

RESEARCH ARTICLE

GROSS BETA COUNTING METHOD IN RADIATION PROTECTION SYSTEM OF ANIMAL PRODUCTION CYCLE

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ABSTRACT

The research investigated the potential application of the gross beta counting method in radiation protection of animal production under conditions of environmental contamination. Mushroom samples, animal and agricultural products as well as samples from proficiency tests were analyzed using both the gross beta counting method and the gamma spectrometric analysis as a control.

The findings revealed that the quantitative measurements of total beta emitters in the tested samples, obtained with the gross beta counting method, can be effectively employed as a triage approach in scenarios involving radioactive contamination. It is important to notice that successful implementation of this method relies on prior knowledge of the average levels of total potassium or its isotope ⁴⁰K in the samples being analyzed.

Control measurements and assessments based on the results obtained from the gamma spectrometric method demonstrated an acceptable uncertainty for the triage of contaminated samples as well as for the implementation of protective measures within the animal production system under conditions of environmental contamination.

Keywords: Beta activity, food production chain, radioactive contamination

INTRODUCTION

Gross beta activity measurements are quantitative analyses used to determine the overall content of beta-emitting radionuclides in a given sample. The method does not provide qualitative results as it detects the impulses or energies released by beta particles (electrons), which are indistinguishable regardless of the specific origin of the beta-emitting radionuclides. Therefore, the results are expressed as the cumulative activities of all beta-emitting radionuclides present in the tested sample (Duong Van et al., 2020).

This method offers advantages such as straightforward implementation, particularly in emergency situations, and suitability for field con-

ditions. These benefits make it a preferred method for rapid assessments of contamination levels in the field (Bari et al., 2011).

However, the measurement uncertainty of this method comes from limitations in analyzing solid samples, including the lack of standardized preparation and measurement procedures to ensure result reproducibility. These limitations become more significant in cases of low-level contamination, where specific laboratory methods such as gamma spectrometry, alpha spectrometry, and radiochemical separation of radionuclides are recommended. Consequently, the measurement of gross beta activity is not currently recommended as an independent approach for monitoring radioactivity in food.

Qualitative results obtained from this method can be significant depending on the context in which they are used. The measurement of gross beta activity in water samples is the official standard method for assessing the radiological safety of drinking water. In accidental situations, this method can provide rapid and efficient triage information regarding the extent and significance of environmental contamination, as well as its impact on animal and agricultural production. Since the majority of biologically important artificial radionuclides (e.g., ^{89}Sr , ^{90}Sr , ^{134}Cs , ^{137}Cs , and ^{131}I) are beta emitters, measuring total beta activity plays a crucial role in rapid field assessments of radioactivity and contamination levels. This method has been extensively used in response to past accidents such as Chernobyl (Hanfi et al., 2020). Beta counters like LARA 5, LARA 10, and LARA 86 were employed in the former Yugoslavia for triage field assessments of food and environmental samples following the Chernobyl accident (Gradašević, 2017).

In situations with high contamination, the results obtained through this method can serve as a basis for implementing protective measures in agricultural and animal production. For applying certain protective measures, understanding the contamination status is essential. Accordingly, field measurements of gross beta activity provide

a rapid and accessible way of obtaining that information (Bari et al., 2011). For that purpose, it should be necessary to provide prompt education and training for veterinarians and other biotechnical experts. The experiences following the Fukushima accident have also emphasized the importance of establishing a system for protection against radioactive contamination, where rapid triage methods such as total beta activity measurements hold a special place (Naletoski et al., 2021).

It is evident that there is a need to enhance existing knowledge and experiences in utilizing this method during emergency situations. While the development of other radioanalytical methods and reduced demand for rapid field measurements of radioactivity have somewhat marginalized this method, it remains a logical choice in response to the existing challenges posed by the increasing risk of nuclear accidents and general political unpredictability surrounding the use of nuclear weapons.

