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Assessment of Heavy Metals Contamination in Groundwater: A Case Study of the South of Setif Area, East Algeria

Lazhar Belkhiri, Ammar Tiri and Lotfi Mouni

Additional information is available at the end of the chapter

Abstract

Heavy metals in groundwater were analyzed and their sources and impacts were identified using multivariate statistical tools and risk assessment. Three significant factors were extracted by factor analysis (FA), explaining 75.69% of total variance. These factors were in turn described by the clusters C3, C2 and C1, respectively, resulting from the cluster analysis (CA). Factor analysis and cluster analysis revealed significant anthropogenic contributions and water-rock interaction effects of the metals in groundwater. The mean values of heavy metal evaluation index (HEI) and degree of contamination (C_{deg}) indices indicated that the groundwater samples were contaminated with high degree of pollution by cadmium (Cd) and lead (Pb). The hazard quotients (via ingestion) of Cd and Pb were found to be higher than the safe limits, posing threat to the consumers. However, no risk related to the dermal contact was associated with the measured metal levels.

Keywords: groundwater, heavy metals, multivariate statistical methods, human health risk assessment, recommendation

1. Introduction

In the context of the management of water resources, the identification of heavy metals is a primary importance because of their influence on the quality of groundwater and consequently on the human being.

Guidance contained in this context should relatively take into account national and international watersheds in terms of policy, planning and management, to support a more effective integration of polluted areas [1].

Groundwater is the principal natural water resources for both drinking and agricultural purposes. Nowadays one of the most important environmental issues is groundwater contamination [2, 3]. In areas where population density is high and human use of the land is intensive, groundwater is especially vulnerable. Virtually any activity whereby chemicals or wastes may be released to the environment, either intentionally or accidentally, has the potential to pollute groundwater. When ground water becomes contaminated, it is difficult and expensive to clean up.

Heavy metals are among the major contaminants of groundwater sources [4]. Some of these heavy metals are essential for the growth, development and health of living organisms, whereas others are non-essential as they are indestructible and most of them are categorized as toxic species on organisms [5]. Nonetheless, the toxicity of heavy metals depends on their concentration levels in the environment. With increasing concentrations in environment and decreasing the capacity of soils toward retaining heavy metals, they leach into groundwater and soil solution. Thus, these toxic heavy metals can be accumulated in living tissues and concentrate through the food chain.

The main objectives of this study are: (1) to determine the spatial variation of heavy metals using multivariate statistical techniques, (2) to assess the potential health risk assessment of heavy metals and (3) take preventive and protective measures.

2. Study area and data analysis

The study area is located in the east of Algeria and the south of Setif (**Figure 1**). It is characterized by intensive agricultural and human activities. The climate of this area is semi-arid, with a mean annual temperature and precipitation of 15.2°C and 296 mm/year, respectively [6].

In the current study, 18 wells were collected (**Figure 1**) and 11 parameters (T, pH, EC, Al, Cd, Cu, F, Fe, Pb, Si and Zn) were analyzed using standard procedures [7]. The electrical conductivity (EC), pH and the temperature (T) were measured by multi-parameter WTW (P3 MultiLine pH/LF-SET). The concentrations of heavy metals were determined by Graphite Furnace Atomic Absorption Spectrophotometer (Perkin-Elmer AAnalyst 700).

