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Assessment of the dynamics of heavy metal contamination in the alluvial aquifer of the Drean plain, El Taref (North-Eastern Algeria) using GIS indices and water quality analysis

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Summary

The study was conducted to assess the quality of shallow groundwater in the Drean plain, with a focus on the concentration of heavy metals. The indices employed included the water quality index (WQI), heavy metal pollution index (HPI), and metal index (MI). Spatial distribution maps of heavy metals, as well as pollution indices, were developed based on twenty samples. The physicochemical parameters, including T, pH, EC, DO, and TDS, were measured using standard techniques. Heavy metals, including iron (Fe), lead (Pb), zinc (Zn), chromium (Cr), manganese (Mn), and cadmium (Cd), were analysed through spectrophotometry. The average concentrations of Mn, Cr, Cd, and Pb exceeded the World Health Organization (WHO) drinking water standards, with respective values of 0.16 mg/L, 0.06 mg/L, 0.28 mg/L, and 0.38 mg/L. According to the water quality index (WQI), which ranges from 21 to 800, approximately 50% of the water samples was classified as highly polluted and therefore not recommended for consumption. According to the metal index (MI), the average value is 6.77, with 80% classified as highly polluted. The average value of the heavy metal pollution index (HPI) was 2201, indicating that the water in the plain is severely polluted for consumption, with 100% of the samples classified as unfit for consumption. The study underlines the urgency of reducing the health risks to the urban population and recommends continuous monitoring of the area to assess the evolution of the pollution.

Keywords

Drean plain • groundwater quality • heavy metals pollution • GIS



1. Introduction

Groundwater, contained in saturated rocks, soil pores, fissures, and crevices, is a vital freshwater resource that is crucial for human needs. Approximately one third of the global population directly relies on this source for consumption [Badra et al. 2024].

However, human activities generate significant amounts of waste that is composed of diverse materials, both biodegradable and non-biodegradable, posing potential risks associated with the presence of extremely hazardous substances [Belkoum et al. 2024]. Recent years have seen a heightened global awareness of health risks caused by metal contamination of the environment. Groundwater pollution from heavy metals has become a major concern due to rapid industrialisation and increasing urbanisation in several regions worldwide [Bougherira et al. 2023]. Heavy metals, such as manganese, zinc, and chromium, show high toxicity and bioaccumulation that can cause severe harm to human health and ecological systems, if they exceed certain limits. Others, like cadmium and lead, pose risks even at lower concentrations [Chaturvedi et al. 2019].

Management of adverse impacts requires regular assessment of the distribution, levels, and potential health risks associated with heavy metals in groundwater. An integral aspect of the study concerns the assessment of groundwater contamination caused by heavy metals, with heavy metal pollution index (HPI) being a commonly used approach in groundwater research [Belkoum et al. 2024].

To evaluate the degree of heavy metal contamination, a number of indices have been suggested, including those created by [Deeksha et al. 2020, Elumalai et al. 2017, Fatima et al. 2022]. However, among them, the heavy metal pollution index (HPI), water quality index (WQI), and metal index (MI) have been more frequently applied to examine heavy metal pollution.

The water quality index emerges as an effective and accessible tool, allowing a comprehensive assessment of water quality based on various parameters. Its objective is to convert water quality data into understandable and usable information for the public, providing a singular indicator of water quality based on critical parameters. In this research, we also employed the metal index (MI) and the water quality index (WOI) to complement our assessment.

Although these indices are less frequently mentioned in the literature, they hold significant importance in the overall characterisation of groundwater quality. By integrating these indices, our study aims for a deeper and more comprehensive understanding of heavy metal contamination in the region, contributing to a robust scientific foundation for the management and prevention of metal pollution. The relevant references for this methodological approach include [Deeksha et al. 2020, Elumalai et al. 2017, Fatima et al. 2022].

The metallic pollution of groundwater has not been explored sufficiently in the study area. Despite previous studies of various sources of contamination [Fernández et al. 2004, Habiba et al. 2023, Horton 1965], the ambition of this study was to address this overlooked issue.

The fundamental objective of this research is to analyse the quality of groundwater, with a particular emphasis on heavy metals, from the perspective of pollution indexing

using three key indices: the water quality index (WQI), the heavy metal pollution index (HPI), and the metal index (MI).

In order to comprehensively assess the groundwater quality in the study area and to identify the areas with different levels of pollution, it is necessary to map the spatial distribution of heavy metal concentrations in groundwater.

