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Influence of Land Use and Sanitation Issues on Water Quality of an Urban Aquifer

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Abstract
In this study, we sought to determine whether there was a relationship between sanitary aspects and land use on nitrate contamination in an urban aquifer of Fortaleza city in the state of Ceará, Brazil. For this, we analyzed land use (constructed area, exposed soil, green area and lagoons) using orbital images with a special resolution of 5 m, as well as sanitary aspects (access to sewage service and use of septic tanks and rudimentary cesspits for domiciles). To study groundwater quality, we collected 30 samples to assess the physical-chemical parameters, including nitrate, nitrite and ammoniacal nitrogen. Sectors with little constructed area produce low concentrations of nitrogenic compounds. Most occupied areas had a significant influence on nitrite concentration, likely due to infiltration from runoff and the low natural recharge from atmospheric precipitation. Areas where >75% of the domiciles were linked to sewage service suffered little nitrate contamination in rainy periods, while areas where <50% of the domiciles were linked to sewage service contribute to a high level of nitrate contamination in wet periods. The study showed that land occupation and sanitation characteristics are important variables in relation to contamination in urban aquifers. Public urban land use polices that promotes the highly effective collection of sewage services and sanitation are critical for preventing urban aquifer contamination.

Keywords Land use · Sanitation aspects · Groundwater · Rudimentary cesspit · Septic tanks

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1 Introduction

Nitrate is one of the main contaminants in the subsurface environment, and it is a problem in rural areas and cities. In cities, nitrate contamination can be derived from commercial, industrial or domestic effluents (Matiatos 2016). Domestic effluents have different components (Huang 2010), but nitrate is the most common because it is the result of the last process of decomposition of organic matter and its ionic form in water and cannot be removed during conventional treatment (Zhao et al. 2012). In addition, nitrate can generate complications in human health, such as cancer, miscarriages, infant mortality, abdominal pain and diarrhea (Ebrahimi and Roberts 2013).

The aquifer study is unconfined and is sited below a sector of consolidated occupation in Fortaleza city in Northwest Brazil. In this study, we assume that there is a very complex integrated relationship between urban characteristics and water quality. Tubau et al. (2017) indicated that groundwater management in urban areas requires detailed knowledge of an adequate tool for predicting water quality evolution. Urban aquifers can be impacted by diverse pollution sources. Urban aquifer pollution includes recharging from the losses of sewage, polluted urban runoff, polluted rivers or other surface water, seawater intrusion, and industrial leaks (Barrett et al. 1999; Lerner and Yang 2000; Foster and Chilton 2004; Manny et al. 2016; Peixoto et al. 2017a; Tubau et al. 2017). Urban aquifers, therefore, have strategic importance in relation to city sustainability and the promotion of important environmental services for residue decomposition.

The intricate situation of urban aquifers includes the socio-environmental complexity that is intrinsic to cities. To study urban aquifers, we must consider and localize soil appropriation, land use and sanitary conditions (Ford and Tellam 1994). Moreover, the vadose zone can act as an efficient filter against urban storm water runoff bacteria during the infiltration of storm water runoff (Voisin et al. 2018). Therefore, hydrogeological features and aquifer vulnerability are important for predicting the comportment of contaminants (Kazakis and Voudouris 2015). Schirmer et al. (2013) state that "There is a need to better relate the city features to environmental and water-related problems on the basis of comparable methodological approaches" (p.289). Methodologies address the need to combine land use data and contamination mechanisms from spatial and temporal perspectives.

The city of Fortaleza is located on an important costal aquifer, which, in the 1960s to 1980s, supported the municipal water supply service (Claudino-sales 2005). There was a necessity to expand the available water supplies because the population has increased by 50% in the last three decades and because of economic increases and industrial diversification (Elias 2003). Thus, the water resource management model adapted to constant droughts prioritized the superficial reservoirs in the detriment of studies and the use and conservation of aquifers (Peixoto et al. 2017b; Campos 2015).

In recent years, the constant droughts inherent to the semiarid climate, the precipitation irregularity and the possibility of climate change the aggravate factor, causing this management model to be questioned (Silveira et al. 2018). For example, currently, the water demand of Fortaleza city is 9.70 m^3/s, (SEUMA 2015), and this volume depends on the water availability in the country area of the state (Ceará), though the effect of climatic irregularity is greater in the coastal zone.

It is necessary to study water quality in urban aquifers to identify the mechanisms of nitrate contamination and to promote action for the use and conservation of an aquifer. Therefore, in this study, we identified whether there was a relationship between sanitary aspects and land use on nitrate contamination in an urban aquifer of the city of Fortaleza, Brazil.

2 Materials and Methods

2.1 Study Area

This study was conducted in an urban watershed in the western area of Fortaleza, in the state of Ceará, in the northeastern region of Brazil (Fig. 1). Fortaleza is the fifth most populous city in Brazil, with more than 2,600,000 habitants (IBGE 2018). Its climate is classified as tropical with a dry summer (Alvares et al. 2014), while 90% of the total area of Ceará state has a semiarid climate. The annual average temperature is between 26 and 28 °C, with maximum temperatures of 31 and 32 °C. The rains are concentrated from February to June, totaling 1500 mm/year, but the high potential evapotranspiration is 2300 mm/year, causing a negative climate hydric balance (Ceará 1992).

There is great difficulty regarding the water supply in large cities, such as Fortaleza, in semiarid regions. However, costal sediments are important alternatives for the water supply. Eighty percent of the study area is on sedimentary rocks, represented by sandstone, conglomerates and levels of silt and clay not lithified (Brandão 1998). Unconsolidated sediments of dunes are present in the northern part of the study area. These dunes have been the best aquifer potential of the area, supporting good flow rates of the wells (5–15 m^3/h) (Cavalcante 1998). Crystalline rocks are present in the southern portion of the represented Ceará Complex, characterized as Proterozoic paragnaiss migmatized. These rocks have poor aquifer vocations

