LEVELS OF RADIOACTIVITY OF NATURAL RADIONUCLIDES AND ^{137}Cs ON CHARACTERISTIC LANDFILLS AND NATURAL HABITATS IN SERBIA

Milena Zivkovic[1], Dragana Krstic[1], Tatjana B. Miladinovic[2], Filip Grbovic[1], Snezana Brankovic[1]
[1] University of Kragujevac, Faculty of Science, R. Domanovica 12, 34000 Kragujevac, Republic of Serbia
[2] Institute for Information Technologies, University of Kragujevac, Jovana Cvijića bb, 34000 Kragujevac, Serbia

Corresponding author e-mail address: milena.zivkovic@pmf.kg.ac.rs (M. Zivkovic)

ABSTRACT:

Recent studies show us that contamination of environment is usually done by estimating soil and grass. Radionuclides in natural and anthropogenicaly degraded habiats are major human and environmental health concern. The goal of this research was to determine the levels of natural and artificial radioactivity in 8 different habitats – 4 samples of substrate of characteristic landfills in Serbia (fly ash deposits, asbestos tailings, lead-zinc-copper mine flotation tailings and sludge dumps) and 4 natural habitats with minimal anthropogenic distrubance (1 sandstone habitat and 3 different salt marshes). A gamma spectrometry was used to measure the activity concentrations. Mean values of specific activity ^{40}K, ^{226}Ra, ^{232}Th and ^{137}Cs were: 392.3 Bq.kg^{-1}, 37.9 Bq.kg^{-1}, 39.0 Bq.kg^{-1} and 13.3 Bq.kg^{-1} respectively. The absorbed dose rate and annual effective dose were calculated. These doses are not harmful to the general public's health.

Keywords: *landfill, gamma spectrometry, radioactivity concentrations*

---

1. INTRODUCTION

The main naturally occurring radionuclides are ^{238}U, ^{232}Th and their decay products, as well as radioactive isotope of potassium, ^{40}K. Gamma radiation emitted from those radionuclides deposited on the ground, is the main external source of irradiation of the human body and acts as a medium of migration for transfer of radionuclides to the biological systems. Natural environmental radioactivity and the associated external exposure due to gamma radiation depend primarily on the geological and geographical conditions, and appear at different levels of radionuclides in the soil of each region in the world [1]. Studying about functions and behavior of natural and anthropogenic radionuclides is extremely important. It gives us the information that helps us apply those radionuclides and prevent the harm to health of human beings and the pollution of the environment [2]. Geographical and geological conditions such as the level of soils of

different regions are the main causes of the different distribution of natural radioactivity. As a result of the anthropogenic activity of one of the most important artificial radionuclides, ^{137}Cs, with a half-life of 30.1 years, has a strong affinity for binding to the soil and is therefore a permanent environmental pollutant [3].
Landfills and waste deposists pose a potential risk to environment and human health because the are very close to rivers, sttlelements, agriguclтural fields and pastures. The aim of this paper is to determine levels of radioactivity in 8 different habitats – four samples of substrate of characteristic landfills in Serbia (fly ash deposits, asbestos tailings, lead-zinc-copper mine flotation tailings and sludge dumps) and four natural habitats with minimal anthropogenic distrubance (one sandstone habitat and three different salt marshes) which represents control sites. A special purpose of this study was to determine the potentially radioactivity of waste materials whose use is planned in construction industry projects in Serbia (such as fly ash).

2. MATERIALS AND METHODS

2.1. Sampling and preparation of soil material

Sampling was realized in the period from October to November 2020 on the 8 different habitats in Serbia, that are located on different geological parent material. Formation of land on these geological substrates are conditioned by a combination of different environmental and pedological factors, as well as significant anthropogenic influences, so that they condition the appearance of different types of soil. Map of the researched area with localities sampling is given in the following figure (Fig. 1).

