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Full Research Article

PRESENCE OF RADIONUCLIDES AND TOXIC ELEMENTS IN FEEDSTUFFS AND FOOD OF ANIMAL ORIGIN

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Abstract

Introduction. The aim of this study was to determine the content of radionuclides and toxic elements in samples feedstuffs and food of animal origin collected between 2007-2017 from two suburban areas of Belgrade, the municipalities of Palilula and Surčin, both areas with intensive agricultural production.

Materials and Methods. Radionuclides (^{40}K and ^{137}Cs) and toxic elements (As, Cd, Cu, Ni, Pb and Zn) were determined in samples of corn, hay, meat, milk and eggs, by gamma ray spectrometry and inductively coupled plasma spectrometry (ICP-OES, Spectro Genesis).

Results and Conclusions. The obtained results showed that natural ^{40}K was present in all investigated samples. The average activity concentration of ^{40}K was 94 Bq/kg in corn, 117 Bq/kg, 108 Bq/kg and 95 Bq/kg in beef, pork and chicken meat, respectively, as well as 61 Bq/kg in cow's milk and 48 Bq/kg in eggs. Anthropogenic radionuclide ^{137}Cs was not detected. The trend for toxic element levels according to the average concentrations found in the studied feed samples (corn and hay) was as follows: Zn>Cu>Pb>As>Ni>Cd. Arsenic was detected in animal feed in both Belgrade municipalities, with the average concentration being 1.08 mg/kg (0.5-1.37 mg/kg), and in corn, the As content was higher than in hay samples. In food of animal origin, only Zn and Cu were detected. It is concluded that the presence of radionuclides and toxic elements in feedstuffs and foods of animal origin does not pose a health risk for either humans or animals.

Key words: radionuclides, toxic elements, feed, food chain, food of animal origin

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INTRODUCTION

Natural radionuclides and toxic elements are integral parts of the environment. Their concentrations in the environment depend on geological and geographical conditions (Tchounwou et al., 2012, Bradl, 2002). Human activities such as mining and smelting operations, phosphate ore processing, coal ash and industrial discharge, vehicular emissions, atmospheric deposition of particles, biosolids and manures, and the application of soil fertilizers (Czarnecki and Düring, 2015, Wuana and Okieimen, 2011, USEPA, 2000) can enhance concentrations of naturally occurring radionuclides and toxic elements in the environment and foodstuffs.

Potassium-40 is a primordial natural radioactive element (half-life 1.3 x 10^9 years), beta and gamma emitter, and is present in soil, rock and foods. Apart natural radionuclides, artificial radionuclides such as ^{137}Cs are present in the environment as a consequence of nuclear disaster.

Toxic elements are considered environmental pollutants due to their toxic effects on plants, humans and food, particularly in areas with high anthropogenic pressures (Pavlović et al., 2016, Tchounwou et al., 2012, Kabata-Pendias and Mukherjee, 2007). Because of their high degree of toxicity, arsenic, cadmium, chromium, lead, and mercury rank among the priority elements that are of public health significance. Depending on their impact on the health of humans and animals, elements are divided into four groups: highly toxic (Cd, Pb, Hg, As, Cr, Ni and F), less toxic (Sn and Al), non-essential (Ba, Al, Li and Zr) and essential, which can be toxic in high concentrations (Cu, Zn, Mn and Fe) (Kochare and Tamir, 2015).

The aim of this study was to determine the content of natural (^{40}K) and artificial (^{137}Cs) radionuclides, as well as toxic elements (As, Cd, Cu, Ni, Pb and Zn) in corn, hay, meat, milk and eggs from the municipalities of Palilula and Surčin. Some of these toxic elements (As, Cd, and Pb) were selected because they are well established as being toxic for living systems, and their effects on humans have been widely documented (Corguinha et al., 2015, Peralta-Videa et al., 2009, Kabata-Pendias and Mukherjee, 2007).

