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ACTIVITY OF NATURAL RADIONUCLIDES IN ANIMAL FEED

Abstract

Radioactive elements like ^{232}Th, ^{235}U and ^{238}U along with their decay products (e.g. ^{226}Ra) are present in most environmental matrices and can be transferred to living beings by different pathways which can lead to the sources of exposure to humans. For these reasons, it was necessary to monitor those natural radionuclides in feed samples in order to assess the potential hazards. Thorium and Uranium content was determined using inductively coupled plasma mass spectrometry. Data analysis was performed by MassHunter Workstation software. The abundance of $^{235}U/^{238}U$ is 0.72%/99.28%. The study included a total of 14 various animal feed samples. The results show that the highest concentrations of the activities of ^{232}Th, ^{235}U and ^{238}U found in phosphates of mineral feed, monocalcium phosphate and phosphosel were significantly higher than the activities of natural radionuclides in other examined samples.

Key words: *potassium, thorium, uranium, feed, ICP-MS*

INTRODUCTION

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 is the first link in the food chain which results in the radioactive contamination of crop and livestock production [1]. Natural radioactivity accompanies Earth evolution and the first beginnings of the development of biosphere. The effect of radioactivity on the development of all biocenoses on the Earth is undoubtedly of fundamental importance. The biggest source of radiation activity is natural radionuclide potassium-40. Other natural radionuclides, which have always been present on the Earth also include radioactive elements like ^{232}Th, ^{235}U and ^{238}U [2]. The exposure to low-level radiation originating from these natural elements has always affected all living beings on the Earth, and it is considered background radiation or natural phon. Radioactive radiation can cause cancerous diseases and other changes in the health of humans and the whole living world in general. Overall, it substantially affects the changes in the environment.

In order to protect consumer health, it is necessary to control the level of toxic and undesirable substances in different types of food for both humans and animals. Nowadays in the countries of the EU, the control of food quality and hygiene is based on modern scientific knowledge, principles and standards defined in the *Codex alimentarius* and EU directives which require the application of Good Hygiene Practice – GHP, Good Manufacturing Practice – GMP, Hazard Analysis and Critical Control Point – HACCP system and Quality Management System – QMS) [3].

When it comes to radiation safety of humans, it is the key that the foods of plant origin have as low content of radionuclides as possible. The content of radioactive substances is the result of the transfer of radionuclides from the air, water and soil, i.e. its direct or indirect contact with plants in the cycle of plant based food production. Livestock production is severely endangered when radionuclide residue is present. This is due to the fact that radioactive contamination lasts for a very long time and the pollutants (radionuclides) very quickly move from one phase of livestock to another, without their own degradation and that way they reach humans in an unchanged form [4]. These are the reasons why agricultural raw materials used for animal feed (hey, corn, wheat, soy) require a special focus when it comes to preventive radiation protection because they enable the natural (primordial) and artificial (produced) radionuclides to reach humans through food chain. Also, mineral nutrients (monocalcium phosphate, monoammonium phosphate feed chalk, animal feed salt, phosphosel) are inorganic compounds used in animal nutrition to satisfy their need for calcium, phosphorus and sodium [5]. By dissolving natural phosphates, uranium, which is present in phosphate oars is deposited in end products - mineral feeds and through animal feed it is reaches humans. A regular control of radioactivity of the ingredients of animal feed, both from domestic production and the imported goods, is a safe way to prevent radioniculides from anthropogenic sources to enter food chain and thus lower the share of "technologically increased natural radioactivity" in the overall radiation exposure of the population [6]. This phenomenon is called TENORM (Technologically Enhanced Naturally Occurring Radioactive Material).

MATERIAL AND METHODS

A total of 14 different animal feed samples were collected. These included mineral and granular nutrients, products of the oil industry, dried plant products, animal nutrients, premixes and complete mixtures for animal nutrition. To determine the activity levels of natural radionuclides, the collected animal feed samples were prepared using wet digestion method in Ethos System, Microwave Labstation, Milestone [7]. Potassium content was determined using the method of AA spectrophotometry on Spectr

AA–10, Varian, at a wavelength of 766.5 nm and using cesium as the ionization-suppressor. In nature, potassium-40 is found in a mixture of natural potassium isotopes (^{39}K, ^{40}K and ^{41}K) with a mass fraction of 0.0119%.The feed samples levels of potassium-40 activity were calculated from total potassium, using the mass activity value for potassium which is 31.561 Bq/g K. The concentrations of thorium and uranium in all the samples were analyzed by a technique of inductively coupled plasma with mass spectrometry, on the Agilent ICP-MS 7700 [7]. The activity levels of uranium-235 and uranium-238 in samples were determined according to the total uranium concentration using mass activity values 0.570 Bq/mgU for ^{235}U and 11.10 Bq/mgU for ^{238}U.

