J Radioanal Nucl Chem (2015) 303:2505–2509
DOI 10.1007/s10967-014-3620-1

Radioactivity in fertilizers and radiological impact

Nataša Todorović · Ištvan Bikit · Miroslav Vesković · Dušan Mrdja · Sofija Forkapić · Jan Hansman · Jovana Nikolov · Kristina Bikit · Miodrag Krmar

Received: 19 June 2014 / Published online: 19 September 2014

Abstract The contents of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in 33 fertilizers and 50 soil samples from Vojvodina region, Serbia, were measured by low level gamma spectrometry. The obtained results showed that the averages of radiation hazard parameters for the fertilizers are higher than acceptable level for radium equivalent activity (Ra_{eq}), representative level index (I_γ), external hazard index (H_{ex}) and absorbed dose rate (D) [1]. Based on the measured values of the activity concentration of radionuclides in soils, the activity concentrations of radionuclides for most commonly grown crops in Vojvodina were calculated.

Keywords Fertilizers · Gamma spectrometry · Dose assessment · Transfer factors

Introduction

Man-made and natural sources of radiation are the cause of radiation that human beings are continuously exposed to [2]. The level of activity concentration of radionuclides in phosphate fertilizer and soil samples provides useful information in the monitoring of environmental contamination [3]. The primordial radionuclides of interest in soil and fertilizers are ^{40}K, ^{238}U, ^{232}Th and the progeny of the uranium and thorium decay series [4, 5]. Processes like mining and transportation of phosphate ores, production and usage of phosphate fertilizers contribute to enhanced exposure of workers, public and the environment to these radionuclides [6].

Radium equivalent activity is an index that has been introduced to represent the specific activities of ^{226}Ra, ^{232}Th and ^{40}K by a single quantity, which takes into account the radiation hazards associated with them [7]

$$Ra_{eq} = C_{Ra} + 1.43C_{Th} + 0.077C_{K} \quad (1)$$

Where C_{Ra}, C_{Th} and C_K are the specific activities of ^{226}Ra, ^{232}Th and ^{40}K in Bq kg^{-1}. The external hazard index is defined as:

$$H_{ex} = (C_{Ra}/370) + (C_{Th}/259) + (C_K/4810) \le 1 \quad (2)$$

Representative level index I_γ used to estimate the levels of γ-radiation hazard associated with the natural radionuclides in specific materials is defined as [8]:

$$I_\gamma = (C_{Ra}/150) + (C_{Th}/100) + (C_K/1500) \quad (3)$$

The total air absorbed dose rate (nGy h^{-1}) due to the mean activity concentrations of ^{238}U, ^{232}Th and ^{40}K (in Bg kg^{-1}) can be calculated using the formula (1), [9]:

$$D = 0.429C_U + 0.666C_{Th} + 0.042C_K \quad (4)$$

To estimate the annual effective doses, account must be taken of the conversion factor from absorbed doses in air to effective dose and the outdoor occupancy factor (1), [10]:

$$\text{Annual effect. dose}\left(\text{mSvy}^{-1}\right) = \text{Dose rate}\left(\text{nGy h}^{-1}\right) \times 8760\,(\text{h}) \times 0.2 \times 0.7\left(\text{Sv Gy}^{-1}\right) \quad (5)$$

The soil-to-plant transfer factor (TF), or the ratio of the concentration of radioactivity in the crop to the radioactivity per unit mass of the soil, is a value used in evaluation studies on the impact of releases of radionuclides into the environment. Transfer factor is retained and used for the

N. Todorović (✉) · I. Bikit · M. Vesković · D. Mrdja · S. Forkapić · J. Hansman · J. Nikolov · K. Bikit · M. Krmar
Department of Physics, Faculty of Sciences, University of Novi Sad, Trg Dositeja Obradovica 4, 21000 Novi Sad, Serbia
e-mail: natasa.todorovic@df.uns.ac.rs

Fig. 1 Sampling locations of agriculture soil from Vojvodina region

Table 1 Mean values, standard deviations, minimum and maximum activity concentrations of radionuclides in fertilzers samples