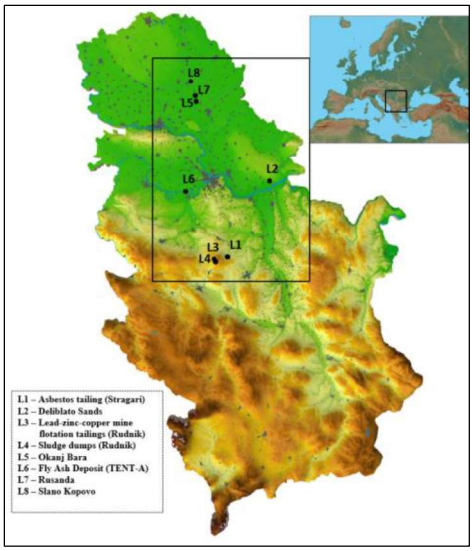

Fig. 1. Studied sites on map of Serbia

Before sampling began, a layer of land that was covered with vegetation and other material was removed superficially. Profile the land of dimensions 1 m × 1 m was then opened with an ax, after which they were separated in layers 5 cm thick (from the surface to a depth of 20 cm) using a spatula, applying the template method [4]. In order to ensured the representativeness of the sample, the land was taken from the vertices of squares and sections diagonals from which a composite pattern is formed. Approximately 1 kg of soil of the sample was packed in a plastic bag and transported to the laboratory. All collected soil samples were additionally cleaned of residual foreign material after being delivered to the laboratory. The samples were dried in a thin layer for 15 days in the open air, and then to constant mass in an oven at 105° C for 1 h. After that, the samples were crushed mechanically by means of a glass mortar. In order to achieve a uniform size particles, soil samples were sieved using a pore sieve with a diameter of 2 mm. For radionuclide determination, samples were placed in marinella containers. Geometry of polypropylene with a polyethylene lid, volume 450 mL. Before placing the samples in the marinade of the vessel, the mass of the empty vessels with the lid was measured, a then the total mass of the samples in the covered vessel from whose difference the mass was obtained sample.

2.2. Gamma spectrometry analysis

Gamma spectrometers are used to investigate radioactivity in the environment and to quickly detect radioactive contamination in a wide area of the ground and estimate activity concetrations from radionuclides. Samples were hermetically sealed in 450 mL Marinelli beakers and kept aside for more than 4 weeks, so that radon gas (^{222}Rn, the half-life 3.825 days) could not escape to avoid disequilibrium problems between ^{226}Ra and its short-lived progeny (^{214}Pb and ^{214}Bi). The activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K, and ^{137}Cs were measured using a coaxial HPGe detector (GEM30-70, ORTEC) with a relative efficiency of 30% and an energy resolution (FWHM) of 1.85 keV at 1.33 MeV (^{60}Co). The computer software MAESTRO 2 was used to perform the spectrum analysis.

2.3. Radiation hazards

The absorbed dose rate is a direct relationship between the radioactivity concentrations of radionuclides and their exposure. The formula (2) was used to measure the mean activity concentrations of ^{226}Ra, ^{232}Th, and ^{40}K (Bq.kg^{-1}) in the given samples at 1 m above the ground surface [1]:

$$D(nGy \cdot h^{-1}) = 0.462C_{R_a} + 0.604C_{T_h} + 0.0417C_K \quad (1)$$

where, D is the absorbed dose rate in nGy.h^{-1}, while C_{Ra}, C_{Th} *and* C_K are the activity concentrations of ^{226}Ra, ^{232}Th and ^{40}K, respectively.

The annual effective dose was calculated by employing the conversion coefficient of 0.7 Sv.Gy^{-1}:

$$D_E(\mu Sv) = 0.7 \cdot D \cdot t \cdot p \quad (2)$$

In the previous formula, t represents the annual exposure time (8760 h) while p is the outdoor occupancy factor of 0.2 for time spent outdoors, implying that 20% of time is spent outdoors [1].

3. RESULTS AND DISCUSSION

In this paper, a gamma spectrometric method of eight soil samples in the Republic of Serbia was performed. The results of the specific activity of natural radionuclides and ^{137}Cs are presented in Table 1.

