# LEVEL OF NATURAL RADIONUCLIDES IN ANIMAL FEED BY GAMMA -RAY SPECTROMETRY 

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#### Abstract

The radionuclides' presence in animal feed is due both to natural radioactivity and radioactive pollution from different sources. Controls of radionuclides in animal feed will reduce the risk of radioactive hazards to animal and human health. The study was carried out in order to detect the natural radioactivity in animal feed and feed additive. Gamma spectrometer Canberra Packard with a high-purity germanium detector and Marinelli beakers (1 1 capacity) were used for the samples measurement. The most prominent gamma energies observed in the spectra belonged to the naturally occurring radionuclides ${ }^{40} \mathrm{~K},{ }^{226} \mathrm{Ra}$ and ${ }^{232} \mathrm{Th}$. Other radionuclides if present occurred infrequently at low activity concentration under the measurable level. The results show that ${ }^{40} \mathrm{~K}$ had the largest contribution to the specific radioactivity in all the samples. The mean activity concentration of the ${ }^{40} \mathrm{~K}$ was highest in feed additive mono calcium phosphate ( $245.03 \pm 17.778 \mathrm{~Bq} / \mathrm{kg}$ ) and lowest activity concentration of the ${ }^{40} \mathrm{~K}$ was measured in concentrate feed for dairy cows (23.20土2.626 $\mathrm{Bq} / \mathrm{kg}$ ). The average activity of the other two detected natural radionuclides in feed samples was lowest and ranged from 0.42 to $5.81 \mathrm{~Bq} / \mathrm{kg}$ for ${ }^{226} \mathrm{Ra}$ and from 0.61 to $2.55 \mathrm{~Bq} / \mathrm{kg}$ for ${ }^{232} \mathrm{Th}$. The data analysis using ANOVA showed statistical significant differences in the radioactivity concentration of ${ }^{40} \mathrm{~K},{ }^{226} \mathrm{Ra}$ and ${ }^{232} \mathrm{Th}$ between feed samples ( $\mathrm{p}<0.001$ ).


Key words: gamma spectrometry, feeds, natural radioactivity.

## Introduction

The radioactive contamination of the animal organisms and body tissues primarily depends on the level of contamination of the food they consume, and to a lesser extent it depends on drinking water and by means of inhalation. With the use of animal feed, animals in their nutrition accumulate natural radionuclides, and to a lesser extent artificial radionuclides $\left({ }^{90} \mathrm{Sr}\right.$, ${ }^{137} \mathrm{Cs}$, etc.). The fast industrial development and human activities in agricultural practices and livestock production significantly increase the environmental pollution, thereby leading to an increase of the concentrations of various radioactive substances such as strontium, cesium and uranium. Radionuclides have very quick migration and it is very difficult to prevent their transmission through the food chain (Petrović and Mitrović, 1994).
According Saračević (1999), the animals can be radioactively contaminated in several ways, however most commonly through nutrition or drinking water ( $80 \%$ ).
An increasing care is dedicated to the radiation level in animal feed because ingestion through the mouth is one of the most frequent ways how radionuclides enter the living organisms. In order to increase the nutritional value of animal feed, substances containing increased levels of radionuclide activity are added, which will increase the concentration of radionuclide's in the feed. Considering that people use poultry meat and eggs as a part of their nutrition, it is necessary to monitor the levels of radioactive contamination in animal feed because it can
easily be deposited in humans through the path of radionuclides in the food chain (Hernandez et al., 2004).
Numerous researches have been performed which enable monitoring of the radioactive contamination of animal feed in order to reduce the risks that can take these quantities to humans (Carvalho et al., 2006; Casacuberta et al., 2009; Shanthi et al., 2009; Tchokossa et al., 2013).

However, in Macedonia there is a lack of data on the concentration of natural radionuclides in animal feed, namely the purpose of this paper was to assess the amount of natural radionuclides in samples of animal feed produced in Macedonia and to compare the values with already measured values of animal feed that was produced in other parts of the world. Bearing in mind that radioactive contamination in animal body tissues primarily originates from the level of contamination of the used animal feed, as well as the feeding water, a preventive measure would be to control the radioactivity of the animal feed being used, that is, if possible, it should be with a lower concentration of radioactive isotopes, which would not harm the animal organism.
Preventive measures for the reduction of the radioactive contamination of animals are also the method of land treatment which is applied for growing crops for animal feed or grazing, changes in the regime of animal management, giving binding agents or analogous products to animals and delayed slaughter of animals (Beresford and Howard, 2011).

