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ACTIVITY CONCENTRATION OF NATURAL RADIONUCLIDES IN CHICKEN FEEDS IN THE REPUBLIC OF NORTH MACEDONIA

KONCENTRACIJA AKTIVNOSTI PRIRODNIH RADIONUKLIDA U HRANI ZA PILIĆE U REPUBLICI SJEVERNOJ MAKEDONIJI

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SUMMARY

The care of radioactivity level in animal feed is very important, as ingestion is one of the most common ways radionuclides enter living organisms. Thus, since humans consume products of animal origin (eggs and poultry, milk and beef), it is important to monitor radiation levels in animal feed, as part of the radioactivity that these animals are exposed to, could be transmitted to humans. Natural radionuclides such as ^{40}K, ^{232}Th and ^{226}Ra find their way into the food chain from soil and air to plants and from plants to animals and humans as well. The goal of this study was to determine the activity concentrations of natural radionuclides in three commercial chicken feeds – starter, grower and finisher. The analyzes were performed by means of HPGe gamma spectrometry - gamma spectrometer (Canberra Packard) with a high-purity germanium detector. The obtained spectra from the measurement were analyzed by using the program GENIE 2000. Based on the performed tests, the mean values for the activity concentrations in the samples were the following 26.03±4.1 Bq kg^{-1} for ^{226}Ra, 37.9±4.8 Bq kg^{-1} for ^{232}Th, and 64.40±6.5 Bq kg^{-1} for ^{40}K. The results obtained from the analyses of chicken feeds were compared to studies in other countries and it was found that ^{40}K was present in the highest amount in all types of samples while the vales for the other natural radionuclides were within the nominal concentrations. Higher concentration of ^{40}K can be associated with plants used in the production of animal feed, which were contaminated with fertilizer during their cultivation. The range of radionuclide concentrations in the samples was within the recommended values UNSCEAR.

Key words: Chicken feed, gamma-ray spectrometry, natural radioactivity

INTRODUCTION

There is a significant need for radiological analysis in the environment, i.e., the soil, human food and animal feed. Natural radiation is the main source of radiation exposure that humans and animals are exposed to. Natural radionuclides are present everywhere in the human environment and are continuously released into the ecosystem from natural and artificial sources.(Anas et al., 2015).

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These elements have a half-life equivalent to the age of Earth and may be present in animal feed. Numerous studies have been dedicated to monitoring the amount of activity of radionuclides in food and dietary supplements, especially those related to human nutrition, in order to determine the risks that those amounts may cause (Carvalho et al., 2006); (Casacuberta et al., 2009).

For this reason, care of radiation levels in animal feed and samples is also very important, since ingestion is one of the most common ways radionuclides enter living organisms.

In order to increase the nutritional value, elements that can actually increase the amount of radionuclide activity in animal feed are increasingly added, which was discussed by (Arruda Neto et al., 1997). It should be noted that phosphorus is one of the elements often added to food and feed, as it is one of the most important minerals for living organisms. It is an essential element for all living cells and a key element for the cellular energy transportation. It affects several important biological processes, such as osmotic pressure and all metabolic reactions. (Casacuberta et al., 2007). In North Macedonia there has been a lack of data on the concentration of natural radionuclides in poultry feed therefore the purpose of this paper was to estimate the amount of natural radionuclides in samples of poultry feed and to compare the obtained results with values from different studies in other parts of the world. It is necessary to ensure prevention and to control the radioactivity of the animal feed which should be with a lower concentration of radioactive isotopes The most commonly used methods of prevention are:cultivation of the soil used for growing crops for animal feed or grazing, changes in the management regime of animals, adding binding agents or analogue products to animals and delayed slaughter of animals (Beresford et al., 2011; Finley et al., 2013).

MATERIALS AND METHODS

Sample preparation

Sample collection was done by interaction with farmers, milling (manufacturing) companies and a few samples directly purchased from the market. The feed samples (27 total) were divided into three groups (Starter, Grower and Finisher feeds). The mass of each sample was approximately 0.5 kg. The samples were homogenized, packed in a 0.5 liters Marinelli beakers, closed and stored for 21 days, in order to achieve a secular equilibrium before the measurements were made.

Sample analysis

Gamma-ray spectrometry was used for the analysis of the samples of poultry feed. The samples were placed in the detection system, composed of a germanium detector with a relative efficiency of 30% and a resolution of 1.8 keV at 1.33 MeV. The detector is enclosed in a shield with a thickness of 10 cm, internally coated with 2 mm copper foil. The detector is connected to a multi-channel analyzer and is connected to a computer system for data acquisition and analysis. GENIE 2000 software was used for data collection and analysis. Efficiency calibration was performed with mixed calibration standard sources MBSS2, supplied from the Czech Metrological Institute, Inspectorate for Ionizing Radiation. In order to determine the background distribution in the detector environment, empty sealed Marinelli beaker with the same geometry was measured at equal counts as the feeds' samples. The analysis procedure included the subtraction of the background spectrum.