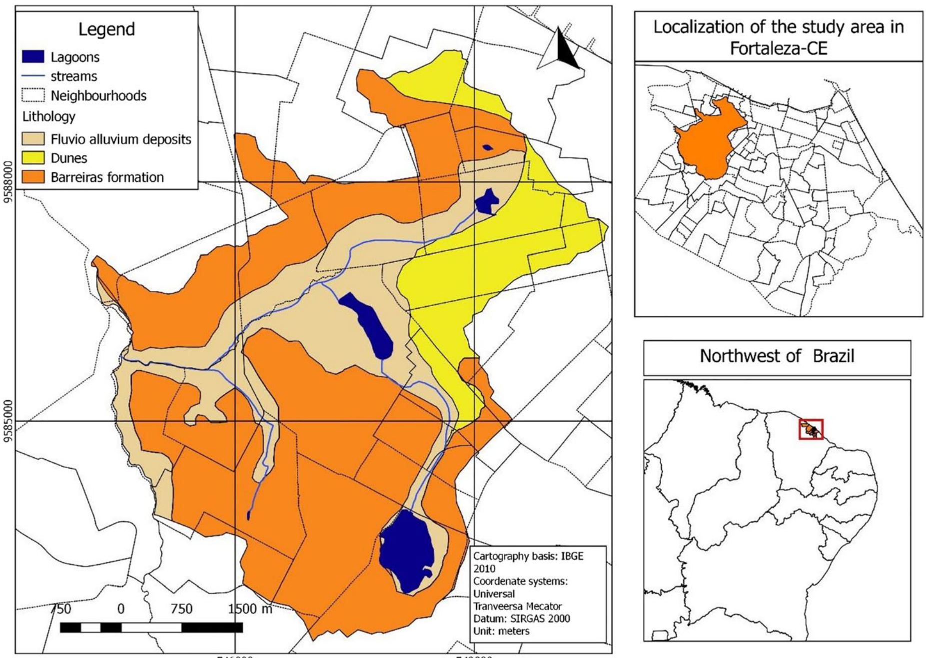

Fig. 1 Study area location

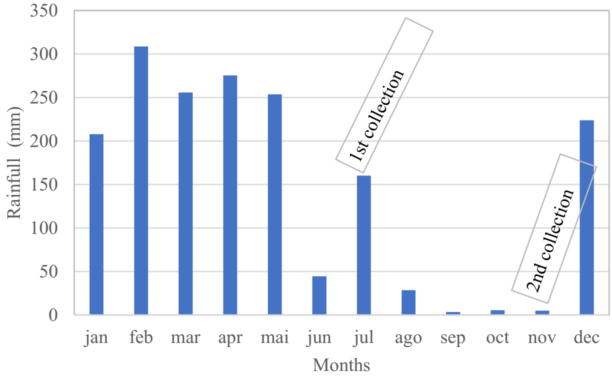

Fig. 2 Annual pluviometry in 2018 in the study area

(Cavalcante 1998). The fluvic-alluvium deposit was composed of interleaved layers of silt, sand and clay and accompanied the river course and lagoons.

2.2 Methodology

2.2.1 Land Use

For land use evaluation, we collected an orbital image of the satellite RapdEye-4, sensor RapidEye Earth Imaging System-REIS, which has the following characteristics: 5 multispectral bands (blue, green, red, red-edge, near-infrared), a spatial resolution of 5 m, and 12 bits of radiometric resolution. The utilized images were obtained from the Ministério do Meio Ambiente (MMA) and dated from June 2015.

In the preprocessing stage, we performed atmospheric correction and geometric correction of the image. Therefore, the processing stage consisted of the composition of bands in the following channels: blue/blue, green/green and near-infrared/red. This composition in false color highlights the green areas. We determined 4 land use classes: green areas, constructed

Table 1 Analytic methodologies applied according to APHA (2012)

Parameter	Methodology applied
Total Alkalinity	Titrimetric
Ammonium	Phenate
Bicarbonate	Titrimetric
Calcium	Titrimetric EDTA
EC	Electrometric
Total Hardness	Titrimetric EDTA
Magnesium	Titrimetric EDTA
Nitrate	Cadmium reduction column
Nitrite	Spectrophotometric
pH	Electrometric
Potassium	Photometric
Sodium	Photometric
TDS	Electrochemistry
Sulfate	Turbidimetric

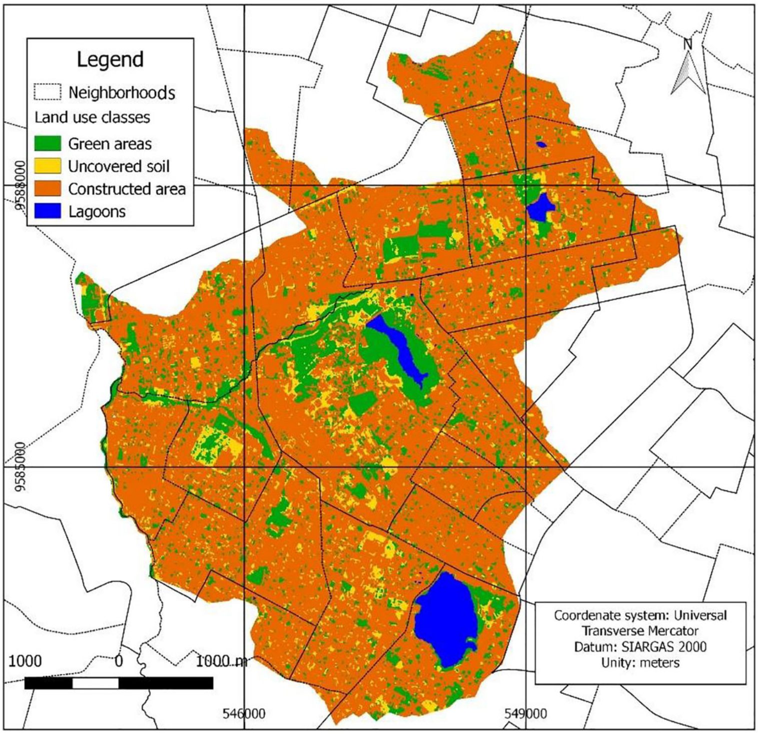

Fig. 3 Land use in the study area

areas, exposed soil and lagoons. We archived a supervised classification by region, utilizing maximum likelihood in Spring 5.5.2 of the Instituto Nacional de Pesquisas Espaciais-INPE.

2.2.2 Sewage Aspects

The sanitary aspects considered in this study were an index of the sewage service index composed of domiciles with sewage service access/total number of domiciles and the quantity of septic tanks and rudimentary cesspits used for domiciles/km^2.

All parameters were obtained from data of the 2010 census conducted by the Instituto Brasileiro de Geografia e Estatística-IBGE (2011). The data were aggregated in sectors and collected through a geographical information system. For this, we used the software Quantum Gis 2.18.26. To conduct comparative analyses with water quality parameters, we calculated the centroid of the represented results by sector-census polygons and produced kriging of the sewage service index and of septic tanks and rudimentary cesspits used for domiciles/km^2.

Table 2 Areas of respective classes

Land use classes (km^2)				
Green areas	Constructed areas	Uncovered soil	Lagoons	Total
3.33	18.76	1.77	0.63	24.52

2.2.3 Hydrochemistry and Water Quality

The quality of water is important to direct the priority of its use, and it is dependent on the hydro-geochemistry of the ground environment and the quality of water recharge in aquifers.

We chose the sampled wells from pre-cadaster made by Gomes and Cavalcante (2017), composed of 434 tubular and bore wells in Fortaleza. Thirty-two wells were selected for sampling and formed our cadaster in our study area. The camp procedure was conducted in 2 stages: the first was between July, 2 and July, 6, 2018 (end of the wet period), when we conducted analyses of 32 samples. The second was between November 22 and November 26 (i.e., the dry period), when we analyzed 30 samples because 2 wells were under maintenance (Fig. 2). We chose to work with 30 samples for each period of this comparative study.

All parameters were analyzed in the Environmental Geochemistry Laboratory/Geology Department/Federal University of Ceará-UFC on the same days they were collected in both periods. We analyzed the major ions of water: bicarbonate - HCO_3^-; chloride - Cl^-; sulphate - SO_4^{2-}; potassium - K^+; calcium – Ca^{2+}; and magnesium – Mg^{2+}. In addition to pH, alkalinity, electrical conductivity, ammonium-NH_4^+, ammonia nitrogen (N-NH_4^-), nitrite-NO_2^-, nitrate-NO_3^-, Fe^{2+}, Fl^{2+}, turbidity, water hardness, and total dissolved solids-TDS, were analyzed according to standard methods (APHA 2012) (Table 1).

