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The occurrence of nitrate and its depuration perspective in the scenario of an urban unconfined aquifer with variable recharge, northeastern Brazil.

A ocorrência de nitrato e sua perspectiva de depuração no cenário de um aquífero não confinado urbano com recarga variável, nordeste do Brasil.

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Abstract: The nitrification of groundwater in urban areas commonly stems from the infiltration of untreated wastewater. This study aimed to characterize the presence of NO₃ and evaluate the possibilities for natural remediation in an urban unconfined aquifer subject to variable recharge. The investigation utilized hydrochemical analysis techniques and numerical modeling to examine the interplay between aquifer thickness variations, recharge fluctuations, urban development, and the configuration of the sanitary sewage system. Four scenarios were devised to assess the decline in saturation levels and its subsequent impact on the aquifer's theoretical replenishment rates, assuming a gradual reduction in nitrogen inputs from infiltrating waters. Over the years 2012 to 2020, findings revealed a steady escalation in nitrate concentrations in approximately 58% of the observed wells, with concentrations trending higher towards the southern reaches of the study area. The estimated residence times, indicative of the potential for water quality restoration, spanned from 6.5 to 12.9 years, with the latter value representing the most critical scenario in terms of recharge dynamics. In this critical scenario, all contributions from losses via the water supply network and wastewater infiltration were assumed to be eliminated.

Keywords: Nitrate; Depuration; Urban Unconfined Aquifer.

Resumo: A nitrificação das águas subterrâneas em centros urbanos normalmente decorre da infiltração de águas residuais não tratadas. Na presente pesquisa, caracterizou-se a ocorrência de NO₃ e perspectivas de depuração natural no cenário de um aquífero não confinado urbano com recarga variável. Para isso, utilizaram-se ferramentas de análises hidroquímicas e modelos numéricos, considerando a relação entre a variação da espessura do aquífero diante da variação da recarga, ocupação do solo e a estrutura da rede coletora de efluentes sanitários. Nesse contexto, quatro cenários foram inseridos nas avaliações do decréscimo das reservas de saturação e consequente impacto nas taxas de renovação teóricas do aquífero, assumindo-se a gradativa diminuição dos aportes nitrogenados das águas infiltradas. Os resultados demonstraram o aumento progressivo das concentrações de nitrato em cerca de 58% dos poços analisados no período de 2012 a 2020, com progressão das concentrações no sentido sul da área de pesquisa. A estimativa de tempo de residência, associado com perspectivas de renovação qualitativa do manancial, variou de 6,5 a 12,9 anos, sendo este último no cenário considerado mais crítico em termos de recarga, com a supressão de 100% da contribuição proveniente de perdas pela rede de distribuição de água e infiltração de águas residuais.

Palavras-chave: Nitrato; Depuração; Aquífero não Confinado Urbano.

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1. Introduction

The pollution of groundwater by nitrate (NO_3), whether in rural regions or urban areas, is a global environmental problem (ABASCAL *et al.*, 2022). Excessively high concentrations of NO_3 in groundwater are associated with various anthropogenic activities, including agricultural, industrial, and domestic activities (HIRATA *et al.*, 2020; PILEGGI, *et al.*, 2021; ABASCAL *et al.*, 2022). These activities produce much wastewater, often discharged directly into the soil or watercourses without proper treatment (WAKIDA E LERNER, 2005; PEIXOTO *et al.*, 2020; PILEGGI *et al.*, 2021).

In this context, the environmental impacts related to groundwater contamination by NO_3 can cause damage to various aquatic systems and, more importantly, health problems such as methemoglobinemia and cancer through the continued ingestion of water with high nitrate levels (AYERS E WESTCOT, 1985; KNOBELOCH e ANDERSON, 2000; TOKAZHANOV *et al.*, 2020). These high nitrate concentration levels interfere with public water supply systems in several countries that depend on groundwater, often requiring the closure of wells or even the need for water dilution. This situation has become a significant concern for managers, especially considering the sixth Sustainable Development Goal (SDG), which addresses ensuring the availability and sustainable management of water and sanitation for all populations (UNITED NATIONS, 2016; UNITED NATIONS, 2021).

In Brazil, nitrate contamination is mainly caused by the infiltration of wastewater (sewage and septic tanks) and, secondarily, by industrial activities and agriculture (HIRATA *et al.*, 2020; PEIXOTO *et al.*, 2020; ABASCAL *et al.*, 2022). The groundwater quality standard (BRAZIL, Ministry of Health Ordinance GM/MS N° 888/2021) stipulates that the nitrate (NO_3) concentration for drinking water should not exceed 10 mg/l, a limit not necessarily observed in large urban centers in terms of potability standards (HIRATA *et al.*, 2020; PEIXOTO *et al.*, 2020).

Another aspect contributing to the contamination of groundwater sources in large urban centers is soil impermeabilization, as urban infrastructure construction alters aquifer recharge rates and consequently hinders the natural dilution of potential contaminations (HIRATA *et al.*, 2020; MENDOZA *et al.*, 2021; CONICELLI *et al.*, 2021).

In the research area, nitrate contamination of the aquifer is attributed to the natural vulnerability of the hydrogeological system and the dynamics of groundwater flow (Sotero, 2016). This situation was substantially aggravated in the post-World War II period, with urban expansion and population densification, without implementing an adequate sewage system for the city. This scenario led to a systematic increase in untreated sanitary effluents infiltrating the rocky substrate and the local aquifer formation (SOTERO, 2016).

In this regard, this research aims to contextualize the occurrence of nitrate and assess the natural attenuation times of this contamination, considering the spatial variability of recharge rates in an unconfined urban aquifer. The study focused on an area of occurrence of the Barreiras Aquifer, located in the northeasternmost region of Brazil (Figure 1). However, it is important to note that the present study addresses the natural conditions of the hydrogeological source (raw water), as opposed to the water supplied to the public distribution system, which is subjected to preventive disinfection and dilution treatments, among other techniques.

