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Original Articles

Assessment of soil salinity status under different land-use conditions in the semiarid region of Northeastern Brazil

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A R T I C L E I N F O

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ABSTRACT

Soil salinization and sodification, caused by inadequate land management, is one of the main threats to the semiarid agroecosystems. It is essential to investigate saline levels under different land-use conditions to maintain the sustainability of agricultural production. The main objective of this study was to diagnose the salinity status of the soil in different land-use conditions in the semiarid region of Northeast Brazil. Soil samples were collected in the surface layer (0-5 cm) of three different land-use conditions: areas of low salinity (native vegetation - Caatinga), areas of different saline levels (cultivated areas), and areas of very high salinity (desertified by salinity), and compared regarding the chemical attributes of the soils by descriptive and multivariate analysis. The results showed that sodium and chloride were the predominant soluble ions in cultivated and desertified areas due to saline waters commonly used in the study region. Increases in electrical conductivity values of 1219 and 23207% were observed for cultivated and desertified areas, respectively, compared to areas of native vegetation. The principal component analysis contributed to identifying parameters responsible for the variation in soil salinity, and the second factor identified that percentage of exchangeable sodium, electrical conductivity, and exchangeable and soluble sodium plays an essential role in soil desertification. However, calcium, magnesium, sodium, and chloride ions correlated positively with variables that degrade soils - EC, ESP, and SAR. We also found that in desertified areas, in addition to chloride, sulfate is also present in expressive levels, and due to the strong associations with calcium, magnesium, and sodium, our study suggests that in addition to the free forms of these ions, the forms of chlorides and sulfates of calcium, magnesium, and sodium also effectively contribute to the degradation of the areas. The combination of multivariate analysis imesgeochemistry \times laboratory techniques was a valuable tool to identify and monitor saline levels in converting uncultivated to cultivated areas and from cultivated areas to desertified areas.

1. Introduction

Currently, soil salinity is considered one of the main threats to the sustainability of agricultural soils and consequently to food security in semiarid regions of the globe (Shrivastava and Kumar, 2015). Regarding the expressiveness of saline areas globally, estimates indicate that approximately 6% of soils worldwide are affected by salinity, of which 54% are affected by sodicity. (Khaime et al., 2018), moreover, almost

20% of the irrigated land (45 million ha) is already salinized (Shrivastava and Kumar, 2015). In addition, soil salinity is considered the second main type of soil degradation after soil erosion (Zaman et al., 2018) and causes annual economic damage of at least US\$27 billion to agricultural production worldwide (Qadir et al., 2014).

The main natural factors contributing to soil salinization (primary salinization) are climate, natural drainage, topography, relief position, geology, source material, and distance from the sea (Akça et al., 2020).

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Concerning anthropogenic factors (secondary salinization), inadequate irrigation practices, insufficient drainage, and poor land management have contributed enormously to accelerating the process of soil salinization (Zhou et al., 2013). However, primary and secondary salinization continue to increase (Liu et al., 2019) at an annual rate of approximately 10% (Machado and Serralheiro, 2017). It is estimated that>50% of arable land will be salinized by 2050 (Abdelaziz et al., 2019), resulting in a drastic reduction in soil fertility, vegetation cover, and biodiversity (Farifteh et al., 2006; Gorji et al., 2017), impacting the ecological functions of the soil (Gorji et al., 2020), leading to its degradation and consequent desertification (Peng et al., 2019).

In Brazil, inadequate irrigation management through the water with high saline content and lack of drainage has contributed to soil salinization. Consequently, saline areas are found mainly in irrigated perimeters located in semiarid regions of Northeastern Brazil (Pessoa et al., 2016). However, despite being a highly expressive problem in these regions, studies addressing the salinity diagnosis in Brazil's soils on a watershed scale are rare or non-existent. Most reports are local or at maximum on irrigated perimeter scale (Freire et al., 2014). Given this context, it is crucial to assess and investigate the salinity status of soils in Brazil, as intensive local soil monitoring is of great relevance for areas subject to high environmental impacts (Pereira et al., 2019), such as those located in the irrigated perimeters of the Brazilian semiarid region, as they present a high risk of salinization and sodification (Castro et al., 2019).

The survey of soil salinity status can provide crucial quantitative information for land management and restoration (Peng et al., 2019) and contribute to the sustainable development of the ecological environment (Liu et al., 2019). However, monitoring soil salinity is difficult due to its spatial and temporal variability, which is strongly influenced by management practices (irrigation, drainage), climate, and the salinity and depth of the water table (Akramkhanov et al., 2011). For this

purpose, the multivariate analysis of saline data can be a helpful tool because this technique can simplify the multivariate dataset (e.g., ionic composition) without a substantial loss of information (Mora et al., 2017). Among the classes of multivariate analysis that can be used for environmental analyses, cluster analysis can be used to separate groups of soils with similar saline and sodic levels (Freire et al., 2014), while principal component analysis can be used to classify observations along axes of variation that represent intrinsic/environmental-derived gradients (Mora et al., 2017).

As soil salinity is also affected by high levels of soluble elements (Carmona et al., 2010; Groenewegen, 1961), often reaching toxic levels in plants (Rengasamy, 2016; Tavakkoli et al., 2010), it is essential to evaluate not only the contents of these elements in the soluble phase of the soil but also the interactions between them, to understand the geochemistry of salinity. However, knowledge of the dynamics and prediction of the species of formed salts is limited. In this work, we aim to investigate (1) soil salinity levels using descriptive statistics, (2) evaluate the main contributing variables to soil salinity and the similarity of saline areas, and (3) identify the main salt species prevalent in the soils in different land-use conditions in the semiarid region of northeastern Brazil – areas of native vegetation, cultivated and desertified by salinity.

