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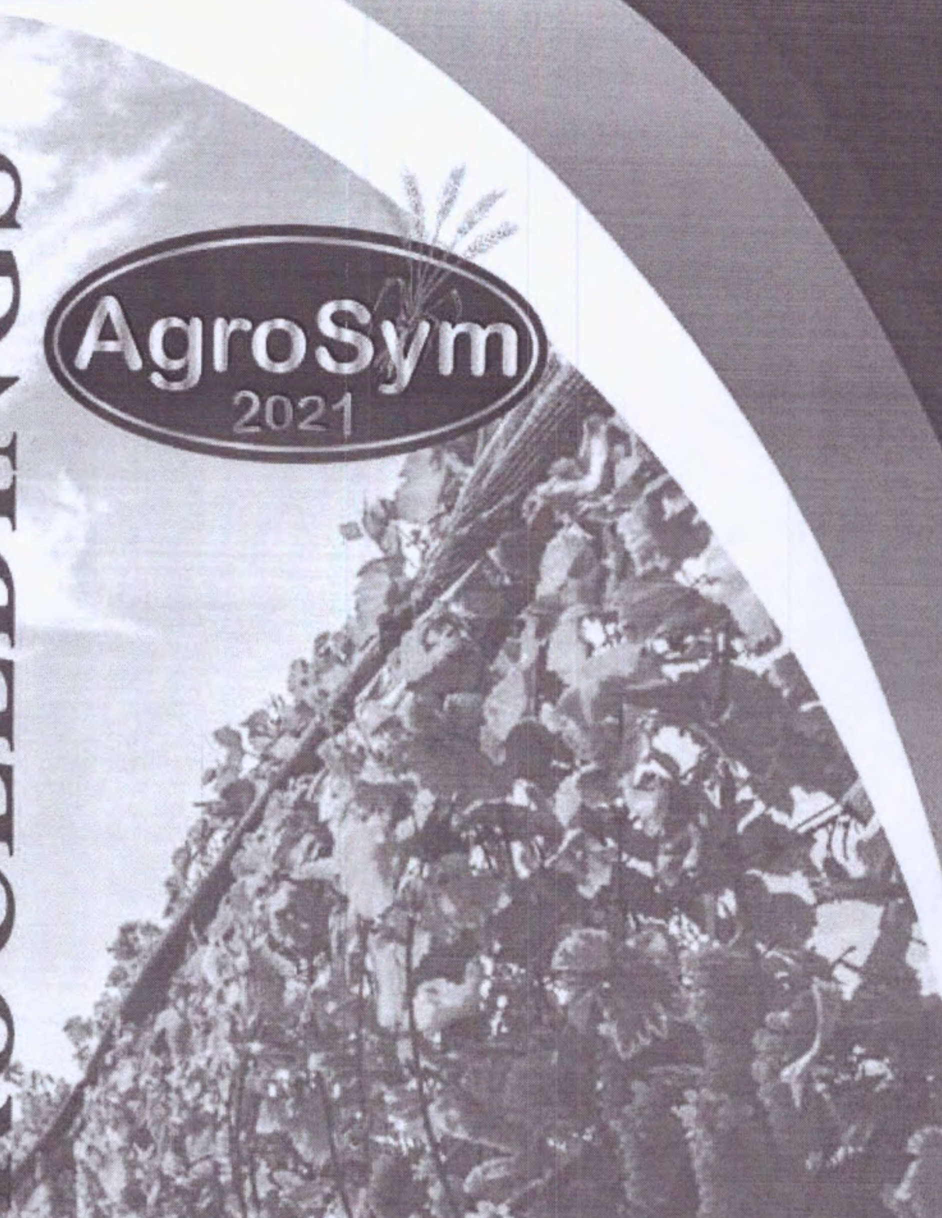

XII International Scientific Agriculture Symposium "AGROSYM 2021"

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RADIATION LEVELS IN SAMPLES OF DICALCIUM PHOSPHATE (DCP) WITH A GAMMA SPECTROMETRY METHOD

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Abstract

Radioactive contamination of living organisms and body tissues primarily depends on the level of contamination of the food they consume, and to a lesser extent on drinking water and inhalation. The aim of this study was to determine the level of radioactivity present in dicalcium phosphate (DCP) samples used as a feed additive. DCP is used as a major source of phosphorus and calcium because it strengthens the skeleton and accelerates the growth of the animal. It is therefore important to familiarize yourself with these levels as part of this radioactivity is likely to be transmitted to humans through the food chain. Measurements were made in order to determine the risks that those quantities may bring. Radioactivity levels are measured using a standard spectroscopic system with a high-resolution HPGe detector. The mean values for the measured activities in the DCP samples were 5.27Bq.kg-1 for 226Ra, 2.87 Bq.kg-1 for 228Th. The mean value measured for 40K was 22.26 Bq.kg-1, respectively. From the results obtained for DCP, it can be seen that the activity of 226Ra is significantly lower than the activity of 228Th, while the value measured for 40K in the samples does not pose a risk to human health, even to animals, because potassium is an essential mineral for living organisms. Considering that the radioactive contamination of the tissues of the animal body originates primarily from the level of contamination of the used animal feed, as well as from the water used for irrigation of the animals, a preventive measure is to control the radioactivity of the feed in their use, i.e. the concentration of radioactive isotopes should be as low as possible, ensuring that they do not pose a threat to the animal organism.