MATERIALS AND METHODS

Sampling sites

Between 2007-2017, samples of animal feed (corn and hay) and foods of animal origin (beef, pork and chicken meat, cow milk and eggs) were collected from two municipalities in Belgrade (Serbia): Palilula and Surčin. Palilula (sampling locations: Ovča, Borča, and Dunavac) and Surčin (sampling locations: Jakovo, and Bojčin forest) are both areas with intensive agricultural production. These sampling sites were selected on the grounds that they have significant areas of agricultural land and are important food sources for Belgrade residents. Palilula is around 15 km from Pančevo city, where the Pančevo Oil Refinery and *HIP-Petrohemija* are located, making it the main industrial hot spot in Serbia. Surčin was used as a control area, because there are no industrial sources of environmental pollution in this municipality.

Analysis

Activity concentrations of ^{40}K and ^{137}Cs were determined by gamma ray spectrometry using a High Purity Germanium detector (ORTEC) with a relative efficiency of 30% and energy resolution of 1.85 keV (1332.5 ^{60}Co). Analysis of the measured gamma spectra was performed using GAMMA VISION® 32 (Ortec, USA) software. Before analysis, samples were homogenized and put into 1l Marinelli beakers. Counting time for the samples and background was 60,000 s. For efficiency calibration, we used commercially available standards with mixed radionuclides:

- ^{241}Am, ^{133}Ba, ^{109}Cd, ^{139}Ce, ^{57}Co, ^{60}Co, ^{137}Cs, ^{54}Mn, ^{113}Sn, ^{85}Sr, and ^{88}Y, dispersed in silicone resin in a Marinelli beaker, density (0.98±0.01) g/cm^3, volume 1l (for corn, meat, milk and eggs);
- ^{241}Am and ^{152}Eu, dispersed in silicone resin in a Marinelli beaker, density (0.182±0.05) g/cm^3, volume 1l (for hay).

Minimum detectable activity levels were 2.5 Bq/kg for ^{40}K and 0.2 Bq/kg for ^{137}Cs.

For toxic element analysis, subsamples were homogenized to make composite samples. Hay was dried to a constant weight at 65º C and ground in a laboratory mill (Polymix, Kinematica AG, screen size 2.0 mm). Samples of animal origin were placed into labeled plastic bags and stored at -18º C prior to analysis. Sample mineralization was conducted through wet digestion in a microwave (CEM, Microwave 39 MDS- 2000), in advanced composite vessels, by using a 3:1 mixture of nitric acid (HNO_3, 65%) and hydrogen peroxide (H_2O_2, 30%). Total concentrations of As, Cd, Cu, Ni, Pb and Zn were determined through inductively coupled plasma spectrometry (ICP-OES, Spectro Genesis). The minimum detectable concentrations for the analyzed elements were as follows (mg/kg): As – 0.011, Cd – 0.001, Cu – 0.001, Ni – 0.001, Pb – 0.018, and Zn – 0.003.

Statistical analysis

The experimental data were subjected to analysis of arithmetic means and standard deviations using Excel (Microsoft Office Excel 2007).

RESULTS

The activity concentrations of ^{40}K (Bq/kg) in corn, meat, milk and egg samples collected in the municipalities of Surčin and Palilula are presented in Figure 1. The activity concentrations of artificial radionuclide ^{137}Cs in the investigated samples were below the minimum detectable activity (0.2 Bq/kg), so results are not presented. The contents of As, Cd, Cu, Ni, Pb, and Zn in the feedstuffs/foods (mg/kg) are presented in Table 1.

