



Implementation of a methodology for analysis of gross alpha and gross beta, as water potability parameters in Brazil, and ^{226}Ra , ^{228}Ra and ^{210}Pb for radiological water monitoring

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Abstract: In order to guarantee the quality and safety of water intended for human consumption in Brazil, the GM/MS Regulation N° 888, of 4 May 2021, of the Ministry of Health, establishes parameters, through laboratory analyses, to monitor substances that pose a risk to human health. As far as radiological standards are concerned, the activity concentration limits must not exceed 0.5 Bq/L for gross alpha and 1.0 Bq/L for gross beta. In the case of alpha/beta values exceeding the maximum permitted value, other natural and/or artificial radionuclides present in the water must be quantified. The radionuclides most likely to be found in water intended for human consumption are radium and uranium, which are the main fractions of the doses received by man from internal emissions derived from radium, ^{226}Ra and its decay products. In order to comply with environmental regulations and to be a conformity assessment organism, competent to carry out ionizing radiation tests, necessary for the monitoring of drinking water in Brazil; the Centro de Biologia Experimental Oceanus laboratory, located in Rio de Janeiro, carried out gross alpha and gross beta analyses, using the gas flow proportional counter technique; a the analyses of ^{226}Ra , ^{228}Ra and ^{210}Pb , using the radiochemical separation technique, followed by alpha/beta counting; in water for human consumption. It was necessary to obtain results on the equipment's operating plateau, background, efficiency and factors related to the attenuation effect of alpha counts. The laboratory has demonstrated the reliability of its results by participating in the National Intercomparison Programme - PNI, and by requesting for accreditation from the Brazilian Network of Testing Laboratories - RBLE, according to the ABNT ISO/IEC 17025 standard; in the category of ionising radiation tests.

Keywords: gross alpha, gross beta, radium, potability, water.



Implementação de metodologia para análise de alfa total e beta total, como parâmetros de potabilidade da água no Brasil, e de ^{226}Ra , ^{228}Ra e ^{210}Pb para monitoramento radiológico da água

Resumo: A fim de garantir a qualidade e a segurança da água destinada ao consumo humano no Brasil, a portaria GM/MS nº 888, de 04 de maio de 2021, do Ministério da saúde, determina parâmetros, através de análises laboratoriais, para monitoramento de substâncias que representam risco à saúde humana. Quanto aos padrões radiológicos, os limites relacionados à concentração da atividade não devem exceder 0.5 Bq/L para alfa total, e 1.0 Bq/L para beta total. No caso de níveis alfa/beta acima do valor máximo permitido, devem ser quantificados outros radionuclídeos naturais e/ou artificiais, presentes na água. Os radionuclídeos mais prováveis de se encontrar em águas para consumo humano são o rádio e o urânio, sendo as principais frações de doses de emissões internas, que os seres humanos recebem, derivados do rádio, ^{226}Ra e seus produtos de decaimento. Para atender as portarias ambientais e ser um organismo de avaliação da conformidade, competente a realizar ensaios de radiação ionizante, necessários ao monitoramento da água potável no Brasil; o laboratório Centro de Biologia Experimental Oceanus, localizado no Rio de Janeiro, implementou as análises alfa total e beta total, através da técnica contador proporcional de fluxo gasoso; e as análises de ^{226}Ra , ^{228}Ra e ^{210}Pb , através da técnica separação radioquímica, seguida de contagem alfa/beta; em água para consumo humano. Foi necessário obter resultados de platô operacional do equipamento, de background, eficiências e fatores relacionados ao efeito de atenuação das contagens alfa. O laboratório comprovou a confiança dos seus resultados em participação no Programa Nacional de Intercomparação – PNI, e através da solicitação de acreditação na Rede Brasileira de Laboratórios de Ensaio – RBLE, pela norma ABNT ISO/IEC 17025; na categoria de ensaios de radiação ionizante.

Palavras-chave: alfa total, beta total, rádio, potabilidade, água.

1. INTRODUCTION

The procedures to assess water quality are carried out in accordance with the resolutions of the National Environmental Council (CONAMA): n° 430/2011 (Brazil, 2011), n° 396/2008 (Brazil, 2008) and n° 357 /2005 (Brazil, 2005) (Xavier, et al., 2022). Regarding potability, the determination of laboratory analyzes to evaluate water must comply with the regulations of the Ministry of Health (MS) (Xavier, et al., 2022). Ordinance GM/MS no 888, of May 4 2021, from the Ministry of Health, establishes procedures for controlling and monitoring the quality of water for human consumption and its potability standard (MS, 2021). The water supply is managed according to defined laboratory parameters and their respective maximum permitted values.

