RADIOACTIVITY DETERMINATION IN ENVIRONMENTAL THICK SAMPLES USING A GAS FLOW PROPORTIONAL COUNTER

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Abstract

Environmental monitoring is essential for public health and environment protection being performed by measuring the radiation levels or radioactivity from environmental samples. A simple, rapid, and cost-effective method commonly used for environmental radioactivity screening is the global alpha/beta counting method. In global beta counting determinations, the detection geometry is crucial, as it must replicate the calibration geometry to increase the results level of accuracy. The present study aims to optimize the detection geometry for thick samples analysed using a gas flow proportional detector to obtain more accurate results in laboratory measurements. For sufficiently thick sources, the number of beta particles interacting with the detector will reach a limit, the counting rate becoming independent of source thickness. To address this aspect, experimental tests were conducted to determine the net counting rate dependence on activity concentration, considering detection geometry with planchets of larger diameter, different from those usually used in routine laboratory measurements. This new calibration will be further used in laboratory for determining global beta activity in soil, sediment, or thick vegetation samples.

Keywords: *Environmental radioactivity monitoring, Global alpha/beta counting method, Calibration, Detection geometry, Counting rate*

Introduction

Human activities such as mining, milling and processing of uranium ores, and mineral sands, smelting of metalliferous ores, manufacture of fertilizers, drilling of oil and gas, transportation, processing and burning of fossil fuels have raised the concentration of naturally occurring radioactive materials, such as U^{238} series, Th^{232} series and K^{40}, in the environment [1].

Environmental radioactivity monitoring is essential for:
- Public health protection: monitoring ensures that the radiation levels in the environmental remain within safe limits to protect the public health, allowing detection of any abnormal increases in radiation;
- Environmental protection: monitoring helps to assess the impact of human activities (nuclear power generation, mining, industrial processes) on the environment;
- Emergency preparedness: monitoring provides early warning of any accidental release of radioactive material into the environment and allows taking of prompt measures to minimize public exposure and to mitigate the consequences;
- Regulatory compliance: monitoring ensures compliance with regulatory requirements related to radiation protection and environmental safety.

According to the European Atomic Energy Community (Euratom) Treaty, art. 35, European Union (EU) member states have the obligation to perform measurements of the radioactivity in their surroundings, and submit the results (radioactivity content in levels in air, water, soil, and food) to the European Commission (EC). The environmental radioactivity monitoring could be continuous or could be assured by collecting samples from the environment and analysing them in laboratory [2, 3]. European Commission (EC) keeps a Radioactivity Environmental Monitoring database of monitoring organizations and facilities, and verifies the monitoring arrangements. The database also includes details on areas in EU member states radioactively contaminated [3]. The Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN Pitesti (Romania) is participating in interlaboratory comparisons regarding environmental radioactivity, being registered in the EC database.

Established in 1962, the Environmental Radioactivity Monitoring National Network (ERMNN) within the Ministry of Environment, Waters and Forests, is a specialized component of the national radiation protection system, which carries out the supervision and control of compliance with the legal provisions on environmental radiation protection and ensures the detection of increased radioactivity levels, warning and alarming the decision-makers in case of events with radiological impact on the environment.

Under the National Environmental Protection Agency (NEPA) coordination, ERMNN is responsible for monitoring the environmental radioactivity in Romania, being the national data provider, both in normal and emergency situations.

ERMNN constantly report information to the public (via NEPA website, www.anpm.ro), to national decision-makers (Ministry of Internal Affairs, National Commission for Nuclear Activities Control, Ministry of National Defence) and international (European Commission on the EURDEP platform). ERMNN distribution (laboratories and automatic stations) covers all landforms from the sea up to the mountain, and all factors of the environment: air, water, soil and vegetation (spontaneous vegetation) [4].

The monitoring program carried out by the Laboratory of Radioprotection, Environmental Protection, Civil Protection and Medical Analysis at RATEN ICN was configured according to the radiation sources nature and size, and considering the existing exposure routes. It includes all the activities needed to determine the radioactivity levels in the environment, as well as their impact on the environment and population health, during normal operation of the nuclear facilities on the site, and in emergency situations [5].

