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ASSESSMENT OF HYDROGEOCHEMISTRY AND GIS-BASED EVOLUTION OF GROUNDWATER QUALITY AND SALINITY IN THE SHALLOW AQUIFER OF SÃO JOSÉ DO NORTE, SOUTHERN BRAZIL

* Hullysses Sabino [1]
Juliana Menezes [2]

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Abstract
Water salinization changes the hydrogeochemistry, affects the quality and quantity of groundwater available for human consumption and its mitigation is time-consuming. A large brackish lagoon and the Atlantic Ocean surround the aquifer of the São José do Norte town (Brazil). Population and economic growth have been demanding more groundwater for domestic and agricultural purposes, stressing the aquifer. Despise the aquifer's social-economic relevance, potential threats on the groundwater have not yet been substantially investigated. The aims of this study were to analyze the hydrogeochemistry of the São José do Norte aquifer, focusing on the search for salinization indicators, and to assess the groundwater suitability for human drinking and irrigation purpose. Ionic ratios, Piper and USSL diagrams were applied, such as new methods, as Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) and the Groundwater Quality Index for human consumption ($GWQI_{HC}$). The results showed that the groundwater is mostly calcium bicarbonate type and did not indicated salinization occurrence. The interior and the northern area had the highest ionic content and the lowest water quality values. Most of the samples contained at least one parameter above the maximum allowed concentration for drinking purpose according to water quality standards. High concentration of alkalinity, iron and hardness reduced the groundwater suitability for human consumption, requiring water treatment before ingestion.

Keywords: coastal assessment, saltwater intrusion, GQISWI, groundwater quality, GWQIHC.

[1] Departamento de Geografia, Instituto de Geociências, Universidade Federal Fluminense, Brasil.
[2] Departamento de Análise Geoambiental, Instituto de Geociências, Universidade Federal Fluminense, Brasil.
**Corresponding author*: Departamento de Geografia, Instituto de Geociências, Av. Gal. Milton Tavares de Souza, s/nº, Niterói, Rio de Janeiro, CEP 24210-346, Brasil. Email: hscontact21@gmail.com

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Introduction

Large urban centers are concentrated in coastal regions around the world. For example, in the European Union, about 206 million inhabitants live in coastal areas (Eurostat, 2013), in the USA, 126 million people (NOAA, 2020), and in Brazil, about 51 million inhabitants live in coastal municipalities (IBGE, 2011). However, coastal water resources are widely contaminated (Bertrand *et al.,* 2016; Khawla and Mohamend, 2020; Marques *et al.,* 2020). Some usual sources of water contamination are uncontrolled urban growth, pollution from upstream drainage systems, lack of a basic sanitation system, improper disposal of solid and liquid waste, fertilizers from agricultural activities, mining, cesspools, inadequate construction and maintenance of wells and groundwater overexploitation bringing saline intrusion into aquifers (Thirumurugan *et al.,* 2019; Scherer *et al*., 2010; Tibbetts, 2002; National Research Council, 1984).

The Intergovernmental Panel on Climate Change stated that global warming will increase the number of coastal storms and sea level, which will intensify the saltwater intrusion processes (Wong *et al.,* 2014). Theses processes occurs when the hydrostatic force of fresh groundwater is reduced, usually due to overexploitation and reduction of recharge, decreasing the freshwater hydrostatic force in the face of sea pressure and favoring the flow of saltwater into the aquifer (Fitts, 2002; Rueda *et al.,*. 2018). According to Custodio and Bruggman (1987), the groundwater becomes salty for consumers if only 3% of the aquifer volume is saltwater, reducing the quality and quantity of groundwater suitable for domestic and agricultural uses.

Discovery and monitoring of groundwater salinization are based on the evaluation of the aquifer's hydrogeochemical behavior (Sangadi *et al*., 2022; Ahmed *et al*., 2022; Gueddari *et al.,* 2022). The ionic composition and hydrochemical faces are usually evaluated by diagrams of Piper (1944) and Stiff (1951) and by ionic ratios (Custodio and Llamas, 1983; Hem, 1985). Tomaszkiewicz *et al.,* (2014) based on an algebraic expression of Appelo and Postma (2005) developed the Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) to assist a preliminary detection of saltwater intrusion (Abu Salem *et al.,* 2022; Idowu *et al.,* 2022; Aladejana *et al.,* 2021).

Human influences, such as discharge of domestic effluents and agrochemicals, change the hydrogeochemical behavior and reduce the suitability of groundwater for drinking purposes. Ingestion of contaminated water can seriously affect human health, including kidney, intestinal and heart problems (WHO, 2011). Groundwater content of physical-chemical and biological parameters needs to be analyzed according to the guidelines of water quality standards, such as the one of Australia (NHMRC, 2011), European Union (CEC, 1980; CEU, 1998), USA (USEPA, 2009), India (BIS, 2012) and Brazil (Ministério da Saúde, 2017), in order for consumers to be assured of their water safety.

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Groundwater suitability for human drinking purposes can be analyzed by quality indices, such as the Groundwater Quality Index for human consumption ($GWQI_{HC}$) (Menezes, 2009; Sabino *et al.,* 2020). The $GWQI_{HC}$ algorithm indexes several physical-chemical and biological parameters and calculates the groundwater potability in a single number. This index applied guide and maximum concentrations values allowed by official water drinking standards, making the water classification in line with the legislation.

Indices such as GQI_{SWI} and $GWQI_{HC}$ produce a single value as a product, which assists the understanding of their results among government officials and the public (Lima, 2019; Sabino *et al.,* 2020). Indexes values may be spatialized through interpolating methods in Geographic Information Systems (GIS) (Sabino *et al.,* 2020). Spatial distribution maps associate the index values to land use, regional hydrogeology, lithology and topographic relief and complementary geosystemic analyzes.

Coastal town of São José do Norte is situated on a sedimentary plain between the mouth of the brackish Patos lagoon and the Atlantic Ocean. According to the Brazilian National Water Agency (ANA, 2010), no rivers suitable for supply flows in this town, which depends on the well supply system. About 27,500 people live in S. J. do Norte (IBGE, 2019). Currently, the well supply system needs to be expanded due to increased water demand (ANA, 2010).

Groundwater in the same coastal area were analyzed and some physical-chemical and biological parameters were find with high concentrations, as fecal coliforms and arsenic (Candiago *et al.,* 2019; Horák-Terra *et al.,* 2019; Michalski *et al.,* 2012). Wells in the free aquifers were built without sanitary protection and close to sources of contamination (Reginato *et al.,* 2008). Given this scenario, the present study aims to analyze the shallow aquifer in São José do Norte (Brazil) to: (i) identify the hydrogeochemical behaver and (ii) GIS-based space-time assessment of salinization and groundwater quality for human consumption and irrigation purpose. This study also intends to expand and disseminate knowledge about the hydrogeological features, to encourage and assist in the management of catchment, qualitative control, and monitoring of groundwater, which lacks research, despite its vulnerability and importance to society.

Materials and methods

Study area

São José do Norte town is localized in the State of Rio Grande do Sul (RS) (southern Brazilian), on a sandspit bounded by the Atlantic Ocean and the Patos lagoon estuary. The average temperature is 17.9 º C, precipitation varies between 1150 - 1450 mm, summer is the driest season, and winter is the wettest season (Machado, 1950). Coastal plain is a successive sand-bodies formed during transgressive-regressive marine cycles since the Miocene (Tomazelli and Villwock, 2005) (Figure 1).