2. Methods

2.1. Study area

The study area is located in the north-eastern Algeria, approximately 600 km east of Algiers, near the Mediterranean coast. The precise location of the study area is defined by its geographical coordinates at 36°41'00" north latitude and 7°45'00" east longitude. This area includes a plain that connects four distinct wilayas, situated at 73 kilometres from Souk-Ahras, 43 kilometres from Guelma, 63 kilometres from El Tarf, and 24 kilometres from the city of Annaba. The study area is integrated into the Seybouse watershed, which is divided into three distinct sub-watersheds, namely the upper Seybouse, middle Seybouse, and lower Seybouse, as illustrated by Figure 1.

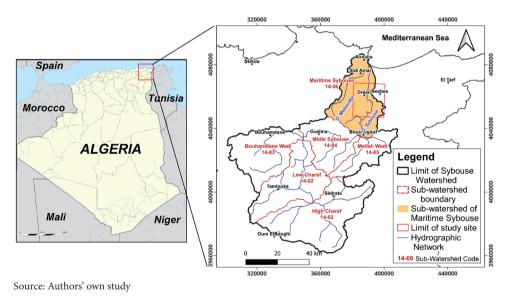


Fig. 1. Geographic location of study area

More specifically, it is located within the middle Seybouse sub-watershed, traversing the maritime Seybouse sub-watershed coded 14-06, according to the classification of National Water Resources [NWR 2019].

The study area is a part of a plain characterised by an alluvial aquifer, mainly comprising the Annaba and the El-Tarf plains. This aquifer is confined by recent and current

alluvial deposits, and its notable permeability is primarily fed by contributions from the Meboudja and Seybouse rivers [Bounab et al. 2023, Kherici and Messadi 1992]. The Mebouja River, which serves as the final tributary to the Seybouse River, forms the hydrographic network of the research area before its discharge into the Mediterranean Sea.

This watercourse facilitates the drainage of Lake Fetzara, the outlet of an endorheic watershed spanning 515 km², through a 14 km long drainage channel.

The available rainfall data were collected from the National Water Resources Agency (NWRA), covering a 20-year period (2003–2023). The area has a Mediterranean climate, with an average annual temperature of 18.5°C and 615 mm of rainfall.

Using the Thornthwaite formula, the actual evapotranspiration is close to 460.3 mm, with runoff and infiltration accounting for 12.60% and 12.55% of the total precipitation, respectively.

2.2. Sampling and analysis

The test areas, identified as the main industrial zones of the city, are located in the maritime Seybouse sub-watersheds. They primarily include the downstream part of the Seybouse River, which serves as the drainage axis for the Seybouse watershed.

Industrial effluents, whether untreated or partially treated, are directly discharged into the Seybouse River, thereby supplying the watercourse that ultimately flows into the Mediterranean Sea east of the city of Annaba.

This worrying situation was highlighted after a concern related to the lack of research on the subject. To address this issue, a comprehensive investigation has been undertaken.

Sampling was conducted on 20 wells from the alluvial aquifer of the study area (Fig. 2). Sampling points were strategically distributed along the downstream part of the Seybouse River, its tributaries, urban areas, and industrial zones that could potentially be sources of pollution. The sampling campaign took place in May 2023.

In May 2023, groundwater samples were collected from wells situated across various industrial zones within the research area. Twenty plastic bottles of one litre each were used to collect these samples. Labelled arbitrarily based on their location, the samples were preserved refrigerated at 4°C, awaiting laboratory analyses for a maximum of seven days.

The positions and elevations of the wells were recorded using the Garmin Stc62 model of the global positioning system (GPS).

Physical parameters such as temperature, pH, electrical conductivity (EC), dissolved oxygen (DO), and total dissolved solids (TDS) were also recorded on-site using a portable multiparameter device (HORIBA). Metal ions including iron (Fe), chromium (Cr), manganese (Mn), zinc (Zn), lead (Pb), and cadmium (Cd) were also quantified.

These analyses were conducted in the laboratory using the flame atomic absorption method with a flame atomic absorption spectrophotometer according to the DIN 38405-D9-2:2008-05 standard.

Each sample was subject to three distinct measurements in order to guarantee optimal accuracy. The average values obtained from these measurements constitute

the final result of this research. Finally, the results of our study, expressed in appropriate units, were thoroughly compared with the standards set by the World Health Organization [Kherici and Messadi 1992].