For the N-NH_4^-determination, a blue compost was formed for the reactions between ammonium and hypochlorite and phenol. The hypochlorite was substituted for sodium dehydrated icloisocyanurate, according Aminot and Kérouel (2004). The NH_4^+ measurement

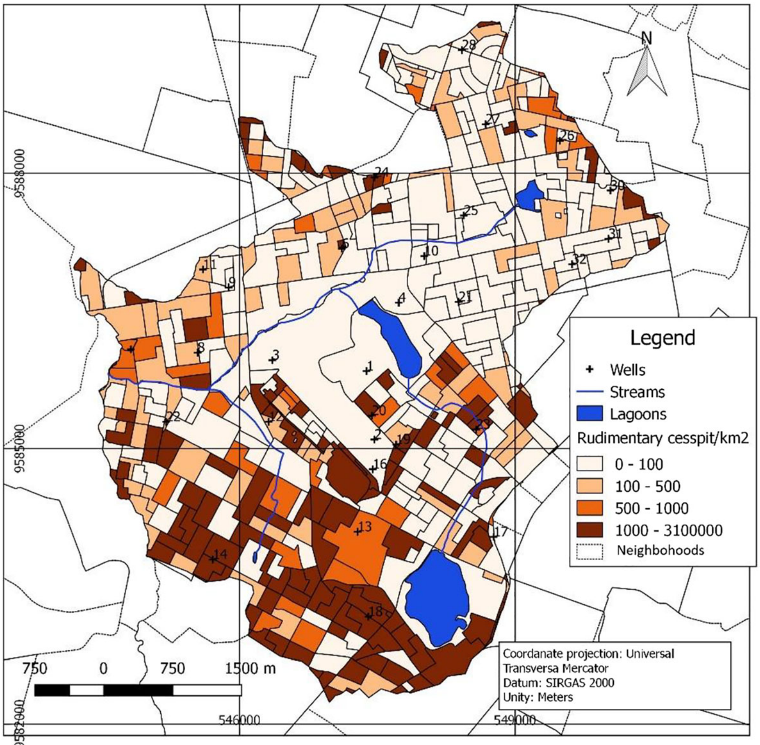

Fig. 4 Rudimentary cesspits in study sectors

began in situ, and phenol and sodium citrate reagents were added just after the collection of the samples. The pH, TDS and EC parameters were determined using a multiparameter probe (Thermo Scientific Orion e model Star A329 Portable).

We conducted hydrochemistry descriptions, classifying salinity and ionic water types using Qualigraf 18 software. We used multivariate statistics in the principal components of the analysis - PCA through Andad 2.0. This process allowed us to identify the associated parameters to be described at subsequent stages, such as parameters with meaningful relations with nitrate concentrations. “PCA utilized is widely used for data reduction in hydrochemical and hydrogeological studies” (Matiatos 2016, p. 804).

The geostatistical analysis was produced by kriging with pH, NO_3^-; Fe^{+2}; and SO_4^-. The spherical adjustment model was applied to describe the variogram. This procedure was performed using GeoR and Suffer 9 software, in which we performed the cross-validation process. We performed a comparative covariance correlation between the N-NO_3^- and sewage service index of septic tanks and rudimentary cesspits used for domiciles/km^2 using the tool “Cov.r” tool of Quantum Gis 2.18.26.

3 Results and Discussion

The characteristics of land use are important for describing its impacts on the environment. The appropriation of natural resources by societies involves not only the resource that can be

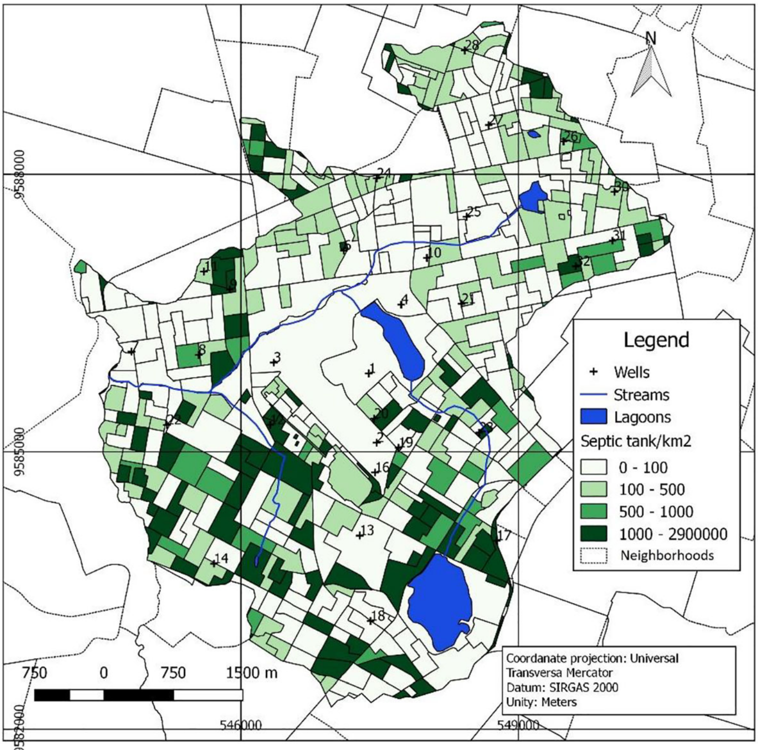

Fig. 5 Septic tanks in study sectors

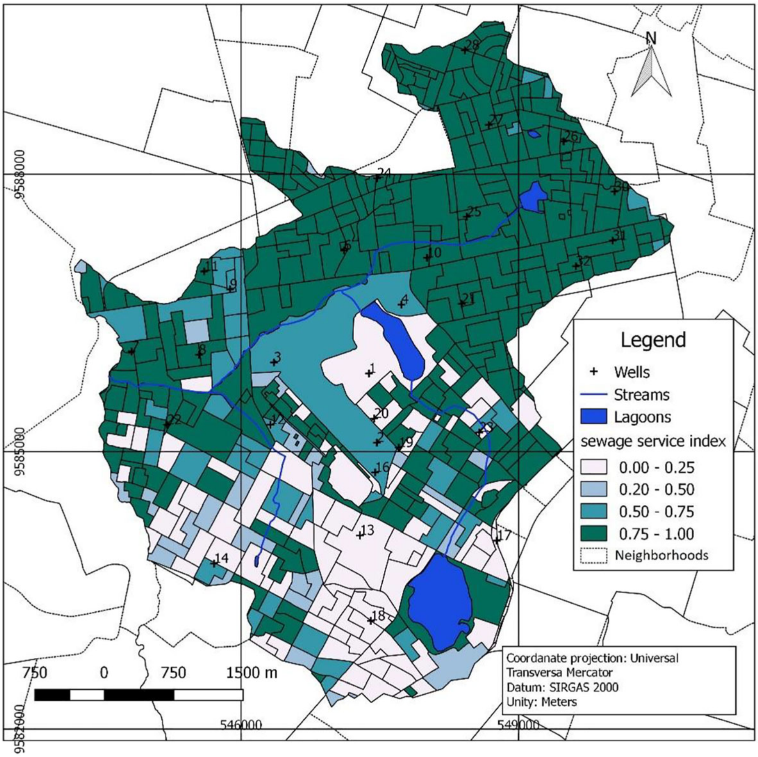

Fig. 6 Sewage service index

allocated but also the geographical space, where soil use promotes important landscape changes. Ford and Tellam (1994) found a strong relationship between NO_3^- and Ba^{+2} pollution and land use. However, the highest salinity, sulfate, chloride, sodium, boron and total heavy metal concentrations are associated with metalworking in industrial sites.

