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Heavy Metals Analysis and Quality Evaluation in Drinking Groundwater around an Abandoned Mine Area of Ouichane (Nador's Province, Morocco)

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ABSTRACT

The eventual polluting of the Ouichane region's groundwater by heavy metals around an abandoned iron mine was investigated. To reach this aim, the research began with a questionnaire survey to assess local people's use of and appreciation for well and spring water, followed by measurement of spatial pollution load of heavy metals: Al, Ag, Fe, Cd, As, Cr, Co, Zn, Pb and Cu for water samples collected from twelve wells and three spring drinks of water using inductively coupled plasma-atomic emission spectrometry method (ICP). Determining the overall quality of spring and well water for human use was also performed by calculating the heavy metal pollution index (HPI). As result, the survey revealed that 44.90% of the households are not connected to the drinking water network, 97.3% of them use well water, which is highly appreciated, and 88.1% of the population consider its quality to be good to excellent. Meanwhile, the ICP analysis showed that all the water samples contain heavy metals. In fact, the maximum concentrations (expressed in 10^{-5} g/l) recorded per element were 9.7 for (Ag), 15 for (Al), 6.9 for (As), 4.5 for (Cd), 5.6 for (Co), 31 for (Cr), 14 for (Cu), 858 for (Fe), 7 for (Pb) and $2.9*10^{-5}$ g/l for (Zn). Moreover, most of the water samples recorded heavy metal values above World Health Organization (WHO) limits, for at least one metal among the ten tested, with high concentrations of iron observed in all samples. The HPI values for the three explored sources (S1, S2 and S3) and for 8 out of 12 wells (P3, P4, P5, P6, P7, P8, P9 and P10) exceed the critical pollution value and identify non-potable water with a high potential of contamination. Consequently, the results of this study raise the question about groundwater around this abandoned mining area, especially in the long term, the use of groundwater could increase because of the succession of years of drought on Moroccan territory and hence may constitute a significant health risk for most of the inhabitants.

Keywords: contamination, pollution index, heavy metals, water quality, mining site, Ouichane region.

INTRODUCTION

From 1914 to 1990, the Ouichane region in Morocco's northeast experienced Morocco's mining golden age. It included several iron deposits, of which the most important were Ouichane, Axara, Imnassen, and Setolazar (Bouabdellah et al., 2012). At various stages of extraction, the mine generated around 65 million tons of iron and 1.5 million m^3 per year of rock waste of various natures (Lakrim et al., 2011), which today form hills of mining waste spread across several sites abandoned without rehabilitation for more than 32 years. This endangers the environment and the health of the residents. Effectively, it was shown that mining operations are diverse and release a large proportion of heavy metals into sediments, soils, and surface and groundwater (Ugochukwu et al., 2022; Wei et al., 2018). These heavy metals are widely distributed in nature and enter the food chain, causing serious environmental problems and health issues (Ali et al., 2019; Liang et al., 2017; Zhang et Wang, 2020).

This distribution is due to chemical and biological oxidation reactions of ores present in the waste rock hills directly exposed to climatic conditions (air, water), These reactions produce an acid mine drainage that leads to a pH decline in the environment and is accompanied by the solubilization of metallic elements (Fe, Pb, Cd, Cu, Cr, Zn...) (Hakkou et al., 2008; Lakrim et al., 2011). These elements are one of the most serious threats to the environment and the most significant sources of impacts on water and soil quality and consequently on wildlife, plants, and human health (Ugochukwu et al., 2022; Wei et al., 2018).

Furthermore, the most well-known heavy metals, like silver (Ag), lead (Pb), mercury (Hg), and arsenic (As), are not required for life, whereas trace heavy metals for instance iron (Fe), zinc (Zn), manganese (Mn), magnesium (Mg), copper (Cu), and nickel (Ni) are certainly required for several biochemical and physiological processes in organisms, but they may be toxic when their concentrations in the environment exceed a certain threshold (Pal et al., 2017).

Several studies have examined the contamination of mining sites and their environs by heavy metals and the environmental effect of these operations. It has been shown that heavy metal contamination may have irreversible effects on the environment if sufficient precautions are not followed. (Chen et al., 1995; Liang et al., 2017).

Two geological studies (Lakrim et al., 2011; Khafouri and Talbi, 2020) had been conducted so far in this area to evaluate the transport of heavy metals from abandoned mine wastes to sediments, surface water, and groundwater through acid mine drainage. Nevertheless, none of these studies focused on the proportion of residents who use these waters in this area, and their assessment of its quality, and neither suggested remediation solutions for this pollution. In this context, this investigation aims, firstly, to determine the frequency of groundwater consumption around this abandoned mine area, as well as the local population's use and perception of the water's quality. Second, a chemical characterization of heavy metals is performed to determine the level of water pollution in order to propose potential solutions.