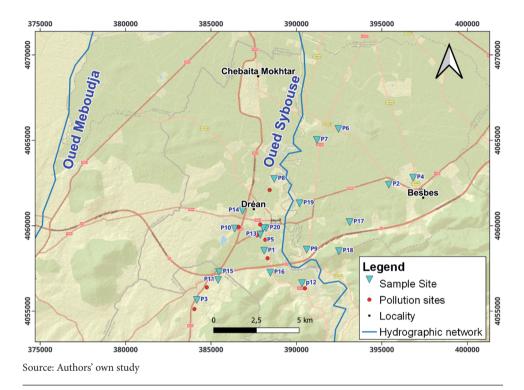


Fig. 2. Location of sample sites

2.3. Groundwater pollution analyses

The heavy metal pollution index (HPI), the water quality index (WQI), and the metal index (MI) were the three indices used to assess the water quality in the research area.

2.3.1. Water quality index (WQI)

The water quality index was calculated taking into account all physicochemical parameters, while the heavy metal pollution index only considered metal concentrations.

The initial formulation of the water quality index dates back to [Mohan et al. 1996]. The WQI is typically used when there is a specific and designated purpose. We took into account the WQI for human consumption in our investigation [Rezaei et al. 2017].

$$Q_i = 100 \left[\frac{V_n - V_i}{V_s - V_i} \right] \tag{1}$$

In equation 1, V_i denotes the ideal value for the n-th parameter, while V_n is the parameter's actual quantity. Each parameter's suggested standard is represented by the letter V_s . All parameter standard values were taken from Kherici and Messadi [1992].

For each related parameter in equation 2, the relative weight (W_i) was determined by taking the inverse of the recommended standard (S_i) .

$$W_i = \frac{1}{S_i} \tag{2}$$

Lastly, the following equation was used to determine the overall WQI:

$$WQI = \sum W_i Q_i \tag{3}$$

2.3.2. Metal index (MI)

The metal index (MI) was proposed by Tachi et al. [2023] and later used by Tachi et al. [2023], Tandel et al. [2011]. This index can be calculated using the expression given by equation 4.

$$MI = \sum_{i=1}^{n} \frac{C_i}{(MAC)^i}$$
 (4)

In this case, MAC stands for the maximum allowed concentration of each element according to the WHO [2017], while MI stands for the metal index. The concentration of each element in the solution is represented by *C*. Lower water quality is indicated by a higher metal concentration in relation to its corresponding MAC value. The warning level is set at an MI value greater than 1.

2.3.3. Heavy metal pollution index (HPI)

The heavy metal pollution index was calculated using the weighted arithmetic mean method. There were three crucial steps in this process: creating a rating scale for each chosen quality characteristic, assigning weights to the selected parameters, and determining the pollution parameters that form the basis of the index [Tamasi and Cini 2013].

We used the following equation to calculate HPI:

$$HPI = \frac{\sum_{i=1}^{n} W_{i}Q_{i}}{\sum_{i=1}^{n} W_{i}}$$
 (5)

The following formula was used in order to calculate Q_i , the *i*-th parameter's sub-index: Q_i is equal to $W_i \cdot n$, where n is the total number of parameters considered in the calculation and W_i is the unit weight assigned to the *i*-th parameter.

$$Q_{i} = \sum_{i=1}^{n} \frac{\left| M_{i} - I_{i} \right|}{S_{i} - I_{i}} \cdot 100 \tag{6}$$

where:

 M_i – the monitored heavy metal,

 I_i and S_i – the ideal and standard values of the *i*-th parameter.

The difference between M_i and I_i ignored the negative algebraic sign. The I_i values were taken from MAC values of the metals, and S_i values were from the standard values set by WHO [2017].

3. Results and discussion

3.1. Physicochemical parameters

Table 1 displays the results of the physicochemical analysis based on the standards suggested by the World Health Organization [Kherici and Messadi 1992].