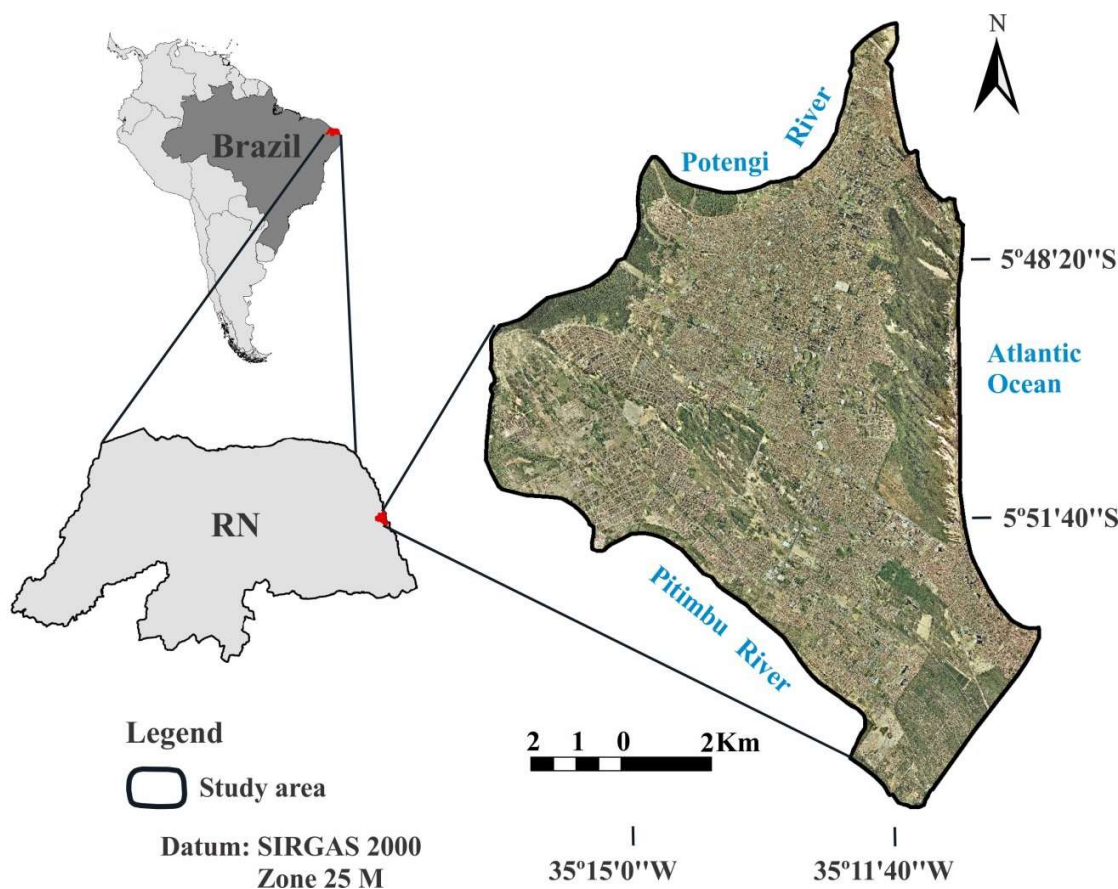


Figure 1 – Location of the study area.

Source: Authors (2024).

2. Geological and hydrogeological contexts

The local non-outcropping stratigraphic is represented by a basal unit composed of Precambrian gneissic-migmatitic rocks and an overlying Mesozoic sedimentary sequence, consisting of sandstones at the base and predominantly carbonate rocks at the top (SOUZA *et al.*, 2019, NUNES *et al.*, 2020).

The outcropping stratigraphic sequence, marked by a regional erosional unconformity over the Mesozoic sequence, is primarily represented by the sedimentary rocks of the Barreiras Formation, dating from the Upper Tertiary to the Lower Quaternary, and Quaternary covers (NUNES *et al.*, 2020, DANTAS *et al.*, 2021). These Quaternary sediments, overlying the Barreiras Formation, mainly involve dystrophic aeolian quartz sands, forming dune belts, stabilized by native vegetation (older dunes) or without vegetation (more recent dunes) (DANTAS *et al.*, 2021).

The regional tectonic-structural compartmentalization is characterized by structural blocks of the graben and horst types, generated by a fault system with preferential directions of N40°-60°E, N40°-50°W, N70°W, and less expressively N350°-10°S. These fault structures significantly influence the structural compartmentalization of regional hydrogeological systems (SOUZA *et al.*, 2019; NUNES *et al.*, 2020).

The hydrogeological context is represented by the Barreiras Aquifer System (SAB), which is predominantly an unconfined hydraulic unit, although with localized semi-confinements, featuring a tabular geometry and horizontal stratification (MELO E QUEIROZ, 2001; NUNES *et al.*, 2020). In lithological terms, and according to the homonymous formation, the aquifer involves sandstones to sandy siltstones and quartz sand sediments in its shallowest portion, associated with various generations of dunes and occasionally integrating the aquifer system. In the study area, these saturated thicknesses vary from 28 to 99 meters (MELO E QUEIROZ, 2001; MELO *et al.*, 2013, NUNES *et al.*, 2020, ALVES E LUCENA, 2022).

The groundwater reserves of the SAB are mainly replenished through the direct infiltration of rainwater. The discharges are primarily natural, with flow towards surface drainages (NNW and SSW) and the Atlantic Ocean (E), in addition to the subtraction of groundwater by well exploitation, especially those forming the water distribution network. The average hydraulic parameters of conductivity and transmissivity are around $3.0 \times 10^{-3} \text{ m}^2/\text{s}$ e $1.0 \times 10^{-4} \text{ m/s}$, respectively (MELO, 1995). Effective porosities on the order of 7.6%, associated with storage coefficients in unconfined aquifers, were obtained by Silva *et al.* (2014) from the processing of thin section images of the aquifer formation, using a methodology similar to that addressed by Lucena *et al.* (2016).

Chemically, the local waters of the SAB are predominantly calcium bicarbonate; however, deeper portions of the aquifer exhibit sodium chloride waters (MELO, 1995; MELO AND QUEIROZ, 2001). The waters are generally acidic, with an average pH of 5.5, low salinity, slightly corrosive, and representative electrical conductivity around $200 \mu\text{S}/\text{cm}$ (MELO, 1995; MELO E QUEIROZ, 2001). In this regard, concerning the physicochemical quality, the water of the Barreiras Aquifer is of excellent quality in its natural conditions (MELO, 1995). However, it currently exhibits a high degree of nitrate contamination on a regional scale.