## Materials and methods

## Sampling

The animal feed samples were collected on the basis of production and consumption in Macedonia. The samples were taken from several producers, and several samples were directly purchased from the market. The animal feed samples were crushed and then homogenized to a fine powder. All samples were left in hermetically sealed plastic containers with approximately 450 g of sample. The containers were stored for 14 days in order to allow radium and thorium to achieve a secular balance with their (daughters) descendants.

## Instrument

The samples are measured on an instrument - gamma spectrometer (Canberra Packard) with high-purity germanium detector. The measurement was performed in containers which were hermetically sealed so that ${ }^{222} \mathrm{Rn}$ produced from decay of ${ }^{226} \mathrm{Ra}$ will not result in gas leak. After the provision of time balance between the successors of ${ }^{238} \mathrm{U}$ and ${ }^{232} \mathrm{Th}$ series ( 21 days), these sealed samples were prepared for an analysis. The obtained spectra from the measurement were analyzed by using the program GENIE 2000. The specific activity of ${ }^{226} \mathrm{Ra}$ is calculated for energy line on $186,1(\mathrm{keV})$ and ${ }^{232} \mathrm{Th}$ through its descendant of decay ${ }^{228} \mathrm{As}$ (second in the decayed sequence), that is, through its three gamma decay energy lines which occur on 338,$4 ; 911,07$ and $968,9(\mathrm{keV})$.

The activities of ${ }^{40} \mathrm{~K}$ were determined from its $\gamma$-line from 1460 keV . The interval of the time for calculation (counting) was 108000 seconds. The natural spectrum was recorded immediately after or before the calculation of the sample.

## Activity calculation

The specific activity $\mathrm{A}(\mathrm{Bq} / \mathrm{kg})$ is determined in accordance with the following formula by Garcia-Talavera (2003):

$$
A=\frac{\frac{N}{t}-\frac{N_{0}}{t_{0}}}{\varepsilon \cdot \gamma \cdot m}
$$

Where, N is clean surface of peak accumulated from a specific radionuclide in analysis of a specific sample (number of readings), $\mathrm{N}_{0}$ 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), $\varepsilon$ is detector efficiency for a given energy (for a specific peak), $\gamma$ is intensity of gamma transition in radioactive decay for a respective radionuclide (\%), and $m$ is mass of the sample (kg).

## Results and discussion

The activity concentrations of ${ }^{40} \mathrm{~K},{ }^{226} \mathrm{Ra}$ and ${ }^{232} \mathrm{Th}$ were assessed and they are presented in Table 1 and in Figure 1.

Table 1. Activity concentration of natural radionuclides in feed samples $(\mathrm{Bq} / \mathrm{kg})$

| Samples | n | ${ }^{40} \mathrm{~K}\left(\bar{x}_{ \pm} S_{\bar{x}}\right)$ | ${ }^{226} \mathrm{Ra}\left(\bar{x}_{ \pm} S_{\bar{x}}\right)$ | ${ }^{232} \mathrm{Th}\left(\bar{x}_{ \pm} S_{\bar{x}}\right)$ |
| :--- | :--- | :--- | :--- | :--- |
| Concentrate feed for pigs | 5 | $226.13 \pm 11.911$ | $1.81 \pm 0.269$ | $0.61 \pm 0.086$ |
| Concentrate feed for dairy cows | 5 | $23.20 \pm 2.626$ | $5.81 \pm 0.982$ | $1.99 \pm 0.281$ |
| Feed additive mono calcium <br> phosphate | 5 | $245.03 \pm 17.778$ | $1.46 \pm 0.138$ | $2.55 \pm 0.217$ |
| Maize | 5 | $119.25 \pm 13.732$ | $0.42 \pm 0.059$ | $0.68 \pm 0.144$ |
| Total | 20 | $153.40 \pm 15.542$ | $2.37 \pm 0.411$ | $1.46 \pm 0.164$ |