2. Material and methods

2.1. Study area

The study area comprises four critical watersheds located in the semiarid region of Pernambuco, Northeastern Brazil – Brígida, Terra Nova, Pajeú, and Moxotó. Fig. 1 shows the location map of the study area and the location of the collected sampling sites for each land-use condition. These watersheds cover 45,000 km² and consist of shallow



Fig. 1. Spatial distribution of the topsoil sampling sites in the four studied watersheds located in the semiarid region of Pernambuco, Northeast Brazil - areas of native vegetation (blue), cultivated (black), and desertified by salinization (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

soils, where high salinity levels are more frequent in Cambisols and Fluvisols located in irrigated areas (Pessoa et al., 2019). The predominant vegetation in the study region is the Caatinga shrubland, a dry tropical forest that covers much of Northeastern Brazil (de Albuquerque et al., 2012).

The main types of cultivation practiced in the region are corn, beans, vegetables, fruits, and pastures, both cultivated in dryland and irrigated areas. In addition, this region is characterized by its climate with welldefined dry and rainy seasons, with average annual temperatures ranging from 23 to 27 °C. The region's average rainfall is low, around 600 mm annually, and the critical dry months occur between October and December. Precipitation is spatially and temporally variable, and the occurrence of multiple years of drought is common (Sampaio, 1995). In the Brígida, Pajeú, and Moxotó watersheds, the Integrated Drought Index varies from moderate to normal; as for the Terra Nova watershed, this Index varies from weak to normal (Cemaden, 2021). The Integrated Drought Index combines the Precipitation Index, the Soil Available Water, the Vegetation Water Supply Index, and the Vegetation Health Index (Cemaden, 2021). It means that the three watersheds - Brígida, Pajeú, and Moxotó - are more prone to soil salinization due to their susceptibility to drought, implying that the driest soils promote the rise of the salts due to the absence of soil moisture.

Irrigated agriculture in Brazil's semiarid region began in the late 1960 s (dos Santos et al., 2011), with the installation of irrigated perimeters designed by the National Department of Works Against Drought - DNOCS, and already in the 1980 s, it was possible to observe the degradation of soils due to salinization (Eschemback et al., 2014). Until the present day, it is common in the study region to use well water for irrigation, which has high concentrations of salts and sodium in their composition and continues to degrade the soils (Fernandes et al., 2015).

There are no records of saline areas in the study region where the native vegetation – the Caatinga vegetation – has not yet been disturbed. Among the cultivated areas, the soils' salinity levels vary significantly between the cultivations and the management of the soil and water adopted. There are also areas where salinity levels are so high that there is no vegetation cover on the land, and these areas are currently desertified and abandoned.

2.2. Soil sampling

Soil samples were collected from 78 sampling sites (Fig. 1), randomly selected in three different land-use conditions: areas of native vegetation (with Caatinga vegetation domain – low salinity), cultivated areas (with agricultural production in irrigated perimeters – different levels of salinity) and desertified areas (degraded due to high levels of salinity) (Fig. 2).

Each sampling site was approximately 400 m². Within each site, 15 to 20 single soil samples were collected from the topsoil (0 – 5 cm deep), which were mixed to form a composite sample representative of the sampling site. This sampling was carried out in the region's dry period, between October and December, when there was a greater concentration of salts on the soil surface.

2.3. Soil analysis

All collected soil samples were air-dried and sieved through a 2 mm mesh. Soil samples were analyzed for pH, exchangeable cations (Ca^{2+} , Mg^{2+} , Na^+ , and K^+), cation exchange capacity (CEC), organic carbon, and soil texture.

Soil pH was measured directly from a 1:2.5 soil:water mixture using a pH meter. After prewashing the salts, exchangeable cations were extracted with ammonium acetate solution $(1 \text{ mol } \text{L}^{-1})$ at pH 7. Ca²⁺ and Mg²⁺ were determined by atomic absorption spectrometry (USSL, 1954), and Na⁺ and K⁺ were determined by flame emission photometry (USSL, 1954). CEC was determined by extraction with sodium acetate (1 mol L⁻¹) at pH 8.2 and measured by flame emission photometry (USSL,



Fig. 2. Land-use conditions at the sampling sites in the study region: (a) area of native vegetation – low salinity; (b) cultivated area – different levels of salinity; (c) desertified area – by the high degree of degradation due to salinity.

1954). The organic carbon content was determined by the methodology proposed by Walkley and Black (1934).

Sand, silt, and clay contents were determined by the pipette method after chemical dispersion with NaOH (1 mol L-1) (Ruiz, 2005), and based on the textural analysis, large textural variability of the collected samples was found (0–5 cm). However, the prevailing textural class was Sandy Loam (46.15% of the total) (Fig. 3). Finally, the exchangeable sodium percentage (ESP) was calculated by the ratio of exchangeable Na⁺ with CEC, according to the equation (USSL, 1954):

 $\mathrm{ESP}\left(\%\right)=\mathrm{Na^{+}/\mathrm{CEC}}\times100.$

2.4. Soil solution analysis

The soil solution was obtained from the saturated paste extract. For



Fig. 3. Diagram of the textural classes of the topsoil samples collected at the sampling sites: areas of native vegetation (blue), cultivated (green), and desertified by salinization (red). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

this, distilled water was added to approximately 500 g of soil passed through a 2 mm mesh sieve with agitation until reaching a condition of complete saturation (USSL, 1954). After 24 h of contact, the soil solution (paste extract) was extracted by suction.

In the soil solution, we measured the concentrations of major cations $(Ca^{2+}, Mg^{2+}, Na^+, and K^+)$ and anions $(Cl^{-}, HCO_3^{-}, CO_3^{2-}, and SO_4^{2-})$. Ca^{2+} and Mg^{2+} were determined by atomic absorption spectrometry, and Na^+ and K^+ were determined by flame emission photometry (USSL, 1954). Cl^- was determined by titration with AgNO₃ solution, HCO_3^- and CO_3^{2-} determined by titration with H^2SO^{4-} solution, and SO_4^{2-} was determined by gravimetry with BaCl₂ (USSL, 1954).

