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Editors
Radoje V. Pantovic
Zoran S. Marković

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Eco-Ist'17

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RADIONUCLIDE CONTENT IN PREMIXES IN 2015 AND 2016

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ABSTRACT

As part of monitoring radionuclides in animal feed produced in the Republic of Serbia in 2015 and 2016, the gamma spectroscopy method was used to analyze 35 premix samples. In all analyzed samples activity levels of ^{137}Cs, were within the detection limits of the gamma spectroscopic device to 2.8 Bq/kg. From the viewpoint of the content of the analyzed radionuclides in animal premixed activity levels (Bq/kg) were: $^{137}Cs < 0.2$ to 2.8 Bq/kg, ^{40}K from 11 to 502 Bq/kg, ^{226}Ra from 2.9 to 49 Bq/kg, $^{232}Th < 1.0$ to 13 Bq/kg, and $^{238}U < 3.2$ to 111 Bq/kg.

Key words: animal feed, premix, radionuclides.

INTRODUCTION

Ionization radiation has accompanied the living world since its formation and it is one of the characteristics of the environment. Radionuclides released into nature through different mechanisms enter the human organism from air, water and food and contribute to human irradiation. Based on systematic measurements of natural radionuclides in the soil, food, animal food, air and water different amounts of radionuclides can be registered in them that through food enter the organism of an inhabitant of a certain area.

Cosmic and terrestrial (primordal) radionuclides are natural sources of radionuclides. Cosmic radiation is a type of natural ionization radiation of the earth surface from the cosmos. Terrestrial radionuclides have a half-life comparable with the age of the Earth (4.5×10^9 years) or longer and have been created by nucleosynthesis at the same time as all substances of the Solar system [1,2].

Most radionuclides that exist on Earth can be grouped into three natural radioactive series. They are the uranium-radium series, uranium-actinium series and the thorium series. A fourth neptunium series was created in laboratory conditions by nuclear reactions. Natural radioactive series are created by the breakdown of three radioisotopes ^{235}U, ^{238}U and ^{232}Th that are still present in nature due to their long half-life. Descendants of these isotopes are also unstable so they break down forming new unstable lighter cores, with a stable isotope ending the series. Levels of natural radioactivity at some point on Earth depend on several factors: content of natural radioactive elements in the biosphere, type of cosmic radiation, geological characteristics of the soil, altitude, geographic latitude and longitude. Natural radioactivity levels differ on the Earth from place to place.

Potassium has three isotopes of which only ^{40}K emits radioactive radiation. Potassium-40 is an essential natural radionuclide that is part of the human organism. Depending on the chemical content of the soil, its structure, pH and moisture, radionuclides bond in the soil, and transfer from soil to plants. The ^{40}K radionuclide has a physical half-life ($T_{1/2}$) of 1.25×10^9 years and a biological half-life in the human body (T_b) of 58 days and represents 0.117% of natural potassium[3].

In nature uranium occurs as a mixture of three long-life isotopes: ^{238}U with $T_{1/2}= 4.5\times10^9$ years and representation of 99.28%; ^{235}U with $T_{1/2}=7.1\times10^8$ years and representation of 0.71% and ^{234}U with $T_{1/2}=2.5\times10^5$ years and representation of 0.006%. All natural uranium isotopes dominantly emit alpha particles and their descendants are beta and/or gamma emitters. Regardless of its chemical form and time of acute exposure uranium in an organism acts as a toxicant as it acts as a source of ionization radiation and can also act as a chemically toxic element. The biological half-life of ^{238}U is between 1 and 500 days depending on the mobility of uranium compounds[3].

Radium is a natural radioactive element that has four isotopes of which ^{226}Ra and ^{228}Ra are the products of radioactive breakdown of uranium. Due to a physical half-life of 1600 years, high chemical activity, low degree of elimination from an organism (T_b=7-45 years) its amount in an organism increases with time. In an organism it follows calcium and is one of the more toxic radionuclides with an extremely expressed cancerous action [1,2].