Table 1. Specific activities of radionuclides in selected soil samples

Sample	^{226}Ra (Bq.kg^{-1})	^{232}Th (Bq.kg^{-1})	^{40}K (Bq.kg^{-1})	^{137}Cs (Bq.kg^{-1})
L1	5.7 ± 0.2	1.6 ± 0.1	13 ± 1	16.5 ± 0.8
L2	17.3 ± 0.9	19.3 ±1.0	380 ± 20	8.1 ± 0.4
L3	37.4 ± 1.9	43.7 ± 2.2	660 ± 40	-
L4	16.6 ± 0.8	63.6 ± 3.2	150 ± 10	-
L5	26.7 ± 1.3	30.4 ±1.5	470 ± 20	42.7 ± 2.1
L6	117.1 ± 5.8	62.2 ± 3.1	430 ± 20	1.5 ± 0.1
L7	38.9 ± 1.9	39.3 ± 2.0	510 ± 30	4.5 ± 0.2
L8	50.6 ± 2.5	51.8 ± 2.6	540 ± 30	6.7 ± 0.3
Min	5.7	1.6	13.0	1.5
Max	117.1	63.6	660.0	42.7
Mean	37.9	39.0	392.3	13.3
SD	32.8	20.0	198.6	13.9

The specific activity was ranged as follows: ^{40}K> ^{232}Th> ^{226}Ra> ^{137}Cs. According to the results from the Table 1, it can be seen that the activity concentration of the mentioned radionuclides in the following range: activity ^{226}Ra in landfill samples is in the range of 5.7–117.1 Bq.kg^{-1}; ^{232}Th activity is in the range of 1.6 to 63.6 Bq.kg^{-1}; value specific activity ^{40}K is from 13.0 to 660.0 Bq.kg^{-1} and ^{137}Cs values are in the range of 1.5 to 42.7 Bq.kg^{-1}. Cesium was formed as a consequence of atmospheric nuclear tests and especially from the Chernobyl accident. Biologically important radionuclides, ^{137}Cs, has a strong afnity for binding to the soil grains making them the permanent environmental pollutants [5].

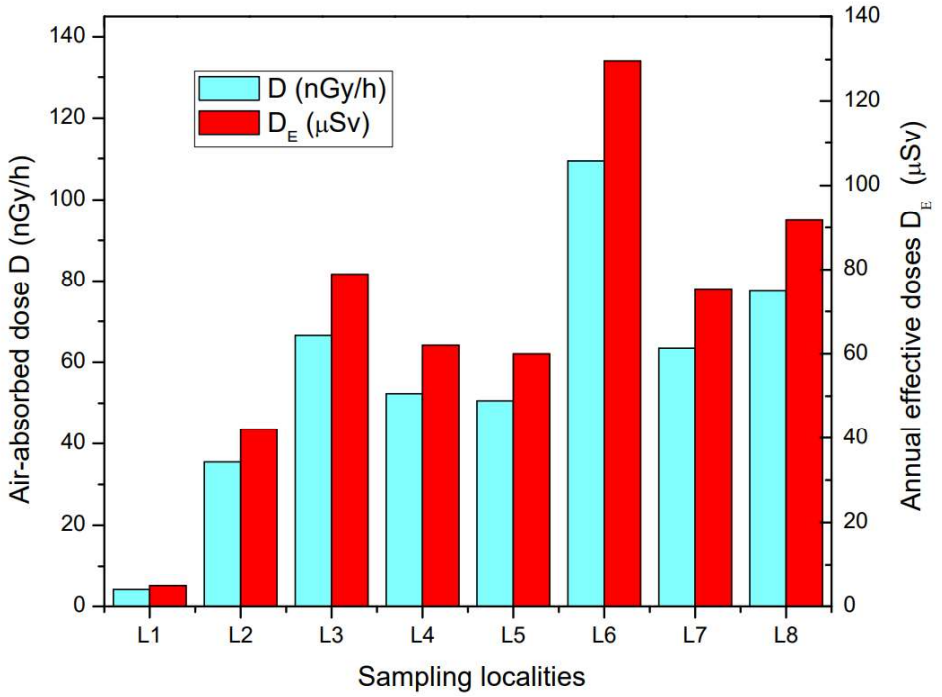

Fig. 1. Air-absorbed dose rates and annual effective doses for eight locality

The absorbed dose rate in air fluctuates in a range from 4.1 to 108.5 nGy/h with a mean value of 57.7 nGy.h^{-1}. This value exceeds the recommended international levels of 59 nGy.h^{-1}. The value of the annual effective dose varies and may be from 5.1 to 130.1 μSv. The mean value is 70.8 μSv and it is greater than the annual average worldwide value of 66 μSv for external exposure to natural terrestrial sources of radiation [2].