The first studies (IPT, 1982) identified wells with nitrate contamination in the central and northern parts of the research area. Other records indicate numerous contaminated wells in the same region over time, even affecting the units of the Public Water Supply System (MELO, 1995). More recent research shows that contamination remains in the central and northern regions, although several other wells have been identified with contamination in the western and eastern parts of the research area (MELO *et al.*, 2011).

This contamination scenario, as reported, is intrinsically related to the high natural vulnerability of the SAB, due to local geomorphological conditions and hydrogeological structures, particularly in terms of the presence of sandy covers of eolian origin (which exhibit high infiltration rates) and the presence of closed sub-basins with lagoons (MELO, 1995). In addition, there is a characterization of the equally high hydraulic connection between the dune sediments and the rocks of the Barreiras Aquifer Formation, which nonetheless represents a unique hydraulic system (MELO, 1995; MELO *et al.*, 2013).

3. Methodology

The proposed methodology for this work focused on investigating the temporal evolution of aquifer contamination and the residence time of the contaminant. For this purpose, samples from 62 wells were used for hydrochemical analyses of nitrogen concentrations, in the form of nitrate, in the period between 2012, 2017 and 2020, considering the dry seasonal period. The data on the wells and the sewage drainage structure were provided by the regional public supply company. Additionally, hydraulic heads were measured in 78 wells, aiming at a preliminary investigation of groundwater flow directions. With this information, a database of these chemical analyses was created, where nitrate concentrations were subjected to basic statistical analysis (mean, median, standard deviation, and simple linear regression) with table and graph constructions. In this way, a preliminary contextualization of the occurrence of nitrate was sought, as well as correlating contamination behavior patterns in the study area. Subsequently, thematic concentration maps were produced using ArcGis10 software employing the Inverse Distance Weighted (IDW) technique for isovalue interpolation.

Numerical models of porous aquifers are based on Darcy's equations for laminar flows and the Law of Mass Conservation, which combined, result in the fundamental equation for laminar flows (equation 1) (ANDERSON E WOESSNER, 1992; HANSON *et al.*, 2014). This study used a mathematical model for the Barreiras Aquifer in the study area previously developed by Campos *et al.* (2023).

$$\frac{\partial}{\partial x} (k_x \frac{\partial h}{\partial x}) + \frac{\partial}{\partial y} (k_y \frac{\partial h}{\partial y}) + \frac{\partial}{\partial z} (k_z \frac{\partial h}{\partial z}) + R(x,y,z,t) = S_s (\frac{\partial h}{\partial t}) \quad (1)$$

Where:

K_x, K_y e K_z - are the values of hydraulic conductivity along the x, y, and z axes, respectively, in (L/T);

R - represents recharge or extractions W (sources and sinks of the aquifer), in (L^3/TL^3);

h - is the variation of hydraulic head in three-dimensional space in (L);

S_s - is the specific storage coefficient (1/L), equivalent to the volume of water released per unit volume of the aquifer (L^3), which is subjected to a unit decrease in hydraulic head (L);

t - is the representation of time.

The thickness of the aquifer was measured in available well profiles and inserted into the calibration of the numerical model (CAMPOS *et al.*, 2023) for four scenarios with different recharge values (Figure 2). The reserves were evaluated

at $4,25 \times 10^8 \text{ m}^3$, $4,17 \times 10^8 \text{ m}^3$, $4,07 \times 10^8 \text{ m}^3$, $4,00 \times 10^8 \text{ m}^3$ e $3,82 \times 10^8 \text{ m}^3$ (Figure 2), respectively for the scenarios of current recharge conditions, scenario 1 (with a 50% reduction in recharge contribution from wastewater), scenario 2 (with a 100% reduction in recharge from wastewater), scenario 3 (with a 50% reduction in recharge contribution from wastewater and water distribution network losses), and scenario 4 (with a 100% reduction in recharge from wastewater and water distribution network losses). Figure 3 provides a summary of the flowchart used in the numerical simulations in the different scenarios reported in terms of natural and artificial recharge (adapted from Campos *et al.*, 2023).

The estimated trends for each recharge scenario were performed using equation 2 (adapted from CASTANY, 1975; BEFUS *et al.*, 2017; PULIDO-VELAZQUEZ *et al.*, 2020), considering the aquifer as having a predominantly unconfined hydraulic character.

$$T=V_s/Q \quad (2)$$

Where:

V_s = Saturation volume;

Q = recharge.

Considering that the saturation volume is given by the product of the aquifer occurrence area (A), its effective porosity (η_e), and thickness (E_s) (CUSTODIO AND LLAMAS, 1983; FEITOSA *et al.*, 2008; NUNES *et al.*, 2020), equation 1 can be rewritten as follows (equation 3) (adapted from CASTANY, 1975; BEFUS *et al.*, 2017; PULIDO-VELAZQUEZ *et al.*, 2020):

$$T=A.\eta_e.E_s/Q \quad (3)$$

The variations in aquifer saturation reserves, resulting from different recharge scenarios, were then compared with renewal rates of the groundwater source, highlighting the perspective of natural nitrate level purging by volumetric variation.