The application of the regulation GM/MS no 888:2021 covers water intended for human consumption, coming from a water supply system, alternative solution for collective and individual water supply and water tankers (MS, 2021). The potability of water is determined by microbiological, organoleptic, radioactive and chemical standards (inorganic, organic, pesticides, disinfectants and secondary disinfectants) that pose a risk to human health (Xavier, et al., 2022). Regarding radiological standards, in Brazil, Regulation GM/MS no 888:2021 establishes limits for the concentration of gross alpha and gross beta activity. For alpha, the activity concentration values must not exceed recommended value of 0.5 Bq/L, and for beta, the activity concentration values must not exceed recommended value of 1.0 Bq/L. In the case of alpha/beta values above the maximum permitted value, other natural or artificial radionuclides that may be present in the water must be quantified. The National Nuclear Energy Commission (CNEN) may assess the potability of water in accordance with the specifications of Regulatory Position 3.01/012:2020 (MS, 2021).

In Europe, Directive no 51 of the Euratom Community of 22 October 2013 establishes requirements for the protection of the health of the general public with regard to radioactive substances present in water intended for human consumption. Considering that drinking water is one of the routes by which radioactive substances are absorbed by the human body, practices that pose a risk of exposure of the general public to ionising radiation must be avoided and controlled in order to keep levels as low as reasonably achievable (EURATOM, 2013). According to Euratom 2013, water monitoring strategies may require the quantification of specific radionuclides or a specific radionuclide if gross alpha and gross beta values exceed the recommended levels of 0.1 Bq/L for gross alpha activity and 1.0 Bq/L for gross beta activity (EURATOM, 2013).

Water in conditions complying with the gross alpha and gross beta limits may be considered to have an indicative dose below the indicator value of 0.1 mSv (EURATOM, 2013). No specific radiological examination is required. It is required only if it is known from other sources of information that specific radionuclides are present in the supply water and are likely to give an indicative dose above 0.1 mSv (EURATOM, 2013).

To enable the monitoring of key parameters in the control of drinking water, ionising radiation testing laboratories are fundamental tools in the process. When they perform their tests in accordance with the requirements of ABNT NBR ISO/IEC 17025 - General requirements for the competence of testing and calibration laboratories; they promote greater confidence. Accreditation enables them to work competently and produce valid results (ABNT NBR ISO, 2017).

The gross alpha and gross beta tests are fundamental parameters for the radiological screening of water, as they guarantee that the reference levels of specific alpha and beta emitters have not been exceeded, in compliance with the maximum permitted value. According to the CNEN, if the gross alpha and gross beta screening levels are exceeded, the contribution of ⁴⁰K must be subtracted, as it is a natural radioisotope of potassium. The site

should be re-sampled for alpha and coarse beta; the persistence of values above the established level will allow the CNEN to request specific analyses of natural and/or artificial radionuclides that may be present in the water (CNEN, 2020).

The radionuclides most likely to be found in water intended for human consumption are radium and uranium. For uranium, aspects of the chemical toxicity of this compound to the kidneys are taken into account (IRPA, 2013). The major dose fractions received by humans from internal emissions are radium, ^{226}Ra and their decay products (Corrêa, 2011). ^{226}Ra has a long half-life of 1600 years and decays to ^{222}Rn radon, which has a half-life of 3.82 days. The decay of ^{222}Rn is followed by the successive disintegration of alpha and beta emitters with short half-lives. This radioactive chain ends with the stable lead ^{206}Pb after several decay steps (Corrêa, 2011). Water is one of the routes by which natural radionuclides, mainly ^{226}Ra and ^{222}Rn , are transferred to humans (Oliveira, 1998).

The radiological aspects of ^{226}Ra are very relevant for studies evaluating the transfer of natural radionuclides to humans, due to its long half-life, although we know that the concentrations of ^{222}Rn present in water can be higher compared to those of ^{226}Ra (Corrêa, 2011). Geological and geochemical factors are directly related to the amount of radium in water, influencing the concentration of the two main isotopes ^{226}Ra and ^{228}Ra , which are daughters of uranium and thorium, respectively (Eisenbud and Gesell, 1997). Research shows that in several cases, especially in well water, the amount of radium exceeds the permissible limits; the most studied elements in water are ^{226}Ra , ^{228}Ra , ^{222}Rn and ^{210}Pb (Corrêa, 2011). ^{210}Pb has an affinity for dense organs, such as the liver and bones, and its degradation chain in the skeleton contributes significantly to the increase in the internal dose received by humans (Costa Júnior, et al., 2013).