When considering potential scenarios of global radioactive contamination, it becomes evident that existing laboratories for radioactivity measurements would be overwhelmed with samples, leading to delays in obtaining results, which has been recognized by other authors (Bari et al., 2011). This delay could result in errors and poor decision-making in the food production process. To avoid this issue, field measurements of total beta activity performed by trained veterinary and other experts could be introduced. It should prevent time gaps and enable timely measures in the food production process. Considering the urgency of information required to implement adequate protection measures, the aforementioned slight unreliability of total beta activity measurement results under conditions of high environmental contamination becomes practically negligible.

Furthermore, in the context of food production protection, the absolute reliability of results is not crucially important compared to the timeliness, considering the wide range of available protective measures. By combining the use of total beta activity measurements with subsequent analyses

using more sophisticated laboratory methods, comprehensive information can be obtained to evaluate the effectiveness of implemented measures and the safety of agricultural and animal products.

The aim of the work is to examine the possibility of using the gross beta counting method in conditions of contamination of animal production. To achieve this, the results obtained under normal circumstances need to be extrapolated and evaluated for use in scenarios involving environmental contamination. Plant samples and animal products with normal and higher activity concentrations of one or more artificial radionuclides would be used as models.

MATERIAL AND METHODS

Sample preparation

Samples of agricultural (pumpkin, potato, grass, corn, spinach, beans, onion) and animal (meat, milk) products as well as wild mushrooms were used for the evaluation of methods used in the research. The levels of artificial radiocesium were in the following ranges: below the detection limit (0.5 Bq kg^{-1}), medium (up to 200 Bq kg^{-1}), and higher (above 200 Bq kg^{-1}). The ranges have been chosen in order to represent the scenarios of low and higher contaminations. Mushrooms were used as the model of feeding stuff according to their significant participation in wild animals' diets.

Water samples were evaporated to dryness, crushed in a mortar, and weighed for planchets. Agricultural and animal samples were mineralised at 400°C , homogenised by crushing in a mortar and approximately 500 mg of the ash was dispersed with acetone on the planchets. The sample preparation procedure for wild mushrooms involved several steps, including drying, homogenization, and packaging. Initially, the samples were crushed using a blender and subsequently subjected to drying at a temperature of 80°C while periodically stirring. Once dried, the samples were further pulverized in the mill until a fine powder consistency was achieved. Completely dried and homogenized samples were

then sieved using a $150 \mu\text{m}$ pore diameter sieve. Following sieving, the samples were appropriately packed based on the specific measurement method. Plastic containers weighing 100 g were utilized for gamma spectrometric measurements, whereas stainless steel planchets were used for measuring total beta activity. Each sample was prepared on a planchet with a mass of approximately 500 mg by slurring with a few drops of acetone to ensure the formation of a uniform layer suitable for counting on the alpha/beta counter.

Measurements

The first group included samples whose radioactivity was coming primarily from the presence of potassium, where isotope ^{40}K produced an average of 31 Bq g^{-1} of total potassium content in the sample (Samat et al., 1997). Those samples were used as a model for regular conditions without significant contamination and whose radioactivity was coming due to the naturally occurring ^{40}K .

The second group consisted of the samples of mushrooms whose radioactivity levels were a combination of potassium (^{40}K) and additional radioactivity related to the presence of ^{137}Cs to different extents (up to 198.6 Bq kg^{-1}). This group was used as the model for low to medium contamination levels.

The third group included higher contaminated mushroom samples as well as samples from different proficiency tests with significant activities of ^{137}Cs (428 to 1823 Bq kg^{-1}), together with background levels of naturally occurring ^{40}K .

The total beta activity was measured using the Ortec MPC 9604 multi-detector alpha/beta counter, which enables the measurement of very low activities. The counter is equipped with Vista 2000 software and allows simultaneous measurement of up to four samples. As stated by the manufacturer, the counter has four pancake-style gas flow proportional sample detectors and a large area gas flow proportional cosmic guard detector. The instrument was calibrated in accordance with BAS EN ISO 9697:2020 standard method, using a solution of potassium chloride (KCl), and a

reference material with known activity of ^{137}Cs in a calcium sulfate (CaSO_4) matrix. Since beta particles emitted by ^{40}K and ^{137}Cs have different energies (1300 keV and 514 keV, respectively), their attenuation through the sample on planchets of varying masses differs. Hence, a calibration procedure was conducted beforehand, involving planchets with masses varying from 100 mg to 1000 mg of the matrix, in order to establish efficiency-mass curves of ^{137}Cs and ^{40}K used in the calculation of activity concentrations in this study (Alharbi, 2018).