Additionally, pH and electrical conductivity (EC) at 25 °C were determined, measured directly by a potentiometer and conductivity meter, respectively. The values of the sodium adsorption ratio were calculated according to the contents of soluble elements, using the following equation (USSL, 1954):

SAR $(mmol_{c}L^{-1})^{0.5} = Na^{+} / [(Ca^{2+} + Mg^{2+})/2].$

2.5. Data analysis

Descriptive statistical analysis was performed to assess the dispersion of the studied variables. Data variability was obtained according to the criteria proposed by Souza et al. (2017) – low (CV < 12%), medium (12% \leq CV \leq 62%), and high (CV > 62%) variability. Data normality was assessed using the Kolmogorov-Smirnov test (Test-KS).

Principal component analysis (PC) was performed to reduce the studied variables for each land-use condition. The data were scaled through standardization, performed as a pre-processing step on the data. To facilitate the interpretation of each component, Varimax Rotation was applied. Additionally, we considered as many principal components as those with a correlation coefficient value between the principal component and the variable above 0.6 in module.

This study used correlation analysis to determine which ions are more positively or negatively associated with saline variables. Pearson's correlation matrix was used to assess the correlations between the studied elements (exchangeable and soluble) and the saline variables (pH, ESP, EC, and SAR). Cluster analysis (CA) was applied to identify similarities between the saline characteristics (pH, ESP, EC, and SAR) of the sites studied in each saline scenery. CA was formulated according to the Ward-algorithmic method, and the squared Euclidean distance was used to measure the distance between groups with similar saline properties. All the descriptive and multivariate analysis were done in the SPSS statistical package (IBM SPSS Statistics for Windows, Version 19.0).

2.6. Geochemical speciation modelling

We used Visual Mintew 3.1 software (Gustafsson, 2011) to assess the speciation of soluble elements. The concentrations of elements in the soil solution (Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , HCO_3^- , CO_3^- and SO_4^-), as well as the pH, were used to calculate the mass distribution of cation and anion species via modeling, for the sampling sites in areas of native vegetation, cultivated and desertified by salinization. We intended to report the most common ion species in the soil solution and how they were changed as a function of soil used in the semiarid region of study.

3. Results and discussion

3.1. Soil chemical properties

The study variable variability was expected since land-use changes impacted the chemical properties of semiarid soils (Martins et al., 2019; Seifu et al., 2020). The evaluated soil chemical properties showed normal distribution, ensuring the results' reliability and eliminating the need for data transformation. In general, the chemical properties of the soil sorptive complex presented CV ranging from medium (12% < CV <62%) to high (>62%), except for soil pH in the areas of native and cultivated vegetation, which presented low CV for this variable (<12%) (Table 1). The soil pH values, the contents of exchangeable cations Ca^{2+} , Mg^{2+} , K^+ and Na^+ , and the ESP were higher in the following order: desertified areas > cultivated areas > areas of native vegetation. These variables were increased, respectively, in the magnitude of 4, 171, 25, 56, 171, and 52% when comparing the areas of native vegetation and cultivated (Table 1). However, when comparing the areas of native vegetation \times desert areas, the increases for these variables were in the magnitude of 8, 322, 113, 60, 909, and 185%, respectively.

Soil pH values were very close among the different land uses, but there was an increase in cultivated and desertified areas – 7.53 and 7.80, respectively. Additionally, by the maximum pH values, alkaline sites were found in the three land-use conditions. Increases in soil pH or alkalinity occur due to an imbalance in the ionic composition resulting from excess sodium (Zhang et al., 2020), as well as due to the presence of carbonate ions, bicarbonates, and hydroxide compounds, which contribute to the increase in pH (Sunkari et al., 2021).

In the soil sorptive complex, the quantitative analysis of the studied elements showed that the abundance of the main cations followed the order $Ca^{2+} > Mg^{2+} > K^+ > Na^+$ for the native vegetation areas, and $Ca^{2+} > Mg^{2+} > Na^+ > K^+$ for the cultivated and desertified areas. Moharana et al. (2019) claim that Ca^{2+} and Mg^{2+} are the main cations present in semiarid soils, corroborating the results verified in our study. However, this order varies widely in semiarid soils according to the management adopted (Moharana et al., 2017).

Regarding organic carbon, reductions of 17.72 and 25.95% were observed in cultivated and desertified areas, respectively, when compared to areas of native vegetation (Table 1). The absence of vegetation cover in desertified areas contributed to the reduction in OC levels. In cultivated areas, intense cultivation and soil disturbance were the main factors contributing to the reduction of organic carbon contents due to the rapid mineralization of soil organic matter. As the organic matter increases the soil's buffering capacity against rapid changes in soil salinity (Darwish et al., 2005), sampling sites in cultivated areas where farmers do not apply organic matter to the soil are more vulnerable to salinization and soil sodification processes.

In the soil solution (paste saturation extract), it was possible to

Table 1

Descriptive statistics of the chemical attributes of the topsoil samples collected at the different sampling sites - sorptive complex.

Sampling sites (n)	Soil attribute	Soil attribute									
	pH	Ca ²⁺	Mg^{2+}	\mathbf{K}^+	Na ⁺	ESP (%)	OC (g kg $^{-1}$)				
		cm	ol _c kg ⁻¹								
Native vegetation areas (19))										
Mean	7.22	2.36	1.47	0.30	0.21	4.30	1.58				
Min	6.40	0.17	0.04	0.08	0.02	0.96	0.26				
Max	9.10	6.24	4.53	0.48	0.55	11.98	4.51				
SD	0.85	2.02	1.59	0.13	0.14	3.47	1.06				
CV (%)	11.79	85.51	108.31	41.67	65.46	80.84	66.95				
Cultivated areas (41)											
Mean	7.53	6.40	1.85	0.47	0.57	6.57	1.30				
Min	5.70	0.14	0.03	0.07	0.04	0.76	0.50				
Max	9.70	28.92	7.93	3.50	7.98	42.33	2.77				
SD	0.76	5.61	1.79	0.55	1.26	9.19	0.68				
CV (%)	10.12	87.75	96.97	116.81	221.31	139.84	52.75				
Desertified areas (18)											
Mean	7.80	9.97	3.14	0.48	2.12	12.27	1.17				
Min	6.50	1.78	0.37	0.09	0.17	1.08	0.36				
Max	10.70	25.57	8.87	1.20	7.80	35.44	2.44				
SD	1.08	5.71	2.37	0.30	2.08	9.74	0.62				
CV (%)	13.84	57.28	75.56	61.24	98.11	79.39	53.01				

*ESP = Exchangeable Sodium Percentage; OC = Organic Carbon.

observe that among the chemical properties studied, only the pH in the areas of native and cultivated vegetation had a low CV (<12%) (Table 2). For the other variables, CV ranged from medium (12% \leq CV \leq 62%) to high (>62%).