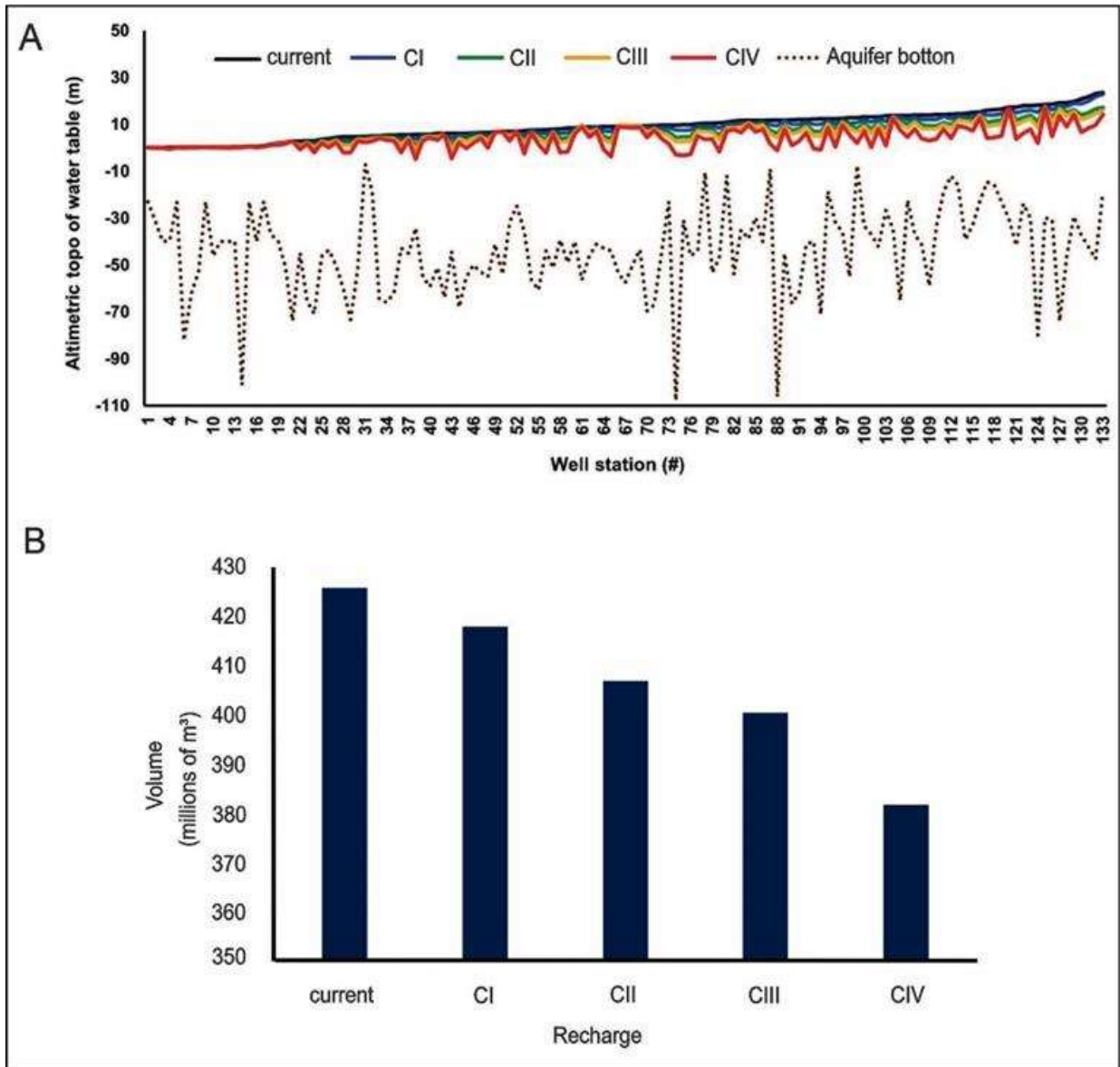


Figure 2 – Graph of potentiometric levels and their variations considering the simulated scenarios (A); estimates of saturation reserves (B).

Source: Adapted from Campos et al., (2023).

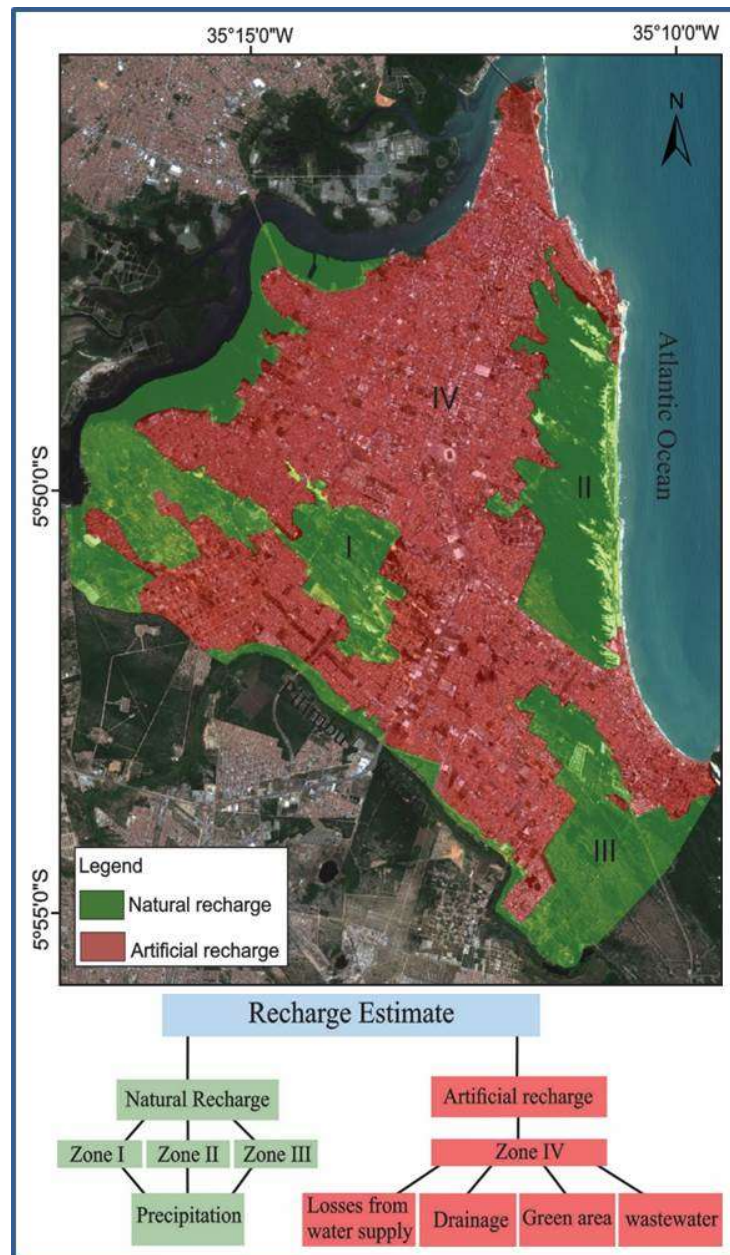


Figure 3 – Summary of the recharge rate composition used in the numerical simulations for the different reported scenarios, based on land use and occupation in the study area.

Source: Modified from Campos *et al.*, (2023).

4. Results and discussion

Hydraulic head values, both measured and interpolated, are shown in Figure 4, highlighting groundwater flows towards the ocean (east) and surface drainages (south-southeast and north-northwest). These characteristics guided the boundary conditions for local numerical models proposed by Campos *et al.* (2023). Irregular patterns in the potentiometric curves suggest that hydraulic heads likely suffer interference due to exploitation as well as varying conditions of groundwater recharge and discharge.