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Coastal wind deposit from the Holocene period forms most of the area (IBGE, 2017). The soil is formed by dunes with smooth-undulating and undulating relief (DN) (IBGE, 2002). Aquifer has a porous domain and fine to medium quartz sand (Q4e), and some sandstones layers compose the lithology (Tagliani, 2002; CPRM, 2020).

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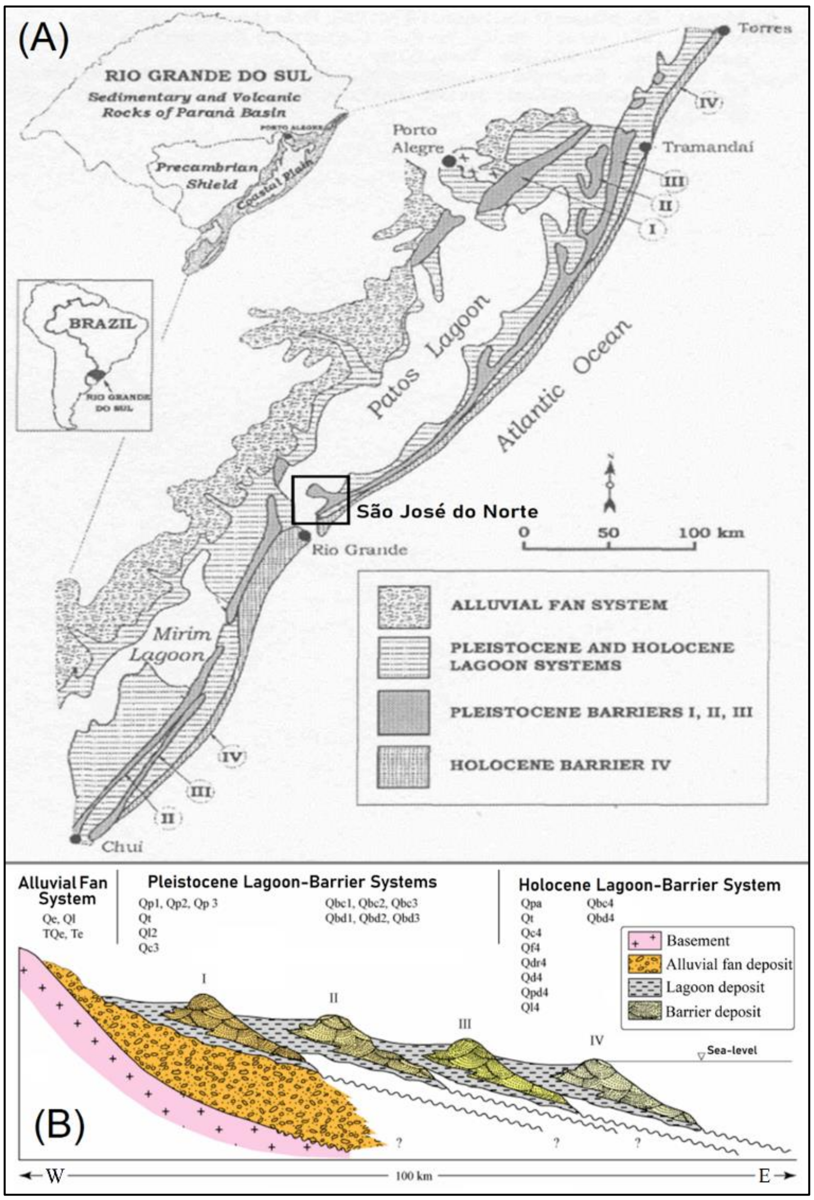

Figure 1. Geological maps of the coastal plains of São José do Norte (RS, Brazil). (A) and (B) maps were adapted from Tomazelli *et al.,* (2000) and Tomazelli and Villwock (2005), respectively.

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Data description

Companhia Riograndense de Saneamento collected the groundwater samples at sites with different population densities, land use and land cover (Figure 2). Lithological and hydrochemical data set were made available on Sistema de Informações de Águas Subterrâneas (SIAGAS, 2020) by Serviço Geológico do Brasil – CPRM (Geologic Service of Brazil). The data set consists of groundwater samples collected in 1992 (P562), 1993 (P563), 2005 (P403, P405, P556, P557, P558, P559, P561), 2007 (P195, P398, P399, P400) and 2010 (P005). These samples were collected from tubular wells managed for domestic supply and belong to the local water supply company (SIAGAS, 2020).

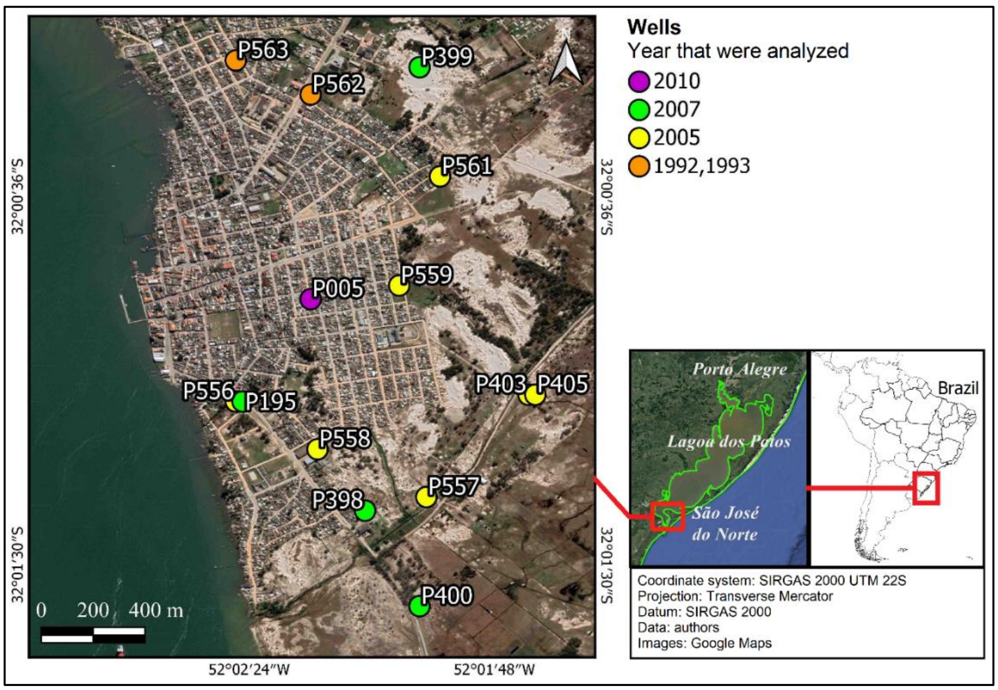

Figure 2. Location of groundwater samples collected from wells in São José do Norte (RS/Brazil).

Hydrochemical tools

Piper's triangular diagram classifies samples in different hydrochemical faces according to the predominant anion and cation (Piper, 1944). Diagram of Durov (1948) is similar to Piper, but total dissolved solids and pH values are plotted by a graphical adaptation. Stiff's radial diagram is a graph with horizontal lines cut by a vertical line, where each side of the horizontal line expresses the ionic concentration of different elements (Stiff, 1951). Schoeller linear diagram demonstrates

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the largest cations and anions on its horizontal axis and the concentration of each element analyzed on the vertical axis (Schoeller, 1956).