The EC values, the contents of the soluble elements Ca^{2+} , Mg^{2+} , K^+ , Na^+ , Cl^- , $CO_3^2^-$, HCO_3^- , $SO_4^2^-$ and SAR were higher in the following order: desertified areas > cultivated areas > areas of native vegetation (Table 2). In the soil solution, these variables increased by 1219, 1525, 343, 277, 8168, 1497, 2, 139, 304, and 1157% in cultivated areas compared to native areas (Table 2). In desertified areas, the increases for these variables were 23207, 18094, 5425, 7160, 88343, 16710, 124 499, 8180, and 13764%, respectively, in relation to areas of native vegetation (Table 2).

Among the soluble ions, the quantitative analysis of the elements

showed the following orders for the studied cations: $Mg^{2+} > Na^+ > Ca^{2+} > K^+$ for the native vegetation areas, and $Na^+ > Mg^{2+} > Ca^{2+} > K^+$ for the cultivated and desertified areas. For the studied anions, the quantitative order was $Cl^- > HCO_3^- > SO_4^{2-} > CO_3^{2-}$ for the areas of native vegetation and cultivated, and $Cl^- > SO_4^{2-} > HCO_3^- > CO_3^{2-}$ for the desertified areas. The predominance of Na^+ and Cl^- in the soil solution of cultivated and desertified areas reflects the low-quality water used in the region, which contains high levels of these elements in its composition (Fernandes et al., 2015). High levels of HCO_3^- verified in cultivated and desertified areas were due to the increase in the partial pressure of CO_2 in these areas, which reacts with water to form HCO_3^- .

The increase in soil salinity (evaluated in this study by EC) verified in cultivated and desertified areas was due to the increase in the cationic (Ca^{2+} , Mg^{2+} , K^+ , and Na^+) and anionic (Cl^- , CO_3^{2-} , HCO_3^- , and SO_4^{2-})

Table 2

Descriptive statistics of the chemical	attributes of the topsoil sam	ples collected at the different san	npling sites – saturation extract
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Sampling sites (n)	Soil attribute										
	pH	EC	Ca ²⁺	Mg^{2+}	\mathbf{K}^+	Na ⁺	Cl ⁻	CO_{3}^{2-}	HCO_3^-	SO ₄ ²⁻	SAR
		$(dS m^{-1})$			mmolc L ⁻¹						(mmolc L ⁻¹)
Native vegetation areas	(19)										
Mean	7.39	0.26	0.93	4.13	0.35	1.22	5.68	0.00	2.96	0.61	0.98
Min	6.60	0.10	0.03	0.10	0.00	0.00	1.00	0.00	1.33	0.04	0.00
Max	7.90	0.37	8.11	7.39	0.78	3.00	12.50	0.00	9.33	7.50	2.86
SD	0.36	0.08	1.83	2.75	0.23	0.60	2.68	_	1.97	1.72	0.68
CV (%)	4.91	31.90	196.10	66.62	67.39	49.37	47.11	-	66.53	284.36	69.27
Cultivated areas (41)											
Mean	7.48	3.43	15.12	18.46	1.32	100.87	90.71	0.02	7.08	2.47	12.32
Min	6.40	0.10	0.09	0.31	0.02	0.29	0.50	0.00	1.33	0.04	0.16
Max	8.30	24.46	228.72	206.67	10.46	1837.12	1425.50	1.00	20.67	20.74	214.09
SD	0.45	5.92	38.27	40.06	1.95	349.60	255.44	0.16	4.94	4.61	37.51
CV (%)	6.01	172.47	253.08	217.09	147.60	346.58	281.59	640.31	69.78	186.28	304.52
Desertified areas (18)											
Mean	7.25	60.60	169.21	228.22	25.41	1079.01	954.86	1.24	17.74	50.51	135.87
Min	5.80	35.07	7.86	1.50	0.93	49.61	60.00	0.00	2.00	13.17	3.69
Max	10.20	100.18	625.00	430.67	207.38	2633.75	1775.00	13.57	93.00	195.56	671.56
SD	0.97	21.32	169.34	165.54	48.89	706.77	410.04	3.59	20.12	52.17	173.47
CV (%)	13.37	35.18	100.08	72.54	192.37	65.50	42.94	290.46	113.39	103.29	127.68

*EC = Electrical Conductivity; SAR = Sodium Adsorption Ratio.

composition of the soil solution. The excess of ions in the soil solution contributes to increasing the ionic strength of the soil solution and its electrical conductivity (Black and Campbell, 1982). It occurs due to chemical inputs, such as fertilizers and correctives, into irrigated agriculture with no drainage, accumulating these ions in the entire soil profile (Liu et al., 2020). In irrigated perimeters in Brazil, the presence of irrigated lands without installing drainage systems is a common practice, contributing to the increase in soil salinization.