Figure 1. Activity concentration of naturally occurred radionuclides in feed samples
The results show that ${ }^{40} \mathrm{~K}$ had the largest contribution to the specific radioactivity in all samples. The mean activity concentration of ${ }^{40} \mathrm{~K}$ was highest in feed additive mono calcium phosphate ( $245.03 \pm 17.778 \mathrm{~Bq} / \mathrm{kg}$ ) and lowest activity concentration of ${ }^{40} \mathrm{~K}$ was measured in feed concentrate for dairy cows ( $23.20 \pm 2.626 \mathrm{~Bq} / \mathrm{kg}$ ). The average activity of the other two
detected natural radionuclides in feed samples was lowest and ranged from 0.42 to $5.81 \mathrm{~Bq} / \mathrm{kg}$ for ${ }^{226} \mathrm{Ra}$ and from 0.61 to $2.55 \mathrm{~Bq} / \mathrm{kg}$ for ${ }^{232} \mathrm{Th}$.
The main source of calcium and phosphorus in the concentrate for pigs is monophosphate (Ševković et al., 1991). This supplement is obtained by processing phosphorus ore, and it can also contain large amounts of uranium, thus being a potential source of radioactive contamination both for animals and people (Mitrović et al., 2011).
The results show that the measured monophosphate has a significant activity in all analyzed radionuclides. Considering the concentration of the radionuclide in the monophosphate brand, one can conclude that a large percentage of the radioactivity measured in animal feed is a result of food that contains this component. However, one cannot determine the level of radioactivity that is directly attributable to monophosphate, since the manufacturer does not specify the amount of monophosphate in nutrition. The presence of anthropogenic radionuclides was not detected, indicating that there was no contamination due to artificial radionuclides.

## Conclusion

The concentrations of activity of natural radionuclides, ${ }^{40} \mathrm{~K},{ }^{226} \mathrm{Ra}$ and ${ }^{232} \mathrm{Th}$, were assessed in this study, in different samples of animal feed in the Republic of Macedonia. The results are similar with other such research. However, the values of the concentrations of the tested radionuclides are within the limits of the permitted values i.e. the transfer of such levels in animal feed and ultimately, in the human, through the radionuclide pathway, will not pose a threat when people will ultimately consume poultry meat, products and eggs of poultry fed with this feed. At the same time, systems should be established to monitor radionuclides in the main foodstuff in order to reduce human exposure to radiation through the consumption of animal products. Prevention is perhaps the best approach, and much attention needs to be paid to reduce radioactive contamination in animals and further in consumers as well. It should be emphasized that continuous monitoring of the level of natural and artificial radionuclides in animal feed is necessary in order to mitigate the amount of radioactive substances that can reach the human organism through the trophic chain.

## References

Beresford, N.A. and Howard, B.J. (2011). An Overview of the Transfer of Radionuclides to Farm Animals and Potential Countermeasures of Relevance to Fukushima Releases. Integ Environ Assess Manage. 7: 382-384.
Carvalho, C., Anjos, R.M., Mosquera, B., Macário, K., Veiga, R. (2006). Radiocesium contamination behavior and its effect on potassium absorption in tropical or subtropical plants. Journal of Environmental Radioactivity 86, 241-250
Casacuberta, N., Masque, P., Garcia-Orellana, J., Bruach, J.M., Anguita, M., Gasa, J., Villa, M., Hurtado, S., Garcia-Tenorio, R. (2009). Radioactivity contents in dicalcium phosphate and the potential radiological risk to human populations. Journal of Hazardous Materials 170, 814-823.
Garcia-Talavera, M. (2003). Evaluation of the suitability of various $\gamma$ lines for the $\gamma$ spectrometric determination of 238 U in environmental samples. Applied radiation and isotopes 59(2) 165-173.
Hernandez, F., Madrid, J., Garcia, V., Orengo, J. and Megias, M.D. (2004). Influence of two plant exstract on broilers performance, digestibility and digestive organ size. Poult .Sci. 83:169-174.
Mitrović, B., Vitorović, G., Stojanović, M., Vitorović, D. (2011). Radioactivity of Phosphate Mineral Products," Vet. Gazette, vol. 65, no. 1-2, 133-140.

Petrović, B., Mitrović, R.(1994): Radijaciona zaštita u biotehnologiji, DP Institut za mlekarstvo, Beograd
L. Saračević, Veterinary radiobiology with radiation hygiene, Sarajevo, Bosnia and Herzegovina, DES Sarajevo, 1999.
Shanthi, G., Kumaran, J.T.K., Raj, G.A.G., Maniyan, C.G. (2009). Natural radionuclides in the South Indian foods and their annual dose. Nuclear Instruments and Methods in Physics Research, doi:10.1016/j.nima.2009.10.068.
N. Ševković, S. Pribićević, I. Rajić, Nutrition of domestic animals, Belgrade, Yugoslavia: Naučna knjiga, 1991.
Tchokossa P., Olomo J.B., balogun F.A. and Adesanmi C.A. (2013). Assessment of Radioactivity Contents of Food in the Oil and Gas Producing Areas in Delta State, Nigeria" International Journal of Science and Technology Vol 3, No. 4, 245-250.