The Oceanus Experimental Biology Center Laboratory, located in the city of Rio de Janeiro, seeks to comply with environmental regulations as a Conformity Assessment organism, competent to perform laboratory analyses necessary for the monitoring of waters

in Brazil. The Laboratory is accredited under no CRL 0306 in the ABNT NBR ISO/IEC 17025:2017 standard for biological, toxicological, metal, organic compound, physicochemical and bacteriological testing (INMETRO: CRL 0306). However, based on environmental compliance studies, the need to perform and accredit the tests for gross alpha, gross beta, ^{226}Ra , ^{228}Ra and ^{210}Pb ionising radiation in water has been verified.

2. MATERIALS AND METHODS

In order to perform the gross alpha and gross beta analyses, the ISO 10704 technical standard, dated February 2019, was used as a reference. The standard defines a methodology for the determination of alpha and beta activities using a detector or alpha and beta particle counting system. that has been previously calibrated with alpha and beta emission standards. The aim is to obtain a fine and homogeneous deposit directly on a stainless steel plate by progressive evaporation of the samples.

To carry out radium and lead analyses, the laboratory used the radiochemical separation technique, followed by alpha and beta countings; after the secular equilibrium of ^{222}Rn and ^{210}Bi , determination of efficiencies and determination of the K and η_{abs} factors. The test begins with the addition of stable Ba and Pb, as carriers of Ra and Pb isotopes, to calculate the gravimetric chemical yield and evaluate the percentage of recovery at the end of the radiochemical separation (Moura, 2018). The radiochemical separation is carried out by precipitating the isotopes in the form of sulphates (BaSO_4 , RaSO_4 and PbSO_4), followed by precipitation of $(\text{Ba}(\text{Ra})\text{SO}_4)$ for the analysis of ^{226}Ra and ^{228}Ra , and precipitation of PbCrO_4 for the analysis of ^{210}Pb . The samples are filtered and the filters, after drying under an infrared lamp, are stored in a desiccator; for alpha and beta counting, after the secular equilibrium of ^{222}Ra and ^{210}Bi .

To obtain the activities of ^{226}Ra , ^{228}Ra and ^{210}Pb , secular equilibrium was ensured after 21 days, since the Ra and Pb counts, according to the technique used, refer to their children ^{222}Rn and ^{210}Bi . In the search for radioactive equilibrium, isotopes can enter secular equilibrium through decay, forming a new radioactive element (Tauhata *et al.*, 2003). This process continues through successive decays, forming smaller and better organised elements. Radioactive families or series are formed by the emission of radiation from the parent element (Tauhata *et al.*, 2003). It is recommended that count measurements are obtained after secular equilibrium, although it is possible to establish a model for correction (Moura, 2018). The laboratory used the work of Costa, 2016, carried out at the Instituto de Pesquisas Energéticas e Nucleares (Institute of Energy and Nuclear Research – IPEN), and Moura, 2018, carried out at the Centro Regional Ciências Nucleares do Nordeste (Northeast Regional Nuclear Sciences Center - CRCN-NE), as references for the stages of radiochemical separation, preparation of curves, determination of efficiencies and quantification of activities in Bq/L.

2.1. Proportional gas flow counter and use of ionizing radiation sources

The laboratory used a gas flow proportional counter for alpha/beta counts, fed with P-10 gas (argon/methane) and calibrated using Apex alpha-beta software (Mirion Technologies, 2017). Background was measured, and standard sources of ^{241}Am and ^{90}Sr were used to determine operating plateaus and zero efficiencies. The sources were collected in electrodeposited geometry, on stainless steel plates, compatible with the geometry of the instrument and certified by Eckert and Ziegler. Considering that a particle may not enter the active volume of the detector depending on several factors such as thickness, density, detector window and energy, the absolute efficiency of the detection system is valid in the considered counting geometry (Knoll, 1989).

Since the alpha particle counting efficiency is directly dependent on the source thickness, it was necessary to estimate the attenuation factor (ISO, 2019). To calculate the

alpha attenuation factor, a known alpha activity curve with mass variation was constructed, using a standard ^{241}Am source with liquid geometry, certified by the Instituto de Radioproteção de Dosimetria (Institute of Radioprotection and Dosimetry – IRD). For the beta efficiency, attenuation phenomena are negligible (ISO, 2019).

