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RADIOACTIVITY OF THE SOIL IN VOJVODINA (NORTHERN PROVINCE OF SERBIA)

Željko Mihaljev[1]*, Dragana Ljubojević[1], Miroslav Ćirković[1], Milica Živkov-Baloš[1], Sandra Jakšić[1], Brankica Kartalović[1], Nadežda Prica[1]

1 Scientific Veterinary Institute "Novi Sad", Novi Sad, Serbia
* Corresponding author: zeljko@niv.ns.ac.rs

Abstract

Peaceful uses of nuclear energy (nuclear weapons testing, nuclear reactor accidents, industrial and medical use of radioactive compounds) and application of phosphate mineral fertilizers in agricultural production lead to substantial environmental contamination. Land contaminated with radionuclides represents the first link in the food chain and hence the radioactive contamination of crop and livestock production. To determine the soil levels of natural radionuclides the samples were collected from 11 localities in the territory of Vojvodina. The measurements were performed applying inductively coupled plasma with mass spectrometry. The values for concentration of radionuclide activity measured in the examined samples do not significantly diverge from standard values for agricultural soils.

Key words: natural radionuclides, soil, ICP-MS

Introduction

Natural radioactivity implicates presence of radioactive elements that have been present in nature since the formation of Earth and the very beginning of formation of its entire living world (biosphere). Abundant researches revealed substantial differences in the levels of natural radionuclides between particular localities at Earth's surface, and their amounts vary according to the locality (Mitrović et al., 1996). The largest source of radiation activity of the soil is a natural radionuclide potassium-40 (Dželalija, 2006). Some other natural radionuclides, which have always been present on Earth, include also uranium-235, uranium-238, thorium-232, radium-226 and radon-222 (Levant, 1996). The exposure to low-level radiation originating from these natural elements has been always affecting all living beings on Earth, and it is considered background radiation or natural phon.

The basic component of the biosphere is lithosphere (Earth's crust), which represents the first link of the ecology chain: soil-vegetation-animals-man. The soil plays a crucial role in the process of radionuclide distribution and transfer, thus, knowing of radioactive contamination of the soil is of great importance for radiation safety issues in biotechnology (Petrović et al., 1994). Radioactive contamination of the soil is either natural (formed without man's activity) or human produced (nuclear testing, reactor accidents, radioactive waste) (Dobrić et al., 2006).

The "technologically enhanced naturally occurring radioactivity" is attributed to uranium, which is brought to the environment through diverse technological procedures and agro technical measures (Rajković, 2001). Technological development resulted in substantial increase of natural soil radioactivity, predominantly through intensive application of agro-technical measures based on the use of artificial phosphate fertilizers that contain substantial amounts of natural uranium. Moreover, mining of ore containing heavy metals (zinc, copper, led) from deep layers of lithosphere and processing thereof, as well as the thermal power stations producing huge amounts of solid waste (ash, cinder) that contain natural radionuclides such as uranium, thorium and their prodigy (Kisić et al., 2013) increase the levels of soil radioactivity. Uncontrolled application of phosphate fertilizers

implicates substantial potential for undermining of ecological balance as they represent the most powerful source of ^{238}U and ^{232}Th content in the soil, and hence in plants and other links within the food chain. In a superphosphate, uranium is deposited as highly water-soluble uranyl sulphate - $UO_2(SO_4)$ and urano-sulphate - $U(SO_4)$ (Mitrović et al., 2011). Contrary to other radioisotopes, radioactive decay of ^{238}U and ^{232}Th results in formation of the series of unstable nuclei, ^{226}Ra and ^{222}Rn being the most dangerous members of uranium chain. ^{226}Ra is a long half-life α-emitter, manifesting affinity for accumulating in bones, while gaseous ^{222}Rn is responsible for internal irradiation of lung tissues. Emissions of those radionuclides into the environment represent potential significant risk factors for the exposure of local inhabitants to ionizing radiation, as well as for increased levels of natural radiation in particular regions.

Involvement of particular radionuclides into biological cycle is associated with plants ability to absorb radioactive elements from the soil via their root systems. The amount of radioactive material from the soil absorbed by plants is directly proportional to the pollution emission density at particular territory (Simić M., 2001). The transfer of radionuclides from the soil into the plants is substantially dependant on the soil type, that is, its physico-chemical properties that significantly affect the resorption rate of radioactive material. The most important physico-chemical properties include chemical composition (concentration of minerals and content of organic matter), structure (mechanical composition), pH, moisture content and crop density.

