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Original Article

Radium Accumulation and Radiometric Assessment of Filtration Membranes in Reverse Osmosis Desalination Systems for Groundwater in Semi-Arid Regions

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Abstract: Water scarcity in the Brazilian semi-arid region makes desalination via reverse osmosis a strategic solution for rural community water supply. However, the interaction of groundwater with geological formations rich in uranium and thorium minerals can favor the natural occurrence of radionuclides, especially radium, in desalination systems. During operation, filtration membranes act as efficient physical barriers, retaining these radionuclides and, upon reaching the end of their service life, generating radiologically significant waste. This study evaluated the presence of the isotopes ^{226}Ra and ^{228}Ra in filtering elements from desalination systems installed in the municipality of Riacho das Almas, Pernambuco, Brazil, and the potential environmental and radiological impacts associated with them. Filters collected from seven wells were calcined and analyzed by gamma spectrometry. ^{226}Ra activities ranged from 0.33 to 7.08 Bq/g, with two wells showing values above the 1 Bq/g limit established by the National Nuclear Energy Commission, and ^{228}Ra was not detected. The results confirm the efficiency of membranes in Ra retention but highlight the need for specific protocols to safely manage and dispose of this waste and mitigate occupational and environmental risks.

Keywords: desalination; reverse osmosis; filtration membranes; radium; radioprotection.

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Acumulação de rádio e avaliação radiométrica de membranas de filtração em sistemas de dessalinização por osmose reversa para águas subterrâneas em regiões semiáridas

Resumo: A escassez hídrica na região semiárida brasileira torna a dessalinização por osmose reversa uma solução estratégica para o abastecimento de água em comunidades rurais. No entanto, a interação da água subterrânea com formações geológicas ricas em minerais de urânio e tório pode favorecer a ocorrência natural de radionuclídeos, especialmente rádio, em sistemas de dessalinização. Durante a operação, as membranas de filtração atuam como barreiras físicas eficientes, retendo esses radionuclídeos e, ao final de sua vida útil, gerando resíduos radiologicamente significativos. Este estudo avaliou a presença dos isótopos ^{226}Ra e ^{228}Ra em elementos filtrantes de sistemas de dessalinização instalados no município de Riacho das Almas, Pernambuco, Brasil, e os potenciais impactos ambientais e radiológicos associados a eles. Filtros coletados de sete poços foram calcinados e analisados por espectrometria gama. As atividades de ^{226}Ra variaram de 0,33 a 7,08 Bq/g, com dois poços apresentando valores acima do limite de 1 Bq/g estabelecido pela Comissão Nacional de Energia Nuclear, e o ^{228}Ra não foi detectado. Os resultados confirmam a eficiência das membranas na retenção de Ra, mas destacam a necessidade de protocolos específicos para o gerenciamento e descarte seguros desse resíduo e para a mitigação dos riscos ocupacionais e ambientais.

Palavras-chave: dessalinização; osmose reversa; membranas de filtração; rádio; radioproteção.

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1. INTRODUCTION

Water is an essential resource for the survival of living beings and for the functioning of society, the economy, and ecosystems, and it plays a strategic role in sustainable development [1]. Despite its importance, water availability is unevenly distributed across the planet, leading to recurring water scarcity, especially in arid and semi-arid regions characterized by low rainfall and high evapotranspiration [2].

In Brazil, the Northeast region is characterized by limited water resources, particularly in semi-arid areas, where irregular rainfall undermines surface water availability. As an alternative, groundwater exploitation has been widely used to meet local communities' demands. However, approximately 51% of the northeastern territory is underlain by crystalline rocks, which favor groundwater salinization through water-rock interactions, often making it unsuitable for consumption without prior treatment [3].

In this context, desalination technologies for brackish or saline water emerge as viable solutions to mitigate the effects of water scarcity in the Northeastern semi-arid region [4]. Among available technologies, reverse osmosis membrane systems are the most widely employed due to their high efficiency in removing dissolved salts and other chemical compounds, thereby enabling the sustainable use of groundwater resources [5].

