

IDENTIFICATION OF GROUNDWATER QUALITY SIMILARITY USING MULTIVARIABLE ANALYSIS¹

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ABSTRACT - A identificação das similaridades na qualidade das águas subterrâneas pode ajudar a reduzir o número de postos de monitoramento utilizados nos corpos hídricos. O objetivo desta pesquisa foi identificar similaridades na qualidade da água subterrânea usando a técnica de estatística multivariada conhecida como análise de agrupamento, no Distrito de Irrigação do Baixo Acaraú (DIBAU), no estado do Ceará. Dez poços rasos distribuídos aleatoriamente no DIBAU foram monitorados regularmente por um período de 27 meses (dez/2003 a nov/2005, nov/2006, mar e abr/2007). Para cada amostra foram analisados pH, Ca²⁺, Mg²⁺, Na⁺, K⁺, Cl⁻, CO₃²⁻, HCO₃⁻, SO₄²⁻, PO₄³⁻, NH₄⁺, NO₃⁻, condutividade elétrica (CE) e razão de adsorção de sódio (RAS). A estatística descritiva, a análise de agrupamento hierárquica e o t-test (1%) foram avaliados utilizando o software SPSS 16.0. No geral, com exceção de dois poços, a água foi classificada como ácida e o pH médio foi menor do que cinco. A concentração de fósforo foi sempre acima do limite recomendado para o consumo humano (0,1 mg L⁻¹). Além disso, a qualidade da água subterrânea foi utilizada para definir quatro agrupamentos que foram independentes da posição geográfica dos poços. Os valores de CE e as concentrações de sódio e cloreto distinguiram dois poços (P1 e P7) dos demais, e o pH, o Mg²⁺ e a RAS determinaram a dissimilaridade dos dois poços entre si.

Keywords: Cluster analysis. Water salinity. Dissimilarity. Water quality.

IDENTIFICAÇÃO DA SIMILARIDADE DA QUALIDADE DE ÁGUA SUBTERRÂNEA UTILIZANDO ANÁLISE MULTIVARIADA

RESUMO - A identificação das similaridades na qualidade das águas subterrâneas pode ajudar a reduzir o número de postos de monitoramento utilizados nos corpos hídricos. O objetivo desta pesquisa foi identificar similaridades na qualidade da água subterrânea usando a técnica de estatística multivariada conhecida como análise de agrupamento, no Distrito de Irrigação do Baixo Acaraú (DIBAU), no estado do Ceará. Dez poços rasos distribuídos aleatoriamente no DIBAU foram monitorados regularmente por um período de 27 meses (dez/2003 a nov/2005, nov/2006, mar e abr/2007). Para cada amostra foram analisados pH, Ca²⁺, Mg²⁺, Na⁺, K⁺, Cl⁻, CO₃²⁻, HCO₃⁻, SO₄²⁻, PO₄³⁻, NH₄⁺, NO₃⁻, condutividade elétrica (CE) e razão de adsorção de sódio (RAS). A estatística descritiva, a análise de agrupamento hierárquica e o t-test (1%) foram avaliados utilizando o software SPSS 16.0. No geral, com exceção de dois poços, a água foi classificada como ácida e o pH médio foi menor do que cinco. A concentração de fósforo foi sempre acima do limite recomendado para o consumo humano (0,1 mg L⁻¹). Além disso, a qualidade da água subterrânea foi utilizada para definir quatro agrupamentos que foram independentes da posição geográfica dos poços. Os valores de CE e as concentrações de sódio e cloreto distinguiram dois poços (P1 e P7) dos demais, e o pH, o Mg²⁺ e a RAS determinaram a dissimilaridade dos dois poços entre si.

Palavras-chave: Análise de agrupamento. Salinidade da água. Dissimilaridade. Qualidade de água.

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INTRODUCTION

Before the 1970s, scientific consensus was that groundwater has a certain level of natural protection against contamination because soil layers and subsoil rocks were believed to act as filters that remove contaminants before they reach the aquifer. Now, however, contaminants are widely understood to sometimes reach the aquifer (BARTON et al., 2006; DOUGHERTY et al., 2009). Groundwater pollution is related to many sources, the most important of which are point-source pollution from municipal sewage and non-point-source pollution from agricultural sources (GULIS et al., 2002; BELPOMME et al., 2007; CHOI et al., 2007). The non-point sources of pollution by agriculture activities depend on land-use management and the time of the year. These sources are related to periods of soil preparation for planting, fertilizing, application of herbicides, fungicides and other treatments, and harvesting (CHOWDARY et al., 2005; FENG et al., 2005).