Thorium isotopes: ^{232}Th with a half-life of 1.4×10^{10} years and ^{228}Th with a half-life of 1.9 years have radio-ecological significance[1]. ^{232}Th is the most significant member of the thorium series of radioactive breakdown. Bones, lungs and liver are its main deposition points.

Two of seven radioactive isotopes of cesium are significant for environment pollution: ^{134}Cs ($T_{1/2}$ = 2.06 years) and ^{137}Cs ($T_{1/2}$ =30.17 years). Both radionuclides were present in fuel elements of the nuclear reactor in Chernobyl (1986) from where they were emitted into the environment. Radiocesium most often reaches plants by dry or wet precipitation. The cesium ion is a chemical and biochemical homologue of potassium and follows its metabolism in an organism. It can be ingested by physical or chemical sorption or ion exchange. It is completely soluble in body fluids and is uniformly distributed in the organism. Due to this property there is no critical organ for cesium and it is an organothropic radionuclide. Its physico-chemical characteristics are such that it actively enters the human and animal food chain through plants. The ^{137}Cs anthropogene radionuclide to the greatest extent entered the environment in Serbia after the accident in the Chernobyl nuclear power plant in the Ukraine in 1986. There were no significant deposits of this radionuclide in Serbia after the accident in the Fukushima nuclear power plant in Japan in 2011[4].

Modern biotechnical production represents a very complex food chain starting from plant production and a series of agrotechnical measures for advancing this production, through livestock production. This food chain ends with final biotechnical products – plant and animal food with man as its basic and most significant consumer[5]. Radionuclides originating from the soil, water and air can enter the food chain by deposition and/or migration. They enter soil and water by migration and collection and via them into plant and animal food.

Internal exposure of animals to radionuclides can occur through skin, inhalation and ingestion. Ingestion is the most common way radionuclides enter an animal organism. The danger of exposing livestock production to radioactive contamination results from the fact that radionuclides migrate very fast in the livestock production cycle. A whole series of factors exist that hamper possible interruption of the transfer of radionuclides in the food chain from animals and their products to man.

High production with minimal production costs are the requirements of modern livestock production. In order to realize this it is necessary to adapt nutrition according to amount and type to a species and category of animal. According to the Rules of Animal Food Quality it follows that animal food is any substance or product that is unrefined or has been partially refined and is intended for feeding animals used for food production in the form of: feed, premix and mixture[6]. Suitable animal feeding is one of the most important conditions for proper growth, development and health of animals and must be adjusted to each animal species and current metabolic needs. Nutrition should give an animal all necessary nutrients in accordance to its physiological state. This to the greatest degree depends on the category, animal age and life cycle phase. Adequate nutrition improves animal health, increases consumption and efficiency of food utilization, changes certain physiological processes in the animal organism and thus stimulates its growth and improves the quality of obtained products. The need for proteins and energy are the highest, but a deficit of only one vitamin or mineral in a feed causes specific disorders leading to deterioration of the animals general state and reduced production. Over 30 nutrients in a suitable amount and ratio need to be provided in animal feed. In order to reach optimum different premixes and additives are added to nutrients. Vitamins, minerals, essential amino-acids, saturated fats, different probiotics and prebiotics are most often added[7-9].

Premixes are products with high content of vitamins, amino-acids and allowed additives that are homogeneously mixed with a carrier. They are used for animal feed combined with nutrients or for making mixtures. Premixes can be:

- *mineral* (contain a mixture of allowed minerals that have no energy value for the organism but are significant as gradient elements in different activities and biological systems as they perform many vital functions in the organism),
- *vitamin* (contain only vitamins – complex organic compounds, physiologically active that regulate the metabolism of matter in the organism. The organism

does not synthesize them or synthesizes them in insufficient amounts that require their substitution through food or vitamin additives. Each individual vitamin has a specific role in the organism that cannot be replaced by any other form of chemical compound),

- *vitamin-mineral* (contains allowed minerals and vitamins) and
- *other premixes* (these are premixes with amino-acids, premixes of non-protein nitrogen compounds).[6]

MATERIAL AND METHODS

Samples of premixes for animals were collected in 2015 and 2016 on the territory of the Republic of Serbia in factories for animal food production, factories for mineral nutrient production, on farms, in retail (agriculture pharmacies) and obtained from individual producers.