During treatment, filtration membranes act as physical barriers that retain various contaminants in raw water, including organic matter, microorganisms [6], and natural radioactive elements such as the isotopes ^{226}Ra and ^{228}Ra, which belong to the natural radioactive series of ^{238}U and ^{232}Th. The mobilization of these radionuclides into groundwater occurs mainly through geochemical processes associated with interactions between water and rock formations, thereby promoting the release of ions into the aqueous environment [7].

Among the natural radionuclides of greatest relevance to water ingestion, ^{226}Ra and ^{228}Ra warrant particular attention, as their incorporation into the water matrix can pose risks to human health and the environment if not properly monitored [8,9]. These isotopes are often associated with high-salinity groundwater, a common characteristic of aquifers developed over crystalline basement rocks [10,11].

The municipality of Riacho das Almas, located in the interior of the state of Pernambuco, Brazil, is entirely situated within this geological context, with 100% of its territory resting on crystalline rocks. Studies conducted in the region have indicated the presence of radionuclides ^{226}Ra and ^{228}Ra in approximately 95% of the wells analyzed [12], highlighting the relevance of the subject and the need for further investigation into the behavior of these radionuclides in the locally used desalination systems.

Given this scenario, it is essential to evaluate the membranes used in reverse osmosis desalination systems, particularly with respect to radionuclide retention and accumulation over their useful life. Although these membranes play a key role in maintaining the quality of the water supplied to the population, the progressive accumulation of radioactive elements can generate radiologically significant waste, necessitating proper management and planning for final disposal.

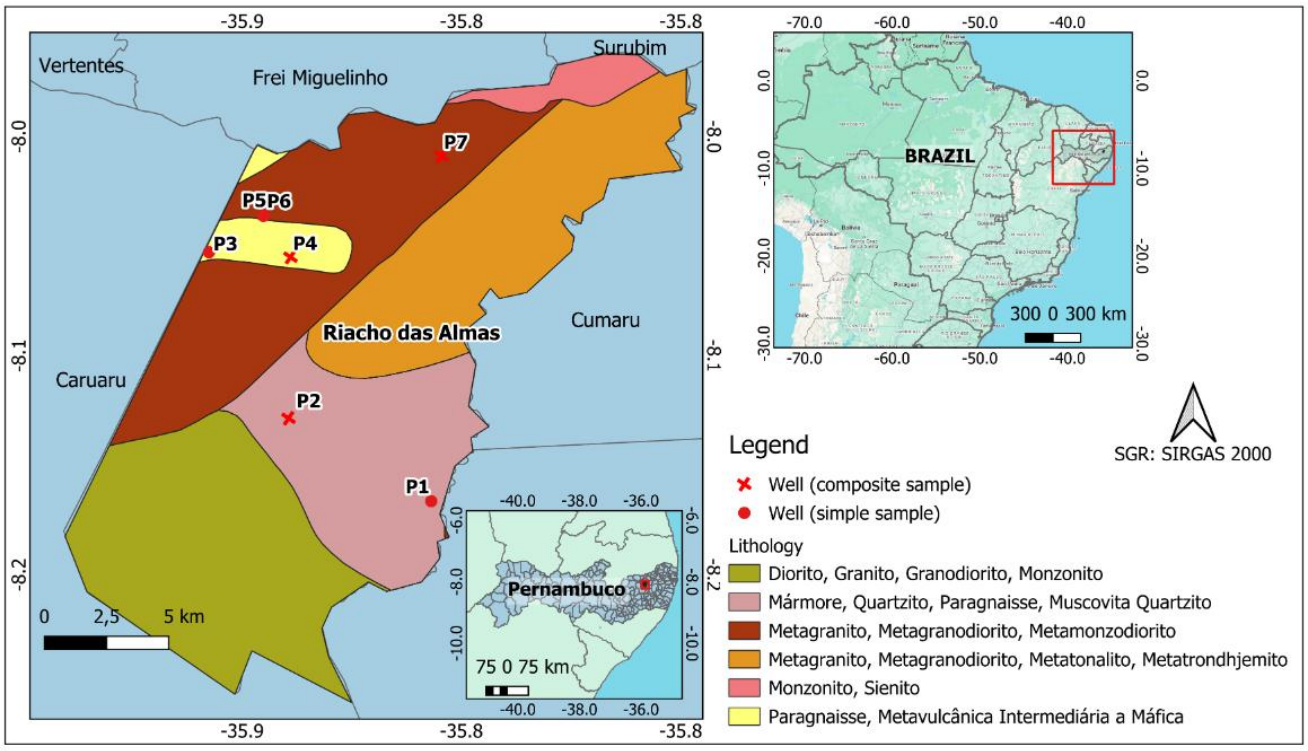
Thus, investigating the presence of ^{226}Ra and ^{228}Ra in desalination filters contributes to establishing technical guidelines for preventive replacement, environmentally appropriate disposal, and strengthening radioprotection measures, thereby promoting greater health and safety and the sustainability of desalination systems in the Brazilian semi-arid region.