In the northeastern region of Brazil, water from thousands of shallow wells is used for irrigation and human and animal consumption. This water represents an important intake in the production chain, although the quality of the wells changes with time and location (SOUSA et al., 2009; HOLANDA et al., 2010; BUARTE et al., 2011). The use of unsuitable water may pose a problem for both human health and the environment, with serious negative social and economic impacts (AYRES; WESTCOT, 1999; MOLINA et al., 2003; NG et al., 2000).

Although shallow wells present a financially viable and feasible water source, they may face serious use restrictions due to groundwater pollution and associated risks to human health when they are built and used without adequate technical and sanitary criteria. Also, in most irrigation districts in Ceará, no sewage or drainage infrastructures exist, and septic cesspools proliferate. Organic and/or pathogenic point sources of contamination result (CHOI, 2007; SHOMAR et al., 2008), accentuating the need for water-quality monitoring.

Monitoring of water quality is done by analysis of multiple water-quality parameters at various sampling stations. These stations are evaluated at different time periods, resulting in a complex matrix that is difficult to interpret (ANDRADE et al., 2009). Furthermore, there is a tendency to analyze these parameters separately and then to make assertions about reality. When a phenomenon depends on many variables, this kind of analysis usually is inconsistent because knowledge of isolated statistical information is not sufficient. Understanding all of the information given by the many variables is necessary.

Multivariate statistical methods are useful techniques for studying large and complicated data sets, in which many variables and experimental units are present. The use of multivariate data analysis in

water-quality studies has been widely used in locating monitoring stations and selecting water-quality indicators (ANDRADE et al., 2008; ANDRADE et al., 2010). One of the most commonly used methods is hierarchical cluster analysis, through which the dependence of the sampling stations on territorial continuity can be ascertained.

The goals of this work are (1) to identify the similarities between water-sampling stations in the Baixo Acaraú Irrigation District using hierarchical cluster analysis, (2) to evaluate the potential for reducing the number of long-term water-quality monitoring stations, and (3) to identify the limitations on water use for human consumption.

MATERIAL AND METHODS

Samples were taken from the Baixo Acaraú Irrigation District (Distrito de Irrigação de Baixo Acaraú-DIBAU) aquifer, which is located in the lower part of the Acaraú and Litorânea watersheds in Ceará, Brazil (Figure 1). The weather in the area is hot and humid with summer-autumn rains; monthly average temperatures are greater than 18 °C. Rainfall distribution is unimodal and strongly concentrated in the autumn months (March to May). As a general rule, more than 80% of the annual rainfall occurs during that period, even though the rainfall season ends in June. Mean annual rainfall is 960 mm, and the potential annual evaporation is 1600 mm. The geology of the area is represented by the Tertiary formation, Grupo Barreiras, which is characterized by a few consolidated deposits.

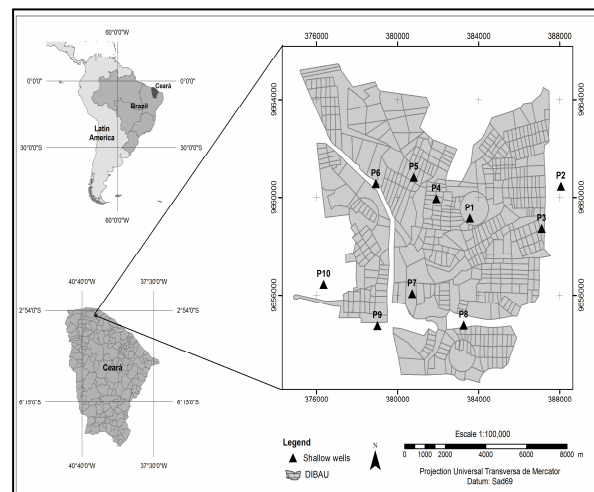


Figure 1. Studied area: location in the Rio Acaraú Basin.