Key words: *DCP, gamma spectrometry, feeds.*

Introduction

Minerals are unavoidable elements in daily animal nutrition, which should be offered in quantities by following animal's requirements. In the same time, the fact that minerals e.g. phosphorus play an important part in environmental contamination should be taken into consideration. Therefore, animal nutritionist should balance animal's daily diet by using high quality phosphorus and in the same time by limiting the excess amount of phosphorus into the environment.

There is a significant need for radiological analysis in the environment, i.e. the soil, food for humans and animal feed. For radioecology, the study of the migration of radionuclides in the soil as a basic resource in agricultural production is of particular interest. In most cases the radionuclides in the soil are tightly bound, however the root of the plant can still absorb them.

Since the first discovery of radioactivity, there has been concern about the damage it can cause to humans, living things and the environment. Many studies have been devoted 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 bring (Tchokossa et. Al., 2013; Zagato et al., 2007).

The process of artificial fertilizer production distributes radionuclides into the environment and in the same time incorporates them into final products and intermediate products (Saueia et al., 2005). The phosphate rock that is usually used for phosphate fertilizer production, contains natural radionuclides from U and Th series (Mazzilli et al., 2000). Phosphoric acid is a starter material for production of: triple superphophate, simple mono superphosphate, monoamonia phosphate and diamonia phosphate, Nitrogen-Phosphorus-Potassium fertilizer and dicalcium phosphate.

Phosphate-based products, such as dicalcium phosphate (DCP) and monocalcium phosphate (MCP) contain approximately 20% phosphorus, and for this reason they are important in the modern production of live raw materials. These compounds are most commonly used in the diet of cattle, pigs and poultry (Arruda et al., 1997), as well as as a dietary supplement in humans. Normally, animal feed is developed on an organic basis and is intended to provide the most complete nutrition possible. 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 et al., (1997). It should be noted that phosphorus is one of the elements often added to food, as it is one of the most important minerals for living organisms, including humans. It is an essential element for all living cells and a key element for the cellular energy transportation (Roessler, 1990). It affects several important biological processes, such as osmotic pressure and all metabolic reactions (Casacuberta et al., 2007).

In this study, radioactivity was measured in a representative number of DCP samples, wherewith specific activities of certain natural radionuclides were calculated, compared with other studies and calculated doses.

Materials and Methods

Sampling

The samples were placed in 0.5 l Marinelli beakers which were fully filled, sealed and stored in order to establish a balance between ^{226}Ra and ^{222}Rn before the measurements are performed. (6) Eighteen samples of DCP were analyzed, which were being used as a feed additive. The measurements were performed at intervals and the background spectrum of the acquisition system is checked on a weekly basis. The process of homogenization of these samples was performed according to the international standard protocol to ensure homogeneity. All studied samples of animal feed were from different farms in the Republic of Macedonia.

Instrument

The samples were measured on an instrument-gamma spectrometer (Canbera Packard) with a high purity germanium detector. The measurement was carried out in beakers that were hermetically sealed so that ^{222}Rn produced by the decomposition of ^{226}Ra would not result in gas leakage. After ensuring a time balance between the successors of the ^{238}U and ^{232}Th series, these sealed samples were prepared for analysis. GENIE 2000 software was used for data acquisition and analysis. The specific activity of ^{226}Ra is calculated for the energy line of 186.1 (keV) and

^{232}Th through its decay descendant ^{228}Ac (second in the decay chain), i.e. through its three gamma decay energy lines which occur at 338.4; 911.07 and 968.9 (keV).
The activities of ^{40}K were determined from its γ-line of 1460 keV, while the activities of ^{137}Cs were determined by means of an estimation of the γ-line at 661.66 (keV). The time interval for calculation (counting) was 108000 seconds.

Activity calculation
The specific activity (A) is determined according the equation

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

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 readings), t is live time of accumulation of the sample spectrum (s), t_0 is live time of accumulation of the phone spectrum (s), ε 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 sample mass (kg) (Garcia-Talaver, 2007).