2. MATERIALS AND METHODS

2.1. Study Area

The study was carried out in the municipality of Riacho das Almas, located in the Agreste mesoregion of Pernambuco, within the semi-arid climate domain of Northeast Brazil (Figure 1). The city covers approximately 314 km² and is characterized by a hot, dry climate, low rainfall, and high spatial and temporal variability in precipitation, all of which intensify regional water scarcity.

Figure 1: Municipality studied with the location of the monitored points



From a geological perspective, the area is predominantly composed of Precambrian basement crystalline rocks, which present low permeability and reduced groundwater storage capacity. These characteristics promote water salinization, compromising its potability and requiring specific treatments for human use [13,14].

The choice of the municipality as the experimental area was based on the significant number of tubular wells equipped with reverse osmosis desalination systems, a technology

widely used to remove dissolved salts and other contaminants. In total, 19 wells operating with this type of system were identified [12]. For this study, 7 points with installed and operational systems were selected. Simple or composite samples were used, depending on the availability of units per point, aiming to increase the representativeness of the results.

2.2. Sample Collection and Preparation

The reverse osmosis membranes were collected directly from the operational sites of the desalination systems, placed in sealed plastic bags, properly identified, and recorded for usage time, collection date, and well of origin.

In the laboratory, the filters were dried in an oven at 70°C for 32 hours, and the ceramic containers used in the process were heated to 105°C for 3 hours to ensure complete removal of residual moisture. After weighing, the samples were calcined in a muffle furnace to eliminate organic matter and obtain a stable mineral matrix. The procedure followed Method A of ISO 3451-1 [15], adapted for polyamide membranes, with gradual heating to 600°C over 6 hours, then holding at this temperature for an additional 2 hours. The resulting ashes were weighed and sent for subsequent conditioning and analysis.

The ashes were placed in glass Petri dishes measuring 80 mm x 15 mm, with standardized thickness ensured by a rubber support, guaranteeing reproducible geometry among samples (Figure 2).

The set was sealed with hose and silicone glue, ensuring an airtight closure. The samples were stored for at least 30 days before radiometric analyses, a period necessary to establish secular equilibrium between radium and its gamma-emitting progeny.

Figure 2: Steps of conditioning mineral ash after calcination

1- Petri dish 2- Addition of mineral ash 3- Addition of rubber material 4- Sealing with silicone glue 5- Fitting the lid to the base 6- Sealing with silicone hose 7- Sealing with silicone glue 8- Sample identification.