The specific goals of this research are:
- The determination of the frequency of groundwater consumption by inhabitants around this abandoned mine area by determining the frequency of homes not connected to the drinking water system, and their source of drinking water.
- The determination of heavy metal concentrations in wells and spring water used by local inhabitants.
- The determination of the HPI by using the heavy metal concentrations.

MATERIAL AND METHODS

Study area

Ouichane (35.124442, -3.024412), Axara (35.118906, -3.022028), Imnassen (35.115711, -3.012064), and Setolazar (35.125965, -2.998997) were the most important iron deposits in the Beni-Bou Ifrour meso-rifan massif (Bouabdellah et al., 2012) as shown in Figure 1. It is characterized by carbonate sedimentation in the Upper Jurassic and detrital with intercalations of volcano-sedimentary rocks in the Berriasian (Bouabdellah et al., 2012). Furthermore, it has a Mediterranean climate with an annual rainfall of 116–430 mm (Lyazidi et al., 2019). According to the 2014 Haut Commissariat au Plan (HCP), this area includes two municipalities (BniBou ifrour and iksan) with a total population of 14835 people (Haut Commissariat au Plan (HCP), 2014).

Survey

In order to determine the consumption of well and spring water and the population's appreciation of water quality in the region, a questionnaire survey was conducted among 412 inhabitants distributed in the two municipalities of BniBou Ifrour and Iksan.

The questionnaire collects general information about the respondents and the connection of their homes to drinking water networks, as well as their opinions on the quality of water from wells and springs in the region, via the following 3 questions:

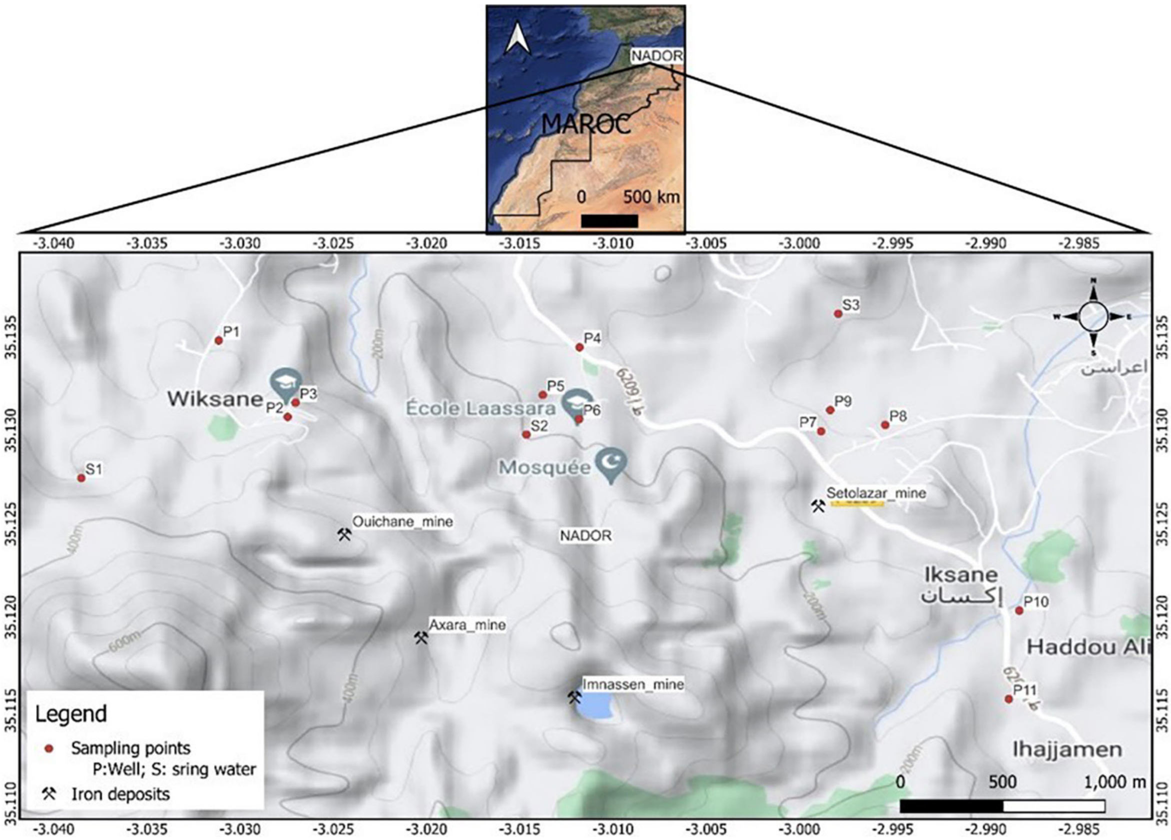


Figure 1. Map of the study area and sampling location, P: well, S: spring water

- Is the home connected to the drinking water system?
- What is the source of drinking water for homes not connected?
- How do you rate the quality of water from wells and springs in the area?