Agriculture and pasture areas occupy 14% and forestry 13% of the São José do Norte land (Projeto MapBiomas, 2020). Thus, the methodology to assess the groundwater suitability must be dynamic and objective, focusing on the data set analyze and understanding. In this way, the diagram developed by the United States Salinity Laboratory (USSL) was applied because it uses the sodium adsorption ratio (SAR) ($Na^+/(Ca^{2+} + Mg^+)$) and the electrical conductivity value to classify water irrigation (Richards, 1954). Data were plotted on the USSL diagram using Qualigraf program (Mobus, 2003). Geochemical data were plotted on the diagrams of Piper, Durov, Stiff, Schoeller using the Geochemist's Workbench software (Aqueous Solutions, USA).

The ions ratio is used to evaluate the hydrogeochemistry and changes in its hydrochemical behavior, such as those caused by marine intrusion or external influence (Hem, 1985). Milliequivalents (mEq/L) unity was applied in the ionic reasons. Custodio and Llamas (1983), FAO (1997), Bear *et al.,* (1999) and Appelo and Postma (2005) discuss some ionic reasons (r), such as: rNa^+/ rCl^-: 0.88 represents sea water, higher values, fresh water; $rCl^-/rHCO_3^-$: the high proportion indicates salt water; $rMg^{2+}/rCa^{+\,2}$: ratios above 5 represent sea water and below 5 indicate fresh water. Fresh water and the increase in this ratio over the years is an indicator of intrusion; and $(rCa^{2+} + rMg^{2+}) / (rHCO_3^- + rSO_4^{2-})$: ratio above 1 is an indication that marine intrusion is occurring.

Seawater Intrusion Groundwater Quality Index

Seawater has higher ionic content of many parameters, such as Cl^-, SO_4^{2-}, Na^+, Ca^{2+}, Mg^{2+}, than fresh water, enabling to distinguish the water type by hydrochemical behavior assessment (Custodio and Llamas, 1983; Hem, 1985; Bear *et al.,* 1999). Thus, the hydrochemical reactions may be deduced from the ratio of the conservative concentrations with those found in the samples. Considering that seawater is the only source of chloride for groundwater and the chloride concentration in seawater is 566 mmol/L, the fraction of seawater (*f*sea) can be found using Equation 1 (Appelo and Postma, 2005).

$$fsea = \frac{Cl\ concentration\ in\ the\ sample}{566} \quad \textbf{Equation 1}$$

Only 3% of saltwater content in the groundwater is sufficient for many consumers consider the water too salty to consume it and only the 5% saltwater content to render the water unfit for human consumption (Custodio and Bruggman, 1987). Thus, in this work, the *f*sea of 3% was considered the maximum ideal value and 5% as the maximum tolerable value of saltwater in the well samples.

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According to Tomaszkiewicz *et al.,* (2014), saltwater contribution in groundwater samples found in Piper diagram could also be obtained using the $GQI_{Piper(mix)}$ (Eq.2). These authors adapted the *f*sea equation of Appelo and Postma (2005) to analyze the fraction of saltwater in the groundwater as an index, developing the GQI_{fsea} (Eq. 3). Finally, Tomaszkiewicz *et al.,* (2014) joined $GQI_{Piper(mix)}$ and GQI_{fsea} and developed the Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) (Eq. 4). The GQI_{SWI} can be categorized from 0 to 100, subdivided into 4 types of water, as seen in Table 1.

$$GQI_{Piper(mix)} = \left[\frac{(Ca^{2+} + Mg^{2+})}{Total\ cations} + \frac{(HCO_3^-)}{Total\ anions}\right] x\ 50\ (meq/l) \qquad \textbf{Equation 2}$$

$$GQI_{fsea} = (1 - fsea) * 100 \qquad \textbf{Equation 3}$$

$$GQIswi = \frac{GQI_{fsea} + GQI_{Piper(mix)}}{2} \qquad \textbf{Equation 4}$$

Table 1. GQI_{SWI} classification of the groundwater sample according to the salinity.

Water type	GQI_{SWI}	
	Min	Max
Freshwater	75	100
Mixed groundwater	50	<75
Saline groundwater	10	<50
Seawater	0	<10

Groundwater Quality Index for human consumption ($GWQI_{HC}$)

Menezes (2009) developed the Groundwater Quality Index for human consumption ($GWQI_{HC}$) to be applied according to the legislations values, as water quality standards. In this way, $GWQI_{HC}$ algorithm has no weighting methods since they are subjective and can mask the groundwater quality assessment. This index uses Boolean Logic, which applies same importance to all parameters indexed.

Toxic parameters has high influence on the water quality, since even low concentrations can represent harm to human health, including organ disorders and carcinogenic risks (WHO, 2011). In this way, the $GWQI_{HC}$ algorithm was developed with two levels, not indexing or indexing toxic parameters. This index has also been developed with a classification approach with only positive values and one also with negative values if toxic parameters are indexed.

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In this study, no toxic parameters were analyzed, so the first level and the rating method with only positive values was employed. For the $GWQI_{HC}$ application, a disjunctive contingency table was developed and its three classes were filled with the selected water quality standards:

- >MAC: higher value than Maximum Acceptable Concentration—MAC;
- ≤ MAC-GV: value equal to or less than the Maximum Acceptable Concentration and higher than the Guide Value—GV; and
- ≤GV: value equal or less than to the GV.

Each indexed parameter applies the following functional logic to fill the contingency table: 1 for the class in which the concentration fits and 0 for the other classes. For example, if the concentration of an indexed parameter was above the maximum acceptable concentration (MAC), then the class "> MAC" for this parameter is filled in with 1 and the other two classes ("MAC-GV" and "≤GV") is filled with 0 (Table 2). Finally, the number of classifications in each class is added individually (total column in Table 3).

Table 2. Classification of the parameters concentration in the disjunctive contingency table.

Concentration		Classification		
		>MAC	MAC-GV	≤GV
If the concentration of the parameter was	above the MAC	1	0	0
	equal or less than MAC and greater than GV	0	1	0
	less than or equal to the GV	0	0	1

Table 3. Example of a complete disjunctive contingency table.

Well	Parameter X			Parameter Y			(...)	Total		
	>MAC	MAC-GV	≤GV	>MAC	MAC-GV	≤GV	...	>MAC	MAC-GV	≤GV
A	n_{11}	n_{12}	n_{13}	n_{11}	n_{12}	n_{13}	...	$\sum n_{11}$	$\sum n_{12}$	$\sum n_{13}$
B	n_{21}	n_{22}	n_{23}	n_{21}	n_{22}	n_{23}	...	$\sum n_{21}$	$\sum n_{22}$	$\sum n_{23}$
C	n_{31}	n_{32}	n_{33}	n_{31}	n_{32}	n_{33}	...	$\sum n_{31}$	$\sum n_{32}$	$\sum n_{33}$
.	.	.	.	.	.	.	.	.	.	.
.	.	.	.	.	.	.	.	.	.	.
.	.	.	.	.	.	.	.	.	.	.

Water quality standards from Brazil (Ministério da Saúde, 2017), European Union (CEC, 1980; CEU, 1998), Australia (NHMRC, 2011), USA (USEPA, 2009) and India (BIS, 2012) were compared and the most restrictive GV and MAC values were selected (Table 4). Applying the most restrictive values ensures that consumers have better water quality for drinking and prevents health problems.

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Table 4. GV and MAC values of water quality standards applied to the Groundwater Quality Index for human consumption ($GWQI_{HC}$).