In 2015 16 premix samples were collected for monitoring, while in 2016 there were 19 samples.

After delivery to the laboratory, the samples were homogenized and packed in Marinelli vessels with a volume of 1 L sealed with paraffin and left to sit for at least four weeks to establish a radioactive balance between ^{226}Ra, ^{222}Rn and their short-life products. Gamma-spectrometric measurements of samples of animal feed were performed using the standard method of the International Agency for Atomic Energy[9,10]. A semiconducting germanium detector with a high purity of the n type produced by ORTEC - AMETEK, USA, with 8192 channels, resolution 1.65 keV and relative efficiency of 34% at 1.33 MeV for ^{60}Co was used for determining radionuclide activity levels. Calibration of the detector energy and efficiency was performed by the Department of Physics, Faculty of Natural Sciences, University of Novi Sad. All samples were measured at 14400 s. Spectrum analysis was performed using the Gamma Vision 32 program[11]. Activity levels of ^{238}U were determined using gamma lines: ^{234}Th (63 and 93 keV) and ^{234}Pa (1001 keV). Activity levels of ^{226}Ra were determined using gamma lines: ^{214}Bi (609, 1120 and 1764 keV) and ^{214}Pb (295 and 352 keV). Activity levels of ^{232}Th were determined using gamma lines ^{228}Ac (338, 911 and 969 keV). Activity levels of ^{40}K were determined using gamma lines at 1460 keV, and for ^{137}Cs gamma lines at 661.6 keV.

In order to secure checks of the measuring device for potential pollution periodical measurement of the background radiation of the detector system was performed. Background radiation measurement was performed just before sample measurement. Minimal detectable levels of radionuclide activity (Bq/kg) were determined using the Currie equation[12]. The relative measuring uncertainty of all results was up to 10% of the activity levels of analyzed samples. Quality control of gamma spectroscopic measurements, based on which investigated samples were analyzed, was performed using a calibration standard and reference materials and regular yearly participation in evaluations between laboratories organized by the International Agency for Atomic Energy.

MEASUREMENT RESULTS AND DISCUSSION

Tables 1 and 2 show minimal and maximal values of activity levels of natural (^{226}Ra, ^{232}Th and ^{238}U) and artificially produced radionuclides ^{137}Cs (Bq/kg) and the number of premixes.

For monitoring purposes in 2015, 16 premix samples were collected used in nutrition of: cattle (2 samples), pigs (4 samples), poultry (8 samples), lambs (1 sample) and all animal species (1 sample). For monitoring purposes in 2016 19 premix samples were collected that were used in nutrition: cattle (4 samples), pigs (8 samples), poultry (6 samples) and all animal species (1 sample).

Table 1. Activity levels of ^{137}Cs, ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U (Bq/kg) in premixes collected in 2015

Sample name (sample number)	^{137}Cs	^{40}K	^{226}Ra	^{232}Th	^{238}U
	(Bq/kg)				
Premix for beef cattle (1)	< 0.2	104	2.9	1.5	< 3.2
Premix for cows (1)	< 0.3	143	8.1	1.9	< 5.1
Premix for pigs (1)	< 0.2	64	6.5	2.2	< 3.6
Premix for fattening pigs (2)	< 0.2	96; 351	< 4.7	< 1.0	< 4.3
Premix for sows (1)	< 0.2	142	4.9	1.7	< 4.2
Premix for chicken (3)	< 0.2	25 - 78	< 4.1	< 1.0 - 2.5	< 4.3
Premikx for laying hens (5)	< 0.3	29 - 98	3.4 - 49	< 1.0 - 2.3	< 3.7 - 100
Premix for lambs (1)	< 0.2	130	4.4	< 1.0	< 3.5
Premix for all animals (1)	< 0.2	75	8.0	2.3	8.6

Table 2. Activity levels of ^{137}Cs, ^{40}K, ^{226}Ra, ^{232}Th and ^{238}U (Bq/kg) in premixes collected in 2016