Considering the typical composition and characteristics of mushrooms, the presence of the other beta-emitting radionuclides (apart from ^{40}K and ^{137}Cs) in significant concentrations was not expected. Therefore, the accredited gamma spectrometry method was used as a comparative analysis for the quantitative determination of ^{137}Cs and ^{40}K activity concentrations and their contribution to the total beta activity with relatively low uncertainty.

The gamma spectrometric analysis was conducted using a BSI (Baltic Scientific Instruments) HPGe gamma spectrometer equipped with a P-type detector. The detector has a relative efficiency of 50% at the energy of 1332 keV emitted by ^{60}Co , and it provides a resolution (FWHM) of 1.9 keV at 1332 keV. The levels of ^{40}K and ^{137}Cs were determined at 1460.8 keV and 661.6 keV, respectively. Quality control procedures were included in the operating activities of the laboratory accredited by BAS EN ISO/IEC 17025:2018.

RESULTS

The results are displayed through tables and graphs arranged by the groups of analyzed samples.

1. Untamined samples

Untamined samples from several villages around Tuzla were analyzed and the results are presented in Table 1. ^{40}K activities were calculated on the fresh masses (f.m.) of the samples.

Table 1 Gross beta activities in animal and agricultural products and water samples with ^{40}K levels measured by gamma spectrometry method (Bq kg^{-1})

	A_{β} (Bq kg^{-1})	^{40}K (Bq kg^{-1})	Ratio ($^{40}\text{K}/A_{\beta}$)	^{40}K % in gross β
Meat 1	83.54±0.25	89.20±6.01	1.068	100
Meat 2	85.98±0.25	81.70±6.67	0.935	99.6
Milk 1	44.74±0.14	43.70±3.57	0.977	99.4
Milk 2	46.53±0.15	45.00±3.67	0.967	99.1
Pumpkin	51.93±0.14	52.90±4.31	1.019	100
Potato	127.27±0.30	128.00±10.50	1.006	100
Grass	181.84±0.58	179.00±12.00	0.987	94.5
Corn	91.75±0.28	96.80±7.91	1.055	100
Spinach	142.08±0.50	130.00±7.16	0.915	98.5
Beans	362.61±0.99	418.00±34.20	1.153	100
Onion	60.82±0.16	61.10±4.79	1.005	100
Water	0.0313±0.0004	0.0441±0.0153	1.409	100

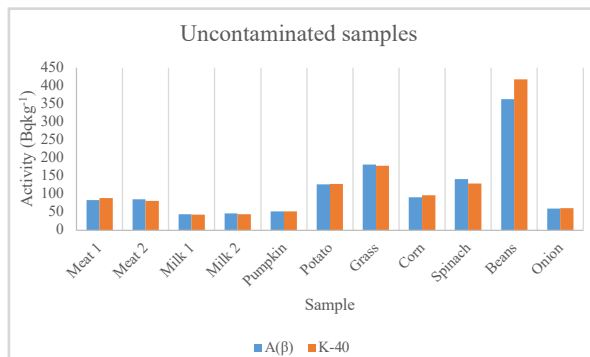


Figure 1 Gross beta activities and ⁴⁰K levels in animal and agricultural samples (Bq kg⁻¹)