To convert equipment counts into ^{226}Ra activity concentration, using the proportional counting technique, it is necessary to use zero alpha efficiency, and obtain factors derived from the ^{226}Ra alpha attenuation curve: $\eta_{\text{abs(Ra-226)}}$ and K , which are the attenuation coefficient and the attenuation constant (Costa, 2016)(Moura, 2018). $\eta_{\text{abs(Ra-226)}}$ was obtained by intersecting the straight line with the ordinate axis (Moura, 2018). The constant K considers the difference between the attenuation coefficients of the 4 α particles that are emitted in the decay of ^{226}Ra (Godoy and Schuttelkopf, 1987). These have the energies: ^{226}Ra (α energy of 4.8 MeV, 94.5%), ^{222}Rn (α energy of 5.5 MeV, 100%), ^{218}Po (α energy of 6.1 MeV, 100%) and ^{214}Po (α energy of 7.8 MeV, 100%) (Costa, 2016). To prepare the ^{226}Ra attenuation curve, a solution of known ^{226}Ra activity was used, using a standard ^{226}Ra source, acquired in liquid geometry, and certified by the Instituto de Radioproteção de Dosimetria (Institute of Radioprotection and Dosimetry – IRD).

In order to convert the number of devices to the concentration of ^{228}Ra and ^{210}Pb activity, it is necessary to obtain the efficiency of these isotopes (Costa, 2016), using solutions of known activity obtained from standard sources of ^{228}Ra and ^{210}Pb . The solutions used were obtained from standard sources with liquid geometry, the ^{228}Ra source certified by ERA Waters and the ^{210}Pb source certified by the Instituto de Radioproteção de Dosimetria (Institute of Radioprotection and Dosimetry – IRD). All sources obtained in liquid geometry were counted on stainless steel plates compatible with the instrument geometry.

2.2. Internal performance evaluation

Replicates of the gross alpha, gross beta, ^{226}Ra , ^{228}Ra and ^{210}Pb assays were performed using samples prepared with standard liquid geometry sources; added at known concentrations. The results were statistically evaluated for accuracy and precision. Accuracy was calculated from the percentage of recovery and the percentage of error to assess trends by combining random and systematic error components (CGCRE, 2020). Precision was calculated from the percentage of repeatability, intermediate precision and reproducibility, expressed as standard deviation and coefficient of variation (CGCRE, 2020).

2.3. External Performance Reviews

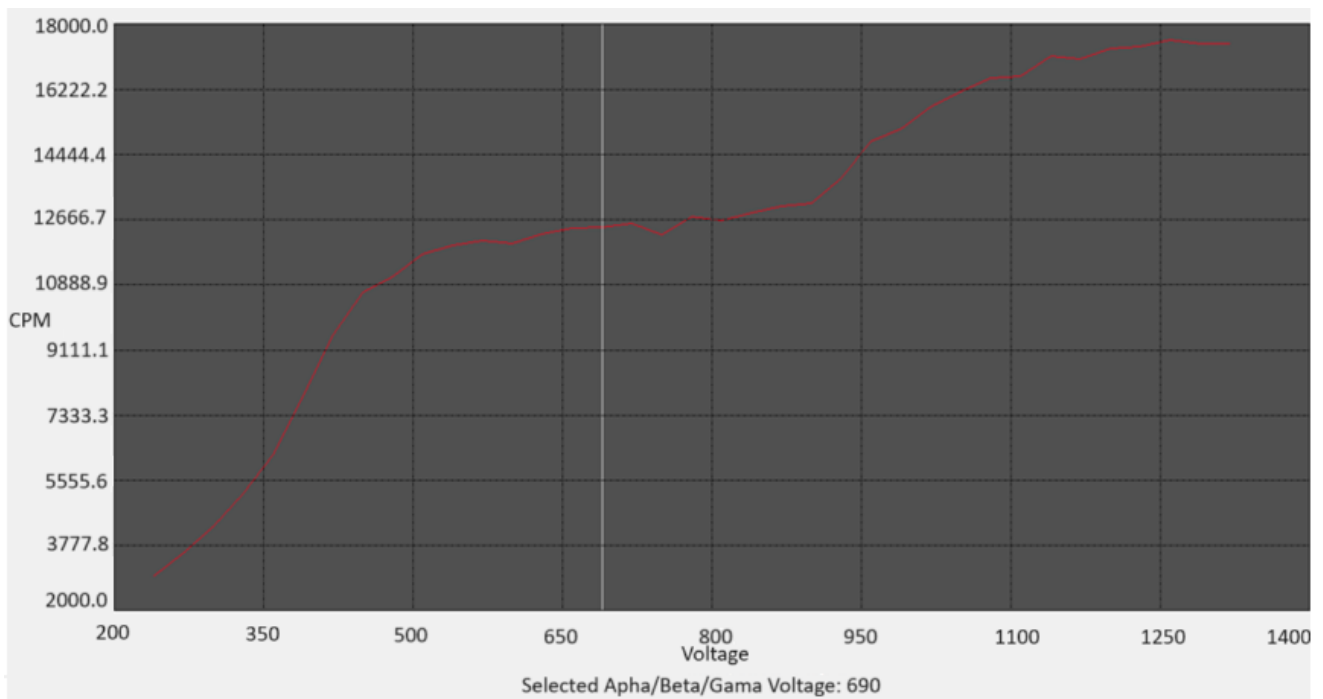
In order to externally validate its results, the laboratory participated in a proficiency test, in the April/2023 round of the National Intercomparison Plan - PNI, provided by the Instituto de Radioproteção de Dosimetria (Institute of Radioprotection and Dosimetry – IRD) (CNEN/IRD, 2021). The Radiation Protection Division of the IRD has been participating in the National Intercomparison Programme - PNI - for the analysis of environmental samples for the determination of radionuclides since 1991, together with the Metrology Department with the area of radionuclide metrology (CNEN/IRD, 2021).