The soil sodicity, evaluated in this study by ESP in the sorting complex and SAR in the soil solution, increased in cultivated and desertified areas due to the considerable increase in exchangeable and soluble Na⁺ levels. The Na⁺ from the irrigation water passes first to the soil solution, increasing the soluble Na⁺ contents. With the increase in the content of this element in the soil solution, its passage to the sorting complex occurs, also increasing the levels of exchangeable Na⁺ by replacing Ca²⁺ and Mg²⁺ in the exchange sites (Robbins, 1984). The increase in exchangeable Na⁺, and consequently in ESP, contribute to physically deteriorating the soil, mainly by affecting its structure and permeability (Qadir et al., 2006).

The low levels of exchangeable and soluble ions, as well as the low values of the saline variables - EC, ESP, and SAR, verified in this study for areas of native vegetation, reinforce that the anthropogenic factor is the leading cause of soil degradation by salinization in the Brazilian semiarid region since the natural contents of the elements found in undisturbed areas were considered adequate.

3.2. Multivariate exploratory analysis of soil properties

Several studies in the literature have applied PCA to evaluate saltaffected soils (Acosta et al., 2011; Moharana et al., 2019; Sunkari et al., 2021), and the findings of this study show that it was possible to establish associations between the studied variables, within each landuse condition. Fig. 4 shows the results of the analysis of principal components (PCs) of the different studied land-use conditions.

Two main components were enough to explain 63.19, 68.01, and 69.41% of the data variability in areas of native vegetation, cultivated and desertified by soil salinization, respectively. In areas of native vegetation, the first principal component (PC1) represented 43.17% of the total variance of the data set (Fig. 4a) and showed positive correlations with the following saturation extract variables: pH, Na⁺, SO₄⁻ and SAR (Table 3). However, negative correlations were verified for the OC and CO_3^{2-} variables. In the second component (PC2), which explained 20.02% of the data variation (Fig. 4a), positive correlations were observed for Ca²⁺ and HCO₃ in the saturation extract, while exchangeable Mg²⁺ presented a negative correlation (Table 3). Changes in the correlation patterns for PC1 and PC2 are related to the changes in the soil composition – soil solution and exchangeable complex - verified in each land-use condition (Table 1 and 2).

In cultivated areas, PC1 explained 45.71% of the data variability (Fig. 4b) and seems to represent an "anthropogenic component" as it is strongly related to soil attributes, mainly in the soil solution (paste saturation extract), which are easily altered according to the adopted management: exchangeable Na⁺, EC, Ca²⁺, Mg²⁺, Na⁺, Cl⁻ (soluble) and SAR (Table 3).

Previous studies have shown that EC, Ca^{2+} , Mg^{2+} , Na^+ , Cl^{-} (soluble), and SAR are rapidly altered in semiarid soils according to land-use change (Yu et al., 2018), especially when subjected to saline irrigation. In our study, this was verified by the increase in the levels of these ions in cultivated areas compared to native vegetation areas (Table 1). In PC2, with 22.30% of the total variation (Fig. 4b), negative correlations with K⁺ (exchangeable and soluble) were verified. Our findings corroborate those obtained by Moharana et al. (2019), who suggest PC1 as the soil properties component that contributes to soil salinization in cultivated areas.

Concerning desertified areas, PC1 explained 43.90% of the total data variation (Fig. 4c) and demonstrated a negative correlation with pH (of



Fig. 4. Exploration of variables by Principal Component Analysis (PCA). The plot of PC1 - PC2 loadings for topsoil samples collected at sampling sites from (A) areas of native vegetation, (B) cultivated areas, and (C) desertified areas by salinization.

Table 3

Varimax rotated principal component loading for the two first components.

Soil attribute	Native vegetation areas		Cultivated areas		Desertified areas		
	PC1	PC2	PC1	PC2	PC1	PC2	
Exchangeable complex							
pH	-0.5196	0.5110	0.5359	-0.0116	-0.8942	-0.0317	
Ca ²⁺	-0.5224	-0.5627	0.4259	-0.4305	0.2741	-0.2868	
Mg ²⁺	-0.4269	-0.6356	0.4782	-0.4267	0.2467	0.3180	
Na ⁺	-0.3834	-0.1970	0.8408	0.2198	-0.1207	0.6728	
K^+	-0.4582	-0.4952	0.0015	-0.7927	0.0638	0.4496	
ESP	0.2978	0.4903	0.4069	0.5643	-0.3817	0.6888	
OC	-0.6309	-0.3279	-0.1784	-0.4724	0.4612	-0.0788	
Saturation extract							
pН	0.6424	-0.0034	-0.2657	0.0889	-0.8696	-0.2812	
EC	-0.0152	-0.5757	0.8136	-0.1846	-0.0685	0.6737	
Ca ²⁺	-0.4036	0.6330	0.7752	-0.0622	0.5114	-0.0276	
Mg ²⁺	-0.5332	0.4533	0.8492	-0.0537	0.5437	-0.0980	
Na ⁺	0.7002	-0.0756	0.9336	0.1451	-0.4640	0.6914	
K^+	-0.1348	0.4893	0.2489	-0.6325	-0.8604	-0.2602	
Cl ⁻	-0.0678	0.3065	0.9536	0.1391	0.0335	0.4712	
CO_{3}^{2-}	-0.6396	0.0903	-0.0365	0.1477	-0.9157	-0.2293	
HCO_3^-	-0.2863	0.7043	0.3630	-0.2504	-0.8103	-0.3861	
SO ₄ ²⁻	0.7113	-0.0917	0.0793	-0.1434	-0.3946	0.2153	
SAR	0.6424	-0.0034	0.8877	0.1809	-0.9291	0.1897	

*PC = Principal Component; ESP = Exchangeable Sodium Percentage; OC = Organic Carbon; EC = Electrical Conductivity; SAR = Sodium Adsorption Ratio.

the soil and the saturation extract), K^+ , CO_3^{2-} , HCO_3^- , and SAR (Table 4). PC2 explained 25.5% of the total variation (Fig. 4c) and showed positive correlations with ESP, EC, and exchangeable and soluble Na⁺ (Table 3). This means that this last component represents variables that are more closely involved in desertifying soils through salinization and sodification.