Material and Methods

Soil samples were collected from 11 localities in the territory of Vojvodina (Table 1) during 2014. All samplings (at each of 11 localities) were performed by collecting soil samples from 10-15 different points at the total surface of $100m^2$. The samples were collected only from flattened soil surface at the depth of 10-20 cm. The majority of soil samples were of "chernozem"-type (Živković et al., 1972). The samples were dried at 105^0C until reaching the constant mass (IAEA, 1989). Subsequently, removal of mechanical impurities (mainly stones and plant particles) has been performed. Dried soil samples were mechanically crushed (ground) to obtain fine powder. The amount of 1g of homogenized sample was weighed and decomposed in HNO_3 and H_2O_2 mixture using the wet digestion method and the system Ethos, Microwave Labstation, Milestone. Uranium content was determined using Agilent 7700x Series ICP-MS, and data analysis was performed by MassHunter Workstation software.

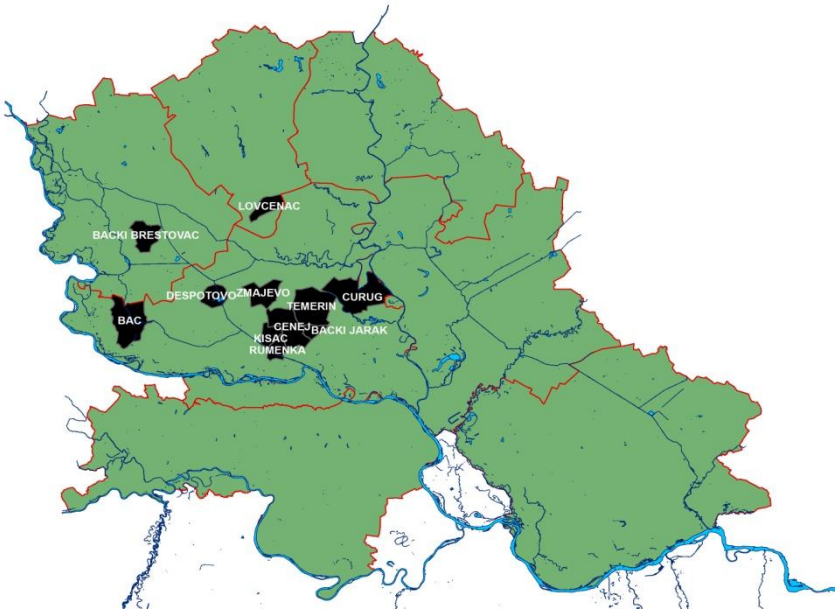

Figure 1. Geographical view of sampling locations

The activity levels of uranium-235 and uranium-238 in soil samples were determined according to total uranium concentration using mass activity values 0.570 Bq/mg U for ^{235}U and 11.10 Bq/mg U for ^{238}U (Eisenbud, 1973). Potassium content was determined using the method of emission spectrophotometry on Spectr AA–10, manufactured by Varian, at wavelength 766.5 nm and using cesium as the ionization-suppressor. The soil levels of potassium-40 activity were calculated from total potassium, using the mass activity value for potassium being 31.561 Bq/g K (Eisenbud, 1973).

Results

The soil in Vojvodina region undergoes radioactive contamination from diverse sources. Above all, emissions from nuclear plant reactors in neighboring regions cause contamination of the air and water in a wide area. Intensive application of phosphate fertilizers with high uranium content potentially causes gradual increase in the activity level of uranium chain in the soil. Moreover, potential contamination of the soil in Vojvodina with depleted uranium caused by NATO bombing in 1999 is widely accepted public opinion. All aforementioned factors strongly suggest the importance of comprehensive assessment of the soil radioactivity status in Vojvodina (Bikit et al., 2010). Table 1 displays the results on potassium and uranium concentrations, as well as the activity levels of natural radionuclides ^{40}K, ^{235}U and ^{238}U.