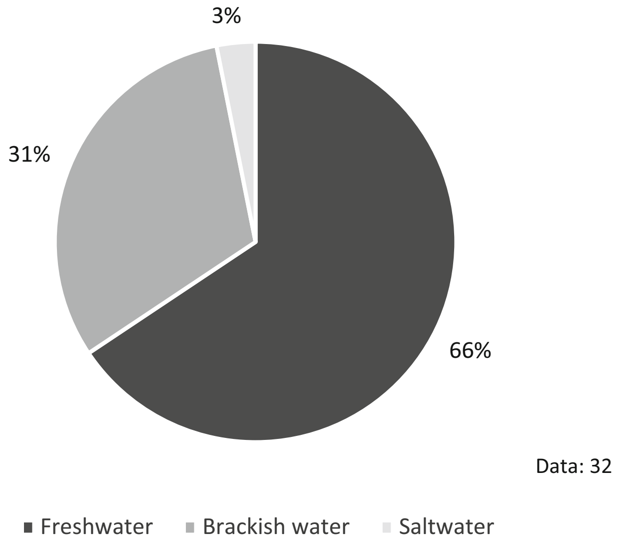

Fig. 7 Salinity water classes

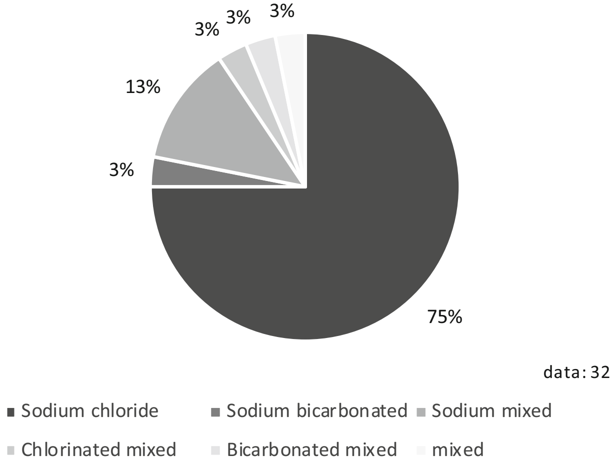

Fig. 8 Ionic water classes

The study area has been used for commercial and residential use, and the constructed area combined both types of use. Occupation is mostly consolidated, and most of the small areas associated with lagoons have not been used for land classes (Fig. 3). For example, a university campus is located in the central area, and the institutional use does not permit residential occupation and is the most conserved sector in this study area.

Constructed areas are largely composed of residential use, with commercial use restricted to buildings. In addition to the main avenues, 76% of the study area is composed of constructed areas. The green area comprises 13%, and the exposed soil comprises 7%. These last typologies are non-occupied areas and likely promote low qualitative and quantitative impacts on the hydrological cycle (Table 2).

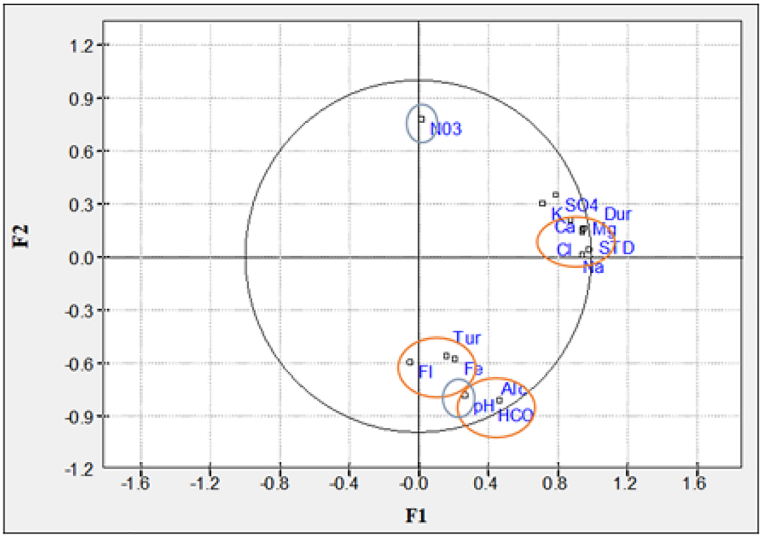

Fig. 9 PCA of the wet periods. Legend: Tur - turbidity; Fl - fluoride; Fe – iron; Alc- alkalinity; K – potassium; Ca – calcium; Mg – magnesium; Dur – hardness; Na – sodium; NO3 – nitrate; SO4 – sulfate; and TDS – total dissolved solids.

Occupied urban areas have many potential contaminants; in the study area, domestic sewage is the principal contamination source associated with this land type because there are no industrial activities in the study area. In contrast, sewage collection systems are found in all study areas, but in cities, there are many residences that are not linked to a sewage system even though a system is available. The sewage in these areas promotes the potential recharge of the urban aquifers, increasing volume but causing declines in water quality. In an urban watershed of 20 km^2 in Fortaleza city, Peixoto et al. (2017a) estimated that 1,07 × 10^6 m^3/year came from domestic effluents. Passarello et al. (2012), who worked for more than 10 years, quantified this aquifer recharge type in 5% of the total recharge in unconfined aquifers. These conditions show intrinsic relationships between water quality and water quantity; thus, the replacement of natural recharge by anthropogenic recharge occurs according to the urbanization process.

3.1 Sanitation Indicators

The distinction between septic tanks and rudimentary cesspits has been discussed in Brazilian standards such as NBR, 7229 (ABNT 1993). The first is a cylindrical or prismatic unit that treats the effluent for horizontal flow through sedimentation, flocculation and effluent digests, while the rudimentary cesspit is the device that does not obey the constructive and operation requisites established in NBR (op. cit). It receives the effluents directly in holes and ditches that are coated or not coated with concrete material, without any mechanism of mitigating the contamination.

The sanitary indicators used to represent the density of septic tanks and rudimentary cesspits are the main contamination potentials in the area. There is a considerable density of rudimentary cesspits in the southern sector (Fig. 4). Septic tanks have demonstrated major dispersion, but the southern sector has also been identified to have a large density (Fig. 5).

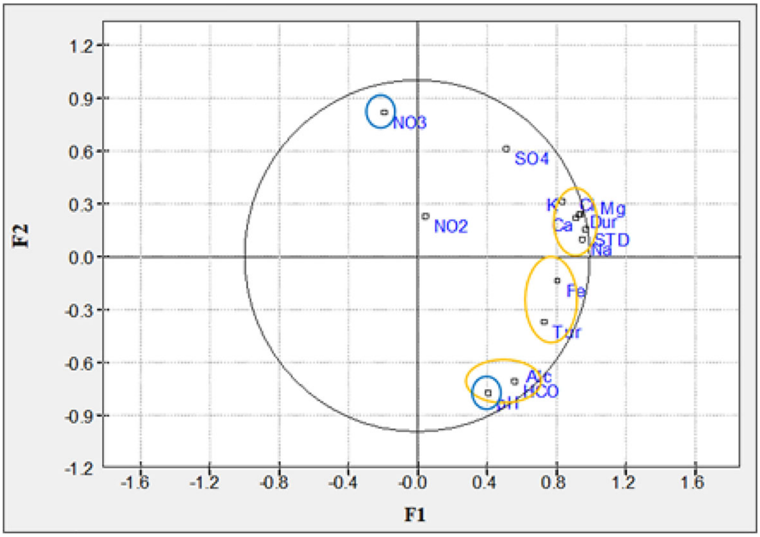

Fig. 10 PCA of the dry periods. Legend: Tur - turbidity; Fl - fluoride; Fe – iron; Alc- alkalinity; K – potassium; Ca – calcium; Mg – magnesium; Dur – hardness; Na – sodium; NO3 – nitrate; SO4 – sulfate; and TDS – total dissolved solids.