Table 1. Physicochemical analyses results (sampling date: May 2023)

Site	T [°C]	рН	EC [μs/sm]	TDS [mg/L]	DO [mg/L]
P1	23	6.47	1205	1408.4	7.6
P2	22.4	6.59	2940	695.5	8.16
P3	24.8	8.17	2360	694.5	8.02
P4	22.19	7.62	1503	849.9	8
P5	22.4	8.12	1909	4050.36	3.3
P6	21.3	8.3	1545	368.5	7.41
P7	23.7	7.39	2630	479.5	7.25
P8	24.1	7.69	1536	100.5	8.3
P9	18.65	8.88	3320	2222.5	8.45
P10	21.45	8.12	2610	2626.5	8.48
P11	22.6	7.47	1980	695.5	7.87
P12	22.55	7.63	2236	2000.6	8.16
P13	20.31	7.55	3977	7096.9	5.32
P14	20.73	6.99	3645	2658.5	9.05
P15	21.36	8.28	1811	2654.5	8.41
P16	20.68	8.16	2690	695.5	8.48
P17	23.5	7.67	1691	6989.5	5.8

Table 1. cont.

Site	T [°C]	рН	EC [μs/sm]	TDS [mg/L]	DO [mg/L]
P18	24.34	8.21	2336	6998.5	8.16
P19	19.49	8.31	3414	1695	8.11
P20	21.05	7.39	1920	2785.2	9.05
Minimum	18.65	6.47	1205	100.5	6
Maximum	24.8	8.88	3977	7096.9	9.05
Average	22.03	7.63	2363.9	2388.28	8.047
SD	1.63	0.60	781.25	2249.28	0.66

The temperature of the groundwater samples in the Table 1 reached an average of 22.03°C and vary from 18.65°C to 24.8°C. The pH of groundwater samples ranged from slightly acidic to alkaline, with values between 6.47 and 8.88, and an average of 7.63. The higher pH levels in groundwater can mitigate heavy metal toxicity. With an average of 2388.28 μ S/cm, the conductivity of samples ranges from 3977 to 2363.9 μ S/cm, which is exceptionally high. Most of the water samples had conductivity values greater than 1000 μ S/cm, complying with the WHO standards. There is a clear correlation between the dissolved mineral concentration in the water and the observed electrical conductivity (EC).

High levels of total dissolved solids (TDS) are associated with higher temperatures, leading to increased water salinity. Higher TDS values indicate a greater concentration of both cations and anions in the water. The average TDS in the groundwater samples was 2388.83 mg/L, significantly exceeding the TDS limits of 1000 mg/L set by [Kherici and Messadi 1992]. These high levels of TDS can be attributed to various factors, including anthropogenic activities such as intensive agriculture, irrigation, industrial discharges, and wastewater infiltration. These practices can result in elevated TDS levels, thereby compromising water quality and safety for human consumption. The extreme TDS values, ranging from 100.5 mg/L (P8) to 7096.6 mg/L (P13), indicate the extent of contamination and underline the need for effective water resource management in order to mitigate these adverse impacts on public health and the environment.

The microbial conversion of nitrate to nitrite and sulphate to sulphites is facilitated by the depletion of dissolved oxygen in water reservoirs [Kherici and Messadi 1992].

The maximum dissolved oxygen (DO) value of 9.05 mg/L was found at the sample site (P20), while the lowest DO value of 3.3 mg/L was found at the sampling site (P5), next to a paint company, suggesting a possible organic microbial contamination. Ten percent of the samples meet the standards for irrigation water quality, while only one percent meet the requirements for drinking water quality.

3.2. Metal pollution analysis

The heavy metal concentrations of the study area are summarised statistically in Table 2. The six heavy metal average concentrations were arranged in the following order: Zn < Pb < Cd < Mn < Cr < Fe.

Figures 3a–3f represent the spatial distribution map using GIS-based inverse distance weighting (IDW) technique for the patterns of cadmium (Cd), chromium (Cr), manganese (Mn), lead (Pb), zinc (Zn), and iron (Fe).

Name	Fe	Zn	Mn	Cr	Pb	Cd
Minimum	0.001	0.13	0.0001	0.03	0.13	0.03
Maximum	0.1	0.96	0.4	0.17	0.8	0.55
Average	0.0511	0.5705	0.160505	0.0665	0.3865	0.284
SD	0.02	0.17	0.12	0.03	0.21	0.284
WHO [2017]	0.3	5	0.05	0.01	0.21	0.18

Table 2. Results of metal concentrations [mg/L] values of groundwater samples

With an average concentration of 0.5705 mg/L, zinc was the highest of the six heavy metals under study, according to Table 2. Zinc levels were highest in sample P4 and lowest in sample P10 (Fig. 2).

Zinc is a mineral that is necessary for healthy human growth and development. Zinc concentrations below the WHO [2017] threshold are considered safe for irrigation and human consumption.