The survey was conducted while maintaining the anonymity of the participants.

Sample collection and physicochemical analysis

In our study area, 12 wells and 3 springs were selected from which three samples were taken. Figure 1 shows the distribution of the selected sampling stations. Sampling was carried out following guidelines for drinking water quality determined by the World Health Organization (WHO) (World Health Organization, 2017). The samples were then labelled, placed in a cooler at a temperature maintained between 0 °C and 4 °C, and accompanied to the laboratory with a form containing the necessary information on the origin, date, and other remarks on the description. It should be noted that the pH, temperature, and electric conductivity (EC) measurements of each sample were performed on-site.

Chemical analysis of samples

Chemical analysis was performed at the CNRST laboratory in Rabat. The chemical parameters were investigated by ICP. It allows us to quantitatively measure the content and chemical composition according to the charge and mass of inorganic elements present in our samples and to detect traces of the following heavy metals: Fe, Ag, Al, Pb, As, Cd, Co, Cr, Cu, and Zn.

Evaluation of contamination

Following the method described by Mohan (Mohan et al., 1996), the HPI was used to evaluate the overall quality of water based on heavy metal content. The HPI shows how the different heavy metals affect the overall quality of the water. It is calculated using the quality-weighted arithmetic mean, which assigns a weight unit (Wi) to each heavy metal. Thus, the HPI is represented as follows:

$$HPI = \frac{\sum_{i=1}^{n} QiWi}{\sum_{i=1}^{n} Wi} \tag{1}$$

where: Qi and Wi are the sub-index and unit weight of the i^{th} parameter, respectively.

Therefore, the sub-index (Qi) is calculated by the following equation:

$$Qi = \sum_{i=1}^{n} \frac{(Mi - Ii)}{(Si - Ii)} \times 100 \quad (2)$$

The weight unit (Wi) is the value inversely proportional to the recommended standard (Si) of the corresponding parameter (Mohan et al., 1996).

$$Wi = \frac{1}{Si} \quad (3)$$

where: Mi, Ii and Si are the monitored heavy metals, ideal and standard values of the i^{th} parameter, respectively.

A value of 100 is the critical pollution index used for water quality in this study (Mohan et al., 1996).

Statistical analysis

The normality test, through the Shapiro-Wilk test, at 5% of significance, to prove homogeneity of variances, to decide the use of univariate statistical analysis (correlation tests), multivariate analysis (similarity tests) and principal component analysis (PCA) used to determine the different correlations existing between the investigated parameters.

RESULTS AND DISCUSSION

Survey analysis

The results of the survey are divided into two parts: the respondent's general information and their use and appreciation of the water quality in the region by the local population.

Respondent's general information

31.55% of our population is aged 18 to 29 years old, and 52.43% are men. 40.53% of people have lived more than 30 years in the Ouichan region, and 35.19% have a secondary school level (Table 1).

The results reveal that 44.90% of households are not connected to the drinking water network, and 97.3% use well water. Even more, both well and spring waters are highly appreciated by 88.1% of the population, who consider their quality to be good to excellent (Table 2).

According to the survey results, the local population often uses these waters for drinking purposes and other domestic activities.

Heavy metal concentrations

Table 3 and Figure 2 show that samples recorded several concentrations above WHO standards while others were below standards, such as Ag, Al, Cu, and Zn.

The concentration of iron (Fe) is very high in all sites with an extreme value of ($8.58 \cdot 10^{-5}$ g/l) in P7 and indicates a contamination from the mine site. Also, cadmium (Cd) levels exceed WHO in all the sources studied and in wells P3, P4, P5, P6, P7, P8, P9, and P10. At a lower level of pollution, chromium (Cr) levels exceeded the standards in all sources and wells P4, P5, P6, P7, P8, and P9; followed by arsenic (As)