Table 3 Basic statistics of parameters

Parameters	min	max	mean	med	SD	Parameters	min	max	mean	med	SD
pH	4,0	7,3	6,1	6,3	0,9	Na^+(mg/L)	27,1	495,9	103,0	91,0	84,8
pH*	4,1	7,5	6,1	6,2	0,8	Na^{+}*(mg/L)	26,0	658,6	104,6	76,3	115,1
EC(ms/cm2)	152,7	4.922,0	797,7	672,1	799,9	K^+(mg/L)	2,7	26,7	11,7	12,0	5,5
EC*(ms/cm2)	373,4	7.830,0	913,9	665,4	1.339,0	K^{+}*(mg/L)	2,5	37,5	12,1	12,3	6,4
Alk(mg/L)	4,0	220,2	71,5	56,6	65,9	N-NO_3^-(mg/L)	0,1	33,2	11,6	10,1	9,0
Alk*(mg/L)	0,0	210,1	80,7	52,6	85,6	N-NO_3^-*(mg/L)	0,4	35,6	10,6	7,6	8,7
HCO_3^-(mg/L)	0,0	246,4	87,3	69,0	80,4	NO_2^-(mg/L)	0,0	1,1	0,2	0,0	0,3
HCO_3^-*(mg/L)	0,0	256,3	98,4	64,1	104,5	NO_2^-*(mg/L)	0,0	0,3	0,1	0,0	0,1
Cl^-(mg/L)	19,0	1.374,6	149,5	111,5	232,8	N-NH_4^+(mg/L)	0,0	3,0	0,7	0,1	1,0
Cl^-*(mg/L)	18,0	2.439,2	185,2	109,0	432,0	N-NH_4^+*(mg/L)	0,0	0,1	0,1	0,1	0,0
Ca^{2+}(mg/L)	0,8	108,0	21,2	19,6	19,0	Si^{2+}(mg/L)	3,4	59,6	17,7	13,4	17,0
Ca^{2+}*(mg/L)	8,0	192,0	25,1	18,0	33,7	Si^{2+}*(mg/L)	2,6	55,3	14,4	8,5	14,4
Mg^{2+}(mg/L)	2,9	218,4	23,0	16,8	36,3	Fe^{2+}(mg/L)	0,1	2,1	0,2	0,1	0,4
Mg^{2+}*(mg/L)	6,7	364,8	27,5	15,8	64,1	Fe^{2+}*(mg/L)	0,1	0,5	0,1	0,1	0,1
Hardn.(mg/L)	14,0	1.180,0	148,9	117,0	193,3	Fl^-(mg/L)	0,2	1,0	0,4	0,4	0,2
Hardn.*(mg/L)	32,0	2.000,0	177,6	113,0	347,6	Fl^-*(mg/L)	0,1	0,9	0,2	0,2	0,1
SO_4^{2-}(mg/L)	0,6	139,8	29,9	26,0	28,1	TDS(mg/L)	146,0	2.653,0	493,3	419,0	429,2
SO_4^{2-}*(mg/L)	0,5	93,8	31,6	31,7	24,0	TDS*(mg/L)	181,0	4.071,0	534,9	394,0	691,9

*Dry period; SD = standard deviation; med = median

Both are alternative ways to dispose of sewage, and the density values are related to a lack of access to the sewage collection system.

To understand the magnitude of these values in Brazil, the installation of septic tanks must not exceed the limit of 10 units/ha or 1000 units/km^2 (ABNT 1993). According to the United States Environmental Protection Agency, the density must not exceed 15/km^2; this limitation considers the indirect relationship with the quantity of contaminants that can be input into the underground environment (Scalf et al. 1997). Seeing the installation limits of septic tanks in Brazil, 40% of the census sectors exceeded the limit of 1000 units/km^2. Though they are more dangerous than septic tanks, rudimentary cesspits do not have any building parameters. These pits are usually coated or uncoated holes in the ground; every censing sector studied had rudimentary cesspits, which totaled 13.545 devices.

In the southern sector, there was less access to sewage services, in which most census sectors had less than 25% of domiciles linked to a sewage network (Fig. 6). The index showed evidence that the study area is a transition between urban and suburban sewage services, as evidenced by the transect from north to south, where the index had a value from 1 to 0.03 in the censing sectors. The deficit of sewage system access was noted in Fortaleza, showing socio-spatial segregation characterized by bad sanitation conditions in some peripheral zones, as reported by Bento (2011) and Peixoto (2017).

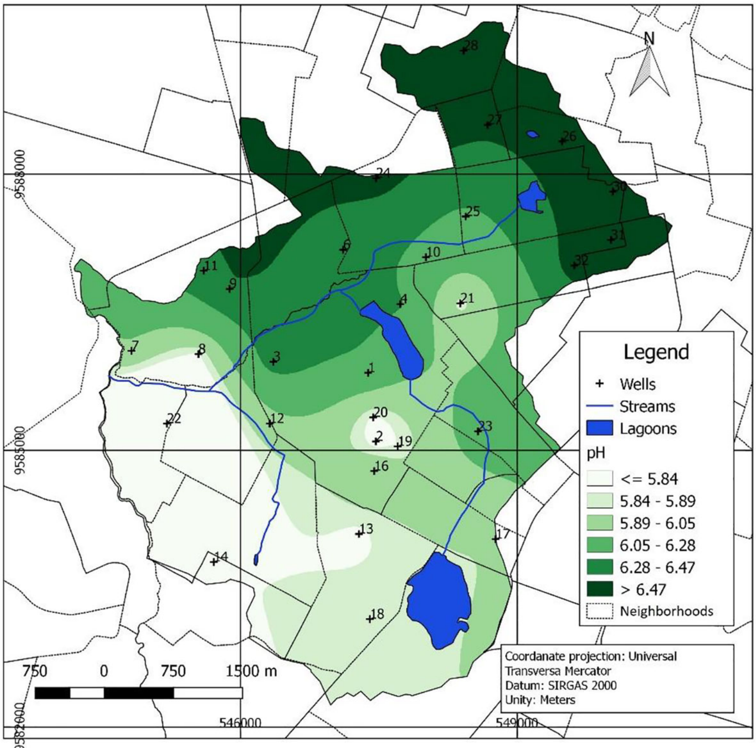

Fig. 11 pH zoning in wet periods

3.2 Water Quality

The water quality was described as 66% fresh water, 31% brackish water and only well number 11 had salt water (Fig. 7). There is good natural water quality in the urban aquifers under Fortaleza (Gomes and Cavalcante 2015). The main water classes according to the pie diagrams are sodium chloride (75%) and sodium mixed (13%), while other classes were found in only one well (Fig. 8).

Main compound analysis, PCA, showed small differences between the dry and wet periods. There was a relationship between all major ions in the wet period, as was also verified by Zhang et al. (2015), who found that bicarbonate (HCO^-) was associated with alkalinity and pH. This result was also observed in dry periods, but the sulfate was dislocated and not associated with any ion. In both periods, we noted an inverse correlation between pH and NO_3^-, as seen in Factor 2 (F2) of PCA, which explained 32,4% of the variance (Figs. 9 and 10). The pH decrease had a biogeochemical relationship with organic matter oxidation and nitrate contamination (Hem 1959). Wilhelm et al. (1996) and Liu et al. (2013) suggested that increases in NO_3^- concentration in aqueous ways are the result of the oxidation of NO_2^- rather than the acidification of waters. Therefore, in this condition, water has a high NO_3^- concentration.