2.3. ^{226}Ra and ^{228}Ra Measurement System

Analyses were performed using high-resolution gamma spectrometry with a high-purity germanium detector with a beryllium window (HPGe-Be; model GX2518, Canberra®), installed at the Laboratory of Radiochemistry and Nuclear Analysis (LABRAN), Department of Nuclear Energy, Federal University of Pernambuco.

Energy calibration of the system was performed in accordance with the technical guidelines of IAEA TECDOC 619 [16], using standard sources of ^{241}Am, ^{137}Cs, and ^{60}Co. Measurements were made for 300 seconds per source, allowing the construction of a calibration curve over the energy range from 59.5 to 1,332.5 keV. Efficiency calibration was performed by preparing secondary standards containing known solutions of ^{226}Ra and ^{228}Ra, which were spiked into a mineral ash matrix obtained from calcined desalination filters from the same study area.

2.4. Secondary Standards, Background, and Sample Blanks

Secondary standards were prepared in Petri dishes containing 1.5 g of mineral ash, with paper filters placed on top, and standard solutions of ^{226}Ra or ^{228}Ra with traceable activities were added. After spiking, the standards were sealed with a rubber hose and silicone glue to ensure physical stability and system tightness, as shown in the samples in Figure 2.

For experimental control, background and blank samples were prepared. The background sample was assembled in accordance with the standards but without the addition of radioactive solutions, thereby allowing correction for the radiological contribution of the constituent materials. The sample blank used the same geometry as the analyzed samples, except that it lacked mineral ash, thereby minimizing systematic errors and increasing the reliability of the results.

2.5. Gamma Spectrometry Analysis and Statistical Treatment

After the secular equilibrium period, the samples were analyzed by gamma spectrometry with a 48-hour acquisition time, optimized to achieve a high signal-to-noise ratio. The activities of ^{226}Ra and ^{228}Ra were determined indirectly from the gamma emissions of their radioactive progeny, notably ^{214}Pb and ^{214}Bi for ^{226}Ra, and ^{228}Ac for ^{228}Ra.

The specific activity (A) was calculated considering the net count (C_{net}), energy efficiency (ε), gamma emission probability for the transition considered (γ), counting time (t), and sample mass (m), applying Equation 1.

$$A = \frac{C_{net}}{\varepsilon.\gamma.t.m} \qquad (1)$$

The detection limit (LD) was determined according to the method proposed by Currie [17], allowing the minimum detectable activity to be calculated using Equation 2, where N_{Bg} represents the number of counts observed in the background spectrum.

$$LD = 2{,}71 + 4{,}66\sqrt{N_{Bg}} \qquad (2)$$

The minimum detectable activity (MDA) was determined using Equation 3, with the same variables previously defined in Equations 1 and 2.

$$MDA = \frac{LD}{\varepsilon.\gamma.t.m} \quad (3)$$

For the calculation of gamma transition efficiencies, gamma energies with the least variation across samples were selected, thereby improving stability in the results.

The data obtained were subjected to descriptive statistics and exploratory analysis, followed by comparison with regulatory limits established by the National Nuclear Energy Commission and the Environmental Protection Agency, enabling assessment of radiological compliance and interpretation of the radiochemical behavior of the analyzed filters.

3. RESULTS AND DISCUSSIONS

3.1. Radium Behavior

The determination of the specific activity of ^{226}Ra in desalination filters from the municipality of Riacho das Almas, Pernambuco, Brazil, revealed significant differences among the wells, indicating high spatial heterogeneity in radionuclide accumulation. Mean concentrations ranged from 0.33 to 7.08 Bq/g, with a total range of 6.75 Bq/g, reflecting the influence of local hydrogeochemical factors and system operating time, as detailed in the descriptive statistics in Table 1.