One of the most commonly used methodology for the environmental radioactivity monitoring is based on sampling and conducting ex-situ determinations in the laboratory [6]. Gross alpha/beta activity measurement is widely applied as a screening technique in the field of environmental monitoring, because is simple, rapid and implies low costs [1]. Gross beta activity concentration in environmental samples is defined as the total radioactivity of all beta emitters in the samples. The presence of gross beta radioactivity in soil and sediments is due to the natural long-lived isotopes K^{40}, Pb^{210} and Ra^{228} [1, 6, 7].

The study mainly targets to optimize the detection geometry for thick environmental samples analyzed using a gas flow proportional counter in order to obtain better accuracy in laboratory measurements.

Materials and methods

Detection equipment

For this research work, MPC 9300 Protean Radiometer was used (see Fig.1). This equipment allows the measurement of gross alpha and beta radioactive concentrations in various environments and for different materials through a gross beta and alpha low background measurement system. Simultaneous or separate alpha plus beta counting modes provide high efficiency detection in a single counting cycle [8].



Fig.1. MPC 9300 low background alpha/beta radiometer

The detection assembly contains a polarization source and a circuit for collecting and processing electrical signals. It can detect various types of ionizing radiation, depending on the detector's constructive characteristics and the radiometer operating mode. MPC 9300 radiometer contains a gas flow proportion counter that can operate both in a windowless configuration and with an 80 μg/cm^2 mylar window. The radiometer is powered using a gas mixture (90% argon, 10% methane), and includes a microprocessor for acquisition, control and data processing. The counting efficiency is 55% for Sr^{90}/Y^{90}, and 42% for Am^{241}, respectively [8].

For gross beta counting, the samples are prepared in two standard geometries: thin-layer geometry, and infinitely thick layer geometry. The results' accuracy significantly depends on the measurement geometry. The thin layer geometry works for submicron samples (e.g., smears for determining loose contamination, liquid effluent samples with a low suspension load etc.). Samples measured in infinitely thick layer geometry must have a mass thickness over 350 mg/cm^2 (tipically, these are soil, sediment or effluent samples with a high suspension load).

Selection of counting geometry

The samples geometry plays a significant role in counting efficiency, so it is important to match the calibration source and the samples geometries. The detection geometry could vary when determine gross beta activity in environmental samples [9]. For soil and sediment, the samples have to be dried, milled and homogenized before being placed in planchets to ensure a high mass thickness. Non-uniformity of the sample's matrix impedes on the method's accuracy and precision. To obtain reliable results, for thicker sources uniform thickness and homogeneous layer of solid material (matrix) is mandatory.

For sufficiently thick sources (mass thickness of samples > 350mg/cm^2, according [9]), the number of beta particles interacting with the detector will reach a limit and thereafter the count rate becomes independent of the source thickness. In that case, the dependence of the net counting rate on the activity concentration should be assessed. For this aim, a series of experimental tests were performed to determine the detector response depending on the activity concentration.

Two detection geometries were selected (see Fig. 2), as follows: ▪ Geometry 1: aluminium planchet of 29 mm in diameter and 7 mm in height; ▪ Geometry 2: stainless steel planchet of 50 mm in diameter and 6 mm in height.

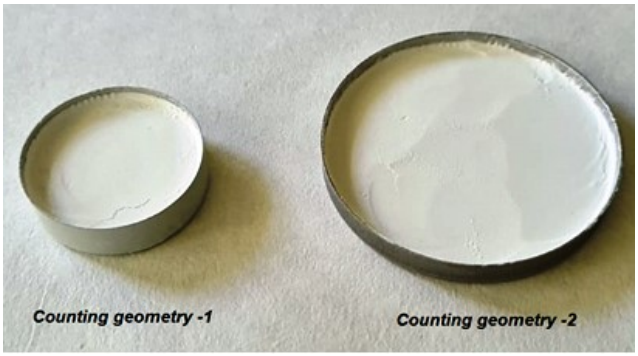


Fig. 2. The selected counting geometries

Another important aspect of the counting geometry is the distance between the sample and the active volume of the detector [9]. For both considered geometries, the solid material's filling height was 2/3 of the planchet walls height (as shown in Fig. 2). For this step, a solid matrix consisting of a known amount of talc powder spiked with radioactive standard solution (pure β emitter 90Sr/Y) was used.