To assess the degree of association between the saline variables and the ions studied, we used Pearson's correlation coefficient (Table 4), and the results found indicate that the pH and the saline variables EC, ESP, and SAR do not correlate in a similar way for the different studied areas. In our study, desertified areas had higher soil pH values (Table 1), but they did not significantly correlate with any exchangeable cation. We attribute this to high correlations with carbonate anions, which contribute to increasing soil pH (Zhang et al., 2018).

Through Pearson's correlation, it was possible to verify that in the areas of native vegetation, there were no positive correlations for the saline variables EC and ESP. Only SAR showed a high positive correlation with soluble Na⁺. ESP obtained negative and moderate correlations with the Mg^{2+} and K⁺ (exchangeable) and soluble ions Mg^{2+} and Cl⁻, while EC did not significantly correlate. These results showed that in areas of native vegetation, the contents of exchangeable and soluble elements, considered adequate (Table 2), are not able to increase the EC and ESP of the soils, which means that in a natural condition, these ions are not directly responsible for the increase in EC and ESP, and consequently do not cause soil degradation by salts or exchangeable sodium.

In the investigated cultivated areas, it is possible to observe that the saline variables already presented stronger correlations with the other studied variables (Table 4). ESP had moderate and positive correlations with exchangeable Na⁺ and soluble Na⁺, and Cl⁻. EC obtained positive and moderate correlations with the exchangeable cations Ca²⁺, Mg²⁺, Na⁺, and high and positive relationships with the soluble ions Ca²⁺, Mg²⁺, Mg²⁺, Na⁺ and Cl⁻. SAR had a high positive correlation with exchangeable Na⁺ ions and soluble Na⁺ and Cl⁻ ions.

Table 4

Pearson's correlation coefficients between ions $ imes$ saline variables in the non-cultivated, cultivated, and desertified areas	Pearson's corre	elation coefficients	between ions \times sali	ne variables in the	non-cultivated, cu	ltivated, and	desertified areas.
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Saline variable	Exchangea	ble cations			Soluble io	ns						
	Ca ²⁺	${\rm Mg}^{2+}$	Na ⁺	K ⁺	Ca ²⁺	Mg^{2+}	Na ⁺	\mathbf{K}^+	Cl	CO_{3}^{2-}	HCO_3^-	SO ₄ ²⁻
Native vegetation a	ireas											
рН	-0.05	-0.04	-0.04	0.11	0.50*	0.53*	-0.26	0.09	0.37	_	0.57*	0.45
ESP	-0.30	-0.52^{*}	0.31	-0.65^{**}	-0.02	-0.56*	0.02	0.22	-0.56*	-	-0.27	-0.01
EC	0.42	0.11	-0.02	0.43	-0.28	-0.43	0.03	0.16	-0.28	_	0.21	-0.39
SAR	-0.24	-0.20	-0.22	-0.12	-0.11	-0.33	0.97^{**}	-0.31	-0.06	-	-0.26	-0.05
Cultivated areas												
рН	0.05	0.15	0.56**	0.10	0.25	0.54*	0.51^{**}	0.26	0.57^{**}	0.18	0.59**	0.13
ESP	-0.23	-0.11	0.66**	-0.20	-0.17	0.20	0.62^{**}	0.04	0.54**	0.02	0.08	0.08
EC	0.53^{**}	0.58^{**}	0.57**	-0.01	0.71^{**}	0.83^{**}	0.67^{**}	0.19	0.68^{**}	0.08	0.26	0.23
SAR	-0.20	-0.32	0.89^{**}	-0.07	-0.38	-0.31	0.98**	-0.18	0.93^{**}	-0.05	0.25	-0.10
Desertified areas												
рН	-0.13	-0.03	0.24	-0.11	-0.50*	-0.43	0.24	0.72^{**}	0.19	0.83^{**}	0.80^{**}	0.55*
ESP	-0.43	-0.17	0.73^{**}	-0.07	-0.35	-0.39	0.89^{**}	-0.02	0.12	0.09	-0.01	0.25
EC	-0.18	0.22	0.82^{**}	0.61^{**}	0.02	0.08	0.67**	0.02	0.74**	-0.02	-0.10	0.64**
SAR	-0.25	-0.27	0.06	0.13	-0.39	-0.55*	0.63**	0.35	0.12	0.85**	0.63**	0.66**

 * Significant correlation to p < 0.05; ** Significant correlation to p < 0.01; ESP = Exchangeable Sodium Percentage; EC = Electrical Conductivity; SAR = Sodium Adsorption Ratio.

Moderate and negative correlations for SAR were verified with Ca^{2+} and Mg^{2+} ions. These findings suggest that Ca^{2+} , Mg^{2+} , Na^+ , and Cl^- ions require attention and must be rigorously monitored in irrigated agriculture in the study region, as these elements are already found at much higher levels than in native areas (Tables 1 and 2) and are correlated with variables that degrade soils - EC, ESP and SAR (Table 4), indicating that increases in the contents of these elements can lead to soil degradation.

In desertified areas, ESP obtained a high positive correlation with exchangeable and soluble Na⁺ (Table 4), which presented considerably high levels in these areas (Table 1). EC had moderate and high correlations with exchangeable Na^+ and K^+ ions and soluble Na^+ , Cl^- , and SO₄²⁻ ions. SAR obtained moderate and high positive correlations with the soluble ions Na^+ , CO_3^{2-} , HCO_3^- , and SO_4^{2-} . This indicates that high levels of these elements, as verified in this study for desertified areas (Tables 1 and 2), in addition to salinizing and sodifying the soils, by increasing the saline variables - EC, ESP, and SAR, in very high levels these ions also lead to land desertification. Additionally, as the Na⁺ cation and the Cl^{-} and SO_{4}^{2-} anions were the dominant ions in the soil solution in the desertified areas, we can attribute that these ions were responsible for the increases in EC and SAR. Previous studies in the literature corroborate our results and attribute the increases in EC and SAR to these ions as they are widely related (Chi and Wang, 2010; Yu et al., 2018).