The concentrations of iron in the studied waters for P4 and P15 were (0.001) and (0.1) mg/L, respectively, and all samples complied with the WHO limits for drinking water. The distribution of this element is illustrated in (Fig. 3d)

At every location, the levels of lead in the groundwater were higher than the WHO thresholds.

The analysis of data from monitoring wells indicates that the lowest concentration is found at site P17 (0.13 mg/L), while the highest is at site P5 (0.8 mg/L). Consequently, water consumption poses an unacceptable risk. The spatial distribution map in (Fig. 3d) shows an uneven distribution of lead concentration, highlighting specific areas where contamination is particularly concerning.

The groundwater contamination in research area is caused by the careless disposal of solid waste that contains lead, including paints, coloured plastics, and lead-based batteries.

According to Table 2, the average chromium concentration in groundwater was 0.0665 mg/L.

The maximum concentration of Cr (0.17 mg/L) was found at sampling site P17, while the lowest value (0.03 mg/L) was found at sampling site P19, which is close to

the construction site. All of the samples in our investigation were higher than the 0.01 mg/L WHO norm for drinking water.

Regarding the spatial distribution, the map in Figure 3b points to a significant variability in chromium concentration, highlighting specific areas with higher contamination levels, notably around the (P20) site, and lower levels near the petrol pump (P19).

The groundwater samples collected in the studied area indicate manganese concentrations with an average of 0.16 mg/L. This value exceeds the limit of 0.05 mg/L, and 90% of the analysed samples exceed this standard, as visualised in (Fig. 3c) The Figure highlights a notable prevalence of elevated manganese concentrations in specific sites of the studied area.

The average cadmium concentration in the region was 0.28 mg/L, with values ranging from 0.03 to 0.55 mg/L.

The sampling site P3, which is close to the tomato industry, had the greatest concentration of Cd (0.55 mg/L), whereas the location P20, which is close to an industrial complex, had the lowest value. The concentrations of all samples that were measured in this study exceeded the limit.

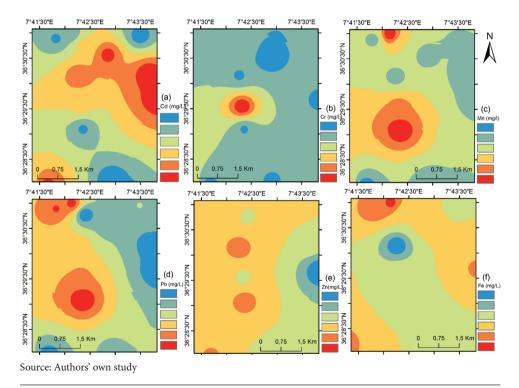


Fig. 3. Spatial distribution of Cd, Cr, Mn, Pb, Zn and Fe (Arcgis)

The zonal distribution analysis of cadmium concentrations in the studied area, as illustrated in Figure 3a, reveals a significant correlation between sampling sites with the high-

est concentrations and their proximity to industrial sources (as observed at site P3 near the tomato industry). This observation confirms the predominant influence of industrial activities on cadmium contamination in groundwater. Additionally, it is worth noting that agricultural practices, including the use of pesticides and fertilisers containing heavy metals, can also contribute to groundwater pollution, particularly near urban areas. Thus, a comprehensive analysis of urban areas is essential to identify high-risk contamination zones and implement preventive measures to protect public health and the environment.

3.3. Water quality indices

The concentration of heavy metals, such as Cd, Cr, Fe, Mn, Pb, and Zn, was included in the calculation of HPI in groundwater samples collected from the study area.

The HPI value ranged from 121 to 5000 with an average value of 2201 (Table 3). Based on the adopted class ranges (Table 4), the HPI can be classified as low (< 100), medium (= 100), and high (> 100). In this study, all groundwater samples were heavily polluted, with HPI values exceeding 100 (Fig. 4a).