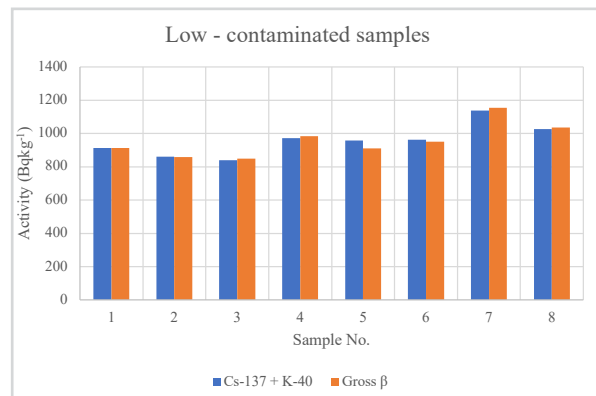


Figure 2 Gross beta activities and the sum of the ¹³⁷Cs and ⁴⁰K activity concentrations obtained by gamma spectrometry method in mushrooms (Bq kg⁻¹)

2. Low-contaminated samples

Mushrooms were chosen considering their affinity for ¹³⁷Cs sorption from the soil and, consequently, their higher levels in observed samples. Taking

into account that the described method is intended for the triage of samples in field conditions, the measurement uncertainty was not shown in the estimated results in Tables 2 and 3.

Table 2 Activity concentrations of ⁴⁰K and ¹³⁷Cs measured by gamma spectrometer and gross beta activities in mushrooms (Bq kg⁻¹ d.m.)

Sample	⁴⁰ K	¹³⁷ Cs	Gross beta	Gross β – ⁴⁰ K (¹³⁷ Cs)
1	754.8 ± 15.6	156.9 ± 2.5	911.8	157.0
2	766.4 ± 16.4	95.4 ± 1.6	858.6	92.2
3	776.7 ± 16.6	63.5 ± 1.2	849.5	72.8
4	781.4 ± 16.7	190.0 ± 3.0	983.7	202.3
5	884.4 ± 18.0	73.0 ± 1.3	909.8	25.4
6	901.2 ± 17.3	62.2 ± 1.0	950.4	49.2
7	940.1 ± 19.9	198.6 ± 3.1	1154.7	214.5
8	1009.7 ± 19.1	16.1 ± 0.4	1036.8	27.1

3. Higher contaminated samples

Higher contaminated samples were used as a model for higher contamination scenarios.

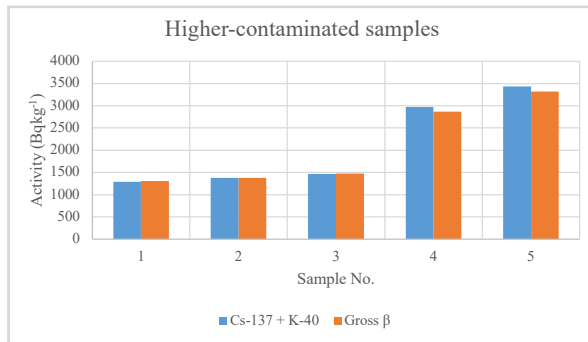


Figure 3 Gross beta activities and the sum of ¹³⁷Cs and ⁴⁰K activity concentrations obtained by gamma spectrometry analysis in samples from the proficiency tests

Table 3 Activity concentrations of ⁴⁰K and ¹³⁷Cs measured by gamma spectrometer and gross beta activities in contaminated samples (Bq kg⁻¹ d.m.)

Sample	⁴⁰ K	¹³⁷ Cs	Gross beta	Gross β – ⁴⁰ K (¹³⁷ Cs)
1	859.0 ± 17.0	428.5 ± 6.4	1305.2	446.2
2	847.6 ± 17.8	529.1 ± 7.9	1374.9	527.3
3.	539.0 ± 32.0	931.9 ± 78.0	1477.8	938.8
4.	1508.3 ± 59.2	1462.8 ± 126.4	2870.6	1362.3
5.	1614.4 ± 63.7	1823.2 ± 149.1	3322.3	1707.9

DISCUSSION AND CONCLUSION

The evaluation of the applicability of the gross beta activity measurement method relies on comparing the obtained results with those derived from the accredited gamma spectrometry method. Generally, for the successful use of the method, it is necessary to know the levels of ⁴⁰K which are the basic parameters of the assessment. The levels of ⁴⁰K should be provided by measurement or calculated from the total potassium content in field conditions.