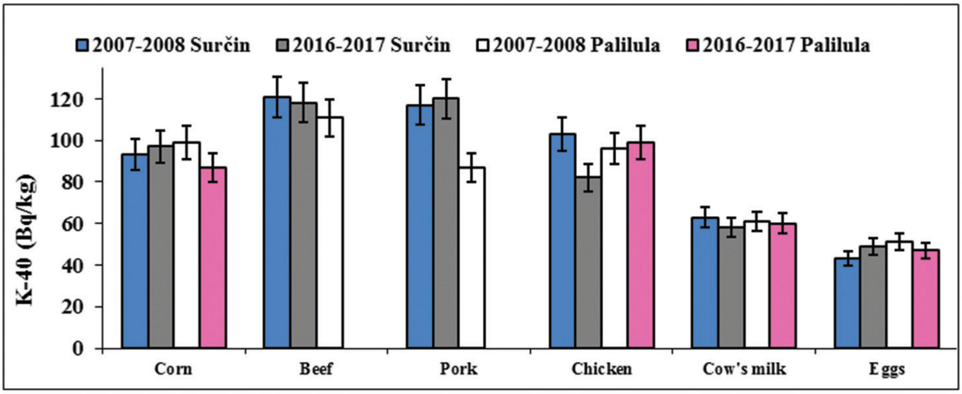

Figure 1. Average activity concentrations of ^{40}K (Bq/kg) in corn, meat, milk and egg samples collected in Surčin and Palilula, Belgrade

Table 1. Toxic element contents (As, Cd, Cu, Ni, Pb, and Zn) in corn, hay, meat, milk and eggs collected during 2006-2017 from the municipalities of Palilula and Surčin, Belgrade. Values are presented as mean ± standard deviation (mg/kg)

Type of feedstuff or food sampled	As	Cd	Cu	Ni	Pb	Zn
			Palilula			
Corn	1.36±0.01	<MDA	4.26±0.32	0.56±0.11	4.01±0.76	20.8±0.9
Hay	0.50±0.04	<MDA	3.31±0.59	0.36±0.07	<MDA	6.04±0.63
Pork	<MDA	<MDA	0.09±0.01	<MDA	<MDA	17.59±0.15
Chicken	<MDA	<MDA	0.65±0.01	<MDA	<MDA	5.69±0.14
Cow's milk and eggs	<MDA	<MDA	<MDA	<MDA	<MDA	<MDA
			Surčin			
Corn	1.37±0.28	<MDA	2.65±0.19	0.48±0.07	2.53±0.25	11.86±0.51
Hay	<MDA	<MDA	2.94±0.12	1.31±0.18	5.94±0.44	11.14±0.59
Beef	<MDA	<MDA	<MDA	<MDA	<MDA	10.89±0.19
Pork	<MDA	<MDA	<MDA	<MDA	<MDA	0.76±0.19
Chicken, eggs and cow's milk	<MDA	<MDA	<MDA	<MDA	<MDA	<MDA

MDA – minimum detectable activity

The results obtained showed that natural ^{40}K was present in all investigated samples (Figure 1). The average activity concentration of ^{40}K was 94 Bq/kg in corn, 117 Bq/kg in beef, 108 Bq/kg in pork, 95 Bq/kg in chicken, 61 Bq/kg in cow's milk and 48 Bq/kg in eggs (Figure 1).

The trend for toxic element levels according to the average concentrations found in the studied feed samples (corn and hay) was as follows: Zn>Cu>Pb>As>Ni>Cd.

Arsenic was detected in animal feed in both Belgrade municipalities, with the average concentration being 1.08 mg/kg (0.5-1.37 mg/kg), and the content of As was higher in corn than in hay samples (Table 1). In foods of animal origin, only Zn and Cu were detected.

DISCUSSION

Radionuclides

Natural radionuclides are normally present in the soil, and ^{40}K is the dominant radionuclide with global average activity concentration of 400 Bq/kg (UNSCEAR, 2000), so the presence of ^{40}K in feed and food is expected. According the IAEA report (2002), the concentration of natural radioactivity in food ranges from 40 to 600 Bq/kg, with ^{40}K being the dominant radionuclide (46 to 649 Bq/kg), while the content of other radionuclides are low in most foodstuffs (0.01-1.16 Bq/kg for ^{226}Ra and 0.02-1.26 Bq/kg for ^{228}Th) (Ramachandran and Mishra, 1989). The present study showed that ^{40}K is present in all samples examined and that its concentration was higher in meat than in milk and eggs (Figure 1).