Table 1. Contents of potassium and uranium and concentration of potassium-40, uranium-235 and uranium-238 activity in soil samples

No.	Locality	K content [g/kg]	^{40}K activity [Bq/kg]	U content [mg/kg]	^{235}U activity [Bq/kg]	^{238}U activity [Bq/kg]
1.	Bač	16.37 ± 0.54	516.6 ± 17.0	4.76 ± 0.24	2.71 ± 0.14	52.84 ± 2.64
2.	Zmajevo	15.08 ± 0.24	476.1 ± 17.6	3.55 ± 0.18	2.02 ± 0.10	39.40 ± 1.97
3.	Despotovo	10.91 ± 0.06	344.3 ± 12.1	3.36 ± 0.17	1.91 ± 0.10	37.30 ± 1.86
4.	Temerin	15.92 ± 0.30	502.4 ± 19.6	2.65 ± 0.13	1.51 ± 0.08	29.42 ± 1.47
5.	Čenej	11.40 ± 0.29	359.8 ± 19.4	3.12 ± 0.16	1.78 ± 0.09	34.63 ± 1.73
6.	Lovćenac	12.84 ± 0.17	405.4 ± 15.3	3.89 ± 0.19	2.22 ± 0.11	43.18 ± 2.16
7.	Čurug	17.72 ± 0.58	559.3 ± 18.6	3.06 ± 0.15	1.74 ± 0.09	33.97 ± 1.70
8.	Kisač	21.88 ± 0.35	690.6 ± 11.0	2.87 ± 0.14	1.63 ± 0.08	31.86 ± 1.59
9.	Bački Brestovac	17.57 ± 0.33	554.5 ± 10.5	2.80 ± 0.14	1.60 ± 0.08	31.08 ± 1.55
10.	Bački Jarak	22.03 ± 0.42	695.3 ± 13.2	2.76 ± 0.14	1.57 ± 0.08	30.64 ± 1.53
11.	Rumenka	23.49 ± 0.45	741.4 ± 14.1	3.24 ± 0.16	1.85 ± 0.09	35.96 ± 1.80
Average value ± SD		16.84 ± 4.28	531.4 ± 134.9	3.28 ± 0.62	1.87 ± 0.35	36.39 ± 6.83

Discussion and Conclusion

The results displayed in Table 1 revealed potassium contents in the examined soil samples ranging from 10.91 to 23.49 g/kg with an average value for all localities being 16.84±4.28 g/kg. These values correspond with the potassium-40 activity concentration in a range 344.3 – 741.4 Bq/kg, with an average activity level for all localities being 531.4 ± 134.9 Bq/kg. The obtained levels of K-concentration and ^{40}K activity are considered common values for the soil and are in full accordance with the results of other authors (Bikit et al., 1990). One can also conclude that potassium-40 is predominant natural radionuclide in the soil as compared to other radionuclides. Obvious variations in potassium content in soil lead to the conclusion that incorporation of radionuclides from the soil into the plants largely depend on pedological properties, soil exploitation, agrotechnical treatments as well as on the type of plants, what is in accordance with the results of our previous researches (Ćupić et al., 2005; Mihaljev et al., 2011).

Uranium contents in soil samples were uniform, ranging within an interval 2.65-4.76 mg/kg with an average value for all localities being 3.28 mg/kg (Table 1). Based on these values, the following activity concentrations for uranium were calculated: ^{235}U = 1.51-2.71 Bq/kg, average value 1.87 ± 0.35 Bq/kg and ^{238}U = 29.42-52.84 Bq/kg, average value 36.39 ± 6.83 Bq/kg. Similar results on uranium concentration in the soil and the activity concentration for radioactive uranium-238 were reported by other authors (Skipperud et al., 2011).

The importance of natural radionuclides as potential pollutants in agricultural and livestock production is addressed in the latest addendum to the Ordinance on the limits of radionuclides in drinking water, food, animal feed, drugs, general use items, building materials and other goods that are placed on the market (Official Gazette of RS, No. 97/13). This addendum introduced the new article, 11a, establishing limits for the contents of natural radionuclides (^{238}U, ^{226}Ra, ^{40}K) in mineral and phosphate fertilizers as potential sources of radioactive contamination of soil.

We also may conclude that ICP-MS (inductively coupled plasma mass spectrometry) proved very sensitive method for quantitative determination of uranium concentration in soil samples (Sahoo et al., 2011). It is therefore an attractive alternative for monitoring uranium because can detect the normal environmentally caused concentrations (Schramel et al., 1997).

Systematic monitoring of radioactivity level of agricultural soil is of paramount importance in Vojvodina, as the region with extremely high potential for production of safe food. Soil contamination with wet or dry atmospheric precipitation and materials characterized by technologically increased level of natural radioactivity can represent a permanent reservoir of radionuclides that significantly contribute to the overall radiation exposure and total population radiation dose.

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