Positive correlations between saline variables (EC, ESP, and SAR) and ions have been reported in previous studies (Buscaroli and Zannoni, 2017; Mahajan et al., 2020). According to Mahajan et al. (2021), when the source of salinity is saline water, cations such as Na⁺ and Mg²⁺ dominate and are better correlated with EC. Our findings indicate that Na⁺ and Cl⁻ will dominate and govern the relationships with saline variables in soil salinization by irrigation with saline water. On the other hand, when soil salinization occurs due to excessive fertilization, in addition to these ions, Ca²⁺, Mg²⁺, K⁺, CO₃²⁻, HCO₃, and SO₄²⁻ will dominate and govern the relationships.

Cluster analysis is a valuable tool for classifying or separating groups so that the degree of association is substantial among members of the same group and weak among members of different groups (Brogueira and Cabeçadas, 2006). In the present study, cluster analysis was applied over a standardized dataset to understand the similarities and dissimilarities between the chemical attributes of the studied sites. The dendrogram obtained by the hierarchical cluster analysis is shown in Fig. 5 and shows the dissimilarity for four groups of the studied sites based on the chemical attributes (saline variables – pH, EC, ESP, and SAR) of the investigated soils.

It is essential to mention that soil texture is an important factor to be taken into account in the salinization of areas since it affects the hydrology and dynamics of salts in the soil. The sandy loam and loamy sand textural classes had the three studied land-use conditions in our research. Although the sampling sites are located in the semiarid region of Brazil, there are differences not only in the soil texture but also in climate, topography, soil management, type of irrigation, and cultivation, which cause differences in the intensity of soil salinization. However, small-scale studies in areas with the same topography, climatic condition, and soil texture have shown that soil texture strongly correlates with the accumulation and distribution of salts in the soil profile (Hu et al., 2011).

Groups I and IV include degraded areas that differ mainly due to the different types of salts present. The management of study sites included in these two groups should prioritize recovery strategies. Group II includes sites in desertified areas and cultivated areas approaching desertification. Desertified sites in this group already require remediation, and management of cultivated sites must carefully avoid soil desertification. For this, one approach that can be adopted is the use of salt-tolerant perennial plants (Zhou et al., 2013). However, it is essential to stop the use of saline groundwater for irrigation and chemical inputs to reduce the input of salts into the soil. It will affect several small farmers in the region where the groundwater is the only water available for irrigation. On the other hand, crop production is already threatened at those sites due to high levels of EC, ESP, or SAR. Thus, low-cost desalination technologies for irrigation systems must be prioritized for those sites, avoiding them reaching desertification.

Group III includes native vegetation sites and cultivated areas. The similarity between these sites occurs because these cultivated sites have little exploration time, are constituted by more sandy soils, and have not



Fig. 5. Dendrogram obtained by the cluster analysis (Ward's method) of the studied variables for the sites sampling from the native vegetation areas, cultivated and desertified areas (NC: Non-Cultivated Areas – Caatinga vegetation; CA: Cultivated Areas; D: Degraded Areas).

yet presented a high accumulation of soluble salts or exchangeable sodium. The management of these cultivated sites must be carefully focused to avoid increasing the salt content to interfere with plant development and harm the soil structure. For this, saline water in irrigated agriculture must be avoided. It is also important to introduce intercropping of main crops with halophytes to reduce soil surface evapotranspiration and prevent salinization (Wang et al., 2008). Furthermore, the introduction of agroforestry – planting shrub trees on farms – is a promising option for salinity control (Zouari et al., 2019), and its effectiveness for these study sites needs to be investigated.

3.3. Ionic speciation of soil solution

In native and cultivated vegetation areas, Ca^{2+} and Mg^{2+} cations occur predominantly in their free forms (Table 5). In desertified areas, free forms of Ca^{2+} and Mg^{2+} occurred in smaller proportions and were reduced to 50.84 and 42.2%, respectively. However, it is possible to evidence that these cations are strongly associated with the Cl⁻ anion, and the species of CaCl⁺ and MgCl⁺ constituted 38.66 and 50.87%, respectively, of the total dissolved in the solution of soils extracted from areas desertified by salinity. CaSO₄ (aq) and MgSO₄ (aq) species were present in smaller amounts – 10.31 and 6.80% - in the soil solution of desertified areas, but these values were increased in relation to native and cultivated areas.

The free form of Na⁺ is present in negligible amounts in areas of native vegetation – 0.15% (Table 5). However, this is the predominant form of this ion in cultivated and desertified areas, with 97.45 and 85.01%, respectively. This increase can be explained by irrigation with water containing high levels of Na⁺, which increase its content in the soil solution (Table 2). In addition, the NaCl (aq) species is present in