Table 1. Characterization of the population under study

Parameters	Subgroup	Frequency		Percentage (%)
		Bni Bouifrour	Iksane	
Age	18–29	58	72	31.55
	30–39	46	41	21.12
	40–49	35	36	17.23
	50–59	31	41	17.48
	Over 60	25	27	12.62
Gender	Man	92	124	52.43
	Woman	90	106	47.57
School level	Illiterate	72	47	28.88
	Primary	44	49	25.97
	Secondary	60	116	35.19
	University	6	18	9.95
Number of years in the region (NYR)	NYR<20	45	57	24.76
	20<=NYR<30	63	80	34.71
	30<=NYR	74	93	40.53

Table 2. Use and appreciation of water quality in the region by the local population

Parameters		Frequency		Percentage
		Bni Bouifrour	IKSANE	
Connection of the house to the drinking water network	Related	110	117	55.10%
	Not related	72	113	44.90%
Water sources for no-related housing	Wells	71	109	97.30%
	Water source	1	4	2.70%
Residents' assessment of well and spring water quality	Very bad	1	5	1.46%
	Bad	3	7	2.43%
	Medium	14	19	8.01%
	Good	66	75	34.22%
	Excellent	98	124	53.88%

concentrations exceeded the standards in wells P5, P8, P9, and P12, and in all sources with a maximum value of ($6.9 \cdot 10^{-5}$ g/l) at S1; also lead (Pb) concentrations exceed the limits in all sources and in tree wells, which are P5, P10 and P11. while cobalt (Co) concentrations exceeded the standards only in the well P6. The comparison of the concentrations of metallic elements measured in wells and springs with the WHO standards for consumption shows a notable excess of concentrations of at least 1 heavy metal out of 10.

Our results differ from those obtained by Lakrim (Lakrim et al., 2011) and Khafouri (Khafouri and Talbi, 2020) in the mining waste of the Nador Mine; This difference can be explained by the fact that these two studies were conducted in different contexts, one of which aimed to examine the mode and the factors influencing the transfer of these metallic elements from the mine wastes to the waters, rather than focusing entirely on the waters of the drinks. In their studies, they chose sampling points that are not used by the inhabitants, such as the lake of Ouixane, as well as a wet period during which the acid mining drainage increases the solubility of the metals. Effectively, during the mine operations, many tailings and rock waste were produced, which were until now exposed to air and water during rainy seasons, enabling the production of acid mine drainage and the release trace elements from sulfites into the surrounding aquatic environments (Cadmus et al., 2018; Förstner et al., 1981). Maanan and Bloundi suggested that trace elements, except for Co, are derived from the same rock source, bound to iron oxides, smectite, or chlorite clays, as reported (Bloundi et al., 2009; Maanan et al., 2015). Other studies suggested that Pb, Cr, Cu, and Zn are derived from geological and anthropogenic sources such as industrialization (Alloway, 2013; Faouzi et al., 2023), and have an impact

Table 3. Descriptive statistics of physicochemical characteristics in well and spring waters

Parameter	Mean	Min.	Max.	1st Qu.	Median	3rd Qu.	Standards (WHO, 2017)
Altitude (m)	188.8	114.0	318.0	126.0	182.0	233.5	-
Temperature(°C)	22.73	20.00	24.00	22.00	23.00	23.50	<25
pH	7.427	7.000	8.100	7.200	7.300	7.600	6.5-8.5
EC(S/m)	0.2817	0.1765	0.3876	0.2390	0.2698	0.3420	0.400
Ag(10^{-5}g/l)	6.5	1.0	9.7	5.5	7.5	9.1	10.0
Al(10^{-5}g/l)	8.2	3.0	15.0	5.7	7.5	10.0	20.0
As(10^{-5}g/l)	1.9	0.3	6.9	1.0	1.1	2.0	1.0
Cd(10^{-5}g/l)	2.0	0.1	4.5	0.7	2.5	2.9	0.3
Co(10^{-5}g/l)	2.3	1.0	5.6	1.1	2.1	2.9	5.0
Cr(10^{-5}g/l)	13.3	1.2	31.0	2.6	15.0	20.0	5.0
Cu(10^{-5}g/l)	7.5	4.7	14.0	6.3	7.3	8.3	20.0
Fe(10^{-5}g/l)	461.1	128.0	858.0	271.5	426.0	660.0	30.0
Pb(10^{-5}g/l)	1.9	0.1	7.0	0.1	0.7	2.8	1.0
Zn(10^{-5}g/l)	1.2	0.5	2.9	0.7	1.0	1.8	30.0