Hydrochemical analyses of samples from 62 wells over the periods of 2012, 2017, and 2020 revealed a progressive increase in nitrate concentrations over time. In 2012, the research area had 24 wells with nitrate concentrations above 10 mg/l (min 0.10 and max 30.15 mg/l), while by 2020, this number increased to 38 contaminated wells (min 0.13 and max 35.19 mg/l), representing a 58% increase in contaminated wells in the study area (Figure 5).

In this context, the wells that showed the greatest increase in nitrate concentration over time, represented by the statistical slope of the annual variation, were: well 8 (slope 1.9), well 7 (slope 1.3), well 38 (slope 1.1), well 23 (slope 0.86), and well 12 (slope 0.82), with no reduction observed in any of these sampled wells (Figure 5).

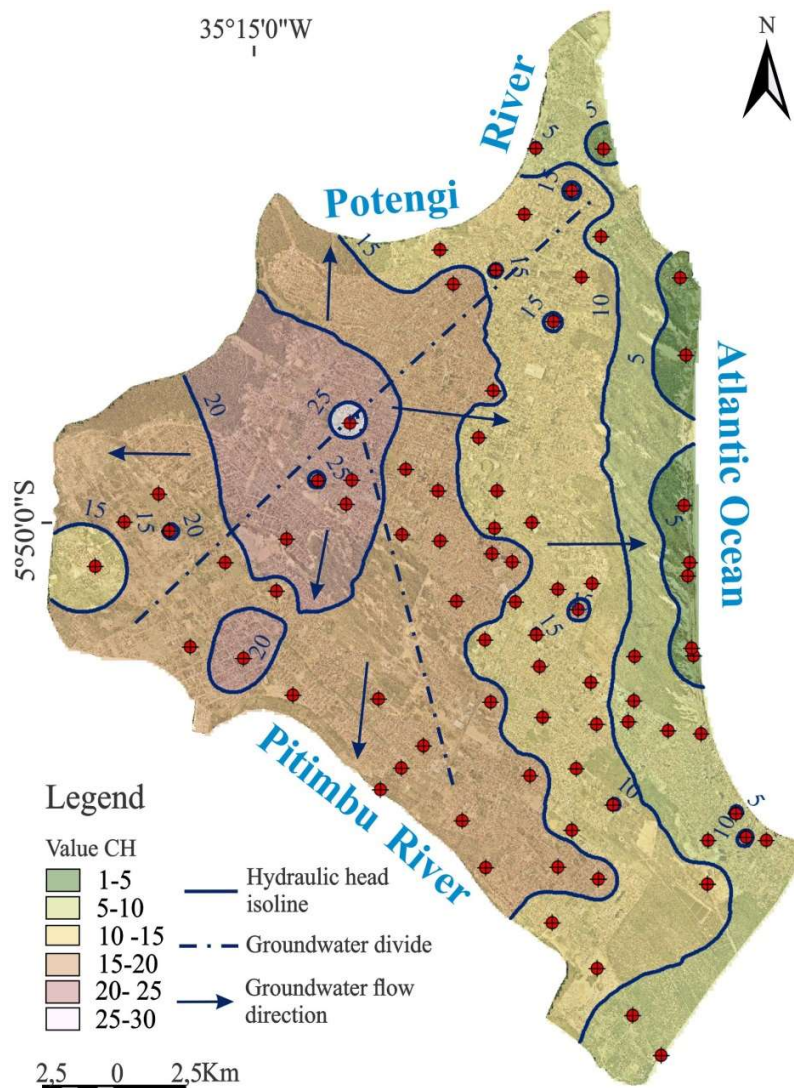


Figure 4 – Hydraulic heads, with contour lines and arrows indicating the directions of groundwater flow in the study area.

Source: Authors (2024).