Radionuclide	C_{av} (Bq kg^{-1})	σ_{av} (Bq kg^{-1})	Range C (Bq kg^{-1})
^{40}K	569	134	43–10,800
^{226}Ra	43	11	4–452
^{232}Th	42	20	8–226
^{238}U	35	8	31–2,640

Table 2 Mean values, standard deviations, minimum and maximum values for radium equivalent activity (Ra_{eq}), representative level index (I_γ), external hazard index (H_{ex}) and absorbed dose rate (D) in fertilzers samples

Radiation hazard parameter	Mean values	σ_{av}	Range
Ra_{eq} (Bq kg^{-1})	428	277	19–1,167
I_γ	3.5	2.3	0.14–10.1
H_{ex}	1.2	0.7	0.05–3.2
D (nGy h^{-1})	287	226	13–1,133

application of fertilizers at rates used in routine agriculture [11, 12].

Since the Vojvodina region in Pannonian Plain is agricultural region, the contents of natural radionuclides (^{226}Ra, ^{232}Th and ^{40}K) in 33 fertilizers samples and in 50 soil samples were measured by low level gamma spectrometry. Based on the measured values of the activity concentration of radionuclides in soils and TF given in IAEA TRS 472 [12] for most commonly grown crops in Vojvodina, the activity concentration of radionuclides in plants were calculated.

Materials and methods

Thirty three samples of fertilizers were collected from importer and local markets and tested for radioactivity by low-level gamma-spectrometry. A typical sample weight was about 400 g. After homogenization, they were transferred to sample holders, cylindrical containers (67 mm diameter and 62 mm height), and sealed. Measurements started at least a month after sealing in order to ensure radon equilibrium. Fifty soil samples from Vojvodina