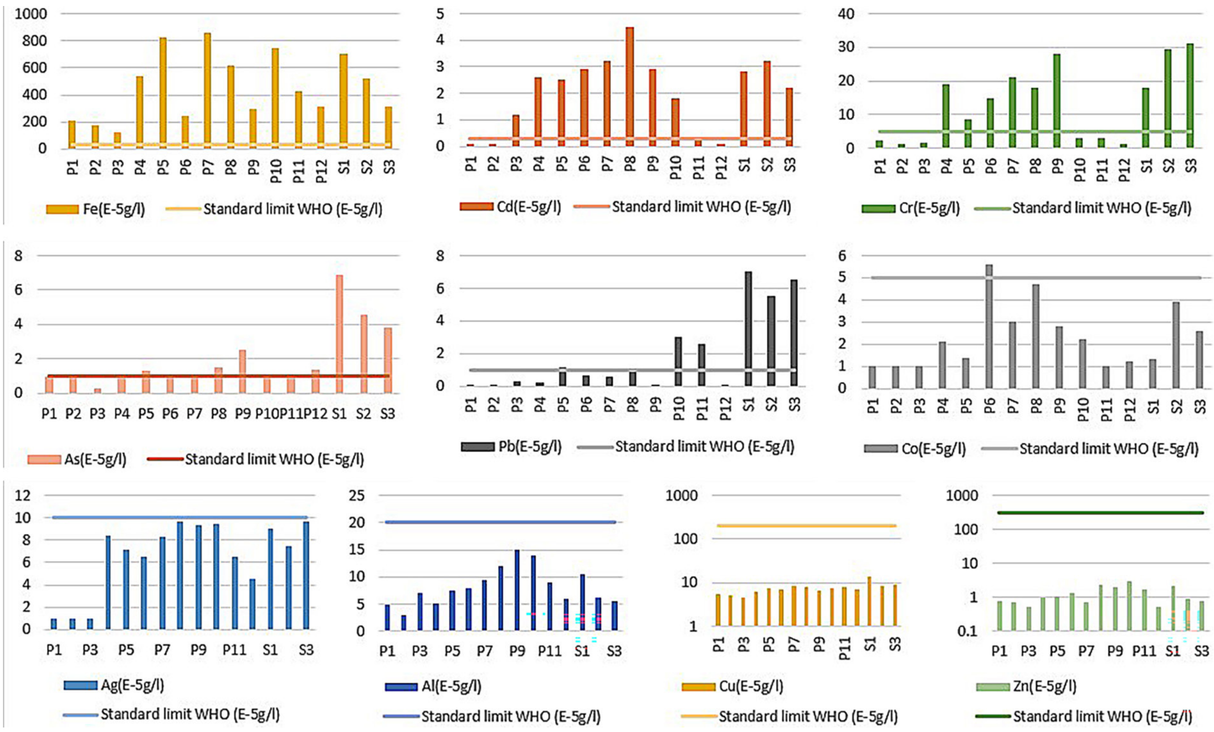


Figure 2. Spatial distributions of ten heavy metals in spring and well water in the study area

on biodiversity and human health (Rezouki et al., 2021; Sanae et al., 2021).

It is important to remember that iron overload causes oxidative stress and is a recognized risk factor for the onset of chronic liver diseases (Bloomer et Brown, 2019; Eaton et Qian, 2002), and yet locals still take this water, believing them to be of high quality.

The heavy metal pollution index

HPI values were calculated globally for each sampling site by using the average concentration values of individual heavy metals, which were compared to the critical value (Mohan et al., 1996), and then presented in Figure 3.

High HPI values exceeding the critical value were observed for all sources (S1, S2, and S3) and wells P3, P4, P5, P6, P7, P8, P9, and P10. These elevated HPI values in the water samples may possibly indicate an impact from anthropogenic mining activities instead of geologic alteration (Uugwanga e Kgabi, 2021). The overall HPI values for these sampling points are high-risk and indicate unsuitable water for drinking. For the remaining sampling points, HPI values are below the critical value, indicating low-risk human-consumable water. (Ghaderpoori et al., 2018). Generally, the HPI value of water from springs and some wells indicates that the water requires treatment.

The normality test proved homogeneity of variances; this allowed us to carry out an analysis using several significant inter-elemental correlations found between metals and HPI in our study (Table 4), such as Ag, Cd and Cr with HPI with a highly significant correlation with indicating that these three metals strongly influence HPI values, also the metals: Co, As and Fe contribute significatively to the rise of this index. Also, a strong correlation between the heavy metals is observed in the cases as follows: Ag with Fe, Cr, and Cd; Al with Zn; As with Pb and Cu; Cd with Cr and Co; Cu with Pb and Fe. this correlation is insufficient to identify the source of the pollution; for this reason, PCA was adopted to facilitate the explanation of the elemental data and find groups of metals with the same derivation. The results of the PCA are presented in Figure 4.