Generally, in dry periods, all major ions had a mean and median higher than those in wet periods. This result is an indication of a pluvial regime of aquifer recharge, a common

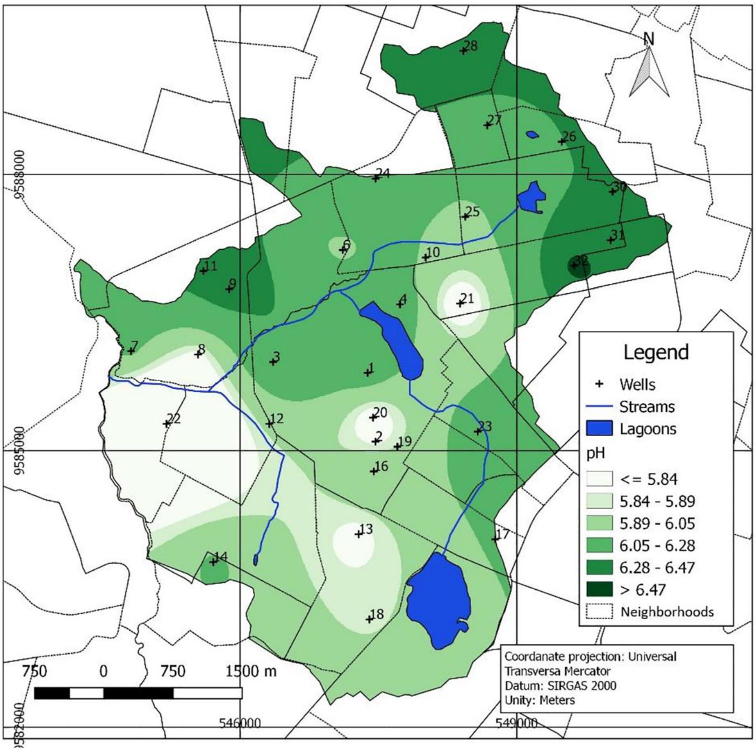

Fig. 12 pH zoning in dry periods

characteristic in unconfined aquifers that presents more major ion dilution because of pluvial recharge addition (Fetter, 1994).

Sulfate (SO_4^-) was the only major ion that lowered the standard deviation (SD) average in dry periods (Table 3). The EC and TDS parameters of well 7 were not considered to be general medians because in both periods, the values were identified as anomalous because they were 6 and 11 times larger than the media in wet periods and dry periods, respectively. This result occurs due to the location of this well, which was built in fluvic-marine sediments with clay layers containing high salt concentrations. Oliveira et al. (2014) identified a tidal influence in the river section in which well 7 is located, and this influence likely causes recharge to occur from marine water in fluvic marine alluvium.

The deep increase in saturation causes a decrease in oxygen and redox potential. When dissolved oxygen is depleted in water, microorganisms reduce NO_3^- to N_2, Fe^{3+} to Fe^{2+} or $SO_4^{2-}$to H_2S (Merkel et al. 2012). Other authors associate the presence of high sulfate concentrations with urban effluents (Postma et al. 1991).

The zoning was conducted through the pH, SO_4^-, NO_2^- and NO_3^- parameters, which revealed different spatial comportments with regard to seasonal conditions. However, pH had an acidic tendency in both periods, with few seasonal differences. The more alkaline pH occurred mainly in the north, with values higher than 6.47, and the acidification tendency occurred in the south (Figs. 11 and 12). This result can be related to the input of elevated nitrate concentrations followed by the denitrification process. According to Hem (1959), natural denitrification in aquifers causes the acidification of groundwater.

Table 2 shows that the average and median sulfate increased in dry periods, although during the rainy period, the highest maximum value and highest variability of data occurred. The major concentration is mainly located in the east and west, with low values in the central area.

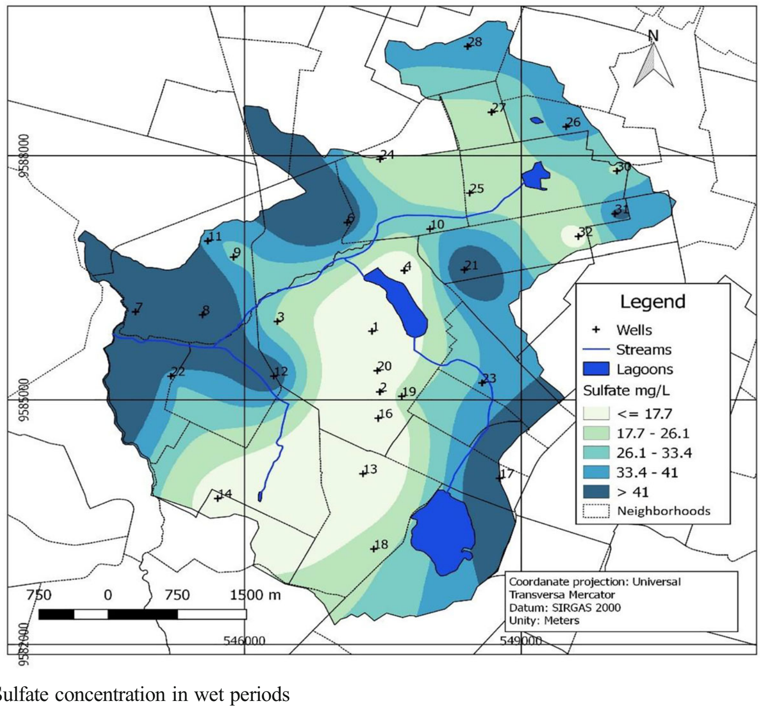

Fig. 13 Sulfate concentration in wet periods

In dry periods, there were decreased concentrations that were focused in the east of the area, with values above 41 mg/L, with a maximum of 139.8 mg/L in well number 7 (Figs. 13 and 14). However, this parameter is a "very important and widespread environmental problem in many irrigated agricultural regions" (Selvakumar et al. 2017 p. 29); however, some high concentrations can be associated with contamination tips of urban sources, such as domestic or industrial sewage (Postma et al. 1991). We understand that this situation has occurred in the study area because there are no sulfate characteristics in local natural waters. Furthermore, the low sulfate concentrations in the center of the area have been related to land use that is slightly intense (Fig. 2).

Nitrite is the intermediary position in a nitrogen cycle. It is important to understand the contamination mechanism because its presence may identify recent contamination. The nitrite had trace concentrations in 40% of the wells in both seasonal periods in the sample (wet and dry). The detected values were between 0.02 and 0.3 mg/L. However, in well 31, there was a relevant concentration (1.1 mg/L), which exceeded the maximum permissible value (MPV) for national consolidation regulation of the Brazilian Health Ministerial n. 5 /2017. Although of low values, the kriging showed a high concentration of nitrite in wet periods with trace values (0.3 mg/L) in samples of wells 9 and 11 (Fig. 15). In dry periods, nitrite was detected only in the extreme northwestern part of the area (Fig. 16).

Wells 9 and 11 were located in this area, and this is likely why rainy infiltration transported contaminants to the water table. In addition, the hydrogeologic conditions of the dunes aquifer, which has high aquifer vulnerability, could facilitate the input of contaminants. According to