Sample name (sample number)	^{137}Cs	^{40}K	^{226}Ra	^{232}Th	^{238}U
	(Bq/kg)				
Premix for beef cattle (2)	< 0.3	30; 71	6.9; 7.2	< 1.0; 2.3	< 4.7
Premix for cows (2)	< 0.2	11; 26	7.8; 9.1	1.6; 1.7	8.8; 9.4
Premix for piglets (2)	< 0.4	119; 153	< 7.8	< 1.5; 6.9	< 7.6
Premix for pigs (4)	< 0.2 - 2.8	15 - 502	3.2 - 44	< 1.0 - 13	< 3.9 - 111
Premix for sows (2)	< 0.3	27; 117	6.7; 9.2	< 1.0; 1.4	< 3.8; 10
Premix for chicken (4)	< 0.3	36; 61	6.5; 7.8	< 1.0; 3.9	< 7.1
Premix for laying hens (2)	< 0.3	25 - 111	< 4.1 - 46	< 1.0 - 1.6	< 3.7 - 103
Premix for all animals (1)	< 0.2	22	8.3	< 1.0	19

The results given in Table 1 indicate that activity levels of ^{137}Cs in premix samples from 2015 were < 0.3 Bq/kg, ^{40}K from 25 to 351 Bq/kg, ^{226}Ra from 2.9 to 49 Bq/kg, ^{232}Th < 1.0 to 2.5 Bq/kg, and ^{238}U < 3.2 Bq/kg to 100 Bq/kg.

The results given in Table 2 indicate that activity levels of ^{137}Cs in premix samples in 2016 < 0.2 to 2.8 Bq/kg, ^{40}K from 11 to 502 Bq/kg, ^{226}Ra from 3.2 to 46 Bq/kg, ^{232}Th < 1.0 to 13 Bq/kg, and ^{238}U < 3.7 to 111 Bq/kg.

Research from 2011[13] show that activity levels of radionuclides in piglet premix (Bq/kg) on the territory of Serbia were for ^{137}Cs < 0.1; < 0.2, ^{40}K 25; 220; ^{238}U < 4.0; 9.6 and ^{226}Ra < 2 to 4.2, in pig premix (Bq/kg) ^{137}Cs < 0.1 to < 0.5, ^{40}K from 31 to 511, ^{238}U < 4.0 to 25 and ^{226}Ra < 2 to 12, in premix for fattening pigs (Bq/kg) ^{137}Cs < 0.1, ^{40}K from 73 to 79, ^{238}U 11 to 507 i ^{226}Ra 1.2 to 111.

Activity levels in premixes from Denmark, Germany and France were for ^{137}Cs < 0.3 to < 0.5 Bq/kg, ^{40}K from 17 to 62 Bq/kg, ^{238}U from< 9 to 718 Bq/kg and ^{226}Ra from < 01 to 326 Bq/kg. Sparse research on the influence of radionuclides on animals warn that animals are chronically contaminated with uranium, as intake of this radionuclide by the animal starts in the earliest period and ends with its slaughter. Also, the degree of the transfer of radionuclides from the digestive tract into the meat depends on the animal species. The calculated transfer coefficient indicates the fact that poultry meat can present a potential danger for human consumption[15].

CONCLUSION

In order to obtain safe and secure food for human consumption monitoring of animal feed samples on the territory of Serbia is necessary.

As part of monitoring radionuclides in animal feed produced in the Republic of Serbia in 2015 and 2016, the gamma spectroscopy method was used to analyze 35 premix samples. In all analyzed samples activity levels of ^{137}Cs, were within the detection limits of the gamma spectroscopic device to 2.8 Bq/kg, indicating that animal feed produced in the Republic of Serbia is not contaminated with this artificially produced radionuclide.

From the viewpoint of the content of the analyzed radionuclides in animal premixed activity levels (Bq/kg) were: ^{137}Cs < 0.2 to 2.8 Bq/kg, ^{40}K from 11 to 502 Bq/kg, ^{226}Ra from 2.9 to 49 Bq/kg, ^{232}Th < 1.0 to 13 Bq/kg, and ^{238}U < 3.2 to 111 Bq/kg.

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