Table 1: Instrument parameters for Agilent ICP-MS 7700

RF Power: 1550 W	S/C Temp: 2 °C
Smpl Depth: 7.4 mm	OctP RF: 180 V
Carrier Gas: 0.9 L/min	Readings/replicate: 3
Nebulizer Pump: 0.1 rps	Detector mode: pulse
Integration time (U): 1 sec/Point	Integration time (Th): 0.1 sec/Point

The activity of thorium was obtained using a specific activity of 4.11 Bq/mgTh for ^{232}Th [8]. The five standard solutions were prepared to 1, 2, 3, 4 and 5 ng/ml, for both elements, Th and U. Concentrations of ^{235}U and ^{238}U, were obtained by calculation, given the known natural abundance of $^{235}U/^{238}U$ is 0.72%/99.28%. The total uranium is the sum of the individual isotope concentrations [9].

RESULTS AND DISCUSSION

Table 2 shows the results of the concentrations of the activities of natural radionuclides ^{40}K, ^{232}Th, ^{235}U and ^{238}U in the examined samples of animal feed. Based on the obtained results, it can be concluded that potassium-40 is the predominant radionuclide in animal feed, decaying by beta particles and gamma photons emission. In the body, potassium is under homeostatic control and is little affected by environmental variations [10]. ^{40}K activity ranged from 36 Bq/kg in chalk feed up to even 823 Bq/kg in phosphosel which is a mineral additive to animal feed intended for all animal types. Phosphosel contains the minimum of 16% phosphorus and maximum of 12% of calcium [11].

Thorium-232 is a metallic naturally occurring element that is radioactive. Because of its insolubility and very low specific activity, thorium can be found in biosphere in very small amounts. This is confirmed by our results as in the largest number of samples the measured ^{232}Th activity was lower than od 3 Bq/kg. Mineral feed with significantly higher specific ^{232}Th activity should be particularly emphasized. They are the following: mineral licking brick (8.39 Bq/kg), monocalcium phosphate **(**14.6 Bq/kg) and specially phosphosel, where the thorium-232 activity amounted even 686 Bq/kg. It is therefore very important to determine the specific activity of this radionuclide in animal feed.

Table 2: The concentration of activities of the Potassium-40, Thorijum-232, Uranium-238 and Uranium-235 in various animal feed samples

No. of sam	**Sample type**	**^{40}K activity [Bq/kg]**	**^{232}Th activity [Bq/kg]**	**^{235}U activity [Bq/kg]**	**^{238}U activity [Bq/kg]**
1.	Meadow hay	565 ± 23	1.28 ± 0.04	0.11 ± 0.01	15.5 ± 0.8
2.	Alfalfa hay	601 ± 20	0.91 ± 0.02	0.02 ± 0.01	0.50 ± 0.01
3.	Corn	125 ± 6	0.06 ± 0.01	0.02 ± 0.01	0.15 ± 0.02
4.	Wheat	162 ± 9	0.04 ± 0.01	< 0.01	< 0.01
5.	Mono-Ca-phosphate	51 ± 3	14.6 ± 0.22	25.2 ± 2.1	486 ± 46
6.	Feed chalk	36 ± 6	0.64 ± 0.09	0.16 ± 0.04	1.69 ± 0.12
7.	Mineral licking brick	587 ± 51	8.39 ± 1.06	0.22 ± 0.08	4.17 ± 1.45
8.	Phosphosel	823 ± 39	686 ± 30	24.5 ± 1.4	657 ± 31
9.	Fish meal	448 ± 47	0.27 ± 0.10	0.05 ± 0.02	0.93 ± 0.14
10.	Soybean meal	540 ± 29	0.23 ± 0.08	0.02 ± 0.01	0.12 ± 0.05
11.	Sunflower meal	183 ± 10	0.04 ± 0.01	0.03 ± 0.01	0.06 ± 0.02
12.	Soy	496 ± 25	0.05 ± 0.02	< 0.01	< 0.01
13.	Premix	321 ± 35	2.62 ± 0.20	1.24 ± 0.09	23.3 ± 2.18
14.	Complete feed for chickens	318 ± 14	0.37 ± 0.12	0.74 ± 0.12	14.5 ± 1.91
Interval of Variation (I.V.)		36 – 823	0,04 – 686	0.02 – 25.2	0.06 – 657