Table 5

Mass distribution of the soluble cations and anions in differ	rent species.
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Component	Species name	% of the total concentration					
		Native areas	Cultivated	Desertified			
Ca^{2+}	Ca ²⁺	95.42	90.64	50.84			
	$CaCl^+$	0.83	6.40	38.66			
	CaSO ₄ (aq)	3.74	2.94	10.31			
	$CaHCO_3^+$	-	-	0.16			
	CaCO ₃ (aq)	_	-	0.01			
Mg^{2+}	Mg ²⁺	95.69	87.88	42.20			
	MgCl ⁺	1.32	9.83	50.87			
	MgSO ₄ (aq)	2.98	2.26	6.80			
	$MgHCO_3^+$	-	-	0.10			
Na ⁺	Na ⁺	0.15	97.45	85.01			
	NaHCO ₃ (aq)	-		0.011			
	NaCl (aq)	0.22	2.31	13.84			
	NaSO ₄	0.15	0.23	1.13			
K^+	K^+	99.58	97.38	84.74			
	KCl (aq)	0.22	2.30	13.80			
	KSO ₄	0.19	0.30	1.45			
C1 ⁻	Cl	98.84	94.32	64.97			
	CaCl ⁺	0.13	1.06	6.85			
	MgCl ⁺	0.96	2.00	12.16			
	KCl (aq)	0.01	0.03	0.36			
	NaCl (aq)	0.04	2.56	15.64			
$CO_3^{2-}/$	CO_{3}^{2-}	-	0.26	0.05			
HCO_3^-	NaHCO ₃ (aq)	-	2.24	9.95			
	$Mg_2CO_3^{2+}$	-	0.03	0.50			
	HCO_3^-	-	82.54	41.33			
	H ₂ CO ₃ (aq)	-	4.48	3.27			
	MgCO ₃ (aq)	-	0.37	0.84			
	$MgHCO_3^+$	-	4.51	19.92			
	$CaHCO_3^+$	-	4.73	22.10			
	CaCO ₃ (aq)	-	0.63	1.50			
	NaCO ₃	-	0.16	0.49			
SO_4^{2-}	SO_4^{2-}	73.68	55.10	9.82			
	MgSO ₄ (aq)	20.18	16.93	30.73			
	CaSO ₄ (aq)	5.70	18.01	34.55			
	NaSO ₄	0.30	9.779	24.16			
	KSO ₄	0.11	0.165	0.73			

considerable amounts in the soil solution of desertified areas – 13.84%. For K⁺, its free form is the one that predominates in the soil solution in the studied areas (84.74 - 99.58%). However, in desertified areas, there was a considerable presence of the species KCl (aq) – 13.80%, possibly due to KCl, which is the main potassium fertilizer used in the region.

Although desertified areas have higher Ca^{2+} and Mg^{2+} in the soil solution (Table 2), the significant reduction in their free forms contributes to sodium having greater activity in these areas, as in addition to very high levels, Na^+ occurs predominantly free in the solution of desertified soils. Furthermore, in these areas, Na^+ significantly contributes to raising the EC, ESP, and SAR of the soils (Table 4). On the other hand, in cultivated areas, although Na^+ also occurs predominantly in the free form, Ca^{2+} and Mg^{2+} also predominate in their free forms and tend to inhibit the activity of the Na^+ ion.

Among the anions, Cl⁻ occurred predominantly in the free form, in native and cultivated vegetation areas. Despite occurring freely in areas of native vegetation, the contents of this element were very low in these areas (Table 2) and did not contribute to degrading the soils in a natural condition, as it does not correlate with any saline variable (Table 4). In cultivated areas, this represents a risk, as Cl⁻ contributes to soil degradation; in addition to the high levels found (Table 2), it is closely correlated with saline variables (Table 4) in these areas, and this implies care with the use of irrigation waters containing high levels of this element, mainly due to its high solubility (Acosta et al., 2011), which can quickly reach toxic levels to plants. There were greater associations with Ca²⁺, Mg²⁺, and Na⁺ in desertified areas, suggesting their presence in the forms of CaCl⁺, MgCl⁺, and NaCl. However, as Na⁺ contents are much higher than those of Ca^{2+} and Mg^{2+} in the solution of desertified soils (Table 2), this means that Cl⁻ single and NaCl are the main contributors to the total chloride concentration in the soils.

Regarding carbonate anions ($\rm CO_3^{2-}$ and $\rm HCO_3^{-}$), it was found that it occurs mainly in the free form in cultivated areas. In desertified areas, in addition to the free form, there are also strong associations with Ca²⁺ and Mg²⁺ in soil solution. This association indicates that the formation of MgHCO_3⁺ and CaHCO_3⁺ in the solution of desertified soils contributes to removing Ca²⁺ and Mg²⁺ from the soil solution (Moharana et al., 2019), increasing the activity of the Na⁺ ion and the consequent sodicity of the soil.

The SO₄²⁻ ion predominated in free form in areas of native vegetation. However, it is strongly associated with Ca²⁺, Mg²⁺, and Na⁺ in cultivated and desertified areas. This revealed that the forms of CaSO₄, MgSO₄, and NaSO₄² are the predominant ones in these areas. However, the high saturation with Na⁺ (Table 2) suggests that NaSO₄² is the main sulfate species formed in desertified and cultivated areas.

4. Conclusions

Through this study, we identified that the contents of elements (exchangeable and soluble) increase significantly due to irrigated agriculture in the region, which requires careful soil and water management strategies to avoid degradation. Ca^{2+} , Mg^{2+} , Na^+ , and Cl^- are the dominant elements in the soil solution of cultivated and desertified areas.

 Na^+ and Cl^- ions were identified as the main contributors to soil desertification due to salinity, and the high levels of these elements verified in cultivated areas reflect the potential and existing risk of degradation in these areas. We also found that in desertified areas, in addition to Cl^- , SO_4^2 anion is also present in expressive levels, and due to the strong associations with Ca^{2+} , Mg^{2+} , and Na^+ , our study suggests that, in addition to the free forms of these ions, the forms of chlorides and sulfates of calcium, magnesium, and sodium also effectively contribute to the degradation of the areas.

Cluster analysis was effective in identifying cultivated sites that are prone to desertification. This is attributed to the unsustainable agricultural practices adopted in the study region, which do not consider the pedological conditions of agricultural suitability of the soils, climatic conditions of the region, use of saline water, absence of drainage systems, excess of chemical inputs such as fertilizers and correctives. In other areas that showed similarities with areas of native vegetation, mainly due to the short period of agricultural exploration, measures and strategies for mitigation and mitigation of salinity are urgently required to maintain the sustainable production of soils.

CRediT authorship contribution statement

Luiz G.M. Pessoa: Conceptualization, Methodology, Investigation, Writing – original draft. Maria B.G. dos S. Freire: Conceptualization, Resources, Supervision. Colleen H.M. Green: Investigation, Writing – review & editing. Márcio F.A. Miranda: Methodology, Formal analysis. José C. de A. Filho: Writing – review & editing, Visualization. Wagner R.L.S. Pessoa: Methodology, Data curation.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

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