According to the PCA, represented in Figure 4, of the Ouichane mine water, the variables were correlated to two principal components, which accounted for 62.88% of the total variance. Because the eigenvalues of the principal components were more significant than 1 (Kaiser, 1960), they were kept. After varimax rotation, three components are obtained. These components were associated with the resources of the elements. The first

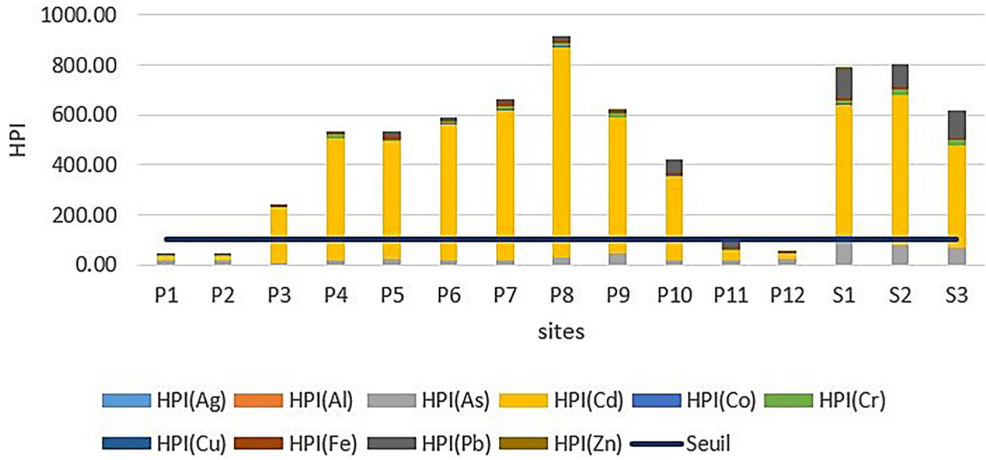
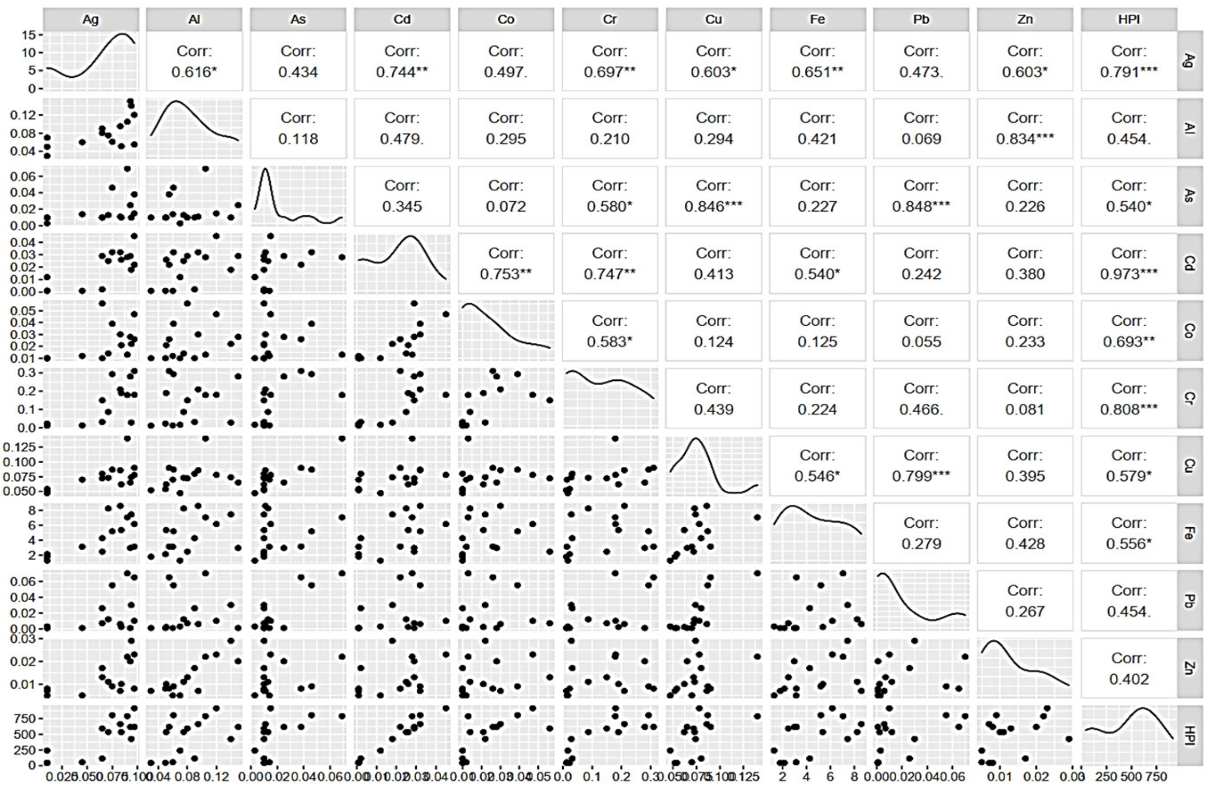