Radium equivalent activity Ra_{eq}
The calculation of the radium equivalent activity (Raeq) is a value for comparing the specific activities of the samples with different contents of ^{226} Ra, ^{232}Th and ^{40}K. The uniformity with respect to radiation exposure was defined in terms of the radium equivalent activity (Raeq) in Bq/kg in order to compare the specific activity of the materials containing different amounts of ^{226}Ra, ^{232}Th and ^{40}K. It is assumed that 370 Bq/kg of ^{226}Ra, 259 Bq/kg of ^{232}Th and 4810 Bq/kg of ^{40}K produce the same gamma-ray dose rate. It is calculated by using the following ratio (Beretka and Methew, 1985).

$$Ra_{eq}(Bq/kg)= A_{Ra}+1.43A_{Th}+0.07A_k$$

A_{Ra}, A_{Th}, A_k - specific activiites (Bq/kg) of ^{226}Ra, ^{232}Th and ^{40}K, respectively. The value of the radium equivalent activity of 370 Bq/kg corresponds to the maximum allowed dose for a population of 1 mSv.

External and internal hazard index
In order to assess the equivalent average of the annual effective dose imposed to the residents of each area, the external hazard index for the soil samples was calculated.

$$H_{eks} = A_{Ra}/370+ A_{Th}/259+ A_k/4810 \leq 1$$

A_{Ra}, A_{Th}, A_k – specific activities (Bq/kg), ^{226}Ra, ^{232}Th and ^{40}K, respectively (Kurnaz et al., 2007)

Results and discussion

The different radionuclide activities in DCP samples could be derived from two causes: the geological characteristics of the phosphate rock; and the industrial manufacturing processes used for its production. According to the obtained results in regard to DCP, it is observable that the activity of ^{226}Ra is higher than the activity of the thorium series. The mean value for ^{226}Ra was

(5.27) Bq.kg-1, while the value for ^{232}Th reached (2.87) Bq.kg-1. According to Silva et al., (2001), ^{226}Ra is the biggest source of radioactivity in phosphogypsum and can enter living organisms because it follows the same biological pathway of calcium. However, the activity for ^{40}K was (22.26) Bq.kg -1, i.e. with the highest value in all samples of DCP, since K is an essential element for living organisms, therefore the radioactivity of ^{40}K cannot be avoided (Abbady et al., 2005). Thus, the results regarding the activity for this radionuclide in this research and for the DCP samples comply with the expectations and do not create any additional concern.

Table 1. Radionuclide concentration of ^{232}Th, ^{226}Ra, ^{40}K in DCP (Bq/kg)

^{40}K ($\bar{x} \pm S_{\bar{x}}$)	^{226}Ra ($\bar{x} \pm S_{\bar{x}}$)	^{232}Th ($\bar{x} \pm S_{\bar{x}}$)
31.3±2.0	1.70±0.3	7.63±2.0
<7.80	0.30±0.1	<4.5
45.24±1.0	6.23±0.1	5.22±2.0
51.1±0.4.	6.82±0.2	6.13±2.0
21.5±2.0	2.04±0.2	<7.1
23.0±1.0	7.73±0,3	3.03±1.5
26.31±2.2	2.55±0.2	4.54 ±1.0
33.37±1.0	1.42±0.1	6.00±1.0
<9	<1.3	<3.1
<5.4	<2.0	<4.20
11.54±2.0	1.66±0.2	2.97±2.1
<6.20	<1.00	<1.0
31.32±2.0	2.13±0.4	4.13±0.2
32.56±1.0	1.65±0.1	4.22±0.2
15.71±1.0	<1.5	<4.20
21.33±2.2	1.12±0.2	<4.5
34.15±2.0	31.3±0.5	4.17±1.0
22.40±2.0	31.3±0.3	3.66±2.0

Table 2. Comparison of the DCP activities in the research literature

Reference	^{40}K	^{226}Ra	^{232}Th
Present work	22.26	5.27	2.87
Casacuberta (2007)	28	/	<148
Saueia (2005)	148	<2	10
Arruda Neto (1997)	/	/	/

The mean value of the radiation risk index H_{eks} is minimum and it amounts to 0.03, and its value is lower than the maximum allowed value which is <1 for H_{eks}. The value of the radium equivalent activity Ra_{eq} is below the maximum recommended limit, i.e. 370 Bq kg^{-1} and it amounts to 10.93 $Bq.kg^{-1}$.

The specific activity values and the calculated radiation risk index (H_{eks}) and radium equivalent (Ra_{eq}) obtained in this study also did not exceed the safety limits, noting the negligible radiation hazard arising from naturally occurring terrestrial radionuclides.

Conclusions

The activity concentrations of natural radionuclides, ^{40}K, ^{226}Ra and ^{232}Th, were assessed in this study, in different samples of DCM used in the Republic of Macedonia. The results were also compared with other similar studies in other parts of the world. However, the values of the concentrations of the tested radionuclides are within the permissible values, i.e. the transmission of such levels to animal feed and, ultimately, to humans, through the radionuclide pathway, will not pose a threat when humans eventually consume poultry meat, poultry products and eggs from poultry fed with this food. However, it would be necessary to establish radionuclide monitoring systems in the main food in order to reduce human exposure to radiation through the consumption of animal products. Prevention would be the best manner to reduce radioactive contamination in animals and further in consumers as well. Monitoring 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 trophic chain is the basis for radiological contamination.

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