To evaluate the similarity of the water quality in the DIBAU aquifer, monthly water samples (Dec/2003 to Nov/2005, Nov/2006, Mar and Apr/2007) were taken from 10 shallow wells (P1, P2, P3, P4, P5, P6, P7, P8, P9, and P10) during a 27-month period. All waters sampled are used for human consumption. Only wells P4 and P5 are directly

influenced by irrigation practices and management. Sampling, preservation and transportation of the water samples to the laboratory was carried out using the standard methods recommended by American Public Health Association (APHA, 1998) and Richards (1954). Samples were analyzed for the following 14 parameters: pH, electrical conductivity (EC), Ca^{2+} , Mg^{2+} , Na^+ , K^+ , Cl^- , CO_3^{2-} , HCO_3^- , SO_4^{2-} , PO_4^{3-} , NH_4^+ , NO_3^- and sodium adsorption ratio (SAR). The pooled data set includes 3780 samples (10 sites x 14 parameters x 27 months of data acquisition).

Similarities in the water-quality data set were estimated using a multivariate statistical technique called hierarchical cluster analysis and processed by SPSS 16.0 software. To perform the cluster analysis, an agglomerative hierarchical clustering was applied to the data set using a combination of Ward's linkage method and the squared Euclidean distances as a measure of similarity (ANDRADE et al., 2008). The water samples were grouped according to their degree of similarity, which was defined by the variation between the coefficients of two consecutive clusters. To reduce the number of errors due to the scales and the units of the selected variables, the data were normalized ($\mu = 0$, $\sigma = 1$) using the following relationship:

$$Y_{ij} = \frac{X_{ij} - \bar{X}_i}{S_i}, \quad (1)$$

where X_{ij} is the original value of the parameter measured, Y_{ij} is the standardized value, \bar{X}_i the average value of variable i , and S_i is the standard deviation of variable i . The average value of each of the analyzed parameters of the respectively formed clusters was submitted to the t-test at a 1% significance level.

RESULTS AND DISCUSSION

The dendrogram output using Ward's linkage method and squared Euclidean distances (Figure 2) does not provide cluster assignments by itself. Therefore, the number of clusters to be formed must be chosen by the user. This flexibility is one of the subjective points in cluster analysis because the user is free to achieve a certain desired result. According to the rescaled distance cluster combination (Figure 2), an adequate "cutting point" is between 4.95 and 8.95 because of the great increase in rescaled distance to form the next cluster (> 15). The dendrogram shows that the clustering procedure formed four groups.

Cluster 1 was formed only by water samples from P7. Cluster 2 was composed exclusively of water samples taken at P1. Cluster 3 was produced by water samples collected in P3 and P10, and Cluster 4 was made up of pooled monthly data from P2, P4, P5, P6, P8 and P9 sampling stations (Figure 3). In a similar study, Andrade et al. (2008) identified

two hydrochemical groupings of water quality by employing multivariate analysis in the hydrochemical characterization of aquifers in the Trussu watershed. Table 1 shows descriptive statistics of the 14 physicochemical water-quality attributes of each generated cluster.

