determination of Pb-210 in this method, for a better precision, is made by measuring the alpha emitter daughter Po-210. The Po-210 is produced by the disintegration of Pb-210 inside of the sediment column where the produced Po-210 can be present in a chemical form which is soluble in water. This study focus on the isotopes behaviour which is used in sediment dating by using Pb-210 method especially the difference between Pb-210 and Po-210. The solubility of Po compounds can produce migration of Po-210 between sediment layers. For a proper chronology, the determination of the behaviour of the radionuclides in sediment column is crucial. Five sediment column with known concentration of the investigated radionuclides (Pb-210, Ra-226, Po-210 and Cs-137) was prepared in laboratory condition, in different chemical condition (Ph, Cl<sup>-</sup> and NO<sub>3</sub><sup>-</sup> ions) and stored 6 months. The columns were sliced again and the radionuclides concentration was investigated.

## **Metrology background of the national radon action plan**

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February 6, 2018 was an important date for the introduction of the Radon Action Plan according with the EU-BSS Recommendation (2013/59 / EURATOM). With several domestic institute participation and the OSSKI leadership the National Radon Action Plan was prepared.

The Action Plan requires uniform measurement methods and the use of verified radon meters. In Hungary the National Institute of Metrology is the BFKH Metrology and Technical Supervision Department. The department has been proving for the task for a number of years with creating a radon chamber in which radon concentration can be varied wely.

The presentation describes the work to be done in the next two years and the tools which are already have been installed and further instruments which still needed to install for the metrological background.

## **EURAMET EMPIR 16ENV10 project: MetroRADON**

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EURAMET is a regional organization of national metrology institutes in European countries. The EU supports the EURAMET research development projects through the EMPIR (European Metrology Program for Innovation and Research) program within the HORIZON 2020 (2014-2020) framework program.

Announcement for 2 main topics annually appears. In 2016, the winning projects were selected from the "Environment" and "Energy" proposals, one of which was the 16ENV10 (MetroRADON) proposal. The aim of this project is to develop reliable techniques and methodologies to enable traceable radon activity concentration measurements and calibrations at low radon concentration.

The main goals of the presentation is to briefly present the EMPIR program and to present the objectives of the MetroRADON project.

## **The effects of hydrogen peroxide solution and ultrasound on the dissolution of electrodeposited uranium oxide**

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The effects of hydrogen peroxide solution and ultrasound with and without high temperature (800°C) on the dissolution of electrodeposited uranium oxide have been studied. Fifty the uranium oxide samples were electroplated onto circular 304 stainless steel plachets. The alpha counting of the samples before and after leaching experiments periods in control (pure water) and 0,1 mmol/L, 1 mmol/L, 10 mmol/L, 100 mmol/L, and 1 mol/L solutions of H<sub>2</sub>O<sub>2</sub> combined ultrasound with and without high temperature was recorded by using an Alpha Ensemble (ORTEC). The results of these analyses have shown that in our experimental conditions the absence of temperature (room temperature ~ 270°C) resulted the decrease of 60% in alpha radiation at [H<sub>2</sub>O<sub>2</sub>] ≤ 1 mmol/L and 50% at control (no H<sub>2</sub>O<sub>2</sub> addition) while at H<sub>2</sub>O<sub>2</sub> concentration > 1 mmol/L the uranium release decreases with increasing H<sub>2</sub>O<sub>2</sub> concentrations. On the other hand, in the presence of temperature of mixture solutions about 800°C, the decrease of 12% in alpha radiation at control was recorded while they ranged from 79% to 98% of the increasing H<sub>2</sub>O<sub>2</sub> concentrations. The over 70% of uranium release was consistently removed in all H<sub>2</sub>O<sub>2</sub> concentrations linked to the significant effect of not only hydrogen peroxide solution and ultrasound but also the temperature on the dissolution of electrodeposited uranium oxide and the decontamination.



## **Radiological characterization of mosses in Western part of Hungary**

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The biological monitoring method is used for observing and monitoring the impacts of pollutants on the environment, which is a very common method nowadays, especially for the investigation of trace elements. Biomonitoring studies are used on lower and higher organisms, these studies provide quantitative and qualitative information about specific features of the biosphere. Moss is widely used as a biomonitoring tool, because of its beneficial properties, for example for its high spreading rates and excellent adaptability. The mosses accumulate the metals without selectivity, for that reason they can be suitable for both quantitative and qualitative analyzes.

In this study 15 moss samples were collected from Central and Western Hungary, the activity concentration of the terrestrial gamma-radiating radionuclides ( $^{226}\text{Ra}$ ,  $^{232}\text{Th}$ ,  $^{40}\text{K}$ ) were determined by HPGE gamma spectrometry, in Marinelly geometry. Based on the measurements the average activity concentration of the  $^{226}\text{Ra}$  in the moss samples was between 9,2 -74,0 Bq/kg, the  $^{40}\text{K}$  activity concentration was between 129,94 – 490,58 Bq/kg and the  $^{232}\text{Th}$  activity concentration was at 3,8 – 33,36 Bq/kg.

These results are correlated with the average activity concentrations of the soils in this region.

## **Investigation of Po-210 content in phosphorous fertilizers, estimation of radiation exposure from their application**

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In recent years there has been an increasing awareness of the radiological impact of non-nuclear industries that makes minerals containing naturally occurring radioactive material (NORM). These industrial activities may result in significant radioactive contamination of products, wastes.

Phosphate fertilisers for agricultural use are derived from phosphate rock (mostly apatites), so they have an increased activity concentration. Thus phosphate fertilizers contain significant amounts of U-238, Th-232, K-40 and Ra-226. These can leach out from the fertilizers used in large quantities for resupplying essential nutrients in the soil. From the soil they can enter into the food chain through plants. Due to this they thereby increasing the internal radiation dose of the affected population.

In our recent studies, the radiological risk (Th-232, U-238) of phosphate fertilizers their leaching behaviours were investigated.

In this part of the study we made full explorations, where we analysed the concentration of the Po-210 in fertilizers.

In the study 8 samples were selected and different techniques (gamma and alpha spectrometry), were used to measuring the leachability.

To characterize the leaching behaviour, two leaching procedures were applied and compared the MSZ 21470-50 and the Tessier five-step sequential extraction method.

According to the results, the activity concentration K-40, Ra-226, 34.4-6959.8 Bq / kg, 2.1-229.2 Bq / kg. of was respectively. Th-232 ranges from 0.4 to 11.3 Bq / kg, while U-238 varies from 0.7 to 40.5 Bq / kg. Based on gamma spectrometric results, the Raeq radiation hazard index were calculated, Raeq is between 145.4 and 694.4 Bq / kg in the fertilizers.

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PE 78/2018