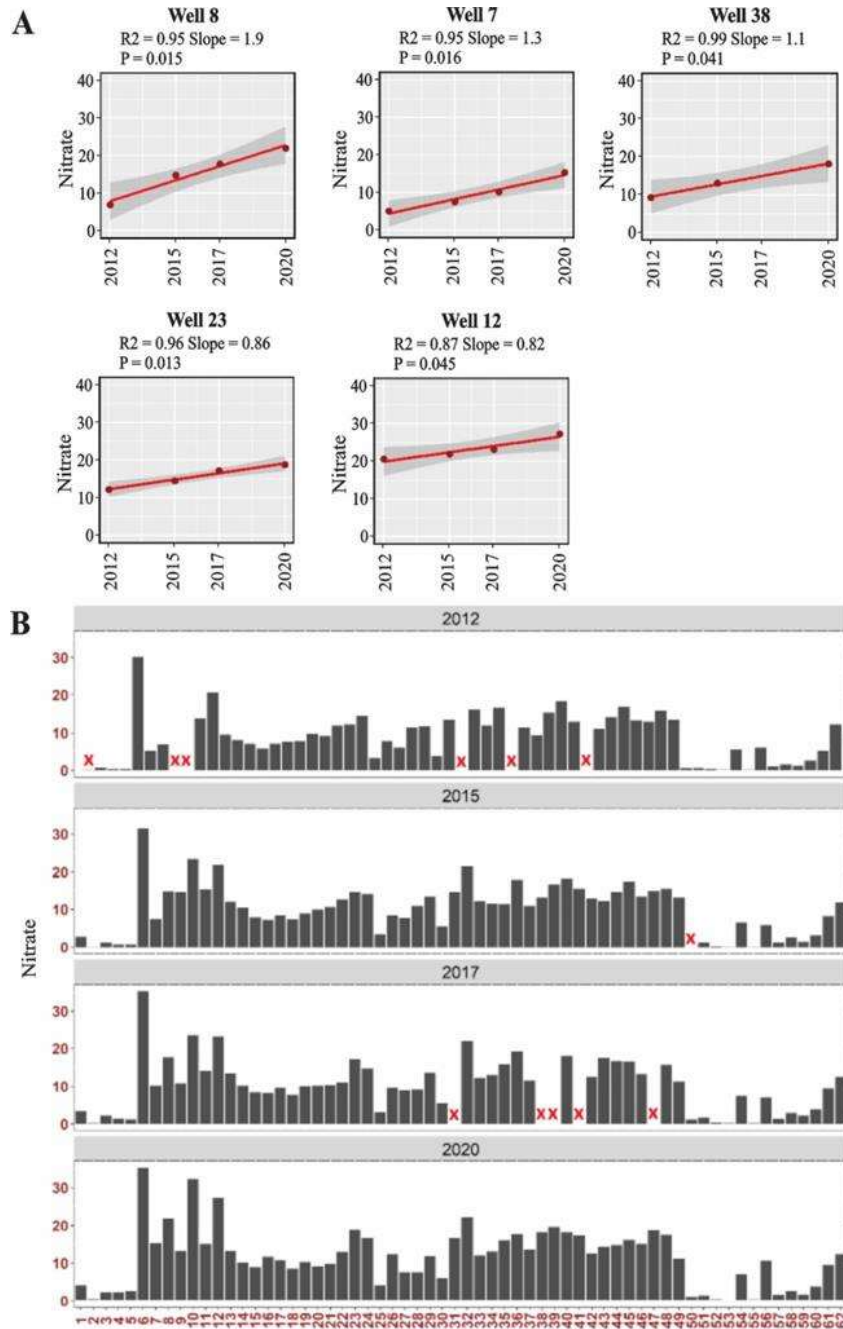


Figure 5 – Relationships of wells that exhibited the greatest increase in nitrate concentrations over time (A); graphic summary of the evolution of nitrate concentrations in sampled wells (B); wells marked with "x" were not sampled in the respective year.
 Source: Authors (2024).

Spatially, the wells with the highest nitrate levels are positioned to the west in the research area, where the greatest contamination advances occurred with nitrate N levels up to 35 mg/l, three times higher than the limit established by

Ordinance GM/MS N° 888/2021. The lowest nitrate concentrations are in the southern region, with values ranging from 0.2 mg/l to 5 mg/l (Figure 6).

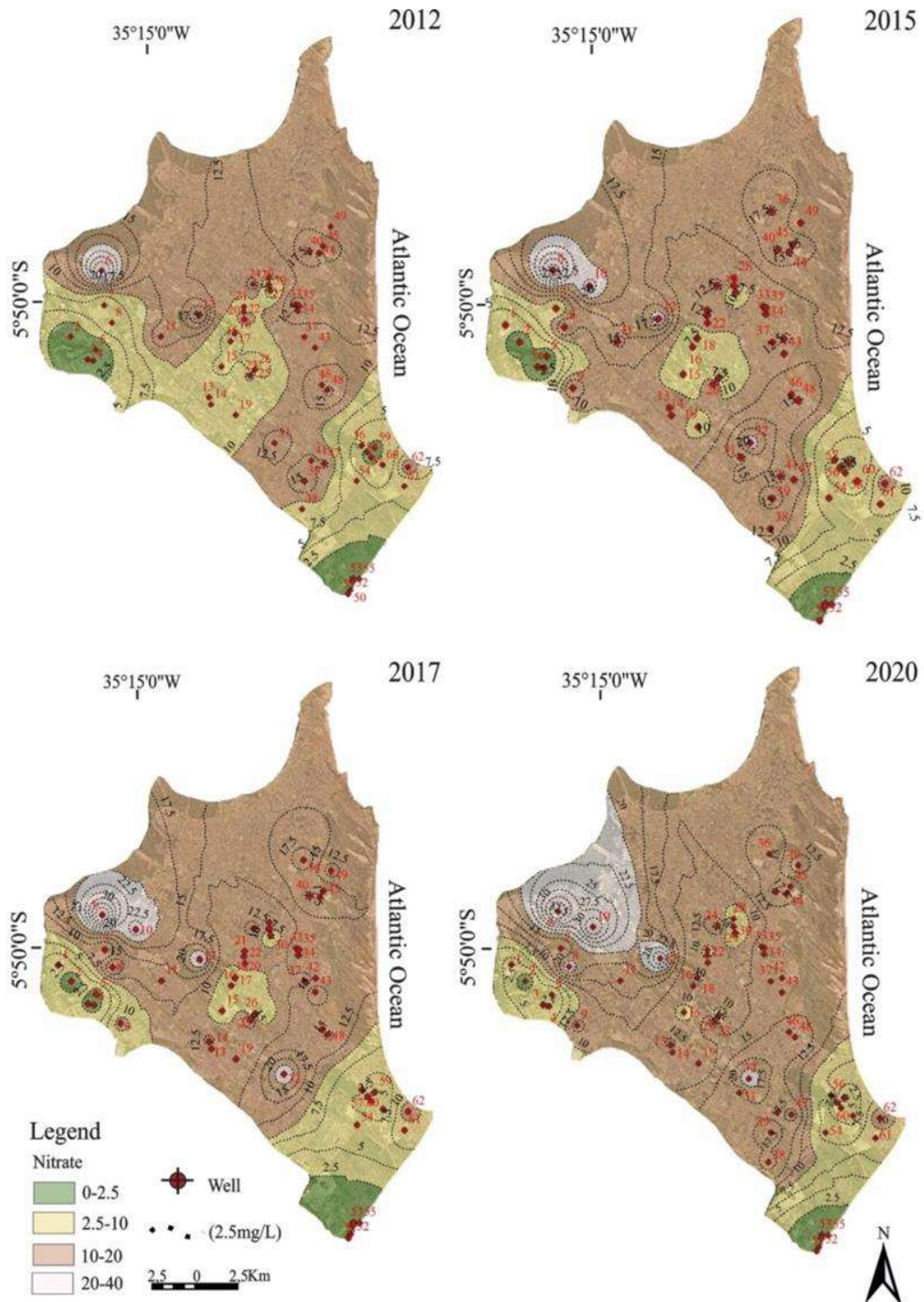


Figure 6 – Isovalue map spatializing the nitrate concentration zones in different sampled periods.

Source: Authors (2024).

In this sense, the southern zone of the research area possibly benefits from the dilution system provided by the sanitary sewage collection network, whereas the western and central parts do not reflect the same situation. This is evident as no reduction in well contamination was observed in these regions, even though some points have a sewage collection network. Therefore, it is considered that these zones of high concentrations are related to three main factors: i) population growth generating a larger volume of wastewater, ii) the insignificant natural recharge affecting dilution, and iii) the inefficiency of the existing sewage system, with higher leakage rates for pipes installed before 1990.