Table 1: Descriptive data by well

WELL	MEAN (Bq/g)	STANDARD DEVIATION	CV (%)
P1	1.08	0.02	1.92
P2	2.94	0.05	1.62
P3	7.08	0.08	1.08
P4	1.03	0.03	2.73
P5	0.61	0.03	4.72

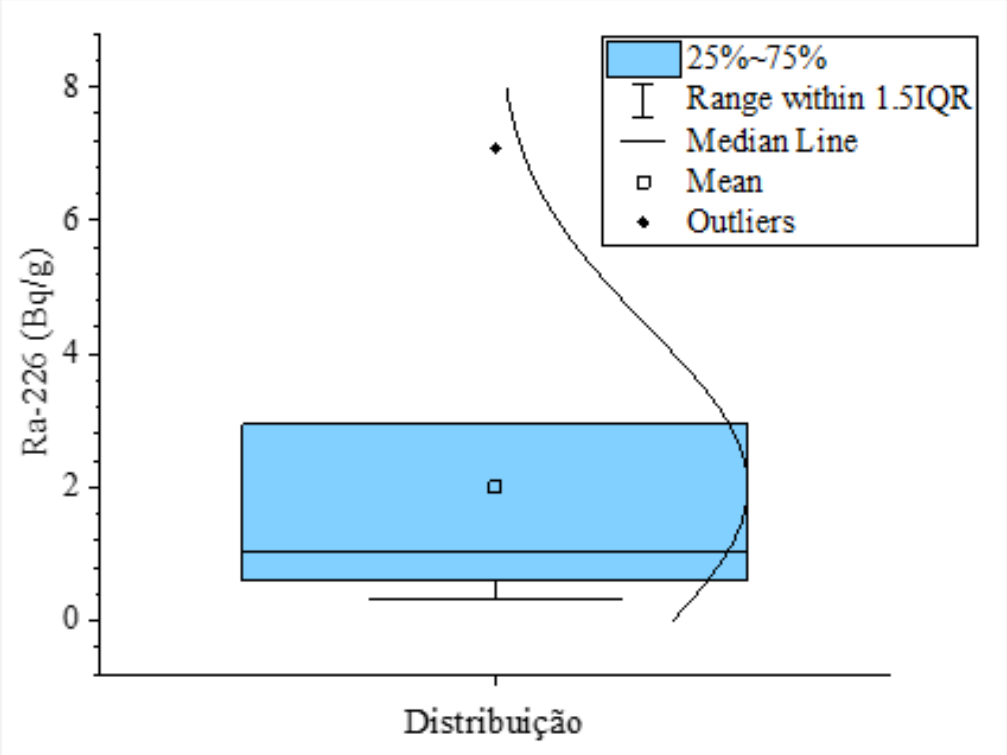
WELL	MEAN (Bq/g)	STANDARD DEVIATION	CV (%)
P6	0.33	0.03	7.08
P7	1.00	0.13	12.95
P1	1.08	0.02	1.92

CV: Coefficient of Variation; MDA ^{226}Ra: 0.05 Bq/g; MDA ^{228}Ra: 0.86 Bq/g.

Statistical analysis shows that most wells exhibited low internal variability (CV < 5%), indicating good reproducibility of measurements. The exception was well P7, whose coefficient of variation of 12.95% suggests greater heterogeneity in the filtering material. Of all samples, approximately 29% showed values below 1 Bq/g, while wells P2 and P3 stood out as critical points, with mean activities significantly higher than the group.

As shown in Figure 3, the frequency distribution of activities confirms the predominance of low values, with 71.4% of samples concentrated in the range of 0.33-2.03 Bq/g, and occasional occurrences of high accumulation.