Calibration sources preparation

The counting system calibration can be realized with a ^{90}Sr/Y standard solution, as indicated in ISO standard no. 18589-6:2019 [10].

To study the dependence of the net counting rate on the activity concentration it was necessary to prepared 4 (four) calibration sources for each of the two considered detection geometries, using amounts ranging between 3 g and 5 g of solid matrix. The solid matrix consisted of a known amount of talc powder spiked with a ^{90}Sr/Y standard solution having the activity concentrations ranging from 2 Bq/g to 22 Bq/g.

The radioactive solid matrix was prepared in 8 glass vials (an empty vial weighing about 14.3 g) which were dried on the sand bath under the infrared lamp. After the total evaporation was achieved, the radioactive solid matrix from the vials was vigorously mixed to ensure the solid material became homogenous. Figure 3 presents the calibration sources during the preparation stage.

Table 1 presents the activity concentration values for eight calibration sources corresponding to the two detection geometries considered in the study.



Fig. 3. The preparation stage of the calibration sources

Table 1. Activity concentrations for the calibration sources

Source ID	Mass of spiked talc powder (g)	Mass of radioactive solution (^{90}Sr/Y) (g)	Activity concentration of radioactive solution (Bq/g)	Activity concentration of spiked talc powder (Bq/g)	Net counting rate (cpm)	Counting geometry
1	2.9894	0.0259	227.00	1.97	37.7	Geometry 1
2	3.0019	0.0841		6.36	105.5	
3	3.0217	0.1540		11.57	163	
4	2.9852	0.2876		21.87	288.58	
A	4.9892	0.0375	439.69	3.30	91.48	Geometry 2
B	4.9690	0.0800		7.08	235.21	
C	5.0184	0.1528		13.39	441.37	
D	5.0848	0.2550		22.05	826.73	

Previously reported gross beta activity concentration measurements using the gas flow proportional counter

According to the National Norms for monitoring radioactivity in the vicinity of a nuclear or radiological installation site [11, 12] (applicable for RATEN ICN which operates TRIGA 14MW Research Reactor), the locations for monitoring are selected as close as possible to the exposure pathway end. Four different categories of locations are considered in the RATEN ICN's Monitoring Program, such as indicator, background, control and supplementary locations. Sampling and measurement of soil and vegetation samples are performed every half-year for the supplementary location, and once a year for the indicator, control and background locations [5, 11, 12].

The beta global activity concentrations for environmental samples collected on the RATEN ICN site between 2014 and 2018, and measured using the MPC 9300 counter, were previously internationally reported in [13]. The results obtained shown that the beta global activity concentrations in soil samples collected from the supplementary locations were by 6.8% up to 37% lower than the average values measured in the other locations. For the vegetation samples the beta global concentrations in the supplementary locations were even lower than those measured in other locations by 38% up to 66%. The study concluded that no negative impact on the environment was registered due to the nuclear activities carried out on the RATEN ICN site.

The beta global activity concentrations from soil and vegetation samples from monitoring locations specified in the Monitoring Program [5] are charted and regularly reported by RATEN ICN Pitesti to the Arges Environmental Protection Agency, to be included in the annual reports of the National Environmental Protection Agency [14-19].

The Laboratory of Radiation Protection (RATEN ICN) participated to the Joint Research Center-EC interlaboratory comparisons within the framework of the Euratom Treaty. Performance of about 75 laboratories in determining activity concentrations of up to 15 natural and anthropogenic radionuclides in soil (^{40}K, ^{90}Sr, ^{137}Cs, ^{212}Pb, ^{212}Bi, ^{214}Pb, ^{214}Bi, ^{226}Ra, ^{230}Th, ^{232}Th, ^{234}U, ^{235}U, ^{238}U, ^{238}Pu, and ^{239}Pu+^{240}Pu) was reported in [20]. Our laboratory determined ^{90}Sr activity concentration by using the low background gross alpha/beta counter MPC 9300. Technical evaluation of REM (Radioactivity Environmental Monitoring) 2019 gross α/β activity concentration in drinking water was performed in [21]. Our laboratory carried out proficiency tests by using multiple counting techniques (gas-flow proportional counter, and liquid scintillation counter) according to standard efficiency calibration procedures.