The anthropogenic radionuclide ^{137}Cs is still present in Serbian soils as a consequence of the Chernobyl nuclear accident (1986) (Mitrović et al., 2016, Mitrović et al., 2014). After the accident, deposition values were 1-10 kBq/m^2 (FCP, 1987) and the activity concentrations of ^{137}Cs in different foodstuffs (milk, meat, fruit and vegetables) in 1986 ranged from 3 to 400 Bq/kg (Popović and Spasić-Jokić, 2006). It was found that the daily intake of ^{137}Cs by the Czech population was about 25 Bq in 1986, but is around 0.1 Bq now (Škrkal et al., 2017). As it happened so far away, the Fukushima nuclear disaster did not lead to contamination with anthropogenic radionuclides in Serbia (Vitorović et al., 2013). The results obtained in this study show that ^{137}Cs is not present in the feed and food examined, as its activity concentration was below the minimum detectable activity.

Toxic elements

For animals, the main route for metal contamination is soil ingestion, while for humans, contamination pathways are inhalation, water and food (Peralta-Videa et al., 2009). Therefore, special attention has been paid to As, Cd, Cr, Hg, and Pb accumulation in plants and animals and their possible transfer to humans via the food web (Corguingha et al., 2015, Jaishankar et al., 2014). Copper, Zn, Mn, Fe, Ni, and Mo are essential micronutrients for plants; however, when present in excess, they can damage the plant or decrease the quality of harvested plant products. Arsenic, Cd, Hg and Pb are non-essential and toxic elements that are almost ubiquitously present at low levels in the environment because of anthropogenic influences. The dietary intake of plant-derived food contributes a major part of potentially health-threatening human exposure to toxic elements, especially to As and Cd (Clemens and Ma, 2016).

Many factors contribute to toxic elements accumulation in animals including behavior, physiology, and diet.

In this study, As was detected only in feed samples, corn and hay, in Palilula and Surčin (Table 1), pointing to the effects of intensive agricultural production, i.e. the use of herbicides and insecticides (Bencko and Foong, 2017), as well as the use of phosphatic fertilizers containing As (Jayasumana et al., 2015). All the feed samples studied contained As at levels within the safe range for animal nutrition according to national regulations (Table 2). In food samples (meat, milk, and eggs), As was not detected (Table 1).

Table 2. Reference values acccording to Serbian legislation for concentrations of toxic elements in food and feedstuffs (mg/kg)

Element	Concentration	
	Food [a] (meat)	Feedstuffs [b]
As	0.1	2
Cd	0.05	0.5
Cu	–	12-250
Ni	–	–
Pb	0-1	10
Zn	–	100-2000

[a]Official Gazette, 23/94 and 28/2011); [b] Official Gazette, 2/90 and 27/90.

Cadmium is often found near sites for metal mining and refining, the production and application of phosphate fertilizers, and waste incineration and disposal. In all investigated feed and food samples, Cd was not detected.

Copper is introduced into the environment through both natural and anthropogenic activities. Natural sources of Cu pollution are volcanic eruptions, windblown dust, and forest fires, while anthropogenic sources include copper mining activities, metal and electrical manufacturing, the agricultural and domestic use of pesticides and fungicides, leather processing, and traffic. An appropriate Cu content in plants is of key importance not only for their health, but also to ensure an appropriate supply of this element to humans and animals. National regulations do not prescribe a norm for levels of this element in foodstuffs of animal origin, while the content of Cu in animal feed was low (Tables 1 and 2).