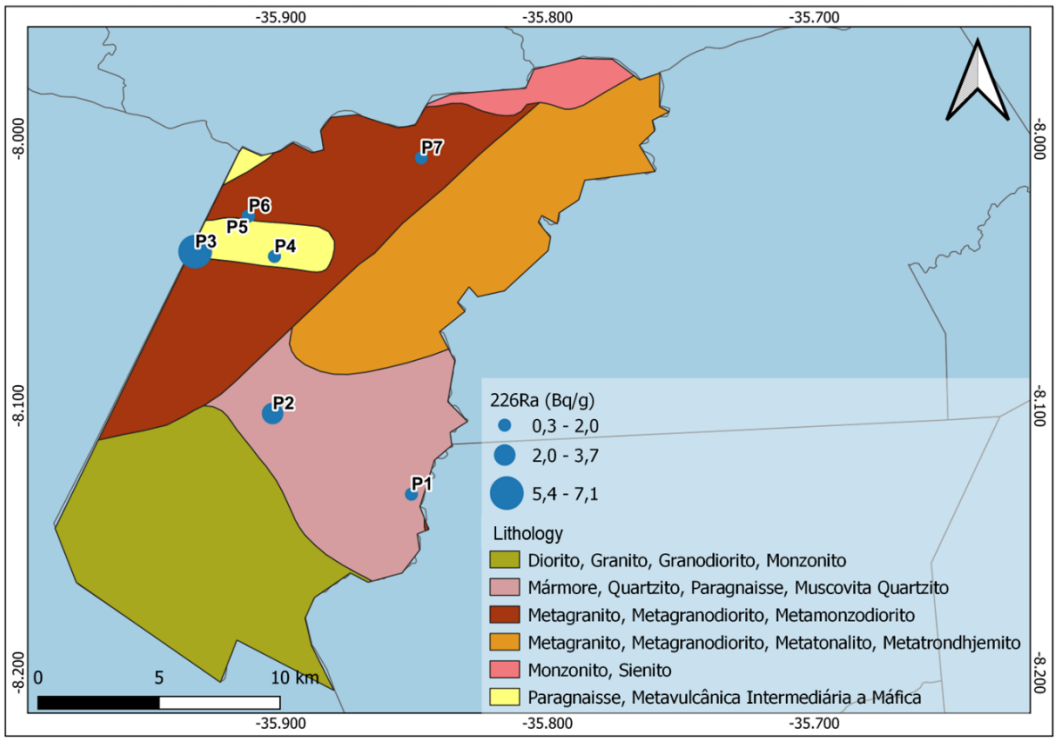
Figure 3: Frequency distribution



The absence of values in the intermediate range (3.75 - 5.45 Bq/g) indicates discontinuity in the accumulation pattern, typical of geologically heterogeneous environments, where lithology, fracturing, and hydrogeological dynamics control radium mobilization.

The spatial analysis of concentrations demonstrates a correlation between ^{226}Ra activity and the lithological context of the wells, as detailed in Figure 4.

Figure 4: Spatial distribution of ^{226}Ra concentrations in filters



Wells installed in metamorphic terrains rich in marble, quartzite, and paragneiss (P1 and P2) exhibited higher values, associated with the presence of uraniferous minerals, whose weathering favors the release of radium into the aqueous environment [18,19]. In contrast, wells in metamorphosed granitic lithologies (P5, P6, and P7) showed activities up to 1 Bq/g, consistent with a lower content of radioactive minerals [20].

The analysis of ^{226}Ra and ^{228}Ra concentrations in raw water, brine, and permeate water was carried out by Oliveira [12], using samples collected from the same desalination systems evaluated in this study. These data serve as a reference for comparison with the results obtained in the filters.

Data from Oliveira [12] demonstrates that filters act as effective barriers, retaining radionuclides that remain below the detection limit in treated water. However, this efficiency results in the progressive accumulation of radioactive material in the filters, making them critical components from a radiological perspective. Radium retained by the membranes is expected to remain predominantly bound to the mineral residue formed during filtration. However, under prolonged operation and depending on the hydrochemical conditions, such as pH and ionic strength, a partial leaching of retained radionuclides may occur. Studies performed in other regions have shown that, even waters subjected to desalination or advanced treatment may still present elevated radionuclide concentrations, as reported in investigations of contaminated groundwater in the Qaseem region, Saudi Arabia [21]. In another study, it is suggested to combine several water treatment methods to obtain better results [22].