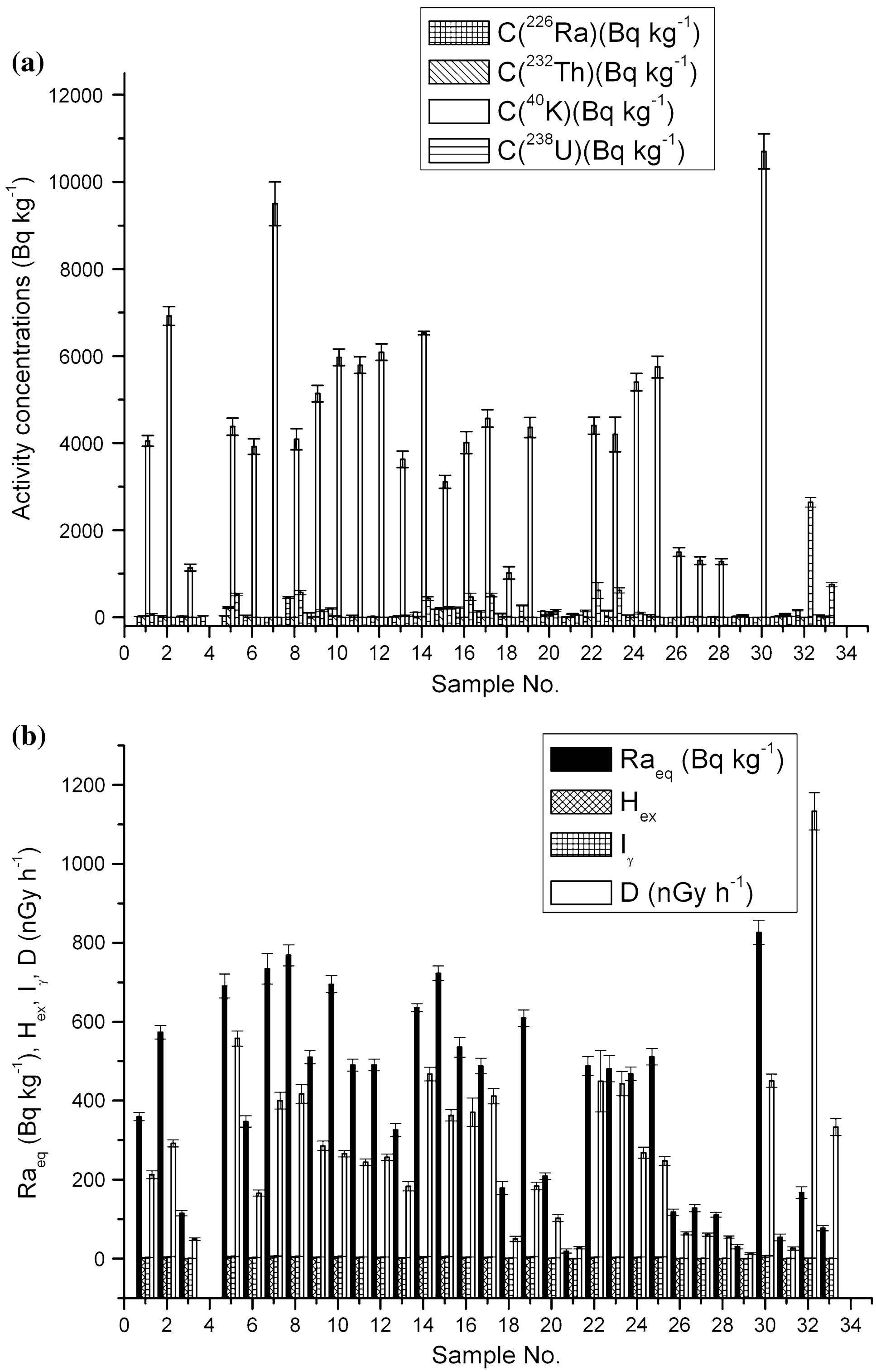

Fig. 2 **a** Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{238}U in fertilizer samples **b** Ra_{eq}, H_{ex}, I_{γ}, D in fertilizer samples

region were collected. Sampling locations are presented on Fig. 1. From each location of an approximately 10 × 10 m area, ten subsamples were collected. The sub sample material was thoroughly mixed and homogenized. The homogenized soil samples were dried at 105 °C to constant mass and transferred to sample holders.

The radionuclide content of the samples was measured using the HPGe extended range ORTEC GMX type detector (10 keV–3 MeV) with nominal efficiency of 32 % and resolution of 1.9 keV. The detector was shielded with the cylindrical 12 cm thick lead shield. The gamma spectra were acquired and analyzed using the Canberra Genie 2000 software. All measurement uncertainties are presented at 95 % confidence level. The detector was calibrated by means of a reference radioactive material in cylindrical geometry (NBS Standard Reference Material 4350B). Self absorption effects due to different densities were taken into account using the ANGLE computer code based on the concept of the effective solid angle. Such careful calibration was necessary in order to ensure low calibration error (<10 %) in the low-energy region (below 100 keV) where the strongest analytical lines of ^{234}Th (direct ^{238}U descendant) are located.

Results and discussion

Activity concentrations of ^{226}Ra, ^{232}Th, ^{40}K and ^{238}U, Ra_{eq}, H_{ex}, I_{γ}, D in 33 measured different fertilizer samples (different chemical compositions and different origin) are presented in Tables 1, 2 and Fig. 2. ^{40}K in fertilizer samples is in the range from 43 (12) to 10,800 (300) Bq kg^{-1}, Fig. 2a. The largest activity concentration of ^{226}Ra is (452 (19) Bq kg^{-1}) and the lowest is (4 (3) Bq kg^{-1}), Fig. 2a. The largest concentration of ^{232}Th is (226 (18) Bq kg^{-1}) and the smallest is (8 (3) Bq kg^{-1}), Fig. 2a. ^{238}U in fertilizer samples is in the range from 31 (20) to 2,640 (110) Bq kg^{-1}, Fig. 2a. The obtained results show that the averages of radiation hazard parameters, calculated according to Eqs. 1–5, for most fertilizers samples are higher than acceptable level 370 Bg kg^{-1} Ra_{eq}, 1 for level index $I_{\gamma r}$, the external hazard index $H_{ex} \leq 1$ and 59 nGy h^{-1} for absorbed dose rate, Table 2 and Fig. 2b [1].