The specific activity of ^{226}Ra is calculated for the power line of 186.1 (keV) and ^{232}Th through its decay product ^{228}Ac (second in the decay sequence), i.e., through its three gamma decay power lines occurring at 338.4, 911.07 and 968.9 (keV). The activities of ^{40}K were determined from its γ-line of 1460 keV. The time interval for calculation (counting) was 108000 seconds.

Activity calculation

The activity concentration A (Bq/kg) is determined in accordance with the following formula (Canberra, USA)

$$A = \frac{\frac{N}{t} - \frac{N_0}{t_0}}{\varepsilon \cdot \gamma \cdot m} \ (Bq \cdot kg^{-1})$$

Where, N is clean surface of peak accumulated from a specific radionuclide in analysis of a specific sample (number of readings), N_0 is clean surface of peak accumulated from the spot of a specific radionuclide without an analysis of sample (number of

Table 1 Concentration of radionuclides in chicken feed samples (Bq kg^{-1})

Tablica 1. Koncentracija radionuklida u uzorcima hrane za piliće (Bq kg^{-1})

Samples / Uzorci	Chicken feed type / Vrsta hrane za piliće								
	Starter			Grower			Finisher		
	^{40}K	^{226}Ra	^{223}Th	^{40}K	^{226}Ra	^{223}Th	^{40}K	^{226}Ra	^{223}Th
A1	55.7±12.	22.5±6.0	37.7±5.0	67.1±7.0	25.7±5.2	42.1±5.0	82.6±5.4	39.6±3.1	42.6±5.0
A2	54.3±8.2	19.7±4.0	25.6±3.2	71.2±5.3	26.5±7.7	34.0±6.5	97.1±8.0	37.1±3.0	31.7±5.0
A3	37.2±12.	21.5±4.2	22.4±4.2	54.3±10	29.0±9.0	29.4±4.1	69.7±4.0	19.2±4.0	31.6±4.2
A4	41.2±9.0	27.3±4.0	35.1±4.0	61.3±5.7	32.2±4.0	51.7±6.5	66.0±4.0	28.7±3.3	39.7±4.0
A5	67.1±8.0	18.7±3.0	21.4±7.2	71.4±6.0	24.1±6.0	26.1±5.0	82.1±6.0	18.1±3.5	36.4±4.2
A6	64.3±8.0	25.4±4.2	30.4±4.2	69.2±5.0	34.2±6.0	48.3±4.2	74.0±5.0	21.6±4.0	46.2±4.3
A7	39.4±8.1	31.1±4.0	21.7±4.0	58.3±5.0	41.1±6.2	26.3±4.0	59.2±5.0	31.0±4.0	36.1±4.0
A8	61.2±7.8	22.0±3.6	41.0±8.0	70.6±5.0	29.7±4.0	53.9±4.2	78.1±5.0	21.0±4.0	41.7±5.0
A9	57.4±8.0	21.5±3	38.±12.0	67.3±4.0	26.6±4.0	49.1±4.0	67.2±5.0	24.3±4.0	37.2±5.0
Mean	**53.0±9.0**	**21.5±3.0**	**30.45±5.3**	**65.63±5.8**	**29.9±5.7**	**40.1±4.8**	**74.5±4.8**	**26.7±3.6**	43.2±4.5

readings), t is live time of accumulation of the sample spectrum (s), t_0 is live time of accumulation of the phone spectrum (s), e is detector efficiency for a given energy (for a specific peak), γ is intensity of gamma transition in radioactive decay for a respective radionuclide (%), and m is mass of the sample (kg).

RESULTS AND DISCUSSION

The concentrations of natural radionuclides obtained in the samples are presented in Table 1. The 40K concentration was in the range of 37.2 ± 12 to 67.1 ± 8.0 Bq kg^{-1} with a mean concentration of 53.0 ± 9.0 Bq kg $^{-1}$ for starter. The results are presented individually and as a mean and standard deviation (M±SD)

The average concentration value of ^{223}Th was 30.45 ± 5.3 Bq kg^{-1} while 21.50 ± 3.0 Bq kg^{-1} was ^{226}Ra. The mean concentration values for grower were 65.63 ± 5.8, 29.9 ± 5.7 and 40.1±4.8 Bq kg^{-1}, respectively, for ^{40}Ka, ^{226}Ra, and ^{223}Th, while in finisher, the values were 74.5±4.8, 26.7±3.6, and 43,2±4.5 Bq kg^{-1}, respectively. However, the ^{40}K concentration is highest in all samples. Considering that K is an essential element for living organisms, the radioactivity of ^{40}K cannot be avoided.