Spectrometric analyses did not show peaks attributable to ^{228}Ra or its progeny, even after secular equilibrium was established, indicating concentrations below the system's detection limit. Although ^{226}Ra and ^{228}Ra have similar geochemical behavior (Ra^{2+}), differences in natural abundance, water salinity, particulate mass, and physical half-lives (1,600 years for ^{226}Ra and 5.75 years for ^{228}Ra) may explain this discrepancy [23-25].

Thus, the non-detection of ^{228}Ra does not imply its absolute absence, but rather reflects analytical limitations and low concentrations. Additional studies with larger sample mass or analysis of solid incrustations are recommended [26,27].

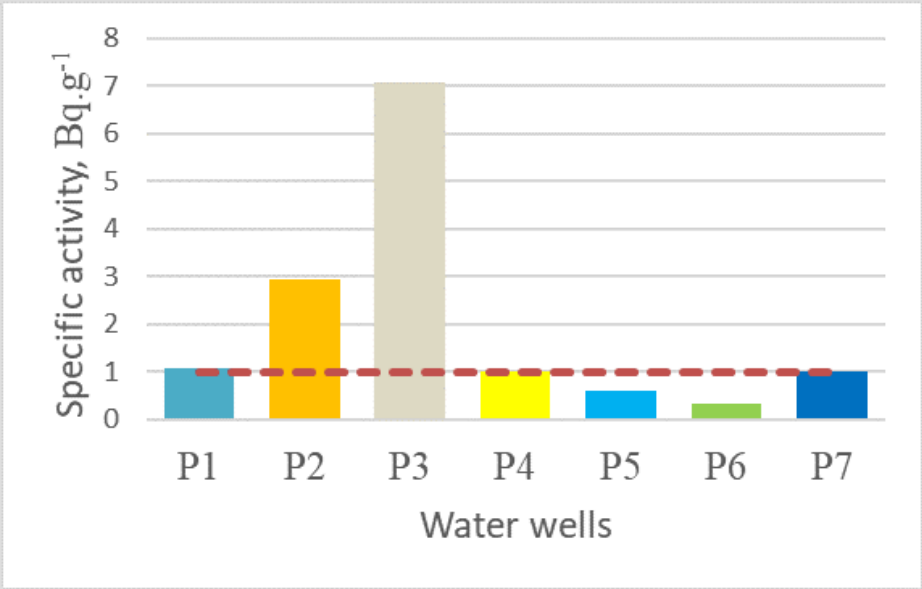
3.2. Regulatory Framework and Radiological Assessment

According to the National Nuclear Energy Commission (CNEN), Standard CNEN NN 8.01, materials containing natural radionuclides at concentrations above 1 Bq/g cannot be released without control and must be classified as radioactive waste. Standard CNEN NN 3.01 further emphasizes that such situations constitute existing exposures that require assessment and control measures [28,29]. Figure 5 presents the specific activities of ^{226}Ra

(Bq/g) determined in desalination filters by well, highlighting the 1 Bq/g limit (red dashed line) as established in CNEN normative references.

Wells P2 and P3 showed mean activities of 2.94 and 7.08 Bq/g, respectively, both significantly above the regulatory limit and indicating greater radiological relevance. Wells P1 and P4 slightly exceeded the limit, while P5 and P6 remained below the reference value. The P7 well was borderline.

Figure 5: Specific activity of ^{226}Ra (Bq/g) in filters



Comparison with international references, such as the WHO [30], Health Canada [31], and EPA [32], shows that the filters accumulate concentrations several orders of magnitude higher than the limits established for drinking water, reinforcing the role of membranes as retention barriers and the need for proper management of this waste [33-35].

3.3. Management of Desalination Filters

The results show that desalination filters serve as compartments for Ra accumulation, requiring specific assessment of their management and final disposal. The retention of radionuclides over time can render filter waste potentially radiologically significant, particularly for systems associated with wells that exceed reference values.