Positive correlations between NO_3 and the parameters Cl^- , Ca^{2+} , Na , Mg^{2+} are consistent with a contamination origin and its progression from the same source, i.e., mainly from the infiltration of untreated wastewater (Figure 7). Such correlations show that the increase of NO_3 is related to contamination from human waste, particularly urea; the increase in the concentration of the Cl^- ion is associated with detergents and household products; and the increase of Ca^{2+} , Na , and Mg^{2+} ions are linked to industrial products, which corroborates previous data from Melo (1995).

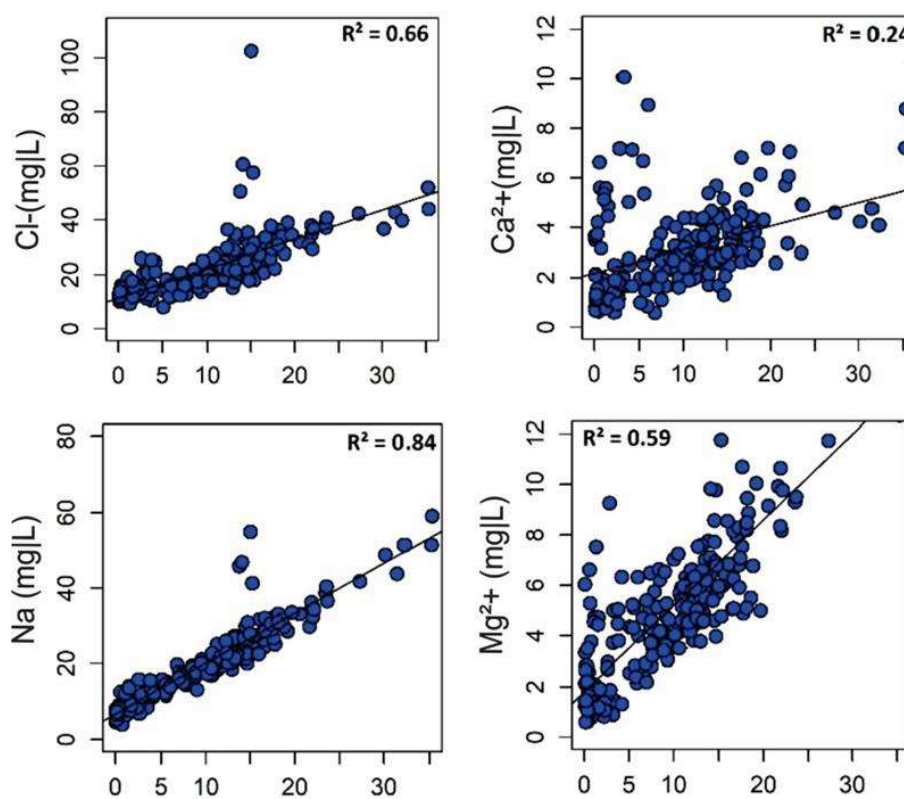


Figure 7 – Correlations between the parameters Cl^- , Ca^{2+} , Na , Mg^{2+} and nitrate, consistent with a single source of contamination associated with the infiltration of untreated wastewater.

Source: Authors (2024).

In this regard, the sewage system infrastructure and the natural recharge zones I, II, and III (see Figure 3) favor the dilution processes and consequently the attenuation of nitrate levels, a crucial factor in keeping the southern region of the area free from contamination over time. The map in Figure 8 shows the areas with sewage systems (blue line) and the areas where the sewage infrastructure exists (red lines) but has not yet been connected to the network. It is observed that a large part of the recharge zones in the north, and a small portion in the southern zone of the research area, have been contemplated by the sewage network.

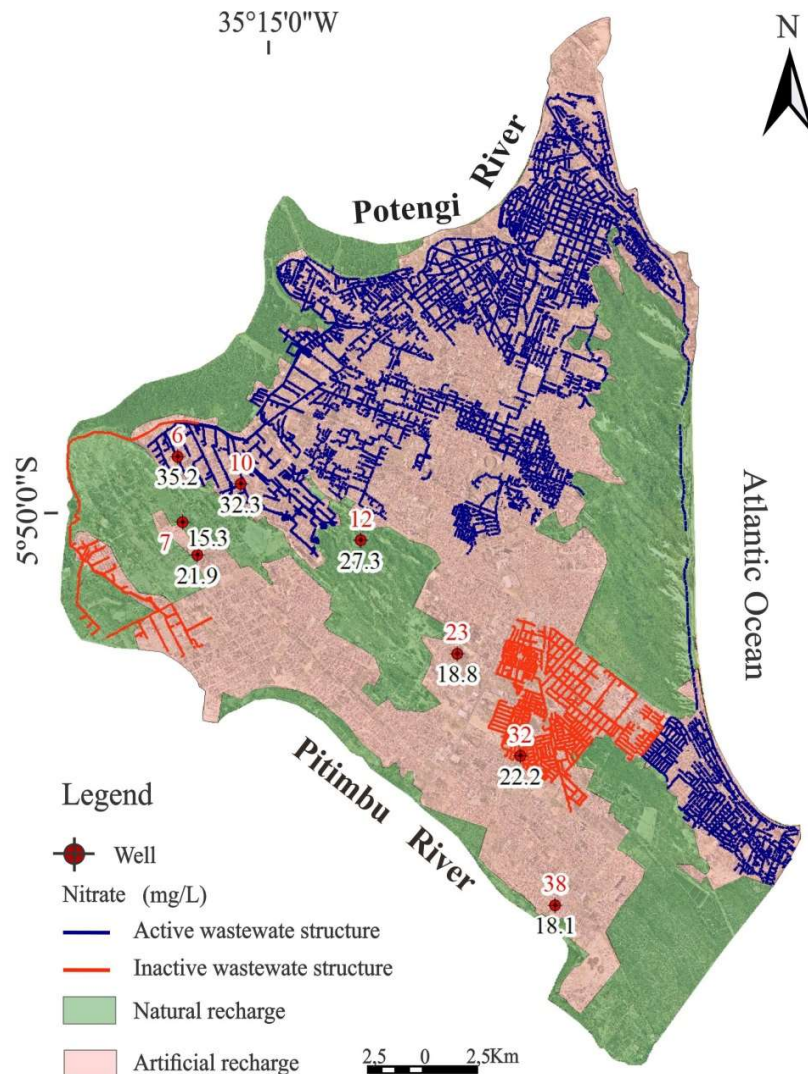


Figure 8 – Recharge zones based on land cover analysis and functioning sanitary sewage structure, and wells that showed the greatest increase in nitrate concentrations during the considered period. Souce: Adapted from Campos et al., (2023).