In order to become a Conformity Assessment Organism - OAC (ABNT NBR ISO, 2021), in the field of ionising radiation, the laboratory applied for accreditation from the Brazilian Network of Testing Laboratories - RBLE (INMETRO/CGCRE, 2024), in the category of ionising radiation testing. The laboratory was evaluated for compliance with the requirements of the ABNT NBR ISO/IEC 17025 standard, in the Quality Management System and in the following tests: gross alpha, gross beta, ^{226}Ra , ^{228}Ra and ^{210}Pb in water. Conformity assessment organism are authorised to provide conformity assessment services, except accreditation (ABNT NBR ISO, 2021).

3. RESULTS AND DISCUSSIONS

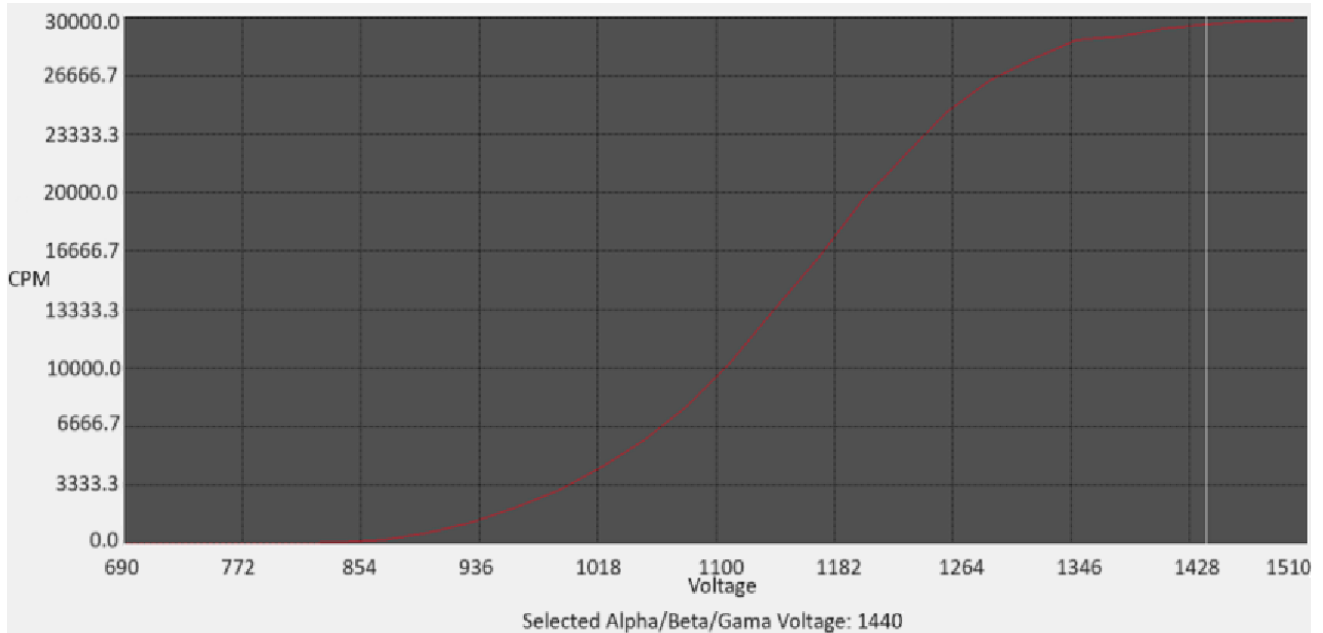
The laboratory performs gross alpha and gross beta counts at different operating plateaus, which improves count rate performance. The alpha operating plateau determined was 690V, obtained from the Apex alpha-beta software using the graph in figure 1 (Mirion, 2017). The determined beta operating plateau was 1440V, obtained from the Apex alpha-beta software using the graph in figure 2 (Mirion, 2017).

Figure 1: Alpha operating plateau determination chart performed by the Apex alpha-beta software. Voltage x counts per minute (CPM).