The radiations can effect human beings internally through the ingestion of radioactive materials into the body [13]. Therefore it is necessary to determine the content of radionuclides in the environment and calculate the potential radiation risk to humans. Mean values, standard deviations, minimum and maximum activity concentrations of radionuclides in 50 soil samples are presented in Table 3. The accumulation of radionuclides in farm crops varies considerably for different soils. The difference in TFs to farm crops for different soils may be one or two orders of magnitude. Soil properties that are likely to affect radionuclide transfer values include mineralogical and granulometric composition, organic matter content, pH and fertility [12]. Soil fertility, the duration of the vegetative period and the character of the root distribution in soil also influence radionuclide TFs. Radionuclides often transfer in greater concentrations to leaves and stems, and in much lower concentrations to the generative parts of plants [12]. Based on the values of TF given in IAEA TRS 472 [12] for

Table 3 Mean values, standard deviations, minimum and maximum activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U in soil samples

Radionuclide	C_{av} (Bq kg^{-1})	σ_{av} (Bq kg^{-1})	Range C (Bq kg^{-1})
^{40}K	569	70	238–1,000
^{226}Ra	35	8	10–43
^{232}Th	43	11	12–71
^{238}U	42	10	5–80

Table 4 Soil to plant transfer factors TF for for most commonly grown crops in Vojvodina for temperate environment (TF taken from [12])

Isotope	Leafy vegetables TF	Cereals-grain	Maize-grain	Non-leafy vegetables	Pasture	Tubers
^{40}K	1.3	7.4×10^{-1}	–	–	7.3×10^{-1}	–
^{226}Ra	9.1×10^{-2}	1.7×10^{-2}	2.4×10^{-3}	1.7×10^{-2}	7.1×10^{-2}	1.1×10^{-2}
^{232}Th	1.2×10^{-3}	2.1×10^{-3}	6.4×10^{-5}	7.8×10^{-4}	9.9×10^{-2}	2×10^{-4}
^{238}U	2.0×10^{-2}	6.2×10^{-3}	1.5×10^{-2}	1.5×10^{-2}	4.6×10^{-2}	5.0×10^{-3}

Table 5 Calculated average activity concentrations of ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U in some agricultural products

Isotope	C_{av} in soil (Bq kg^{-1})	Leafy vegetables	Cereals-grain	Maize-grain	Non-leafy vegetables	Pasture	Tubers
		C_{av} in plant (Bq kg^{-1})					
^{40}K	569 ± 70	740 ± 91	421 ± 52	–	–	415 ± 51	–
^{226}Ra	35 ± 8	3.2 ± 0.7	0.60 ± 0.14	0.084 ± 0.019	0.59 ± 0.14	2.5 ± 0.6	0.38 ± 0.09
^{232}Th	43 ± 11	0.052 ± 0.013	0.090 ± 0.023	$(2.7 \pm 0.7) \times 10^{-3}$	0.03 ± 0.01	4.3 ± 1.1	$(8.6 \pm 2.2) \times 10^{-3}$
^{238}U	42 ± 10	0.84 ± 0.20	0.26 ± 0.06	0.63 ± 0.15	0.63 ± 0.15	1.9 ± 0.5	0.21 ± 0.05

most commonly grown crops in Vojvodina (Table 4), the activity concentration of radionuclides in plants were calculated and presented in Table 5. During the calculations the average values of TF for all group of soil were used.