Based on the results shown in Table 2, it can be concluded that the activity of ^{238}U in the examined samples of alfalfa hay, corn, wheat, soy, fish meal, soybean cake, sunflower meal, feed chalk and mineral licking brick is very low and amounts < 5 Bq/kg. Significantly higher activities of ^{238}U compared to other samples were found in phosphate mineral feeds, particularly in monocalcium phosphate (486 Bq/kg) and phosphosel (657 Bq/kg). An increased activity of ^{235}U was also found in these mineral feeds and it was 25.2 Bq/kg in monocalcium phosphate and 24.5 Bq/kg in phosphosel. Some other authors have shown similar results [12]. These results can be explained by the technological processing of phosphorus ores which can contain high concentrations of uranium. By dissolving natural phosphate using sulfuric acid, more than 90% of uranium in phosphates turns into phosphoric acid. Mono and dicalcium phosphate are obtained as end products of processing phosphorus ores and are used in animal nutrition as a source of calcium and phosphorus. They can be contaminated by high concentrations of uranium [13]. This means that safe animal feed is the first and most important step in providing safe food for humans.

It is extremely important for Vojvodina, as an area with a great potential for production of healthy and safe food, to systematically monitor the level of radioactivity of the environment, because if it is contaminated with substances with a technologically elevated level of natural radioactivity, the doses of radioactive radiation that the population receives may increase.

CONCLUSION

It can be concluded that potassium-40 is a predominant natural radionuclide in animal feed as compared to other radionuclides.

The measured values of the concentrations of ^{232}Th, ^{40}K, ^{238}U, ^{235}U activities in all examined samples are below maximum allowable level (Official Gazette of the Republic of Serbia, No. 36/2018). A natural radionuclide U-238 should be particularly emphasized since its activity ranged from 0.06 Bq/kg (sunflower meal) to 657 Bq/kg (phosphosel).

The obtained results show that continuous control of radioactivity of mineral additives is necessary both for animal feed originating from domestic production and the import. It is a safe way to prevent radionuclides from anthropogenic sources from entering food chain and thus lower the share of "technologically increased natural radioactivity" in the total radiation exposure of the population .

Method ICP-MS (inductively coupled plasma mass spectrometry) proved to be very sensitive for quantitative determination of Th and U concentration in the tested samples. Therefore, ICP-MS offers an attractive alternative for monitoring of thorium and uranium in environmental samples.

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AKTIVNOST PRIRODNIH RADIONUKLIDA U HRANI ZA ŽIVOTINJE

Abstract

Radioaktivni elementi kao što su ^{232}Th, ^{235}U i ^{238}U zajedno sa proizvodima njihovog raspada (npr. ^{226}Ra) prisutni su u većini matrixa iz životne sredine, prenose se na različite načine do živih bića i mogu predstavljati izvore ozračivanja ljudi. Iz tih razloga, neophodno je kontrolisati prirodne radionuklide u uzorcima hrane za životinje radi procene potencijalnih opasnosti. Sadržaj torijuma i urana je određen pomoću induktivno spregnute plazme sa masenom detekcijom. Dobijeni rezultati analizirani su pomoću softverskog programa MassHunter Vorkstation. Odnos ^{235}U / ^{238}U iznosi 0,72% / 99,28%. Ukupno je ispitano 14 različitih uzoraka hrane za životinje. Dobijeni rezultati ukazuju da su izmerene koncentracije aktivnosti ^{232}Th, ^{235}U i ^{238}U u fosfatnim mineralnim hranivima, monokalcijum-fosfatu i fosfozelu, značajno veće od aktivnosti prirodnih radionuklida u ostalim ispitivanim uzorcima.

Ključne reči: *kalijum, torijum, uranijum, hrana za životinje, ICP-MS*

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