Considering the fact that potassium is homeostatically controlled in biological systems, its average contents in agricultural and animal samples do not extremely vary (except the mushrooms) and it is well described in the literature. Therefore, literature values can be used for the assessment of the activity concentration of ⁴⁰K in those samples. That could be done by simply multiplying the average potassium content from literature, e.g. 1.5 g in cow milk (Nogalska et al., 2020) by 31, which is an average activity of ⁴⁰K in 1 g of natural potassium (Samat et al., 1997).

The results presented in the first two columns of Table 1 show values of gross beta activity measured by alpha/beta counter and values of ⁴⁰K obtained by accredited gamma spectrometry method. It was obvious that obtained results were very similar for both methods, indicating that in the case without contamination, gross beta values correspond to the naturally occurring ⁴⁰K in solid samples. An average level of ⁴⁰K contributed to the gross beta activity of more than 94.5%. Statistical analysis proved the null hypothesis about the similarity of the results (t-test: two tails P- values equal - 0.923; Pearson coefficient r²=0.98, significance p < 0.05).

In the presence of other beta-emitting radionuclides, gross beta activities would show higher levels than those recorded in the above samples, whilst the percentage of the ⁴⁰K should be lower depending on the activities (mass) of the additional beta emitters. For the samples of water, the same approach could not be applied considering the fact of low ⁴⁰K levels in drinking water. The national legislation requires the limit value of gross beta activities that should not be exceeded in drinking water (1 Bq l⁻¹) and should

be used as the limit value for consideration of the water samples (“Regulation on the monitoring of radioactivity in the environment”).

Results in column 5 (right) of Table 2 represent the levels of ^{137}Cs obtained from the gross beta activity after subtracting the ^{40}K levels. Comparing those values with values of the ^{137}Cs obtained by gamma spectrometry measurements (column 3), it is obvious that they are similar if the uncertainties of the ^{40}K results would be considered. The exemption in sample 5 was probably due to differences in used efficiencies for ^{40}K and ^{137}Cs and their specific ratio in the observed sample. It could appear in cases in which the yields of ^{40}K might vary in a wide range like in mushrooms (Falandysz and Borovička, 2013), and should be anticipated. For other types of agricultural and animal samples, the potassium content is more stable with no possibility for significant under- or over-estimation of its concentrations (Nasreddine et al., 2008). Additionally, the yields of the rest of the radionuclides potentially present in the contaminated samples (^{90}Sr , ^{131}I , ^{134}Cs , etc.) during the radioactive contamination should be available from the general assessment of contamination.

The values in Table 2 showed compliance among the used methods. Results for ^{137}Cs measured by gamma spectrometry method were in well accordance with those obtained after subtracting ^{40}K activity concentrations from the gross beta activities. Statistical analysis of the gross beta activities and the sum of the values for ^{137}Cs and ^{40}K obtained by the gamma spectrometry method (Figure 2) indicated equal values (t-test: P value 0.972). The results approved the possibility of using the described method for the estimation of the ^{137}Cs levels in contaminated animal and agricultural samples.

Higher contaminated samples were used as a model for higher contamination scenarios. Generally, all samples with gross beta activities above average activity concentration of ^{40}K might be considered as contaminated except for samples significantly rich in potassium (e.g. dried products: mushrooms, bananas, potatoes, etc.).

Results presented in Table 3 showed values above 1000 Bq kg⁻¹ of gross beta activities (1305.2 – 3322.3). The higher values were recorded due to the high levels of ^{40}K (847.6 – 1614.4 Bq kg⁻¹) as well as the presence of significant activities of ^{137}Cs shown in column 3 (428.5 – 1823.2 Bq kg⁻¹). Calculated levels of ^{137}Cs that were obtained by subtracting the ^{40}K from the gross beta activities (column 5-right) corresponded to those obtained by the direct gamma spectrometry measurements. Small differences in activities between calculated and measured levels of ^{137}Cs were in the ranges of measuring uncertainties of gamma spectrometry results for ^{137}Cs and ^{40}K .