The activity concentrations of ^{232}Th, ^{226}Ra, and ^{40}K in poultry feed samples, found in this study, were compared with reports from other authors from different countries in Table 2.

Table 2 Comparison of radionuclide results with other results in the literature

Tablica 2. Usporedba rezultata analize radionuklida s drugim rezultatima iz literature

Country / Država		Activity concentration / koncentracija aktivnosti (Bq kg^{-1}) ±SD		
		^{226}Ra	^{232}Th	^{40}K
Egypt (Harb et al., 2010)		0.35-1.17	0.27-1.07	60.5-91.2
Brazil (Filho et al., 2016)		0.23-1.51	0.29-1.63	236-402
Nigeria (Ageda et al., 2017)		5.0-34.7	0.9-51.6	43.6-196.8
Korea (Choi et al., 2017)		0.026	0.0127	58.5
This study / Ova studija	Starter	21.5±3.0	30.45±5.3	53.0±9.0
	Grower	29.9±5.7	40.1±4.8	65.63±5.8
	Finisher	26.7±3.6	43.2±4.5	74.5±4.8

The obtained concentration of ^{40}K is similar to that obtained in Egypt (Harb et al., 2010), but higher for ^{226}Ra and ^{232}Th. In Brazil, higher concentration of ^{40}K was reported in poultry feed (Filho et al., 2016). The range of radionuclide concentrations in the samples was within the recommended values (UNSCEAR, 2000). Our hypothesis of increased activity of ^{226}Ra and ^{232}Th measured in the samples taken is due to the addition of monocalcium phosphate (MCP) to this type of animal feed. (Filho et al., 2016). However, farmers should be careful when feeding broilers with this chicken feed which may expose poultry to an elevated level of ^{226}Ra and ^{232}Th, which in the long-term may cause several health effects in the poultry.

CONCLUSIONS

The activity concentrations of natural radionuclides, ^{40}K, ^{226}Ra and ^{232}Th, were assessed in this study, in different samples of poultry feed in the Republic of North Macedonia. The results, except for ^{40}K, are not similar to other such studies.

In our study the values of the concentrations of the tested radionuclides are within the permitted values (UNSCEAR, 2000). At the same time, radionuclide monitoring systems should be maintained in the food chain in order to reduce human exposure to radiation through the consumption of animal products. Prevention is perhaps the best approach and much care is required, to reduce radioactive contamination in animals and further in consumers. It would be best to continuously monitor the level of natural and artificial radionuclides in animal feed in order to mitigate the amount of radioactive substances that can reach the human body through the food chain.

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SAŽETAK

Praćenje razine radioaktivnosti u hrani za životinje od izrazite je važnosti, budući da je oralna konzumacija jedan od najčešćih načina unosa radionuklida u živuće organizme. S obzirom na ljudsku konzumaciju proizvoda životinjskog porijekla (jaja, perad, mlijeko i govedina), važno je pratiti razine radijacije u hrani za životinje budući da dio ukupne radioaktivnosti kojoj su životinje izložene može prijeći na ljude. Prirodni radionuklidi poput ^{40}K, ^{232}Th i ^{226}Ra ulaze u hranidbeni lanac iz zemlje i zraka u biljke te u životinje i ljude. Cilj ovog istraživanja je odrediti aktivne koncentracije prirodnih radionuklida u tri komercijalne vrste hrane za piliće – starteru, groweru i finišeru. Analize su provedene uporabom gama spektrometra visoke rezolucije, odnosno gama spektrometrom (Canberra Packard) s germanijskim detektorom visoke čistoće. Dobiveni mjerni raspon analiziran je uporabom programa GENIE 2000. Temeljem izvedenih ispitivanja uzoraka, prosječne vrijednosti aktivnih koncentracija su 26.04±4.1 Bq kg^{-1} za radionuklid ^{226}Ra, 37.9±4.8 Bq kg^{-1} za radionuklid ^{232}Th i 64.40±6.5 Bq kg^{-1} za radionuklid ^{40}K. Rezultati navedenih analiza hrane za piliće uspoređeni su s istraživanjima provedenim u drugim državama, a zaključak istog je da je radionuklid ^{40}K prisutan u najvišim količinama u svim vrstama analiziranih uzoraka dok su zabilježene normalne koncentracije ostalih prirodnih radionuklida. Više koncentracije radionuklida ^{40}K mogu se povezati s biljkama upotrjebljenima u proizvodnji hrane za životinje, a koje su prethodno bile kontaminirane gnojivima tijekom kultivacije. Utvrđeno je da je raspon koncentracija radionuklida u analiziranim uzorcima unutar vrijednosti koje preporuča UNSCEAR.

Ključne riječi: hrana za piliće, gama spektrometrija, prirodna radioaktivnost