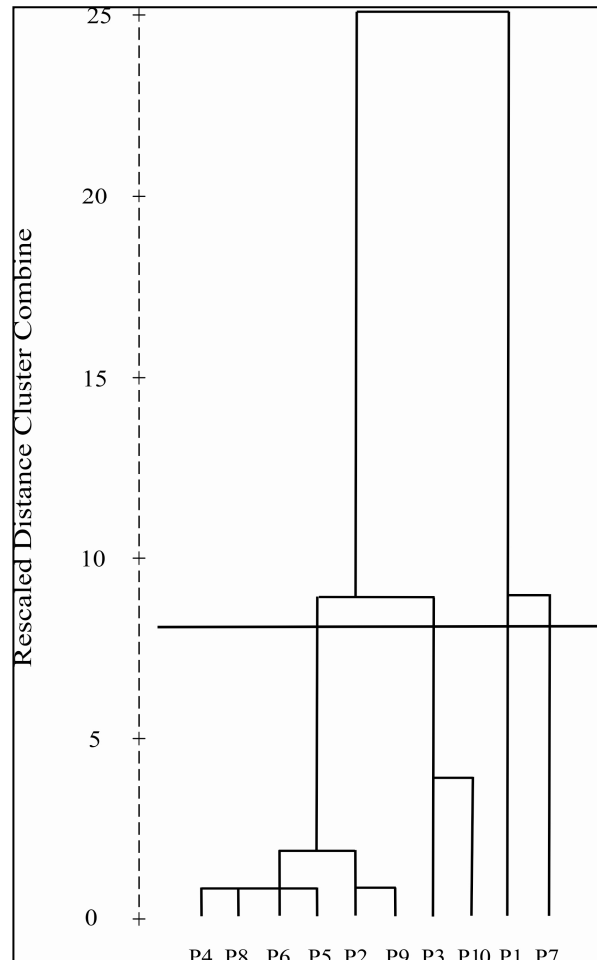


Figure 2. Dendrogram of clustered wells.

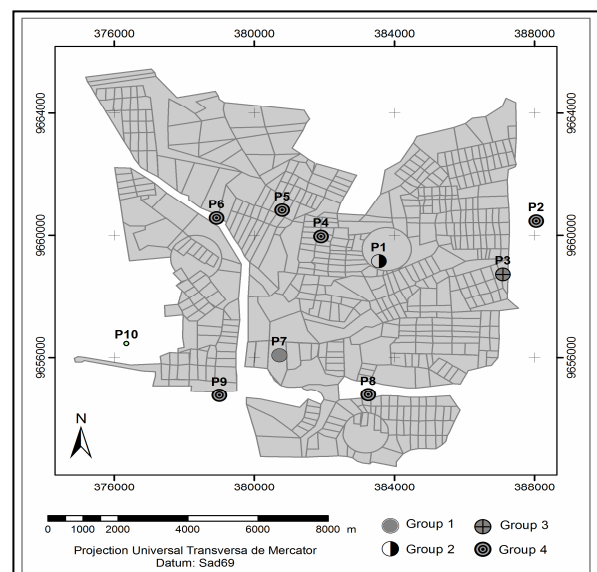


Figure 3. Well distribution in the clusters.

Table 1. Average, minimum, and maximum values for water quality parameters for the groups of wells in DIBAU.