Fonte: Software Apex Alpha/Beta.

Figure 2: Beta operating plateau determination chart performed by Apex alpha-beta software. Voltage x counts per minute (CPM).



Fonte: Software Apex Alpha/Beta.

After determining the plateaus, the background radiation was measured. The alpha and beta backgrounds used to determine zero efficiency are shown in table 1.

Table 1. Alpha and beta background rate results. Where CPM= Count per minute; and U= counting uncertainty.

BACKGROUND	CPM	U
Alpha	0.0111	0.0333%
Beta	0.689	0.326%

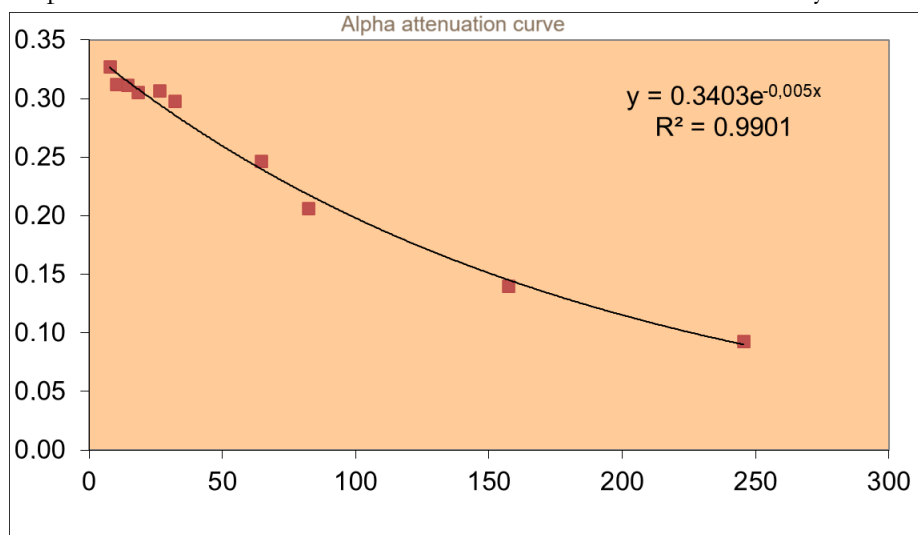
After the approval of the backgrounds, the zero alpha efficiency was calculated with a standard source of ^{241}Am and the zero beta efficiency was calculated with a standard source of ^{90}Sr . The results are shown in table 2. The approval criterion was determined by the technical qualification of the equipment carried out by a company representing the manufacturer of the proportional gas flow meter.

Table 2. Results of zero alpha and zero beta efficiency determination.

ANALYSIS	ZERO EFFICIENCY	CRITERION
Alpha	37.8%	> 32%
Beta	47.6%	> 45%

After obtaining the zero alpha efficiency, it is necessary to estimate the attenuation factor for gross alpha in order to determine the alpha efficiency of each sample according to the mass analysed. The number of alpha particles depends directly on the thickness of the sample (ISO, 2019). The alpha attenuation curve was obtained after mass variations with known activity. The curve obtained to study the attenuation effect of alpha radiation is shown in figure 3. The curve obtained is accepted with $R^2 > 0.99$. In this study, it was used to obtain the alpha efficiency per mass for each sample, using the graph equation based on linear and angular coefficients. Where x is the mass of each sample used to construct the curve and y is the efficiency associated with that mass.

Figure 3. Alpha attenuation curve obtained from the variation of efficiency versus mass (mg).



Fonte: Oceanus Experimental Biology Center.

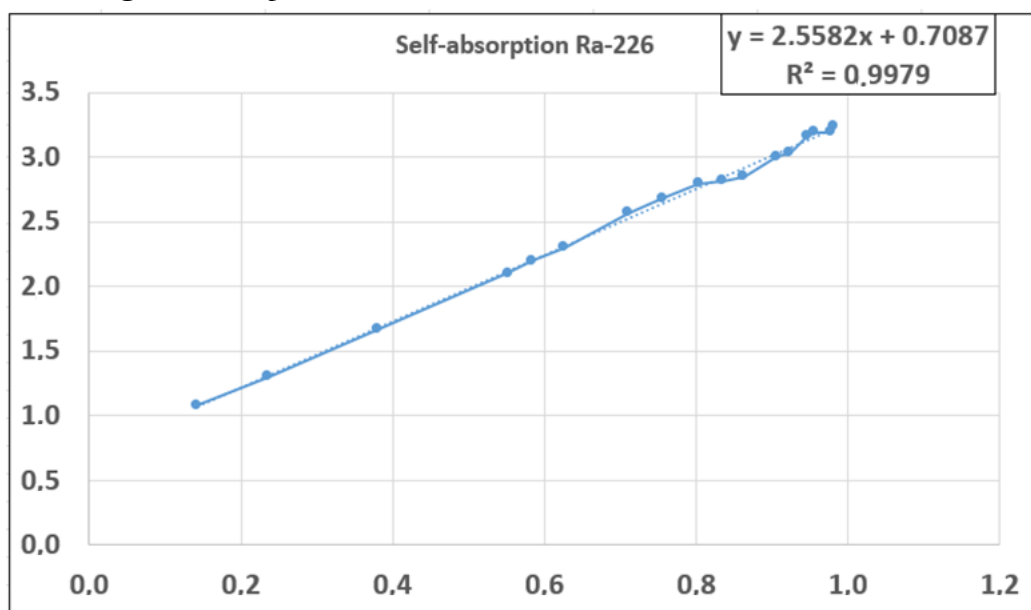
By calculating the concentration of gross alpha and gross beta activity in Bq/L, using the zero efficiencies of alpha and beta and the alpha attenuation factor obtained from the