Experimental tests and results

Assessment of the net counting rate dependence on the activity concentration

For each of the two considered counting geometries, four planchets with the same thickness were prepared using known amounts of the radioactive powder, as follows: Source id: 1, 2, 3, 4 for Geometry 1, and Source ID: A, B, C, D for Geometry 2. It was also considered that the calibration sources filling level for a considered geometry had to be identical, so that during the counting the distance between sample and detector remains constant.

Each calibration source was counted with the gas flow proportional counter system for a long enough time to assure that the uncertainties due to statistics are satisfactory. Twenty measurements for each of the 4 calibration sources for Geometry 1, and also for Geometry 2, were performed.

The evolution of the averaged values of the measured count rates evolution with the activity concentration, for each calibration source, is depicted in Fig. 4 (Geometry 1-left, Geometry 2 – right).

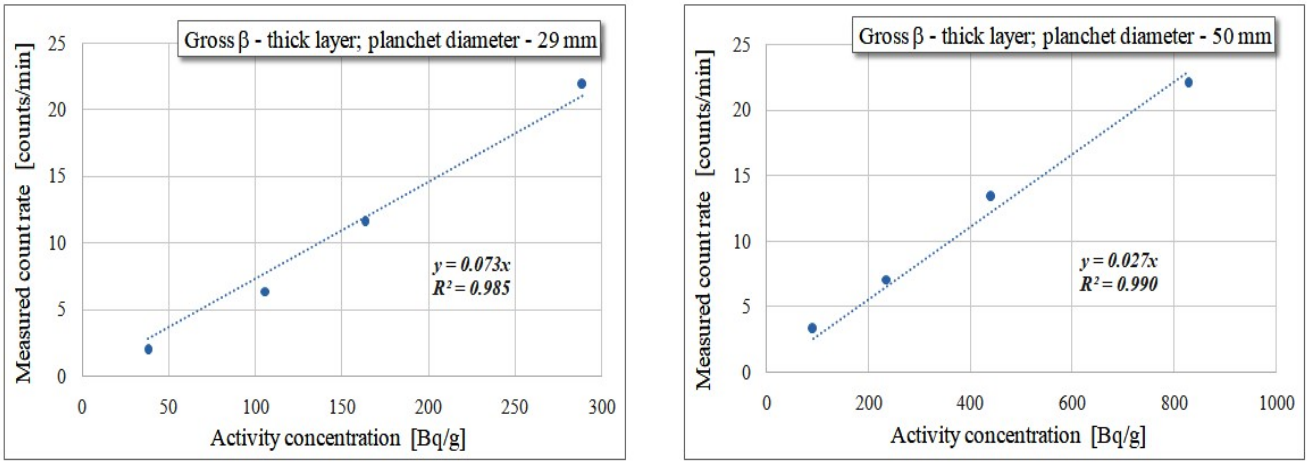


Fig. 4. Dependence of the net counting rate on the activity concentration for Geometry 1 (left) and Geometry 2 (right)

Methods validation

The gross β activity concentrations of five (5) soil samples were determined, using the two counting geometries of interest. The samples were selected from the existing control samples with known activity concentration, the selection assuring that the samples were characterized by different activity concentrations.

The dry, milled and homogenized soil samples with different activity concentrations were placed within the two types of counting planchet, in the same geometry with the calibration sources used for efficiency detection, and a total of 10 samples were obtained. The samples were counted using the gas flow proportional detector for one hour, sets of 10 measurements being performed for each sample and the final values being averaged over each measurements set.

Table 2 shows the results obtained using the two detection geometries, and also the calculated relative differences using the formula given below:

$$\text{Rel. diff} = (C_{\beta i} - C_{\beta 0})/C_{\beta i} \quad , i=1, 2$$

where $C_{\beta 0}$ = known β gross activity concentration of the control sample, and $C_{\beta i}$ = β gross activity concentration of control samples in Geometry 1 (i=1) or Geometry 2 (i=2).