In this context, considering variations in saturation reserves and applying equation 3, results concerning the calculation of residence time based on volumetric variations and renewal rates imply a decrease in the depuration of nitrogen levels. In Scenario I, an estimated time of 6.5 years was determined, considering a 50% reduction in recharge contribution from wastewater ($6.45 \times 10^7 \text{ m}^3/\text{year}$) and a saturation volume of $4.17 \times 10^8 \text{ m}^3$. In Scenario II, a time of 7.4 years was estimated, considering a 100% reduction in recharge contribution from wastewater ($5.50 \times 10^7 \text{ m}^3/\text{year}$) and a saturation volume of $4.07 \times 10^8 \text{ m}^3$. The third scenario showed a time of 7.7 years, assuming a 50% reduction in both wastewater and losses from the water distribution network ($5.17 \times 10^7 \text{ m}^3/\text{year}$), with a saturation volume of $4.00 \times 10^8 \text{ m}^3$. The last scenario, with the most critical prediction regarding recharge deficit, obtained a residence time of 12.9 years, assuming a 100% reduction in both wastewater and losses from the water distribution network, with a saturation reserve value of $3.82 \times 10^8 \text{ m}^3$ (Figure 9).

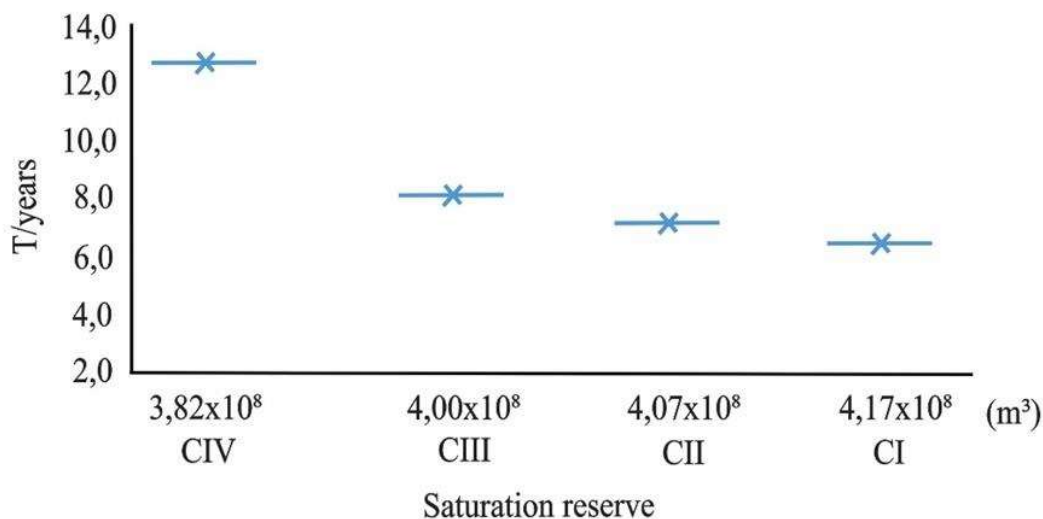


Figure 9 – Simplified graphical analysis of the evolution of residence time, associated with groundwater renewal rates, concerning the decrease in recharge in the various simulated scenarios.

Source: Authors (2024).

Given the results presented, there is a medium- and long-term perspective for the reduction of nitrate levels, although associated with a decrease in saturation reserves due to reduced artificial recharge (partly credited to the infiltration of untreated wastewater). The inconsistent reduction of these levels, even in subareas already covered by wastewater collection, is certainly due to the continued clandestine nitrogen inputs or the migration of these compounds through underground flows (mainly advective, convective, dispersive, or diffusive flows), considering they are conservative contaminants. This aspect is suggested for the discharge area in the northwest-central part of the area (see Figures 4, 6, and 8).

Thus, the studied context should consider additional management measures, which should be jointly conducted to improve groundwater quality. Among these measures are the reevaluation and readjustment of the installed capacity of local production wells and the identification of other water sources, including surface ones, that can be added to the public supply system, minimizing losses and drawdowns associated with decreased wastewater infiltration and improvements in the water distribution network. Additionally, there is a need to deactivate septic tanks, which are certainly still in use, as the sanitary effluent collection network becomes operational and/or expands.

5. Final considerations

The Barreiras Aquifer System presents high nitrate concentrations throughout almost the entire study area, despite having a sanitary sewage system in a portion of it. This link between nitrate concentrations and the sanitation structure is because the contamination comes from the infiltration of untreated wastewater. This observation is supported mainly by direct correlations between the parameters Cl⁻, Ca²⁺, Na, Mg²⁺, and nitrate in local well water samples, consistent with a single source of contamination associated with the infiltration of these wastewaters. In the current situation, it was found that thirty-eight (38) of the sixty-two (62) sampled wells already exhibit nitrate N levels above 10 mg/l, representing an increase of about 58% since the beginning of the considered period (2012).

The upper lithological substrate, composed mainly of aeolian sandy sediments with high infiltration rates, favors the vertical leaching of pollutants, particularly nitrogen compounds, towards the aquifer. In this scenario, the context of green and built-up perimeters in the urban area in question contributes substantially in terms of infiltration rates, in addition to the sanitary effluent collection network still being expanded.

This gradual decrease in saturation reserves in the simulated scenarios associated with decrease in artificial contributions to the aquifer recharge implies longer estimated renewal times for groundwater, exceeding twelve (12) years for the most critical reported scenario. This analysis, although preliminary and in volumetric terms, emphasizes

that the gradual reduction of this contamination is associated with the decrease in artificial recharge, although potentially impacting the dynamic equilibrium in the aquifer.

In this qualitative-quantitative context of the underground water source, the need for increased adoption of water resource management measures for the region is emphasized. Given the importance of groundwater, planning actions should prioritize public supply, considering the possibility of external sources outside the investigated area and the re-adjustment of the installed capacity of production wells. Simultaneously, there is a need for continued qualitative monitoring, with an emphasis on nitrogen compounds, as well as the saturation reserves associated with variations in saturated thicknesses, to refine consistent mathematical models of the aquifer.

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