The results presented in Figure 3 were statistically equal (t-test: P value 0.95) with a statistically significant Pearson correlation coefficient ($r^2=0.99$, $p < 0.05$).

In the case of presence of other beta emitters, more accurate estimates could be possible if efficiency calibration would be performed on existing radionuclides and the data about the quantitative-qualitative composition of contamination would be available.

During environmental radioactive contamination, the food production chain is especially endangered; therefore, appropriate protective measures must be applied. The selection of the appropriate protective measures depends on the contamination levels and it is specific for each production line. The best solution in such circumstances is to have appropriate methods for fast assessment of the contamination levels in field conditions. Therefore, the study was carried out in order to estimate the use of the gross beta counting method for the approximative evaluation of food chain contamination by artificial radionuclides in field conditions.

The results for the first group showed that gross beta counting method provided values that statistically corresponded to the naturally occurring ^{40}K in uncontaminated samples of animal and agricultural products. The possible use of the gross beta counting method in such conditions would be based on the ^{40}K levels, which means

that every significant excess of the gross beta activities above the average ^{40}K values should be considered as possible contamination. The levels of ^{40}K in specific samples could be calculated from the average potassium content (grams) provided by literature and multiplied by 31. The instrument should be calibrated by ^{40}K standard.

The samples contaminated by the low levels of ^{137}Cs (second group) exhibited excellent accordance among the gross beta activities and the sum of the levels of ^{137}Cs and ^{40}K obtained by the gamma spectrometry method. Almost the same statistical significance was obtained among the levels of gross beta activities and high levels recorded in proficiency testing samples which were spiced with the mixture of ^{137}Cs and ^{40}K .

Therefore, the use of the method in contamination circumstances should be based on the subtracting of the ^{40}K levels from the gross beta activities and the rest would be considered as a mixture of artificial beta emitters (mainly Cs, Sr, I). For more accurate assessments in field conditions, it would be necessary to provide the two important

requirements: prepared efficiency calibration for the artificial radionuclides of interest (Cs, Sr, I, etc.) as well as general information about the quantitative-qualitative composition of the contamination.

The findings proved the assumption that the gross beta counting method could be used for fast triage in conditions of low and high artificial radioactive contamination. Obtained results could be used as milestones for decisions about protective measures in food production processes.

CONFLICT OF INTEREST

The authors declared that there is no conflict of interest.

CONTRIBUTIONS

Concept – NG; Design – NK, NM, NG; Supervision – NG, NM; Resources – NG, LT; Materials – LT, NK; Data Collection and/or Processing – NM, NK, LT; Analysis and/or Interpretation – NG, NM, NK; Literature Search – LT, NG, NK; Writing Manuscript – NG, NK; Critical Review – NG, NM.

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METODA MJERENJA UKUPNOG BETA ZRAČENJA U SISTEMU ZAŠTITE OD ZRAČENJA U ANIMALNOM CIKLUSU PROIZVODNJE

SAŽETAK

U studiji je ispitana mogućnost primjene metode mjerenja ukupnog beta zračenja u radijacionoj zaštiti animalne proizvodnje u uslovima kontaminacije okoliša.

Rezultati su pokazali da se kvantitativna mjerenja ukupnih beta emitera u testiranim uzorcima koji su dobijeni metodom određivanja ukupne beta aktivnosti mogu uspješno koristiti u trijaži u slučajevima koji uključuju radioaktivnu kontaminaciju. Važno je primijetiti da se uspješna primjena ove metode zasniva na prethodnom poznavanju prosječnih koncentracija ukupnog kalija ili njegovog izotopa ^{40}K u analiziranim uzorcima.

Kontrolna mjerenja i procjene zasnovane na rezultatima istraživanja korištenjem gama spektrometrijske metode su pokazala prihvatljivu nepouzdanost u trijaži kontaminiranih uzoraka. Korištenjem ove metode u terenskim uslovima omogućila bi se primjena ostalih mjera zaštite sistema animalne proizvodnje tokom radioaktivne kontaminacije okoliša.

Ključne riječi: Beta zračenje, lanac proizvodnje hrane, radioaktivna kontaminacija