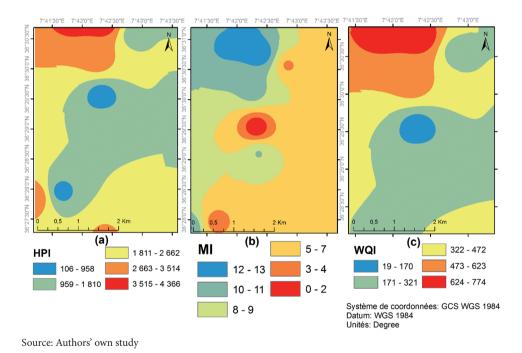


Fig. 4. Spatial distribution of: a. heavy metal pollution index (HPI), b. metal index (MI), c. water quality index (WQI) (Arcgis)

Furthermore, higher levels of Zn and Pb detected at the locations of monitoring wells may explain the observed increases in HPI (Fig. 4a). The classification of all the

studied groundwater resources in the high pollution class of the HPI suggests that the groundwater is significantly polluted with heavy metals and poses a risk to human consumption.

Table 3. Value fluctuations for the metal index (MI), water quality index (WQI), and heavy metal pollution (HPI)

Pollution index	Standard/Class	No. of samples in each class	Standard source references	
Heavy metal pollution index (HPI)	< 100 Low		Sobhanarda-Kania et al. [2016]	
	= 100 Medium			
	>100 High	20		
Metal index (MI)	< 0.3 Very pure			
	0.3-1 Pure	3	Danasi et al [2017]	
	1-2 Slightly affected			
	2–4 Moderately affected	1	Rezaei et al. [2017]	
	4–6 Strongly affected	8		
	> 6 Seriously affected	8		
Water quality index (WQI)	< 50 Excellent water quality	3		
	50–100 Good water quality	1	Tandel et al. [2011]	
	100–200 Poor water quality	1		
	200–300 Very poor water quality			
	> 300 Unsuitable for drinking purpose	10		

The water quality index (WOI) calculated in Table 3 ranges from 21 to 800, with an average of 382. The calculated WQI values showed that 50% of the collected samples are unsuitable for consumption, while the remaining ten samples were classified as highly polluted (25%), polluted (5%), and good (5%). Three groundwater samples (15%) were determined to high quality (Fig. 4b). The average value of this pollution quality index was 382, which is an indication of a highly polluted groundwater.

The calculated average MI values give a mean of 6.77 with a range of 0.36 to 12.64, as shown in Table 3. Furthermore, the metal pollution index (MI) classification, distributing values into four classes within the six categories, was established as shown in Table 4.

The results revealed that 80% of samples (16) was severely affected, representing 40% of the total, while 40% were highly affected, with values ranging from 4 to 6, and beyond 6. The remaining four samples were found to contain a moderate amount of metals, accounting for 5% of the total samples, while 15% of the samples exhibited signs of severe pollution, distributed among two to three samples, respectively.

The calculated average value of 6.77 for the entire area indicates a serious metal pollution.

Indices	НРІ	WQI	MI
Minimum	121	21	0.36
Maximum	5000	800	12.64
SD	2201	382	6.77

Table 4. Evaluation of heavy metal pollution, HPI, MI and WQI value

4. Conclusion

The results of this study indicate that the groundwater in the alluvial aquifer of the study area is significantly impacted by industrial discharges. Physicochemical analyses have shown elevated levels of total dissolved solids (TDS) and conductivity, exceeding recommended limits. Furthermore, the presence of heavy metals such as lead (Pb), chromium (Cr), manganese (Mn), and cadmium (Cd) was detected, with average concentrations of 0.38 mg/L, 0.06 mg/L, 0.16 mg/L, and 0.28 mg/L respectively, exceeding WHO guidelines for water quality. Spatial analysis using GIS-based techniques has revealed different distributions of heavy metal concentrations, highlighting specific areas with elevated contamination levels, often associated with industrial and urban activities. The prevalence of heavy metal pollution poses substantial risks to both the environment and public health, calling for urgent intervention and remediation measures.

The assessment of heavy metal pollution using indices such as HPI, MI, and WQI has justified a high level of concern, with average values of 2201.6, 77, and 382 respectively, indicating widespread contamination. In terms of water quality indices, only 5% of the samples were classified as satisfactory based on WQI, while 15% were classified as having a low level of pollution, considering both the level of contamination and the MI.

This study underlines the urgent need for corrective measures to mitigate metal contamination in groundwater resources in the Drean plain, in order to ensure the safety and quality of drinking water for its residents. These results highlight the need for urgent action to mitigate heavy metal contamination and ensure access to safe drinking water. This could involve measures such as controlling sources of industrial pollution, improving agricultural practices, and implementing appropriate water treatment technologies to reduce heavy metal concentrations in groundwater sources. However, achieving these improvements will require coordinated efforts and efficient management practices.

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