Nickel was detected in corn and hay from both locations, but Ni was not found in foods of animal origin. The major sources of anthropogenic Ni input are metal processing plants, coal and oil combustion, phosphate fertilizers, and sewage sludge (Kabata-Pendias, 2001; Sridhara Chary et al., 2008). Serbian national regulations (Table 2) do not prescribe a norm for levels of Ni in food or feed.

Lead is found in the earth's crust and has been mobilized in the environment by recent anthropological activities. In samples of animal origin, Pb was not detected, while in animal feeds, concentrations of Pb were lower than the levels permitted for animal feed (10 mg/kg), (Tables 1 and 2).

The main sources of Zn pollution are related to the non-ferric metal industry and to agricultural practices. Some fertilizers, and in particular superphosphate, can contribute significantly to Zn levels in soil (Kabata-Pendias and Mukherjee, 2007). In the studied samples of corn and hay, Zn levels were within, or below, the range for global concentrations for cereal grains, which vary between 18 and 33 mg/kg (Kabata-Pendias and Mukherjee, 2007). Serbian national regulations (Table 2) do not prescribe a norm for levels of Zn in food.

CONCLUSION

Research in suburban areas of Belgrade city was conducted with the aim of determining the content of radionuclides and toxic elements in feed and products of animal origin. These locations contain significant areas of agricultural land used for food production for Belgrade residents. The results obtained show that industrial activities localized in Pančevo city do not have a detrimental effect on agricultural production in Palilula. It is concluded that the presence of the examined radionuclides and toxic elements in feedstuffs and food of animal origin does not pose a health risk for either humans or animals.

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Authors contributions

BM, BV, MM, OK samples collected; BM, BV analyzed samples by gamma spectrometry; MM, OK, VP analyzed samples by coupled plasma spectrometry (ICP-OES, Spectro Genesis); PP, MM, BM prepared manuscript.

Competing interests

The author(s) declare that they have no competing interests.

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RADIONUKLIDI I TOKSIČNI ELEMENTI U HRANI ZA ŽIVOTINJE I HRANI ANIMALNOG POREKLA

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Kratak sadržaj

Uvod. U radu su prikazani rezultati određivanja sadržaja radionuklida i toksičnih elemenata u uzorcima hrane za životinje i hrani animalnog porekla prikupljanih na području grada Beograda, opštinama Palilula i Surčin, u periodu od 2007-2017. godine.

Materijal i metode. U uzorcima kukuruza, sena, mesa, mleka i jaja određivani se radionuklidi (^{40}K i ^{137}Cs) i toksični elementi (As, Cd, Cu, Ni, Pb i Zn) metodama gama

spektrometrije i indukovane kuplovane plazma spektrometrije (ICP-OES, Spectro Genesis).

Rezultati i zaključak. Dobijeni rezultati su pokazali da je u uzrocima hrane za životinje i hrani animalnog porekla ^{40}K bio dominantni radionuklid. Prosečan sadržaj ^{40}K u kukuruzu je bio 94 Bq/kg, 117 Bq/kg u junećem mesu, 108 Bq/kg u svinjskom mesu, 95 Bq/kg u pilećem mesu, 61 Bq/kg u kravljem mleku i 48 Bq/kg u jajima. U hrani za životinje (kukuruz i seno) detektovani su toksični elementi u opadajućem trendu: Zn>Cu>Pb>As>Ni>Cd. Arsen je detektovan u hrani za životinje na oba ispitivana lokaliteta, prosečan sadržaj je bio 1.08 mg/kg (0.5-1.37 mg/kg), u uzroku kukuruza sadržaj arsena je bio veći nego u senu. U hrani animalnog porekla detektovani su Zn i Cu, dok je sadržaj ostalih toksičnih elemenata bio ispod praga detekcije. Na osnovu dobijenih rezultata može se zaključiti da radionuklidi i toksični elementi u hrani za životinje i hrani animalnog porekla ne predstavljaju rizik po zdravlje ljudi i životinja.

Ključne reči: radionuklidi, toksični elementi, hrana za životinje, lanac ishrane, hrana animalnog porekla.