The relative differences calculated for both detection geometries show that the gross beta activity concentrations determined using the Geometry 2 are very close to those of the control (up to 8% higher for 3 control samples, and up to 3% lower for the other 2 control samples) comparatively with the values determined using Geometry 1 (up to 17% - 30% higher), as can be seen in Table 2.

Table 2. Gross beta activity concentration of soil samples

Sample	Gross beta activity concentration (Bq/kg)			Relative difference (%)	
	Control sample	Geometry 1 * (D_1 - 29 mm)	Geometry 2 * (D_2 - 50 mm)	Geometry 1 vs Control sample	Geometry 2 vs Control sample
Sample 1	825.54	1181.78	863.04	30.14	4.35
Sample 2	750.17	955.90	818.44	21.52	8.34
Sample 3	1130.04	1532.66	1100.71	26.27	-2.66
Sample 4	3007.07	3662.07	3020.87	17.89	0.46
Sample 5	9509.47	11478.65	9396.30	17.16	-1.20

* Detection geometries considered:
Geometry 1 – planchet with diameter D_1= 29 mm and height H_1=7 mm;
Geometry 2 – planchet with diameter D_2= 50 mm and height H_2=6 mm

Conclusions

Environmental monitoring involves measuring of the level of radiation or radioactivity in environmental samples, aiming to assess the exposure of the population and the impact on the environment. The most used methodology for monitoring environmental radioactivity is based on sampling and conducting ex-situ determinations, in the laboratory, using the gross alpha/beta counting method.

Source thickness is important in determinations of gross beta activities in soil, sediments or water samples with high residue content because of self-absorption and energy losses. Alpha and beta particles emitted at small angles from thin samples placed in deep planchets will lose energy in interaction with the planchet walls.

In this study, the net counting rate dependence on the activity concentration was assessed considering two detection geometries, the difference residing in the size of the planchets.

The gross beta activity concentrations in five soil samples were analyzed taken into account two different detection geometries, the calculated relative differences ranging from 12% to 24%, values acceptable according to previous measurements and calibrations performed in the laboratory. However, the quality of measurements can be checked and improved in the framework of proficiency.

Increasing the detection surface area between the source and detector implicitly leads to better detection efficiency, taking into consideration rather small differences registered between the concentration measurements using Geometry 2 and the known values for selected control samples.

The results obtained recommend using of Geometry 2 and the new calibration for gross β activity determination in thick layer environmental samples (such as soil, sediment or vegetation samples), considering participation of the RATEN ICN laboratory in international comparison tests and the regularly reports towards NEPA.

References

[1] Engelbrecht, R., Schwaiger, M. (2008) "State of the art of standard methods used for environmental radioactivity monitoring", *Applied Radiation and Isotopes*, Vol. 66, pps. 1604– 1610

[2] European Parliament ″Euratom Treaty″, official website, https://www.europarl.europa.eu/about- parliament/en/in-the-past/the-parliament-and-the-treaties/euratom-treaty

[3] Joint Research Center ″Environmental radioactivity monitoring″, European Commission official website, https://www.remon.jrc.ec.europe.eu/About/Environmental/Monitoring

[4] ″Environmental Radioactivity Monitoring National Network″, National Environmental Protection Agency official website, www.anpm.ro

[5] Romanian Government, Ministry of Environment (2023) "Monitoring Program on environmental radioactivity ICN-FCN", annex regarding the "Revised Environmental Authorization for National Society Nuclearelectrica S.A., Nuclear Fuel Factory Pitesti, 2023", HG nr. 568/7 July 2023, published in Official |Bulletin no. 633bis partI, www.monitoruljuridic.ro/act (in Romanian)

[6] US Environmental Protection Agency, US Department of Defense, US Department of Energy, US Department of Homeland Security, US Nuclear Regulatory Commission, US Food and Drug Administration, US Geological Survey, National Institute of Standards and Technology (2004) "Multi-Agency Radiological Laboratory Analytical Protocols Manual (MARLAP)", Vol. II, Chapter 15 "Quantification of Radionuclides", NUREG-1576, EPA 402-B-04-001B, NTIS PB2004-105421, www.epa.gov/radiation/marlap, United States