The results of this study indicate that there are no significant difference in the content of radionuclides in natural (control) habitats (sandstones and salt marshes) and anthropogenically modified habitats (mine flotation tailings, sludge dumps, fly ash deposits, asbestos tailings). However, it is clear that fly ash (L6) had the highest content of radionuclides in relation to other substrates in different habitats [7]. indicated that fly ash created in the Nikola Tesla TPPs boilers was characterised by the increased concentration of the natural radionuclides content compared to coal – so called technologically enchaced natural radioactivity of industrial waste.

Based on the observations of this study, one may conclude that this studied region does not display a notable health threat. However, special care must be taken in case of using larger amounts of fly ash in conctruction industry in Serbia, due to potential radioactivity of this material.

4. CONCLUSION

Natural and artificial radioactivity levels of eight selected localities in Serbia were investigated using gamma spectrometry. The average values of specific activities of ^{226}Ra, ^{232}Th, ^{40}K and ^{137}Cs were 37.9 Bq.kg^{-1}, 39.0 Bq.kg^{-1}, 392.3 Bq.kg^{-1}, and 13.3 Bq.kg^{-1}, respectively. As a result, the study shows that the tested samples have no noticeable radioactivity except for 40K. The absorbed dose rates and the annual effective doses, radium equivalent activities, external hazard indexes were also estimated with the mean values of 57.7 nGy.h^{-1} and 70.8 μSv, respectively.

4. LITERATURE

[1] Unscear. (2008), United Nation Scientific Committee on the Effects of Atomic Radiation to the General Assembly. United Nations, New York.

[2] Monged, M.H.E., Hassan, H. B., & El-Sayed, S. A. (2020). "Spatial Distribution and Ecological Risk Assessment of Natural Radionuclides and Trace Elements in Agricultural Soil of Northeastern Nile Valley, Egypt", *Water, Air, & Soil Pollution*, Vol. 231, No. 7, 231-338.

[3] Keum, D.K., Lee, H., Kang, S.H., Jun, I., Choi, Y.H., Lee, C.W. (2007), "Predicting the transfer of 137Cs to rice plants by a dynamic compartment model with a consideration of the soil properties", *Journal of Environmental Radioactivity*, Vol. 92, No. 1, 1-15.

[4] IAEA. (2004), Soil sampling for environmental contaminants. IAEA-TECDOC-1415 92–0–111504–0. https://www.iaea.org/search/google/Publication%202004

[5] Yamaguchi, N., Eguchi, S., Fujiwara, H., Hayashi, K., & Tsukada, H. (2012), "Radiocesium and radioiodine in soil particles agitated by agricultural practices: Field observation after the Fukushima nuclear accident", S*cience of The Total Environment,* Vol. 425, 128-134.

[6] Al-Alawy, I.T., Nasser, H.J. & Mzher, O.A., (2020), "Natural radionuclide activity concentrations and radiological hazard in tea, coffee, wheat flour, and powder milk consumed in Iraqi markets", *2ND INTERNATIONAL CONFERENCE ON MATERIALS ENGINEERING & SCIENCE (IConMEAS 2019),* Available at: http://dx.doi.org/10.1063/5.0000459.

[7] Kisić, D.M., Miletić, S.R., Radonjić, V.D., Radanović, S.B., Filipovic, J.Z., & Gržetić, I. A. (2013), "Natural radioactivity of coal and fly ash at the Nikola Tesla B TPP.", *Hemijska industrija*, Vol. 67, No. 5, 729-738.