Parameters	(mg/L)		Source	
	GV	MAC	GV	MAC
pH *in situ*	≥ 6.5- ≤ 8.5	< 6.5- > 8.5	B,C,D,E	B,C,D,E
Total dissolved solids	500	2000	D,E	E
Total hardness ($CaCO_3$)	≥ 60 - ≤200	<60 - >200	C	C
Electric conductivity (µS/cm)	≤ 2500	> 2500	F	F
Alkalinity	200	600	E	E
Ca^{2+}	75	200	E	E
Na^{+}	20	180	B	C
Fe^{2+}	0.05	0.2	B	B,F
Mg^{2+}	30	50	B,E	B
Cl^{-}	25	250	B	A,C,D,F
F^{-}	1	1.5	E	A,B,C,E,F
SO_4^{2-}	25	250	B	A,B,C,D E,F

A: Brazilian Ordinance n.5/2017 (Ministério da Saúde, 2017); B: European Directive 80/778/EEC (CEC, 1980); C: Standard of Australia (NHMRC, 2011); D: USA gov. (USEPA, 2009); E: Standard of India (BIS, 2012); F: European Directive 98/83/EC (CEU, 1998).

The $GWQI_{HC}$ values are ranked into 5 categories, from 0 to 100, as seen in Table 5. Excellent water quality comes from the "great" class, that mean that the concentration of all indexed parameter are under de Guide Value (GV) of the standard water quality. Groundwater under "good" class is a water that is not great for human drinking purpose because one or more parameter had the ionic content above de GV, but its content is below the Maximum Acceptable Concentration (MAC). Groundwater falling in the "Requires level 1 treatment" and "Requires level 2 treatment" classes indicated that it has one or more parameter above the MAC, so this water is not fit to drinking *in natura*. Finally, an "Improper" water has all indexed parameters above the MAC.

Table 5. Ranking of the Groundwater Quality Index for human consumption ($GWQI_{HC}$).

$GWQI_{HC}$ value	Water quality	Description
100	Great	All concentrations according to GV. Perfectly drinkable water.
87-99	Good	The concentration of one or more parameters is above the GV, but below the MAC. Water acceptable to drink.
24-86	Requires level 1 Treatment	It has one or more parameters with a concentration above the MAC. Water needs to be treated before it is drunk.
1-23	Requires level 2 Treatment	
0	Improper	All parameters are above the MAC. Water should not be drunk.

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Spatial distribution maps
Geographic Information System (GIS) is a computational system capable to visualize, create, analyze and edit spatial data and its geographic phenomenon. Therefore, GIS is widely applied in the field of geosciences, including hydrological and hydrogeological studies (Tsihrintzis *et al.,* 1996; Aladejana *et al.,* 2021; Horák-Terra *et al.,* 2019; Bertrand *et al.,* 2016). Spatial interpolation process is one of the most applied methods on GIS due its ability to construct values of environmental variables from sampled points to non-sampled areas (Burrough and McDonnell, 1998; Sangadi *et al.,* 2022; Sabino *et al.,* 2020). Interpolation changes the scale of analysis from local to a regional assessment, improving understanding of the studied phenomena. Inverse Distance Weighted (IDW) is an interpolation method that uses a weighting power influence related to the distance and resemblance of the surrounding point values, assuming that closest sampled points have greater similarity than the farthest ones.

Contour, also called isoline and isovalue, on maps is a line that connect points or sites with the same value. They always have the same interval value on a map, but maps has different interval according to the nature of the studied phenomenon. Contours are used to understand the spatial distribution, since the closer they are to each other, the greater the variation of the phenomenon's values in the geographic space. In the same way, remote contours indicate a space with little variation and homogeneous features.

The QGIS version 3.0.1 were used for data geoprocessing and mapping (QGIS.org, Open Source Geospatial Foundation Project). IDW method was applied in this study for the spatial distribution and mapping of *f*sea, GQI_{SWI} and $GWQI_{HC}$ values. The IDW weighting power influence applied was 2. Spatial distribution maps of the index values were created for all sampled years except for 2010, which has only one sample (P005).

Results and discussion

Aquifer hydrogeochemistry
Physical and chemical data of groundwater samples from São José do Norte (Rio Grande do Norte (RS)/Brazil) are seen in Table 6. Some samples (P403, P405, P561, P195, P005) did not have all parameter to be plotted on Piper, Durov, Stiff and Schoeller diagrams. Electrical conductivity (EC) values of P403 and P405 samples were estimated from the concentration of total dissolved solids (TDS), following the equation “TDS = EC x 0.55” proposed by Hem (1985).

Piper diagram revealed calcium bicarbonate was the predominant water type (Figure 2). A few samples was classified as sodium bicarbonate (P563 and P557) and sodium chloride (P562 and P399) type. In common, all samples indicated that sodium was an influential anion. This result was expected since the aquifer is in a depositional environment of lagoon, alluvial and delta

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(Tomazelli and Villwock, 2005) with high seawater influence. Durov diagram demonstrated that the pH values were mainly clustered between 7.6 and 8.2 and the TDS content was concentrated up to 250 mg/L (Figure 3). The P562 has the most unusual TDS content (2801.9 mg/L), almost 6 time greater the mean value (478.7 mg/L).

Table 6. Physical and chemical analysis of groundwater collected in São José do Norte (RS/Brazil). Chemical analysis and TDS in mg/L, EC in µS/cm. *No data available.

Well	pH	Hardness	Alkalinity	TDS	EC	Ca^{2+}	Mg^{2+}	Na	K	Fe^{2+}	HCO_3^-	SO_4^{2-}	Cl^-	F^-
P562	7.30	440	412	2801.9	4675	110.2	41.3	1039	16.2	0.0	502.6	123.9	1170	0.3
P563	7.80	216	234	518.1	865.7	52.5	20.7	98	20.5	0.1	285.5	0.7	140	0.4
P403	8.2	63	129	213	117.15	14	7	28	*	0.42	157	0.52	14	0.197
P405	8.40	75	297	1052	578.6	14	10	365	*	0.0	362	*	350	0.31
P556	7.60	76	86	168.1	1380	25.8	2.8	16	2.6	76	97.6	0.2	15	0.1
P557	8.10	63	110	212	272	20.4	2.9	31	9	0.1	117.1	1.3	14	0.1
P558	7.80	63	76	157.7	335	21.2	2.4	3.8	3.8	0.1	87.8	5.2	9	0.2
P559	7.90	66	101	182.5	533	19	4.7	21	8	0.1	123.2	0.4	15	0.5
P561	7.90	63	91	145	364	19	4	15	*	0.0	111	*	9	0.3
P195	8.10	222	177	356	638	66	14	*	*	*	216	49.9	50	0.08
P398	8.0	106	113	191	349	33	6	24	5	0.12	138	4.2	28	0.1
P399	8.30	61	1	180	267	20	3	30	7	0.83	109	0.98	23	0.2
P400	8.10	174	244	273	455	64	3	22	2.3	0.13	298	1.3	20	0.2
P005	7.6	73	135	252	284	20	6	22	*	0.0	165	0	13	0.3
Max	8.4	440	412	2801.9	4675	110.2	41.3	1039	20.5	76	502.6	123.9	1170	0.5
Min	7.3	61	1	145	117.15	14	2.4	3.8	2.3	0	87.8	0	9	0.08
Mean	7.9	125.8	157.6	478.7	793.8	35.7	9.1	131.9	8.3	6.0	197.8	15.7	133.6	0.2
SD	0.3	107.5	106.5	709.7	1161.1	27.9	10.6	288.9	6.3	21.0	122.1	36.8	312.1	0.1

Stiff diagrams demonstrated that among the cations (Na^++K^+, Ca^{2+} and Mg^{2+}), the predominant ions were Na^++K^+, especially the first one (Figure 4). The HCO_3^- was the highest concentration anion among Cl^-, $HCO_3^-$and SO_4^{2-}. Most of parameters had low standard deviation, such as pH, Ca^{2+}, Mg^{2+}, K^+, Fe^{2+}, SO_4^{2+} and F^-, revealing that groundwater samples were under similar geochemical influences (Figure 4). The P562 sample had more ionic content than the other samples, indicating the possibility of hydrogeochemical anomaly or external influence.