[7] Martın Sanchez, A., Saenz Garcıa, G., Jurado Vargas, M. (2009) "Study of self-absorption for the determination of gross alpha and beta activities in water and soil samples", *Applied Radiation and Isotopes*, Vol. 67(5), pps. 817-820, DOI: 10.1016/j.apradiso.2009.01.040

[8] "MPC 9300 Operations Manual", Protean Instrument Corporation, https://proteaninstrument.com/ products/multi-detector-systems

[9] Dulama, C., Toma, A., Dobrin, R. (2012) "Radiation measurement methods with application in radioprotection", ISBN 978-606-560-299-1, Ed. Univ. of Pitesti (in Romanian)

[10] International Organization for Standardization (2019) "Measurement of radioactivity in the environment - Soil - Part 6: Gross α and gross beta activities - Test method using gas-flow proportional counting", ISO 18589-6:2019, Ed.2.
[11] National Commission for Nuclear Activities Control (2023) "Norms for the monitoring of environment radioactivity in the vicinity of a nuclear or radiological facility", CNCAN President order no.275/2005, published in Romanian Official Bul. No. 923/2005
[12] National Commission for Nuclear Activities Control (2000) "Fundamental Norms for Radiological Safety, NSR01", CNCAN President order no. 14/2000, published in Romanian Official Bul. No. 404/2000
[13] Tudor, G.I., Paslaru, G.M., Valeca, M. (2019) "Gross Beta Activity Concentration Monitoring in Soil and Vegetation", Proc.of the 12th Int. Conf. on Sustainable Development through Nucl. Research and Education Nuclear 2019, Pitesti, Romania, incl. in IAEA INIS database, https://inis.iaea.org/collection/NCLCollectionStore/Public/52/020/52020010.pdf
[14] Arges Environmental Protection Agency (2018) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), incl. in "Report of Indicators Year 2018", Ministry of Environment, Waters and Forestry and National Environmental Protection Agency, https://anpm.ro/documents/12220/2209838/Indicatori+RSM+2018+-+EN.pdf/7875bbc5-b9e4-4e83-821c-f7a8fc9ac487
[15] Arges Environmental Protection Agency (2019) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), incl. in "Report of Indicators Year 2019", Ministry of Environment, Waters and Forests and National Environmental Protection Agency, https://anpm.ro/documents/12220/2209838/Raport+indicatori+2019_EN.pdf/6853413b-0463-4601-a303-8a8e8d4b0c08
[16] Arges Environmental Protection Agency (2020) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), incl. in "Report of Indicators Year 2020", Ministry of Environment, Waters and Forests and National Environmental Protection Agency, https://anpm.ro/documents/12220/2209838/Raport+2020_EN.pdf/84d75a1b-6fb5-426e-8e66-d88e439c3723
[17] Arges Environmental Protection Agency (2021) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), incl. in "Report of Indicators Year 2021", Ministry of Environment, Waters and Forests and National Environmental Protection Agency, https://anpm.ro/documents/12220/2209838/RSM+2021_EN.pdf/d8edf4ca-ef95-4846-a296-283b6f4c9ae7
[18] Arges Environmental Protection Agency (2022) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), to be incl. in "Report of Indicators Year 2022", Ministry of Environment, Waters and Forests and National Environmental Protection Agency, https://anpm.ro/ raport-de-mediu
[19] Arges Environmental Protection Agency (2023) "Monitoring Report on Environmental Radioactivity on the INR platform and its vicinity" (in Romanian), to be incl. in "Report of Indicators Year 2023", Ministry of Environment, Waters and Forests and National Environmental Protection Agency, https://anpm.ro/ raport-de-mediu
[20] Meresova, J., Watjen, U., Altzitzoglou, T. (2012) "Evaluation of EC Interlaboratory Comparison on Radionuclides in Soil", Joint Research Center-European Commission report JRC71619, EUR 25360 EN, ISBN 978-92-79-25111-5, Belgium
[21] Jobbagy, V., Dupuis, E., Emteborg, H., Hult, M. (2021) "REM 2019 proficiency test on gross alpha/beta activity concentration in drinking water", Joint Research Center-European Commission report JRC121498, EUR 30822 EN, ISBN 978-92-76-41388-2, Belgium.