	Statistics	Cluster 1	Cluster 2	Cluster 3	Cluster 4
pH	Average	3.79 ± 0.35 c	4.63 ± 0.47 b	6.46 ± 0.62 a	4.68 ± 0.75 b
	Minimum	2.81	3.62	4.70	3.35
	Maximum	4.36	5.30	7.50	6.30
EC (dS m ⁻¹)	Average	2.30 ± 0.40 b	2.79 ± 0.17 a	0.60 ± 0.20 c	0.54 ± 0.22 c
	Minimum	1,75	2.39	0.34	0.21
	Maximum	3,30	3.00	0.83	1.00
SAR	Average	13.88 ± 3.87 a	8.28 ± 3.87 b	3.71 ± 1.22 d	5.27 ± 2.26 c
	Minimum	9.58	2.91	1.70	1.89
	Maximum	24.22	10.07	6.51	11.25
Ca ²⁺ (mmol _c L ⁻¹)	Average	0.38 ± 0.26 b	1.45 ± 0.37 a	0.48 ± 0.27 b	0.20 ± 0.24 c
	Minimum	0.06	0.95	0.10	0.01
	Maximum	1.05	2.21	0.95	1.03
Mg ²⁺ (mmol _c L ⁻¹)	Average	2.73 ± 0.83 b	6.42 ± 0.93 a	1.11 ± 0.68 c	0.77 ± 0.50 c
	Minimum	1.09	4.69	0.23	0.24
	Maximum	4.21	8.33	1.97	3.08
Na ⁺ (mmol _c L ⁻¹)	Average	16.88 ± 3.67 a	17.11 ± 1.85 a	3.27 ± 1.52 b	3.33 ± 1.56 b
	Minimum	11.57	14.20	1.47	1.48
	Maximum	23.91	20.75	5.32	5.47
K ⁺ (mmol _c L ⁻¹)	Average	0.19 ± 0.04 c	1.11 ± 0.11 a	0.44 ± 0.11 b	0.27 ± 0.12 c
	Minimum	0.16	0.93	0.22	0.13
	Maximum	0.33	1.31	0.64	0.65
Cl ⁻ (mmol _c L ⁻¹)	Average	21.43 ± 3.45 a	22.78 ± 1.37 a	4.29 ± 1.70 b	4.44 ± 1.90 b
	Minimum	14.70	20.00	1.70	0.70
	Maximum	28.00	24.80	6.30	8.50
CO ₃ ⁻ (mmol _c L ⁻¹)	Average	0.00 ± 0.0 a	0.00 ± 0.0 a	0.00 ± 0.0 a	0.00 ± 0.0 a
	Minimum	0.01	0.00	0.00	0.00
	Maximum	0.01	0.00	0.00	0.00
HCO ₃ ²⁻ (mmol _c L ⁻¹)	Average	0.09 ± 0.26 b	0.09 ± 0.09 b	1.53 ± 0.55 a	0.11 ± 0.12 b
	Minimum	0.01	0.01	0.54	0.01
	Maximum	0.95	0.31	2.45	0.56
SO ₄ ⁻ (mmol _c L ⁻¹)	Average	0.07 ± 0.03 bc	0.06 ± 0.03 c	0.29 ± 1.13 a	0.12 ± 0.08 b
	Minimum	0.05	0.01	0.12	0.01
	Maximum	0.15	0.10	0.58	0.38
PO ₄ ³⁻ (mg L ⁻¹)	Average	1.00 ± 1.27 a	1.01 ± 1.26 a	1.89 ± 1.74 a	1.14 ± 1.60 a
	Minimum	0.07	0.10	0.07	0.01
	Maximum	5.00	5.00	6.00	6.00
NH ₄ ⁻ (mg L ⁻¹)	Average	0.62 ± 0.34 b	0.59 ± 0.36 b	4.00 ± 4.00 a	0.79 ± 0.74 b
	Minimum	0.10	0.10	0.10	0.10
	Maximum	1.30	1.20	12.10	3.70
NO ₃ ⁻ (mg L ⁻¹)	Average	3.37 ± 1.48 b	50.58 ± 10.43 a	1.47 ± 2.13 b	4.90 ± 7.16 b
	Minimum	0.60	28.80	0.10	0.10
	Maximum	5.90	67.30	7.60	41.80

Average values followed by different lowercase letters are significantly different by a level of 1% (t-test)

The determining attributes for Cluster 1 were pH, EC, SAR and Mg^{2+} . These variables differed statistically at the level of 1% from the other clusters (Table 1). This group was formed by the wells with the lowest pH (2.81 to 4.36), indicating high acidity waters. This behavior can be explained by the low levels of dissolved CO_3^{2-} that contribute to the formation of carbonic acid. In general, groundwater does not contain suspended material, and the pH varies between 6.5 and 8.0 in a buffered solution due to the presence of dissolved CO_2 and HCO_3^- (HOLANDA et al., 2010).

The elevated EC values in the water samples in Cluster 1 (Well P7) may be related to the high concentrations of chloride and sodium (Table 1). These two ions were not in conformance with the standard for human consumption throughout the entire period of the study. According to Regulation N^o 518/2004 (BRAZIL, 2004) and World Health Organization (WHO, 1998), the acceptable limits of sodium and chloride for human consumption are 200 mg L⁻¹ and 250 mg L⁻¹, respectively. The average concentrations were 21.43 mmol_c L⁻¹ (759.69 mg L⁻¹) for chloride and 16.88 mmol_c L⁻¹ (388.24 mg L⁻¹) for sodium. These concentrations are worrying because consumption of sodium is currently used as an indicator for cardiovascular diseases (MOLINA, 2003). The aquifer contamination source may be human action because this well is located in a community settlement. Another factor that can explain the high EC values for waters of this well may be the local geology characteristics, Grupo Barreiras, which is characterized by a few consolidated deposits.

Another determining parameter in Cluster 1 was SAR, which was statistically different at the level of 1% from the other clusters, distinguishing this cluster in relation to the others. The SAR expresses the risk for generating alkalinity and infiltration problems (AYRES; WESTCOT, 1999; BUARTE et al., 2011). This risk increases when the Ca^{2+}/Mg^{2+} ratio is less than 1.0. According to the data in Table 1, the water samples from Cluster 1 present the greatest risk for generating infiltration problems. This fact is explained by the elevated sodium value and the low Ca^{2+}/Mg^{2+} ratio of 0.13.