Figure 3. Variation in heavy metal pollution index (HPI) for all sites statistical analysis

Table 4. Spearman correlation analysis of target metals in well and spring water from the Ouichane area



Note: significant codes: 0 '***' 0.001 '**' 0.01 '*' 0.05 '.' 0.1 ' '.

two dimensions of the analysis express 62.88% of the total inertia of the data set, which means that 62.88% of the total variability of the cloud of sampling stations is explained by design. This percentage is relatively high; therefore, the first design represents the variability of the data. This value is higher than the reference value, which equals 43.99%, so the variability explained by this design is significant.

Dimension 1 contrasts sampling stations such as P8, S1, S2, and S3 (on the right side of the graph, showing a very positive axis value) with sampling stations such as P2, P1, P3, and P12 (on the left of the graph, showing a very negative axis value). The group in which sampling station P8 is located has high values for the variables EC, Cd, Al, Ag and Co and low values for the variables altitude and pH value. This indicates the significant effect of acid mine drainage that occurs at low-elevation sites. The group in which sampling stations S1, S2, and S3 are located shares high values for the variables Pb, As, Cu and Cr, and

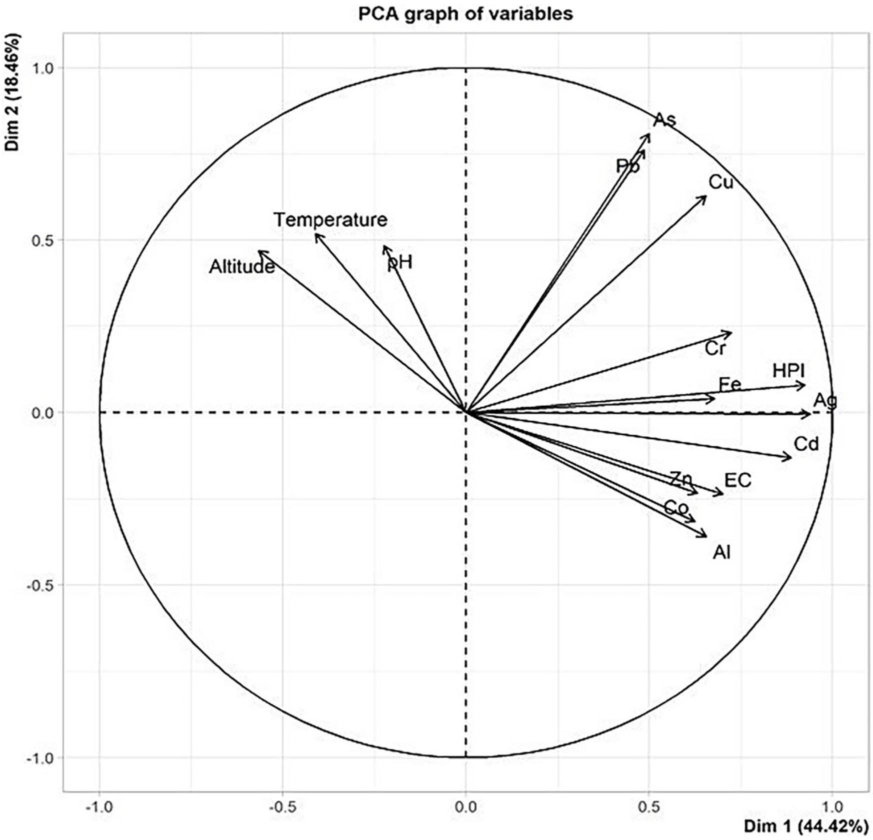


Figure 4. Correlation circle of variables on the Dim 1 × Dim 2 factorial design

this may be influenced by the high piezometric level in this area. Meanwhile, the group in which sampling stations P2, P1, P3, and P12 are located shares low values for the variables HPI, Cd, Ag, Cr, EC, Co, and Fe.