According to international guidelines for waste from drinking water treatment containing radioactivity [36], most of the analyzed filters fall within intermediate activity ranges, for which disposal in sanitary landfills is technically feasible, provided that dehydration procedures and measures to contain radon release are implemented. These results indicate that, although average levels are relatively low, management cannot be done indiscriminately.

In contrast, filters from wells P2 and P3 exhibited significantly higher activity levels, falling within ranges that require individualized assessment and differentiated disposal, preferably at facilities licensed for low-level radioactive waste. These cases represent situations of greater radiological criticality, in which the adoption of stricter protocols for control, conditioning, and traceability is essential.

Within the national regulatory context, materials containing natural radionuclides from the uranium and thorium series, with concentrations above 1 Bq/g, must be classified as radioactive waste and subject to controlled management [28]. These cases require segregation, conditioning (for example, encapsulation in appropriate containers), traceability, and subsequent transfer to facilities licensed to dispose of low-level radioactive waste. Therefore, the end-of-life management of the membranes must follow the same radiological control principles applied to other naturally occurring radioactive materials (NORM), ensuring environmental safety and regulatory compliance.

The comparative analysis between international references and Brazilian regulations shows convergence on the importance of controlling and tracing solid waste generated by desalination systems. In the municipality of Riacho das Almas, the results indicate that a portion of the filters require additional management procedures, with those associated with wells P2 and P3 prioritized for specialized disposal.

In addition to meeting regulatory thresholds, it is important to consider the potential environmental impacts of improper disposal of filters with elevated radium concentrations. Although the treated water supplied to the population consistently showed radium activity

below detection limits, thereby preventing direct exposure, the improper disposal of used membranes may lead to soil contamination. Once in the soil, radium can be taken up by plants, transferred to grazing animals, and ultimately reach humans through the food chain. This reinforces the need for proper classification and management of filters exceeding 1 Bq/g. These procedures effectively prevent environmental dispersion and ensure that no radiological impact reaches the general population.

These findings reinforce the importance of incorporating solid-waste management practices that contain radium as an essential step in the radiological evaluation of desalination systems, complementing analyses that focus exclusively on treated water quality.

4. CONCLUSIONS

The results obtained confirm that the filters used in reverse osmosis desalination systems in the municipality of Riacho das Almas, Pernambuco, Brazil, exhibit measurable accumulation of natural radionuclides, with a predominance of ^{226}Ra. The specific activities measured varied widely across the evaluated wells, with values exceeding the reference limits established by national and international standards, particularly for wells P3 (7.08 Bq/g) and P2 (2.94 Bq/g), which were notable for their radiological significance.

^{228}Ra was not detected in the analyzed samples, indicating concentrations below the method's detection limit and a distinct behavior compared to ^{226}Ra in the studied systems.

Comparison of the results with the regulatory criteria of the CNEN Standard NN 8.01/2025 and the EPA guidelines shows that some filters, at the end of their service life, do not meet the criteria for unrestricted release and require controlled management, proper conditioning, and final disposal compatible with their radiological classification.

Although reverse osmosis is effective in removing Ra from treated water, the process concentrates this radionuclide in the solid residues generated, which must be considered

critical elements in the radiological assessment and environmental management of desalination systems in regions with natural radionuclide occurrences.

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CONTRIBUTORSHIP

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Supervision: RSA, MBCFB

Validation: ZHF, JMNS, FAM

Writing – review and editing: ZHF, JMNS, FAM, JSR, APC, JDB

Visualization: JSR, APC, JDB

CONFLICT OF INTEREST

All authors declare that they have no conflicts of interest.

DATA AVAILABILITY STATEMENT

Interview transcripts and qualitative data are not publicly available due to confidentiality commitments made to research participants. Aggregated data supporting the findings are available from the corresponding author upon reasonable request and subject to participant privacy protections.

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