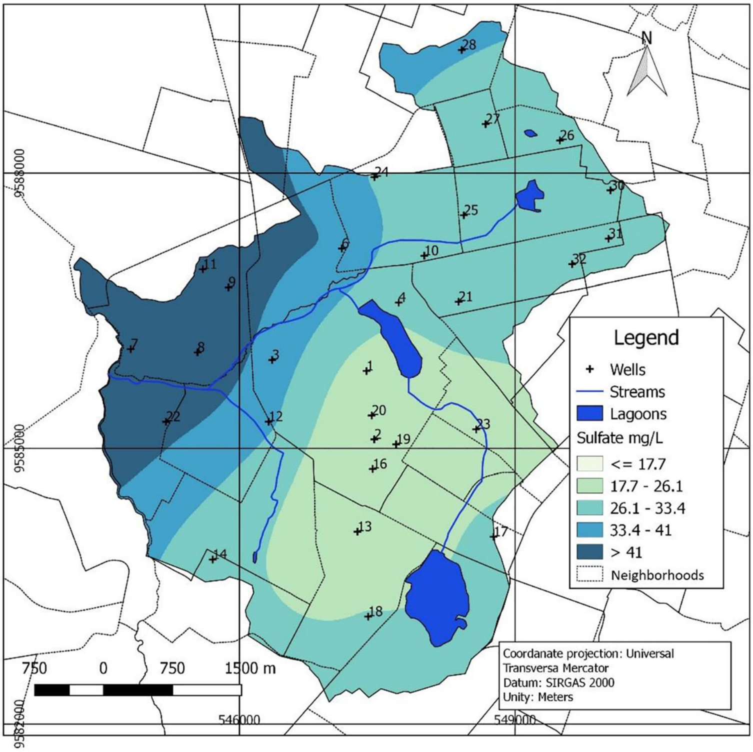

Fig. 14 Sulfate concentration in dry periods

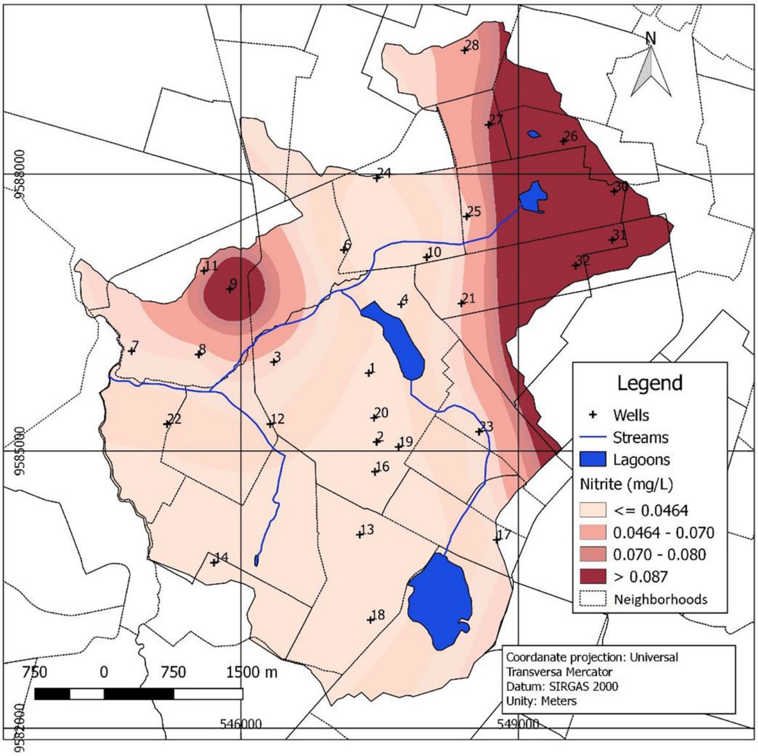

Fig. 15 Nitrite zoning in wet periods

Chitsazan et al. (2017), in urban means, contaminant concentration variations and land use, and the characteristics of sediments, underground hydrodynamic parameters, and hydrogeological situations affect the contamination conditions.

Nitrate is the most problematic parameter in the study area. There are 16 and 14 samples in the wet and dry seasonal periods, respectively, that exceeded the regulatory limits of the World Health Organization (10 mg/L for $N\text{-}NO_3^-$). In other words, 53 to 46% of the samples were contaminated. Consequently, if this water was consumed by human beings, it may cause health risks. However, this water can be used for other purposes, such as cleaning, general domestic services, industrial uses and garden irrigation.

The dilution of nitrate-rich water is the most commonly used method used to achieve the potability standard. However, when water for dilution is not available, other tactics may be used, e.g., reverse osmosis and anionic resin. In a system with similar hydrogeology as that of the aquifer studied, Melo et al. (2010) identified nitrate contamination processes in aquifers below Natal, the capital city of Rio Grande do Norte, also in Northeast Brazil. This contamination was advanced for two decades because the sewage system comprised the majority of septic tanks and rudimentary cesspits and because the water supply company utilized a dilution process to maintain acceptable water quality.

In wet periods, the mean concentration of nitrate tended to be higher than that in dry phases (Table 2). The recharge can be related to this contaminant. In dry periods, the contamination level reached its peak value (35 mg/L) but was present in a more concentrated and punctual form. This result indicates a greater input of domestic contamination than that in wet periods.

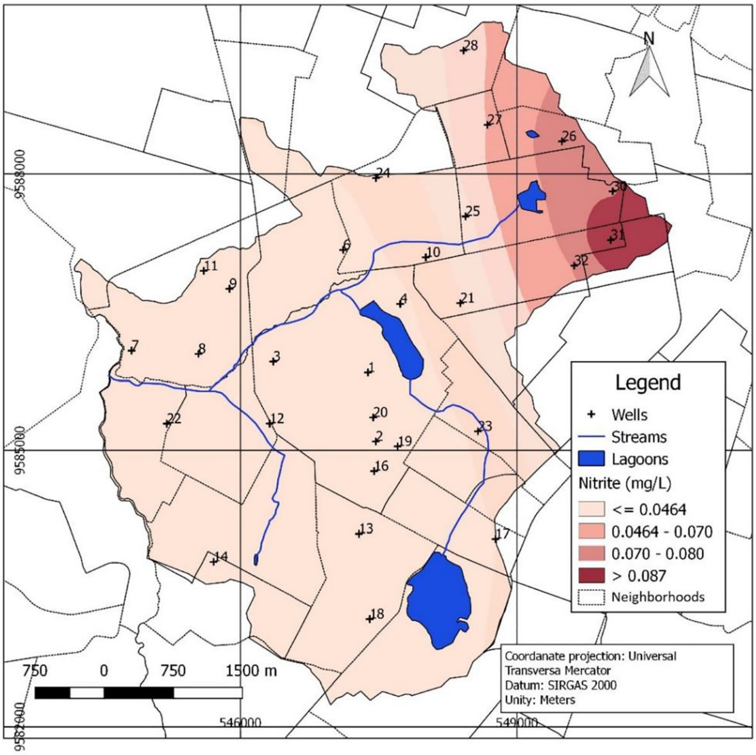

Fig. 16 Nitrite zoning in dry periods

The generalized contamination observed in rainy periods was characterized by contamination in the southwest and in the south, where the concentrations were above 13.5 mg/L. The same contamination level was present in the central-north area, observed in well numbers 21, 25 and 27 (Fig. 17). In dry periods, major nitrate concentration was present in the northeast, as observed in well 21 with 35.6 mg/L, and well numbers 25, 26, 31, and 32. High concentrations also occurred in the extreme south of the area in wells 18 and 17 (Fig. 18).

The differences between Figs. 17 and 18 are related to land use and sanitary aspects. First, where there is an agglomeration of green areas, the NO_3^- and SO_4^{-2} concentrations were lower. Naturally, low residential occupations denote a low potential contamination sources. Regarding sanitation aspects, areas with sewage service indices smaller than 0.50 are most susceptive to seasonal changes in nitrate concentration. Therefore, this area is largely constructed, and only the high density of septic tanks and rudimentary cesspits can explain this behavior. There are no agricultural activities that can be alternative sources of contamination. Based on the nitrogen isotope data, Matiatos (2016) verified that industrial and urban zones are mainly characterized by the most enriched δ15N–NO3 values, indicating that septic and industrial wastes were likely the sources of nitrate contamination. Zhang et al. (2015), in a study in the Peral River Delta in southern China, also demonstrated that domestic wastewater and industrial wastewater were the main sources of groundwater nitrate pollution. Therefore, as there are no industrial areas in the study area, we can conclude that sanitary aspects are the main factors controlling the nitrate content of groundwater.