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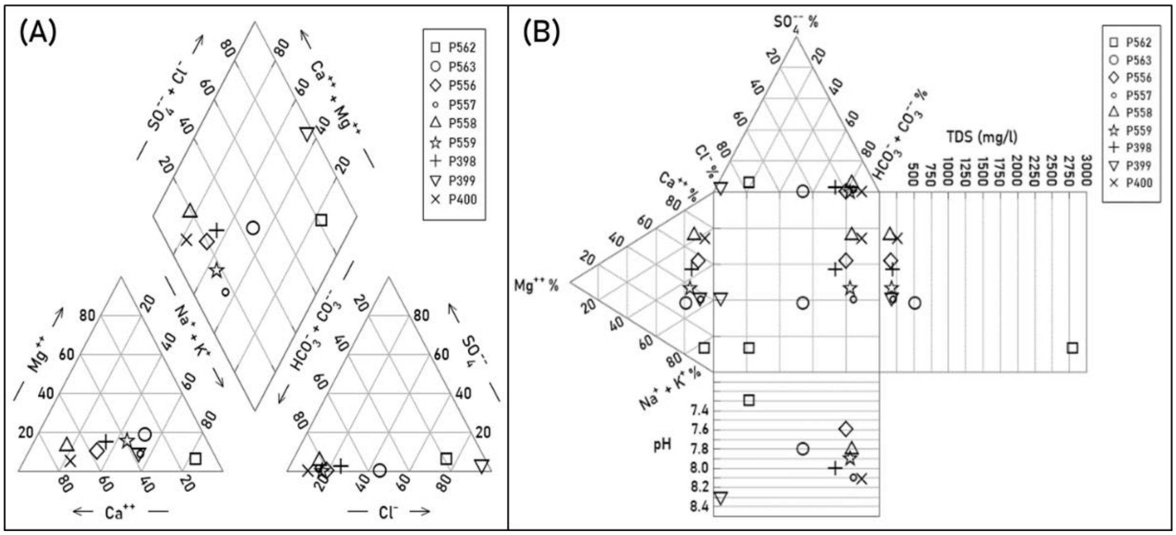

Figure 3. Ionic composition of groundwater samples from São José do Norte (RS/Brazil) in the Piper (A) and Durov (B) diagram.

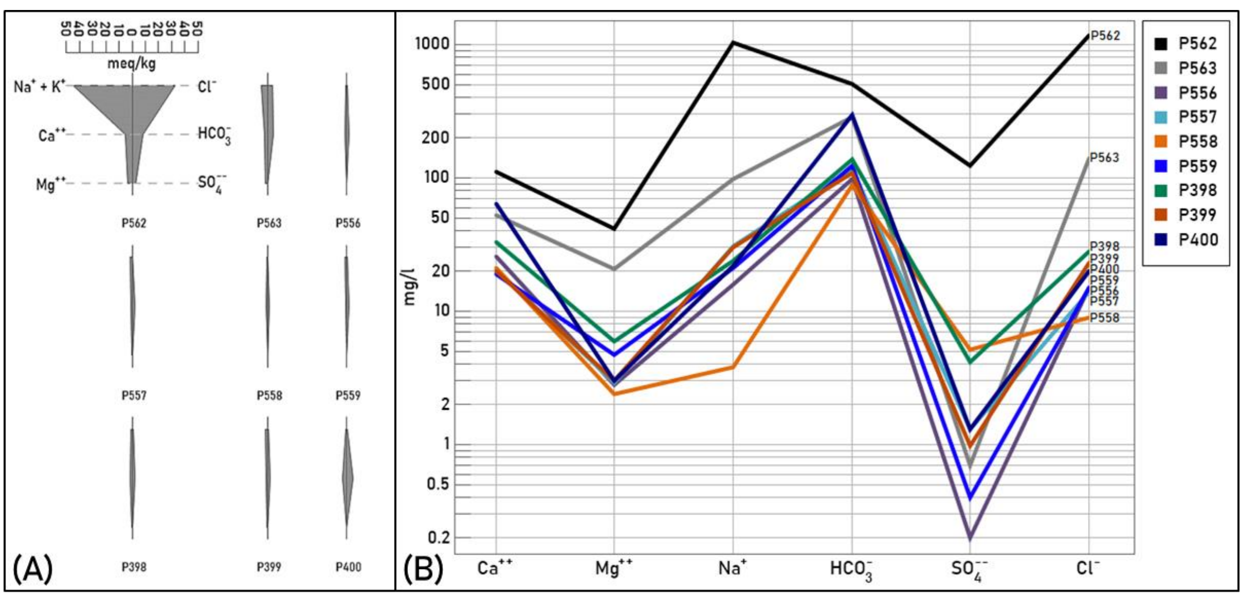

Figure 4. Stiff (A) and Schoeller (B) diagram of groundwater samples from São José do Norte (Brazil).

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Groundwater suitability for irrigation purpose
The USSL diagram classifies the water quality for irrigation according to the danger of salinity from C1 (low salinity) to C4 (very high salinity), and by the danger of sodification from S1 (low sodium concentration) to S4 (very high sodium concentration) (Cordeiro, 2001). The Qualigraf program adds another class to the USSL: S5 (extremely high sodium concentration). The P195 sample was not analyzed because it does not contain all ions data.

Samples were mostly categorized as C2-S1 (n = 8; 62%), that is medium salinity and low sodium water (Figure 5). Just class S1 describes 85% (n = 11) of all samples analyzed. The P562 sample was the only one classified as C4-S4, a high salinity and sodium content scenario, revealing again a unusual behavior. The P405 sample was qualified as C2 and S3, a water with medium salinity and high sodium content. However, spatially close to P405 sample, the P403 sample was classified as C1-S1, the best suitability for irrigation, since its low salinity and sodium concentration. In this way, the groundwater in the north of the town was less suitable and the water in the south the most fitting for irrigation.

Sampled wells were build mainly to human water supply. Still, this study approached water quality for irrigation because water use can be converted at any time and the town lacks qualitative studies, even though agriculture is an important financial source (Zabaleta, 1998).