Conclusion

In the paper the activity concentrations of the natural radionuclides (^{226}Ra, ^{232}Th, ^{40}K and ^{238}U) in 33 fertilizers samples and in 50 soil samples from Vojvodina region, Serbia, were determinate by low level gamma spectrometry. The obtained results show that the averages of radiation hazard parameters for most fertilizers samples are higher than acceptable level 370 Bg kg^{-1} Ra$_{eq}$, 1 for level index $I_{\gamma r}$, the external hazard index $H_{ex} \leq 1$ and 59 nGy h^{-1} for absorbed dose rate [1]. Activity concentration of ^{40}K in agriculture soil samples is within values characteristic of the region [14]. Its higher concentration can be explained by the use of fertilizers with large content of potassium. Activity concentration of ^{238}U in soil samples are within the typical values of Vojvodina soil [14]. Slight deviations and increased levels have been observed in some samples could be explained by phosphate fertilizers used. Based on the values of TFs [12] for most commonly grown crops in Vojvodina, the activity concentration of radionuclides in plants were calculated. General conclusion of this study is that in soil samples and analyzed agriculture products did not find increase radioactivity, which could threaten food production or give negative influence on human health.

Acknowledgments The authors acknowledge the financial support of the Ministry of Education, Science and Technological Development of Serbia, within the projects No. 171002, No. 43002 and No. 43007.

References

1. UNSCEAR (2000) Sources, effects and risks of ionizing radiation. Report of the United Nations scientific committee on the effects of atomic radiation to the general assembly. United Nations, New York
2. Yalcin P, Taskin H, Kam E, Taskin H, Teryi M, Varinlioglu A, Bozkurt A, Bastug A, Tasdelen B (2012) Investigation of radioactivity level in soil and drinking water samples collected from the city of Erzincan, Turkey. J Radioanal Nucl Chem 292:999–1006
3. Ghosh D, Deb A, Bera S (2008) Measurement of natural radioactivity in chemical fertilizer and agricultural soil: evidence of high alpha activity. Environ Geochem Health 30(1):79–86
4. Tufail M, Sabiha-Javied N, Akhtar J (2010) Assessment of annual effective dose from natural radioactivity intake through wheat grain produced in Faisalabad, Pakistan. J Radioanal Nucl Chem 283:585–590
5. Nenadović S, Kljajević Lj, Nenadović M, Omerašević M, Obradović Arsić D, Lješević M (2011) Vertical distribution of ^{226}Ra and radiological hazards indices of soil samples. J Radioanal Nucl Chem 290:479–484
6. Ajmal PY, Bhangare RC, Tiwari M, Sahu SK, Pandit GG (2014) External gamma radiation levels and natural radioactivity in soil around a phosphate fertilizer plant at Mumbai. J Radioanal Nucl Chem 300:23–27
7. Beretka J, Mathew PJ (1985) Natural radioactivity of Australian building materials, industrial wastes and by-products. Health Phys 48(1):87–95
8. Viruthagiri G, Ponnarasi K (2011) Measurement of natural radioactivity in brick samples. Adv Appl Sci Res 2(2):103–108
9. Ahmed NK, El-Arabi AGM (2005) Natural radioactivity in farm soil and phosphate fertilizer and its environmental implications in Qena governorate, Upper Egypt. J Environ Radioact 84:51–64
10. Hamideen MS, Sharaf J (2012) Natural radioactivity investigations in soil samples obtained from phosphate hills in the Russaifa region, Jordan. Radiat Phys Chem 81:1559–1562
11. Mheemeed AK, Najam LA, Hussein AK (2014) Transfer factors of ^{40}K, ^{226}Ra, ^{232}Th from soil to different types of local vegetables, radiation hazard indices and their annual doses. J Radioanal Nucl Chem 302:87–96. doi:10.1007/s10967-014-3259-y
12. IAEA (2010) Handbook of parameter values for the prediction of radionuclide transfer in terrestrial and freshwater environments. Technical Reports Series No. 472
13. Rajeshwari T, Rajesh S, Kerur BR, Anilkumar S, Krishnan N, Pant AD (2014) J Radioanal Nucl Chem 300:61–65
14. Bikit I, Slivka J, Conkić Lj, Krmar M, Vesković M, Žikić-Todorović N, Varga E, Curcić S, Mrdja D (2005) Radioactivity of the soil in Vojvodina (northern province of Serbia and Montenegro). J Environ Radioact 78(1):11–19