Cluster 2 is defined by EC, SAR, Ca^{2+} , Mg^{2+} , K^+ , and NO_3^- values registered in the samples from P1, which differ statistically from the other clusters upon application of the t-test at a 1% significance level. Furthermore, water samples from Cluster 2 show the second-highest electric conductivity average values (2.79 dS m⁻¹), which is in accordance with the cation parameter values (Na^+ , Ca^{2+} , Mg^{2+} , K^+) and high SAR. The EC values registered in Cluster 2 may promote temporary diarrhea in cattle and watery excrement in birds (AYRES; WESTCOT, 1999). Due to the fact that Well P1 is located in an undisturbed area, the increase in these cations is due to deposition of residues originating from human activities in the local community and from the direct influence of a bovine corral located next to the well. Additionally, the predominant soil type at Well

P1 is classified as dystrophic grayish clay soil, which, according to Embrapa (2006), has low activity clay that contributes to a great loss of exchangeable bases due to percolation processes.

Water samples from Cluster 2 show the highest values of Na^+ and Cl^- , although these values are not significantly different from those of Cluster 1 upon application of the t-test at the level of 1% significance (Table 1). Nonetheless, these values are not within the acceptable limits for human consumption (BRAZIL, 2004; WHO, 1998). The average concentration of sodium for Cluster 2 was 17.11 mmol_c L⁻¹ (393.53 mg Na L⁻¹), and the chloride had an average concentration of 22.78 mmol_c L⁻¹ (808.69 mg L⁻¹). These concentrations present a concern because, as stated before, the ingestion of water with high concentrations of sodium salts may compromise human health, and sodium is associated with cardiovascular diseases (MOLINA et al., 2003). Wells P1 and P7 were the first wells in the data set to be built, and they are located in communities where septic tanks are prevalent.

Even though water samples from Cluster 2 contained the highest values for sodium, the SAR values are statistically different from those registered in Cluster 1. This result may be due to the higher concentration of calcium in the water samples of P1. The ratio of Ca^{2+}/Mg^{2+} was higher than in Cluster 1, resulting in a relative reduction of the risk of alkalinity and infiltration problems in the soil.

Cluster 2 presents the highest concentrations of nitrate (Table 1), with a significant difference from the other clusters at the level of 1% significance. These waters are not used for human ingestion. They are used for personal hygiene, laundry, washing domestic utensils, and for livestock water supply. During the entire period of the study, water samples from P1 always showed nitrate concentrations above 28.80 mg L⁻¹, with an average value of 49.59 mg L⁻¹. This value is above the standard limit for human consumption (10 mg L⁻¹) (BRAZIL, 2004; WHO, 1998). These high concentrations may be explained by water pollution originating from human and animal excreta because the well is located near a settlement without sewage treatment. In this region, small domestic animals like chickens and pigs are often raised, and a cattle corral, a potential source of nitrate, is nearby. During the rainy season, these wastes percolate easily through the sandy soil, typical of the area, move toward deeper layers, and rapidly reach the water table (BARTON et al., 2006; ANDRADE et al., 2010). Septic tanks, located near wells and left open sometimes, are associated with a lack of sanitary protection and often cause nitrate contamination (CHOI, 2007; SHOMAR et al., 2008).

The ingestion of water with high nitrate levels is strongly related to gastric cancer in humans (GULIS et al., 2002; BÉLPOMME et al., 2007; DOUGHERTY et al., 2009). In children, nitrate in high concentrations can be transformed to nitrite, which connects to hemoglobin and limits the trans-

port of oxygen to other cells in the body, causing methemoglobinemia (NG et al., 2000), also known as blue baby syndrome.

Cluster 3 is formed because of the similarities between the water samples of P3 and P10. This cluster shows a geographical discontinuity in the water-quality grouping. The attributes pH, SAR, K^+ , HCO_3^- , SO_4^{2-} , and NH_4^+ define the cluster composed of P3 and P10. Samples of this cluster had an average pH of 6.46, which is near-neutral. The higher pH value recorded for the water that composes this group is associated with the higher values of HCO_3^- and SO_4^{2-} observed in these wells because pH values tend to be higher in the presence of HCO_3^- . According to Regulation N^o 518/2004 (BRAZIL, 2004).