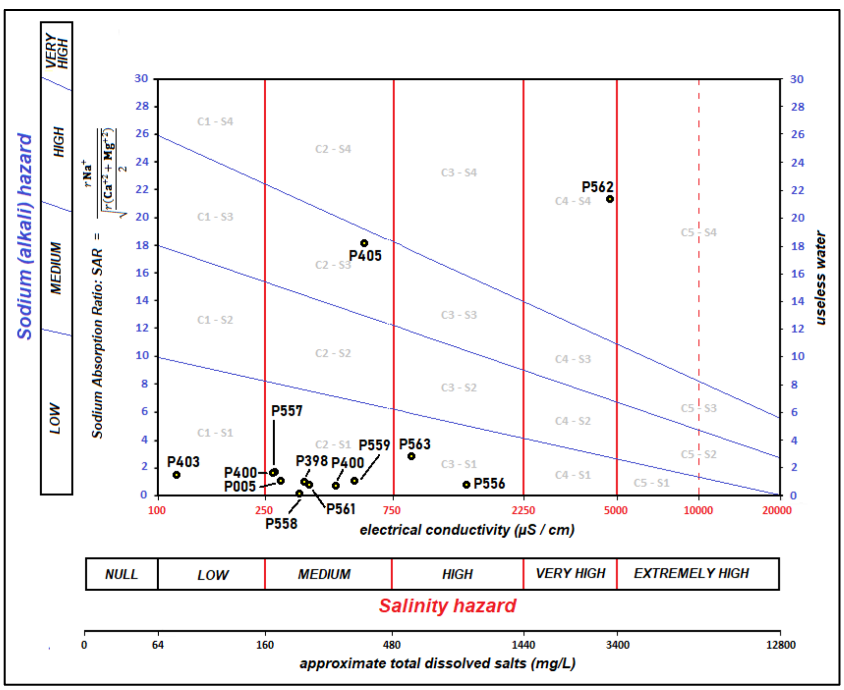

Figure 5. Water suitability classification for irrigation according to the USSL diagram.

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Salinity assessment

Most of the samples (n = 13; 93%) did not demonstrated a Na^+/Cl^- ratio of a salinization process, only indicated in P558 sample. The $rCl^-/rHCO_3^-$ ratios ranged between 0.1 and 1.7, but the 4.0 ratio of P562 sample stood out. Even so, all samples were classified as continental waters. The rMg^{2+}/rCa^{2+} ratios also ranked the samples as continental waters. The ratio of $(rCa^{2+} + rMg^{2+})/(rHCO_3^- + rSO_4^{2-})$ classified no sample as under salinization process. However, the values of 0.93 (from P563 sample), 0.95 (P556), 0.97 (P195) and 0.91 (P398) highlighted due to the high ratio, indicating that in future scenarios of environmental degradation, groundwater salinization may happen.

Samples collected in 2005 (P556, P557, P558) and in 2017 (P195, P398) had low ionic ratios increase over time, reinforcing that salinization was not occurring, instead initially may be indicating water freshening. All ionic ratios are shown in Table 7.

Table 7. Ionic ratios (mEq/L) of groundwater samples.

ID	rNa^+/rCl^-	$rCl^-/rHCO_3^-$	rMg^{2+}/rCa^{2+}	$(rCa^{2+}+rMg^{2+})/(rHCO_3^- + rSO_4^{2-})$
P562	1.4	4.0	0.6	0.83
P563	1.1	0.8	0.7	0.93
P403	3.1	0.2	0.8	0.50
P405	1.6	1.7	1.2	*
P556	1.6	0.3	0.2	0.95
P557	3.4	0.2	0.2	0.65
P558	0.7	0.2	0.2	0.81
P559	2.2	0.2	0.4	0.66
P561	2.6	0.1	0.4	*
P195	*	0.4	0.4	0.97
P398	1.3	0.3	0.3	0.91
P399	2.0	0.4	0.3	0.69
P400	1.7	0.1	0.1	0.70
P005	2.6	0.1	0.5	0.55

**No data.*

Fractions of seawater (*f*sea) in groundwater samples are seen in Table 8 and were spatialized according to the analysis year, as observed in Figure 6. In this figure, the 1992 and 1993 sample were grouped as 1990s' samples. Most of the sample (n= 13; 93%) had the *f*sea value in agreement to maximum tolerable (*f*sea < 5%) and maximum ideal value (*f*sea < 3%). Only P562 sample exceeded the maximum tolerable *f*sea value for human consumption, with *f*sea = 5.83%. Then the water was not palatable for the consumers.

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Table 8. Fraction of salt water (*f*sea) and GQI_{SWI} values of groundwater samples.

ID	*f*sea	GQI_{SWI} Value	GQI_{SWI} Class
P562	5.83%	55.86	Mixed groundwater
P563	0.70%	75.05	Freshwater
P403	0.07%	*	*
P405	1.74%	*	*
P556	0.07%	86.33	Freshwater
P557	0.07%	81.54	Freshwater
P558	0.04%	90.63	Freshwater
P559	0.07%	84.16	Freshwater
P561	0.04%	*	*
P195	0.25%	*	*
P398	0.14%	84.10	Freshwater
P399	0.11%	79.53	Freshwater
P400	0.10%	91.55	Freshwater
P005	0.06%	*	*

**No data.*

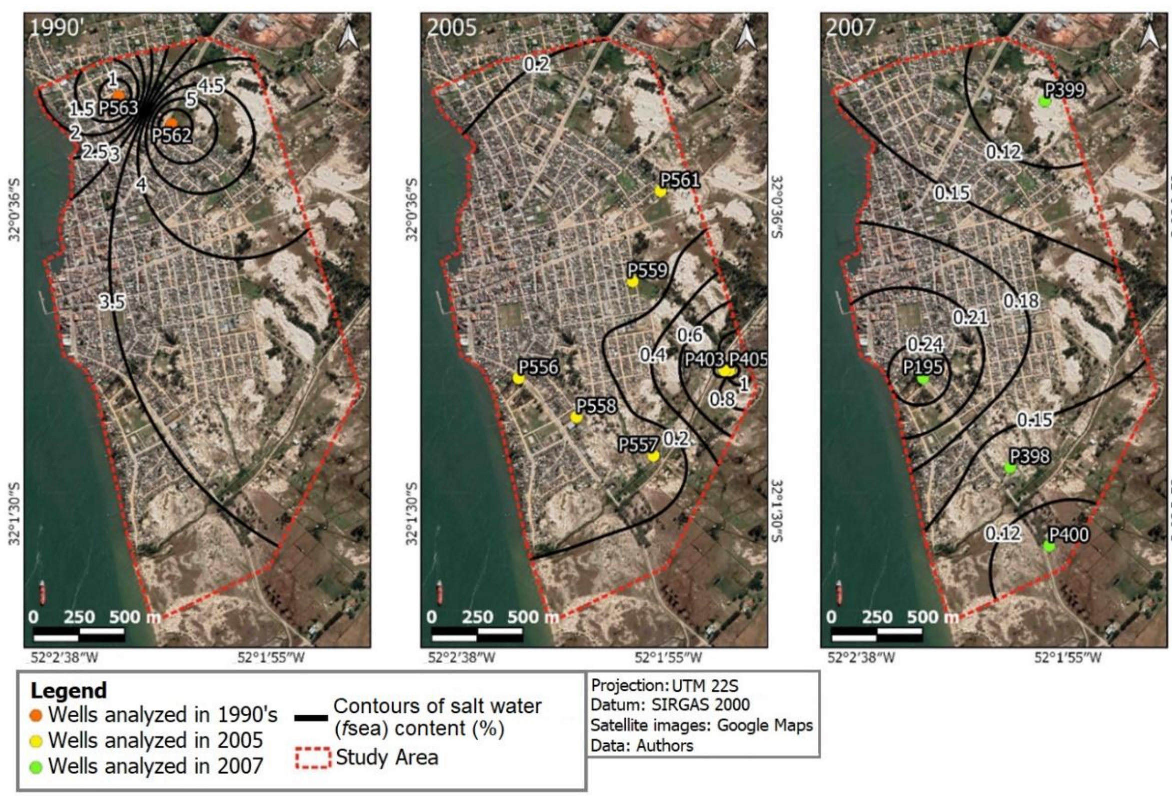

Figure 6. Geographic distribution of fractions of seawater (*f*sea) content (%) in the shallow aquifer of São José do Norte (RS / Brazil). Contour interval of 0.5, 0.2 and 0.3 point for the map from left to right, respectively.