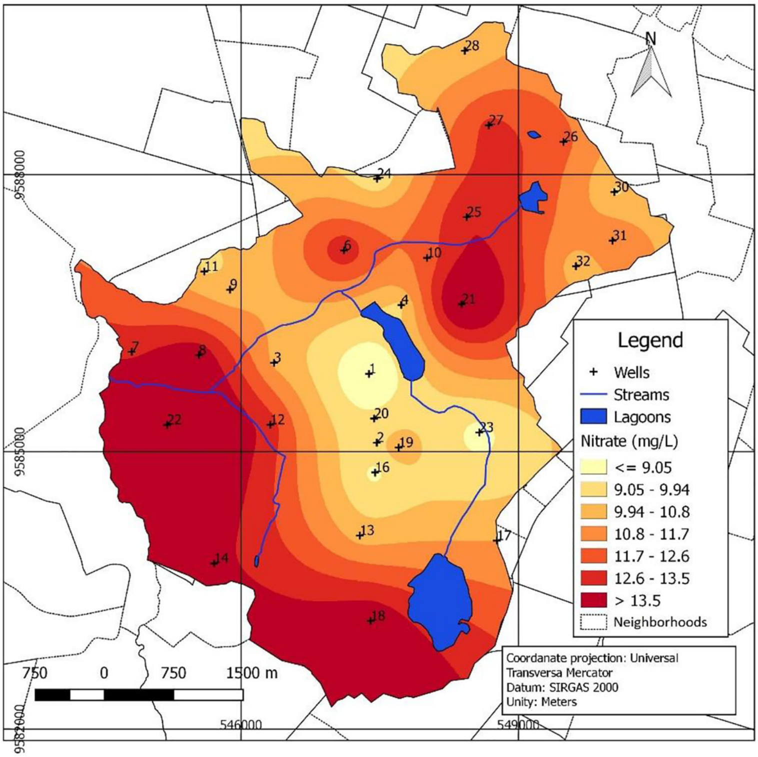

Fig. 17 Nitrate concentration in wet periods

The volume of effluent facilitates mobility conditions in rainy periods, when water infiltration leads to contaminates in the sub-saturated zone. Conditions of large impermeabilization and inefficient drainage systems in the northwestern area may promote nitrite concentrations above 0.08 mg/L. Thus, in this sector, nitrate contamination has been mostly associated with recharge from domestic effluents and water infiltration from runoff that can suffer quality changes depending on land use. Burant et al. (2018), in a study in Madison in the USA, showed that there were organic contaminants that provide information on similarities and differences in organic contaminants in urban runoff derived from different land uses.

It is perceptible that an inverse spatial association exists between pH and nitrate. The correlation of the co-variogram showed that this correlation was higher in wet periods (Table 4).

The covariance between septic tanks, rudimentary cesspits and nitrate showed that the density of rudimentary cesspits was more decisive for the increase in nitrate concentration in wet periods. Tubau et al. (2017), who studied urban aquifer recharges in Barcelona, Spain, showed the efficiency of water use data, population density, per capita allocation, and land use in the absence of more direct data with recharge. Thus, urban aspects are associated with this condition in the study area, in which the analyzed characteristics showed that the sources of these effluents are relative to the density of septic tanks and rudimentary cesspits above 500 per km^2. Furthermore, in the northern part of the area, there is dune aquifer occurrence, where the permeability is high (20%) (Cavalcante 1998), which has promoted the recent

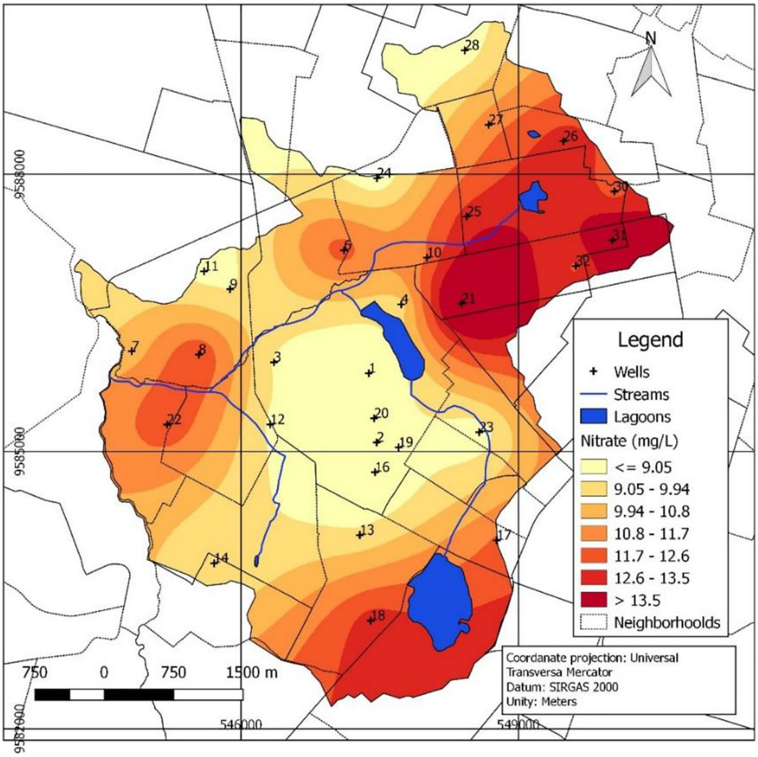

Fig. 18 Nitrate concentration in dry periods

contamination. In addition, the intrinsic vulnerability of the dunes aquifer may be influenced by major groundwater contamination.

4 Conclusion

There is a diversity of land use in the study area, but residences and urban infrastructure form the greatest type in constructed areas. Most specific land use occurs in the center of the area, with agglomerated green areas. This configuration induces different levels of nitrate and sulfate in groundwater.

The natural composition of the analyzed water showed good natural quality because only one sample was identified as saltwater, even though it was situated in a costal aquifer. Sodium

Table 4 Covariance between water quality and sanitary aspects [a]

Covariance				
	Septic tanks	Rudimentary cesspits	pH	pH*
Nitrate	0.1697	**0.17984**	**−0.53**	
Nitrate*	−0.1262	−0.1136		−0.46

Although not statistically significant, values in bold represent a seasonal temporal comparison between periods, and show a greater relationship between the rainy season and nitrate contamination and pH acidification chlorite waters represent 75% of the samples. ACP showed a common relationship between major ions that was similar in the two seasons that were studied.

A negative correlation among nitrate and pH shows the oxidation mechanism of the transformation of N-NO_3^- to N_2. Moreover, the contamination of nitrate in 53 and 47% of the samples in wet and dry periods, respectively, showed that this is the most problematic contaminant in the study area.

The comparative study exposed the determinant influence from sewage aspects and land use:

– In unconfined aquifers, nitrogen compound oxidation is very fast. We observed low concentrations of ammonia nitrogen (NH_4^+-N) and NO_2^-.
– Sectors with low constructed areas produced wispy pollution. Most occupied areas were most significant in their influence on nitrite concentration, likely due to infiltration from runoff and low natural recharge from atmospheric precipitation.
– Areas in which over 75% of the domiciles were linked to sewage services suffered little nitrate contamination in rainy periods, while areas where less than 50% of the domiciles were linked to sewage services have been influenced by large nitrate contamination in wet periods. Septic tanks were found at a density greater than 1000 units/km^2 in many of these census sectors.
– The infiltration of pluvial water in the southern area brings the best conduction of potential contaminants, and the dissolved oxygen promotes the rapid oxidation of part of the organic compound to nitrate. In this case, mixed recharge occurs from natural and anthropogenic sources.

Conditions of association between urban characteristics and urban aquifer contamination require further research. Urban sustainability depends on the rational use and conservation of urban aquifers. For this, the study provides evidence for the necessity of integrated urban land use and environmental policies to preserve green areas for environmental services. In addition, sanitation intervention is required in strategic sectors, mainly those with larger contaminant potential and high aquifer vulnerability.

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