Even though the highest values of SO_4^{2-} among the studied wells were detected in Cluster 3 wells, they were always less than 1 mmol_e L⁻¹ (48.03 mg SO_4^{2-} L⁻¹), well-below the limit of Regulation N^o 518/2004 (250 mg SO_4^{2-} L⁻¹ or 5 mmol_e L⁻¹), which is the limit for human consumption. This fact supports the hypothesis that the water from the DIBAU aquifer tends to be naturally acidic (Table 1), and no external source of sulfur is present.

The water samples from Cluster 3 showed the lowest concentrations of Na^+ (3.27 mmol_e L⁻¹ or 75.21 mg L⁻¹) and Cl^- (4.29 mmol_e L⁻¹ or 152.29 mg L⁻¹) and the highest concentrations of HCO_3^- (1.53 mmol_e L⁻¹ or 93.35 mg L⁻¹) compared to the water samples of the other clusters.

The high concentrations of phosphorus and ammonia in this group suggest point-source and non-point-source pollution of the water due to the excreta from humans and small domestic animals (chicken, pigs, dogs, etc.) because P3 does not have any kind of protection (Figure 4) and receives recharge through surface runoff.

Cluster 4 is formed by the six remaining wells (P4, P8, P6, P5, P2 and P9) and defined by the following attributes: SAR, Ca^{2+} and SO_4^{2-} . Like Cluster 3, Cluster 4 is composed of isolated wells, indicating that the similarity between elements does not depend



Figure 4. Image of the lack of protection in Well P3.

on geographic continuity but rather on similar localized conditions.

The pH of the water samples from Cluster 4 was similar (at 1% significance level) to that of Cluster 2 (Table 1). These waters are not in compliance with the standards for human consumption according to Regulation N^o 518/2004 (BRAZIL, 2004) and World Health Organization (WHO, 1998). These organizations establish the suitability for human consumption as pH values between 6 and 9. The maximum pH value registered for this group was below the lower limit, highlighting the acidity of the Baixo Acaraú aquifer. This acidity may be explained by the low concentration of HCO_3^- .

Average EC values in this cluster were lower than 0.7 dS m⁻¹, indicating low quantities of dissolved salts, which are not restrictive for irrigation, animal, confined birds and human consumption (AYRES; WESTCOT, 1999; WHO, 1998). The low EC values may be explained by the fact that these waters come from the drainage of sandy soils. SAR was statistically different from the other clusters at the 1% level of significance. Even though the SAR values from this cluster were lower than those from Cluster 2, the risk for infiltration problems in the soil is similar because the Ca^{2+}/Mg^{2+} ratios in both clusters are basically the same (0.22 for Cluster 2 and 0.26 for Cluster 4).

Although the average nitrate concentrations (Table 1) in the water samples of the wells that make up Cluster 4 are below the recommended limit for human consumption (10 mg L⁻¹), they present a concern because one standard deviation from the average value is above 10 mg L⁻¹, and a maximum value of 41.80 mg L⁻¹ was detected. This tendency for contamination in the water samples of Cluster 4 is related to the fact that the wells are located inside the irrigated areas (P4 and P5), and analysis of the three last samples detected nitrate concentrations above 16.00 mg L⁻¹. This increase may be due to the sandy texture along the entire soil profile and the application of nitrogen and organic mineral fertilizers in concentrations of 80 kg N ha⁻¹ and 430 kg N ha⁻¹, respectively. This nitrate cumulative effect was found only in the wells located in the irrigated areas.

CONCLUSIONS

The waters of the Baixo Acaraú Irrigated District aquifer were classified into four clusters, which do not depend on the geographic position because the clusters were not formed by neighboring wells. The similarities in the waters from Wells P2, P4, P5, P6, P8 and P9, which compose Cluster 4, allow a reduction in the number of wells that must be sampled, minimizing the monitoring costs. The high EC values and the concentrations of sodium and chloride in Wells P1 and P7 distinguished them from the other ones. pH, Mg^{2+} and SAR distinguished these two wells, one from the other.

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