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Groundwater in southern area did not performed considerable variations of *f*sea values over time, with a small downward trend in the southernmost part. In the northern region, despite one sample (P562) had high salinization, another two samples (P563 and P399) proved low *f*sea values. This scenario indicated that P562 values may be related to poor well conservation or external influences. A minimal increase in *f*sea values was detected between 2005 and 2007 in the central part of the study area. However, the 2010 *f*sea value (0.006%) indicated a sharp drop compared to 2007.

The P405 *f*sea was about twenty-five times greater than P403 even they were spatially close. P403 was collected in the wettest period (August) and P405 in the driest period (November) of 2005. Thus, drought and evapotranspiration may have concentrated the ions in the groundwater or the P405 sample well had external influence. In general, the *f*sea and ionic ratios results corroborated the hypothesis that, a priori, salinization was not occurring during the sampling period.

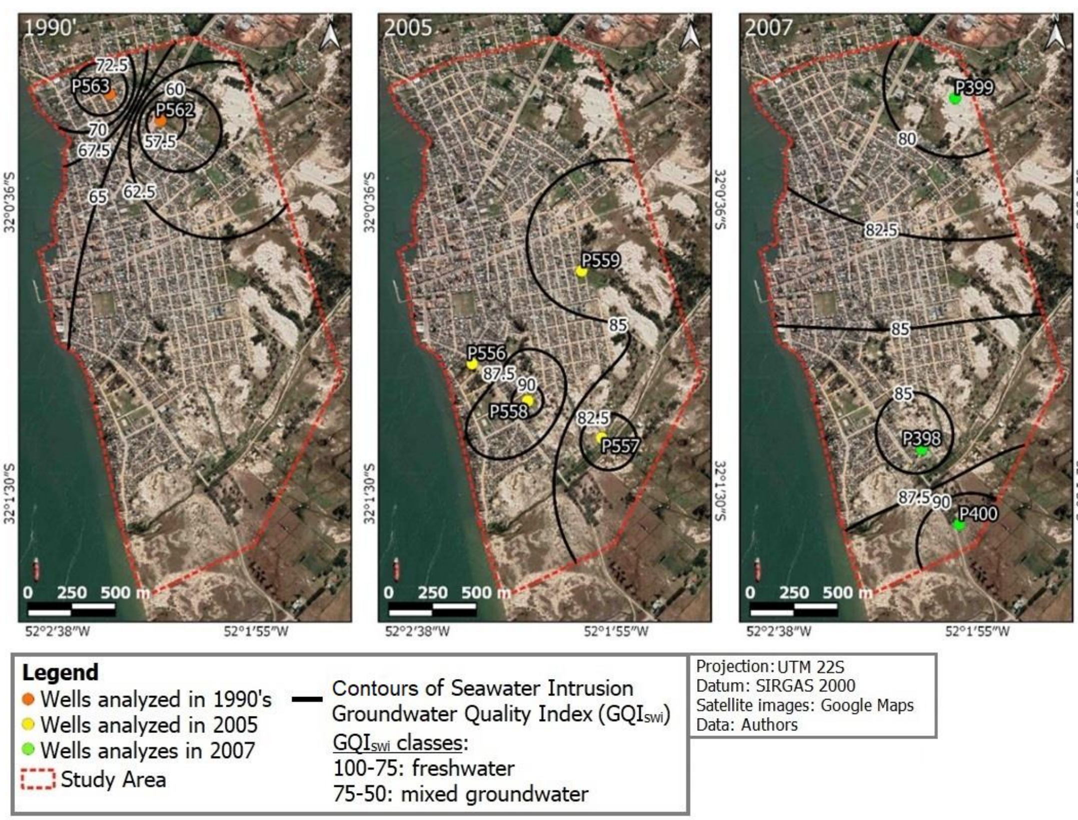

Figure 7. Spatial distribution of Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) values over the 1990's, 2005 and 2007 in São José do Norte aquifer (Brazil). Contour interval of 2.5 points for all maps.

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Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) classified all samples as “fresh water”, except P562 (Table 9). This sample was classified as “mixed groundwater”, indicating a mixture between different types of water. Even so, high GQI_{SWI} values close to the sea confirmed groundwater composition as freshwater and reinforces the no intrusion process hypothesis (Figure 7). In general, the saltier groundwater (lowest GQI_{SWI} values) was in the north of the town. The GQI_{SWI} displayed a tendency to freshening over the years.

Groundwater suitability for human drinking purpose

Disjunctive contingency table of the Groundwater Quality Index for human consumption ($GWQI_{HC}$) was filled and its classification is observed in the Table 9. The P561, P405 and P195 samples did not have all parameters indexed in the $GWQI_{HC}$, so they were not analyzed by this index. P005 sample parameters were assessed and discussed, but as it is the only one 2010 sample, spatial analyzes and maps were not performed. Sabino and Menezes (2019) performed an initial assessment of the groundwater quality for the 2005 samples. These authors considered different data set (another samples, parameters and analysis period). Considering all these changes, the qualitative assessments performed in this paper are distinct and more detailed.

The better $GWQI_{HC}$ classification (98) belonged to the P400 because it had no parameters above the Maximum Allowed Concentration (MAC) (0 in the "> MAC" column in Table 9). Thus, this samples was ranked as “good” for human drinking consumption. In this sample, only sodium and iron concentrations were between the maximum allowed and the guide value (class MAC-GV).

All the other samples had at least one parameter above the MAC (as seen in “>MAC” column in Table 9) and were ranked as “Needs Treatment Level 1”. Most of the samples were not fit for human drinking as they had only one parameter above the MAC. The worst $GWQI_{HC}$ rating (36) was recorded for P562 sample due to the high content of chloride, hardness, electrical conductivity, sodium and total dissolved solids.

Geographic distribution of $GWQI_{HC}$ illustrated that requires treatment groundwater is observed mainly in the northern area, as indicated by 1990’ and 2007 samples (Figure 8). The low quality in north was extra influenced by the P562 sample, which had the worst groundwater classification and unfitness for human consumption. More suitable groundwater is located in the southern area, especially in the 2007 when a sample was under the “good” class (P400).

Just in the 1990’ samples the parameters total hardness, chloride, electrical conductivity, sodium and TDS had high content (>MAC). Iron and alkalinity were higher than the MAC in other years. Almost half of the parameters indexed (pH, calcium, fluorite, magnesium and sulfate) was under the MAC in all samples, most of them according to VG.

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Table 9. Parameters concentration classes and groundwater ranking according to the Groundwater Quality Index for human consumption ($GWQI_{HC}$).