curve in figure 3, the laboratory was able to quantify all the activities added to the test samples with accuracy and precision.

Since ^{226}Ra is quantified through alpha counts, it is necessary to perform a curve to determine the alpha attenuation effect of ^{226}Ra (Moura, 2018). The preparation of the ^{226}Ra curve allowed the laboratory to obtain two curves. The curve representing the attenuation (self-absorption) is shown in figure 4, and was obtained by monitoring the variation in alpha counts of samples prepared with the same ^{226}Ra activity. From the curve in figure 4, $K = 3.6$ and $\eta_{\text{abs}} = 0.7$ were obtained from the angular and linear coefficients, the curve being approved with $R^2 > 0.99$. The curve in figure 5 was obtained from the same samples and the same counts as the curve in figure 4, and allowed the secular equilibrium of ^{222}Rn in the samples to be monitored for 3 weeks. Secular equilibrium was reached after 21 days.

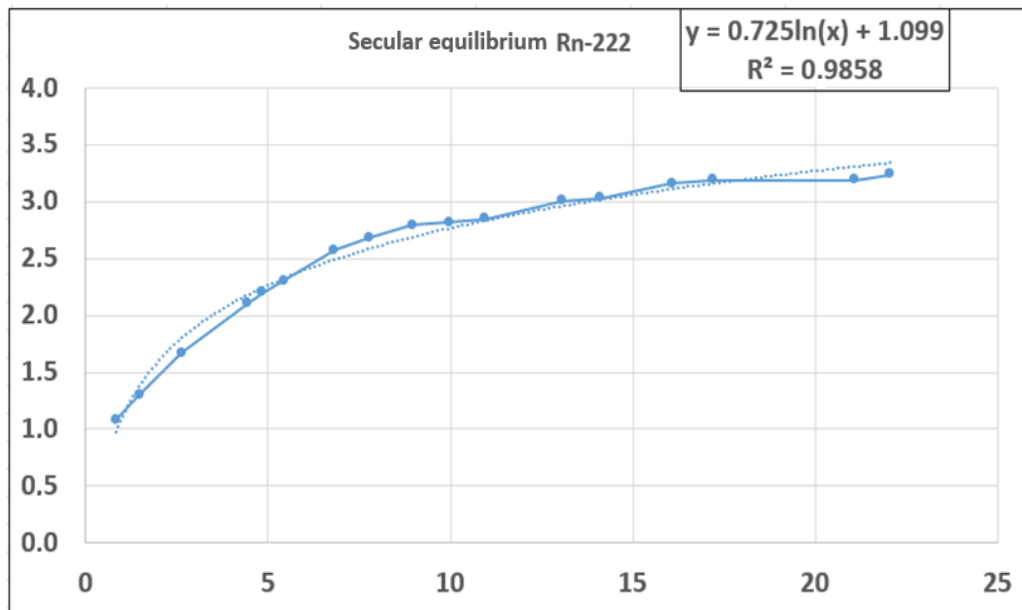
By calculating the alpha activity of ^{226}Ra using the factors obtained from the curve in figure 4, the laboratory was able to quantify all activities added to the test samples with accuracy and precision. The construction of the graphs in figures 4 and 5 was based on the work of Moura, 2018.

Figure 4. Graph of the ^{226}Ra curve to obtain the constants K and η_{abs} .



Fonte: Oceanus Experimental Biology Center.

Figure 5. Graph of the ^{226}Ra curve to monitor the secular equilibrium of ^{222}Rn .



Fonte: Oceanus Experimental Biology Center.