Dimension 2 contrasts sampling stations such as S1, S2, and S3 (at the peak of the graph, showing a strong positive coordinate on the axis) with sampling stations such as P8 (at the bottom of the graph, indicating a strongly negative coordinate

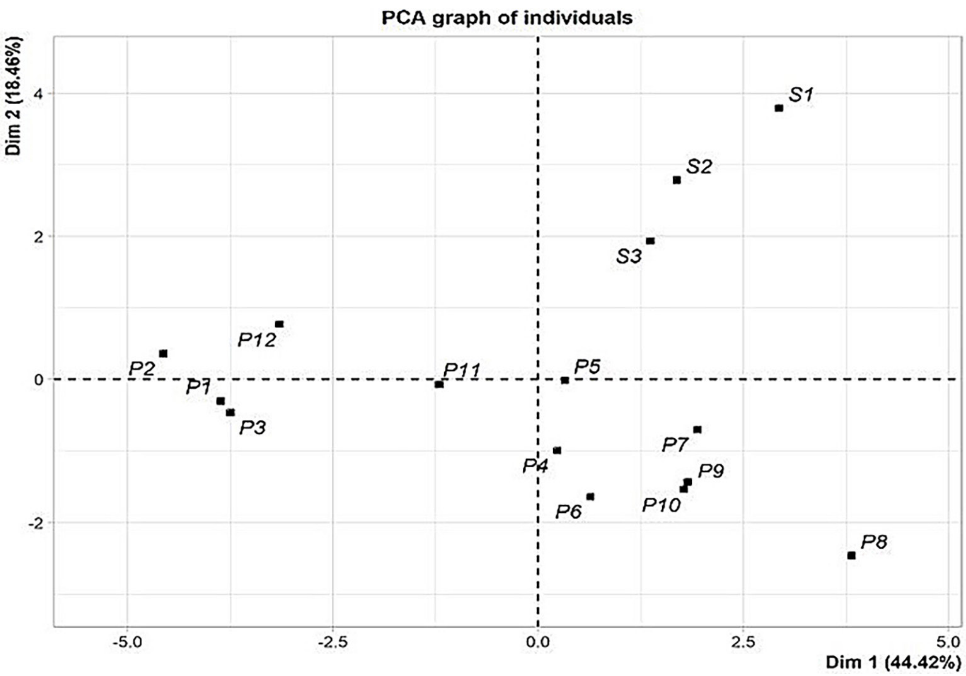


Figure 5. The plot of the distribution of analyzed water samples and stations in the Dim 1 × Dim 2 design

on the axis). The group in which sampling stations S1, S2, and S3 are located shares high values for the variables Pb, As, Cu, and Cr, and the group in which sampling station P8 has located shares high values for the variables EC, Cd, Al, Ag, and Co and low values for the variables altitude and pH.

From this analysis, we can therefore conclude the existence of three groups of stations, the first of which is represented by the stations P1, P2, P3, and P12, characterized by low values of heavy metals and HPI; the second group is represented by the stations P4, P5, P6, P7, P8, P9, and P10, characterized by high values in EC, Cd, Al, Ag, and Co; and the third group is represented by the stations S1, S2, and S3, characterized by high values in As, Pb, Cr, Cu, Fe, and HPI.

HPI, Ag, and Cd are strongly correlated, verifying what we found thanks to the first analysis.

CONCLUSION

The Nador region (northeastern Morocco) was home to one of the world's largest iron ore mines, which has been intensively mined and abandoned without replanning.

The herein study aimed to evaluate local people's use of and appreciation for well and spring water around an abandoned iron mine in the Ouichane region located in the northeast of Morocco, followed by measurement of spatial pollution load of trace metal elements in different water samples gathered from groundwater in order to estimate the water pollution level by heavy metals in the mentioned region. Thus, following the results of the survey, the local population appreciates and regularly uses water from wells and springs for drinking and other household purposes, regardless of the fact that heavy metals contaminate these waters.

Indeed, the water from wells and springs in the vicinity of the abandoned Ouichane mine contains high concentrations of certain heavy metals that exceed the WHO's limits for drinking water, making it unsuitable for consumption by humans. In addition, the calculations of HPI for the source water have identified high values that exceed the critical pollution index value of 100. Exceedance of the critical HPI value was also found in eight of the twelve wells sampled, indicating an impact from anthropogenic mining activities rather than geologic alteration.

Hence, this environmental pollution of the Ouichane mining site is the cause of the alteration of the water quality.

At the end of these findings, an ecol-toxicological and epidemiological examination of the metal pollution of the terrestrial trophic chain will be required in the near future, and rehabilitation measures of the Ouichane mining site and the treatment of its waters are necessary to immobilize the residual metallic pollutants of the mining discharges to prevent public health issues.

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