Well	>MAC	MAC-GV	≤GV	$GWQI_{HC}$	Category
P562	5	3	4	**36**	Requires level 1 Treatment
P563	1	4	7	**81**	Requires level 1 Treatment
P556	2	0	10	**73**	Requires level 1 Treatment
P557	1	2	9	**84**	Requires level 1 Treatment
P558	1	1	10	**85**	Requires level 1 Treatment
P559	1	2	9	**84**	Requires level 1 Treatment
P403	2	1	9	**71**	Requires level 1 Treatment
P398	1	3	8	**82**	Requires level 1 Treatment
P399	2	1	9	**71**	Requires level 1 Treatment
P400	0	2	10	**98**	Good
P401	1	2	9	**84**	Requires level 1 Treatment
P005	1	1	10	**85**	Requires level 1 Treatment

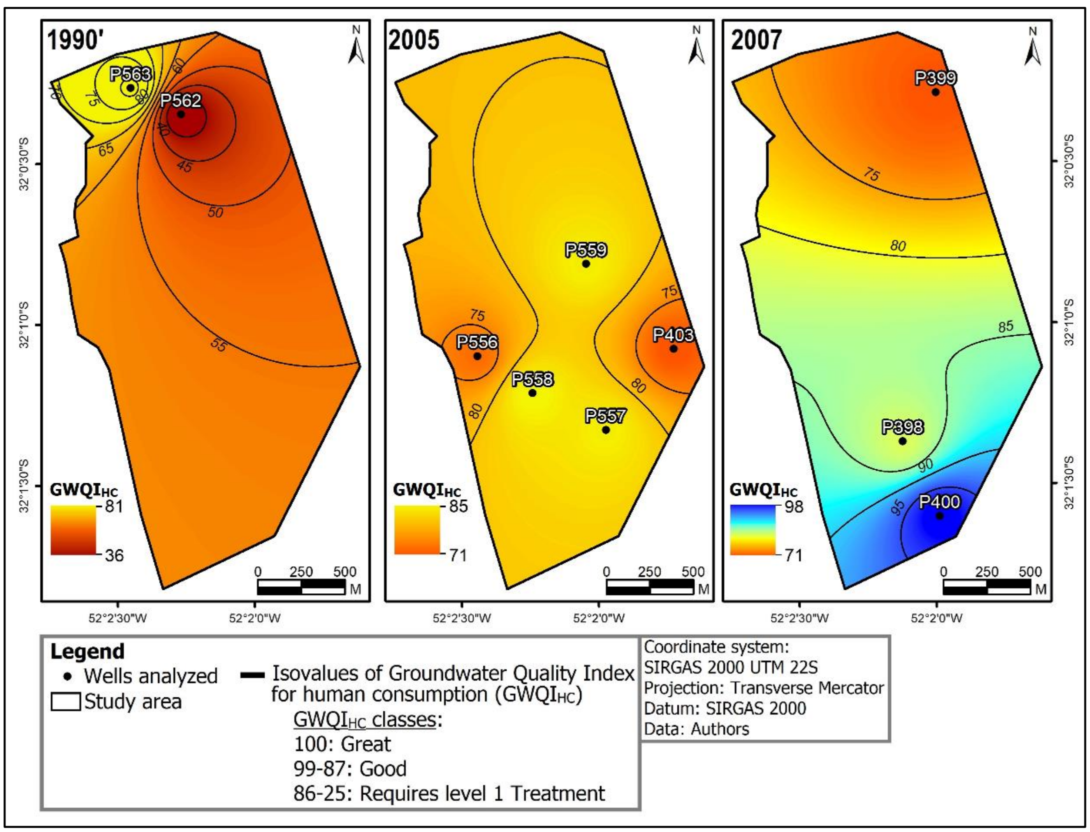

Figure 8. Spatial-temporal distribution of Groundwater Quality Index for human consumption ($GWQI_{HC}$) values in São José do Norte (RS/Brazil). Contour interval of 5 points.

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Alkalinity was the most frequent parameter above MAC (n= 9; 64%), followed by iron (n =3; 21%) and hardness (n= 2; 14%). Alkalinity is the ability of water solutes to neutralize acids and it is related to bicarbonate and carbonate content (Chapman and Kimstach, 1996). The main carbonate source for groundwater is the fraction of CO_2 gas in the atmosphere contained in the soil, biological production of carbon dioxide and biological reduction of sulfates and carbonate minerals (Hem, 1985; DNR-DW, 1996). Calcium and alkalinity content contributes to the stability and control of corrosivity in tubes and machines (WHO, 2011).

Iron concentration may be related to anthropogenic influence, such as sewage and oxidation of well liners and tubes (PNS, 2018). Many wells in the town region had not followed the construction technical standards and were close to several contamination sources (Reginato *et al*., 2008). Iron can create a bitter taste in the water, a redwish appearance and stains on porcelain tiles and fabrics (Swenson and Baldwin, 1965). Thus, the population may refuse to drink the water with high iron content.

High levels of hardness may be related to the lithology formed by layers of sandy and sandstones containing carbonate shells (SIAGAS, 2020). Shell is a calcium source for the environment, which in turn increases the hardness value (Santos, 2008). The high hardness value makes soap production difficult and may reduce the population's interest in drinking this water due to the unpleasant taste (PNS, 2018).

Potassium was not indexed to $GWQI_{HC}$ because not all samples had this element analyzed, as seen in Table 1. However, the potassium concentration draws attention. According to the European Union Water Quality Standard for human consumption (CEU, 1980), the GV for potassium is 10 mg/L and the MAC is 12 mg/L. The P562 and P563 samples had potassium concentration of 16.2 mg/L and 20.5 mg/ L, respectively. Three samples (P557, P559 and P399) demonstrated potassium levels below, but close to the GV limit. Possible sources of potassium in town groundwater are residential sewage and urban runoff (PNS 2018; Chapman and Kimstach, 1996). The higher potassium concentration produces a salty taste in the water (PNS, 2018). People with diabetes, heart diseases as hypertension, kidney diseases and hyperkalaemia should be careful before drinking this water. (PNS, 2018; WHO, 2011).

High electrical conductivity and chloride, sodium, sulfate, potassium can initially indicate salinization and saltwater intrusion (Custodio and Llamas, 1983; Spechler, 1994; Almeida and Silva Júnior, 2007; PNS, 2018). Some samples had chloride and sodium content classified in the MAC-GV class, indicating that the water suitability for consumption has been influenced, but this class is not an intrusion indicator. Only in the P562 sample the electrical conductivity, chloride, sodium, sulfate, potassium demonstrated high concentration for human drinking purposes, but not as higher or close to the seawater contents.

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Conclusion

Chemical analysis, ionic ratios, equation of the fraction of saltwater in the groundwater (*f*sea) and the Seawater Intrusion Groundwater Quality Index (GQI_{SWI}) proved different yet complementary results. These tools' results were in line with themselves, which reflects the consistency of the data set and methodology. These tools quickly categorized the groundwater samples according to its hydrogeochemistry and salinity level.

The GQI_{SWI} is a newly developed instrument that had great potential to assist this salinization study. Groundwater Quality Index for human consumption ($GWQI_{HC}$) compiled several physical-chemical data and water quality standards values into a single value. This streamlined understanding of the groundwater suitability for human drinking purpose. Results of $GWQI_{HC}$ demonstrated that groundwater was unfit for human consumption, requiring treatment measures for human supply. Groundwater was regular for irrigation purpose.

Mapping of *f*sea, GQI_{SWI} and $GWQI_{HC}$ values enabled the understanding of its distribution in the geographic space and allowed the association with natural and anthropogenic influences. Salinization process was not detected during the sampling period in the free coastal aquifer by the methodology applied in this study.

It is recommended that future studies observe the possible effects of climate seasonality on hydrogeochemical behavior and the presence of human external influences, such as runoff and poor well conservation, as influencers on hydrogeochemistry. As a way to complement the current study, it is suggested that other salinization detection methodologies be employed, especially those applied *in situ* through the seasons. Continuous hydrogeochemical monitoring including more wells mainly in the northern region is indicated to assist in the shallow aquifer management.

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