As ^{228}Ra and ^{210}Pb are quantified from beta counts, it is necessary to determine the beta efficiencies of these isotopes (Costa, 2016). The results of the efficiencies of ^{228}Ra and ^{210}Pb are given in table 3 and were obtained after secular equilibration of ^{222}Rn and ^{210}Bi in samples prepared with known activity. By calculating the beta activity of ^{228}Ra and ^{210}Pb with the efficiencies obtained, the laboratory was able to quantify all the activities added to the test samples with accuracy and precision.

Table 3. Results of beta efficiency determinations for the isotopes ^{228}Ra and ^{210}Pb .

ISOTOPE	EFICIENCY
^{228}Ra	46%
^{210}Pb	59%

The results reported by the Radiochemistry Laboratory in the National Intercomparison Programme - PNI of the Institute of Radiation Protection and Dosimetry - IRD (CNEN/IRD, 2021), in the April/2023 round, were approved by the IRD through report n°: R-PQ 2023 04 PNI. Performance was assessed using the

normalised deviation and is described in table 5. The closer the normalised deviation is to zero, the better the performance of the participating laboratory (IRD, 2023).

Table 5. Results of the evaluation of the Oceanus laboratory in the April/2023 round of the National Intercomparison Program – PNI.

WATER ANALYSIS	NORMALIZED DEVIATION	LABORATORY ASSESSMENT
Gross alpha	<2	Approved
Gross beta	<2	Approved

The Oceanus Experimental Biology Center Laboratory applied for accreditation from the General Coordination of Accreditation - CGRE/INMETRO, as a Conformity Assessment Organism - OAC, of the Brazilian Network of Testing Laboratories - RBLE; in the category of ionising radiation testing. The laboratory was evaluated in May 2024 and approved by the evaluation team to continue the accreditation process. It was necessary to demonstrate compliance with the requirements of the ABNT NBR ISO/IEC 17025:2017 standard; for the Quality Management System and for gross alpha, gross beta, ^{226}Ra , ^{228}Ra and ^{210}Pb analyses in water. The laboratory is in the process of accreditation in the field of ionising radiation, awaiting finalisation for publication of the approved scope. The Inmetro General Coordination of Accreditation (CGCRE) is the only accreditation body recognised by the Brazilian government to accredit conformity assessment organism. (MDIC/INMETRO, 2024).

4. CONCLUSIONS

It is concluded that the ionising radiation analyses performed and requested for laboratory accreditation are fundamental for the radiological monitoring of waters in Brazil, with great importance for the environment and human health.

The certified standard sources of ^{241}Am and ^{90}Sr were satisfactory for the determination of operating plateaus, zero efficiencies and alpha attenuation factor. The zero efficiencies determined for alpha and beta meet the approval criteria established by the manufacturer of the proportional gas flow counter, demonstrating the performance of the equipment based on the sources used.

The ^{228}Ra efficiency, the ^{210}Pb efficiency and the K and η_{abs} values obtained from the ^{226}Ra curve guaranteed the quantification of the activities of ^{226}Ra , ^{228}Ra and ^{210}Pb . When used to convert alpha and beta counts into activity concentrations of ^{226}Ra , ^{228}Ra and ^{210}Pb in Bq/L, they allowed isotope quantification with accuracy and precision.

The approval of the gross alpha and gross beta results of the Oceanus Laboratory in the April/2023 round of the National Intercomparison Programme - PNI/IRD proves its ability to quantify gross alpha and gross beta in Bq/L, through the counting rates of the proportional counter equipment of gas flow. Compliance with the ionising radiation analyses required by the GM/MS n° 888:2021 regulation, as screening parameters for the radiological potability of water.

The Radiochemistry Laboratory of the Oceanus Experimental Biology Centre had to demonstrate its competence to carry out ionising radiation tests in water and to prove its compliance with the ABNT NBR ISO/17025:2017 standard. The laboratory is awaiting the completion of the accreditation process in order to publish the scope of ionising radiation testing requested by the General Coordination of Accreditation - CGCRE. The laboratory considers accreditation to be very important as it creates confidence at an international level. Accreditation is the formal recognition of the competence of conformity assessment organism (OAC) to meet previously defined requirements and to carry out their activities with confidence (MDIC/INMETRO, 2024).

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CONFLICT OF INTEREST

Christina Ferreira declares that there is no conflict of interest. Christina Ferreira, Felipe Dias, Bruna Sá, Mariana Monteiro, Edson Ladeira and Hamilton Mendes receive a salary for this work and form the technical team of the radiochemistry laboratory. Richard Guimarães and Ronaldo Guimarães are directors of the Oceanus Group laboratory network. The other authors have no competing interests to declare.

We have no competing interests to declare.

All authors declare that they have no competing interests.

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