3. Statistical analysis

The descriptive statistics of heavy metals in the wells of the study area are demonstrated in **Table 1**. Minimum and maximum values of electrical conductivity are 830 and 2730 μS/cm with a mean value of 1451 μS/cm. The measured water temperatures varied from 14 to 18°C with a mean of 16°C. The pH values of the groundwater samples vary from 6.9 to 7.9 with a mean of 7.4 indicating that the waters were generally neutral to slightly alkaline. pH does not show significant positive correlation with any heavy metals, while it shows negative correlation with Fe, Zn and Cd (**Table 2**). This indicates that influence of pH on heavy metals was different in groundwater of the studied area. The mean concentration of Al, Cd, Cu, F, Fe, Pb, Si and Zn was 0.05, 0.066, 0.241, 0.129, 0.255, 0.087, 21.6 and 0.148 mg/l, respectively. Moreover, the mean values of the heavy metal contents in the groundwater follow the decreasing order: Si > Fe > Cu > Zn > F > Pb > Cd > Al.
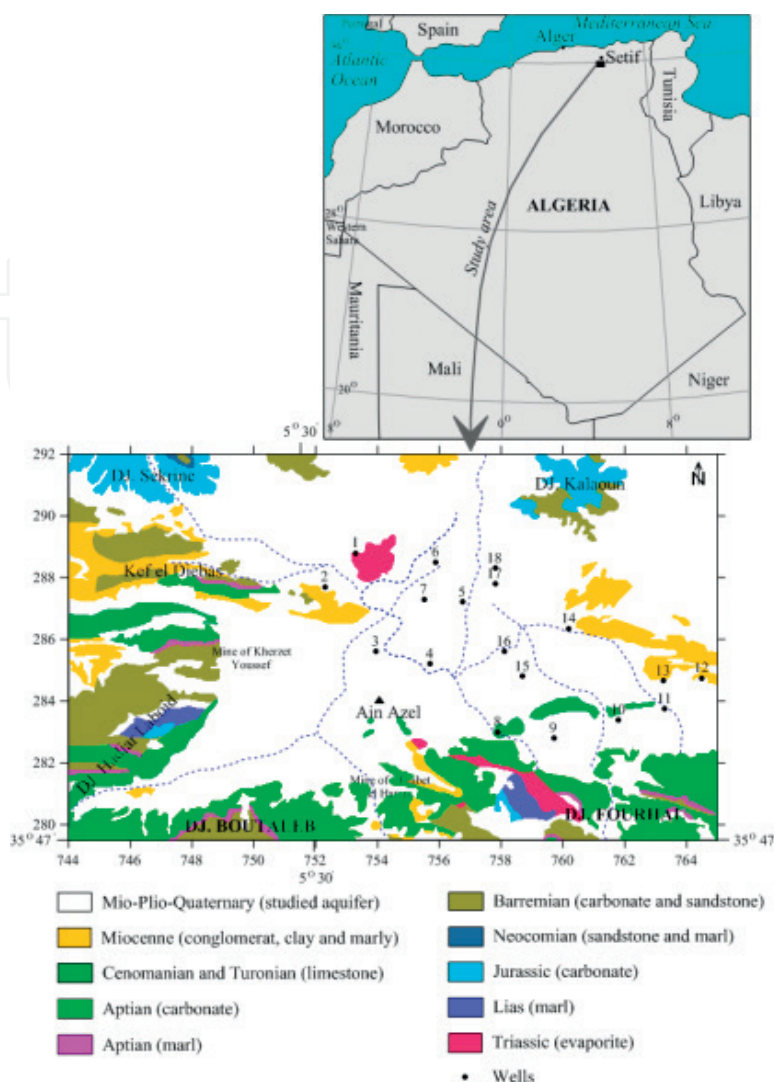
Figure 1. Location of the study area and the samples.

In the present study, factor analysis (FA) and cluster analysis (CA) were used to evaluate the concentrations of heavy elements in groundwater samples.

3.1. Factor analysis

Factor analysis was employed to find and interpret the structure of the underlying data set through a reduced new set of orthogonal (non-correlated) variables (principal components, PCs), arranged in decreasing order of importance. Besides considerable data reduction, PCs can explain the entire multidimensional data set variability without losing much original information. FA with Varimax rotation of standardized component loadings was conducted for extracting and deriving factors, respectively, and those PCs with eigenvalue >1 were retained [8–10]. The distribution manner of individual association of element in groundwater was determined by principal component method (results are shown in **Table 3**). Statistical treatment of these data indicates their association and grouping with three factors explained most of the variability (total variance explained was about 75.69% variance for the groundwater data). The relations among the heavy metals based on the first three factors are illustrated in **Figure 2** in three-dimensional space.

	Min	Max	Mean	SD	CV
EC	830	2730	1451	557	38
T	14	18	16	1.4	8.6
pH	6.9	7.9	7.4	0.3	3.5
Al	0.01	0.09	0.05	0.02	43.69
Cd	0.009	0.165	0.066	0.045	67.646
Cu	0.056	0.43	0.241	0.102	42.248
F	0.017	0.358	0.129	0.111	86.222
Fe	0.055	0.499	0.255	0.116	45.563
Pb	0.017	0.292	0.087	0.069	79.323
Si	12.2	33.3	21.6	7.2	33.2
Zn	0.045	0.276	0.148	0.06	40.466

Remarks: All values are in mg/l except pH, T (°C) and EC (µSiemens/cm). "Min": minimum; "Max": maximum; "SD": standard deviation; "CV" (in %): coefficient of variation.

Table 1. Statistical summary of physicochemical parameters in groundwater samples.

The first factor shows 39.92% of total variance with high loading on Al, F, Pb and Si. These metals were predominantly contributed by the water-rock interaction effects and anthropogenic sources. Aluminum was the most abundant element found in the earth's crust [11] and from the result obtained from its analysis, the minimum concentration of aluminum detected in the groundwater samples is 0.01 mg/l with the maximum concentration being 0.09 mg/l. All samples exceeded the desirable limit of Al for drinking water (0.03 mg/l)

	EC	T	pH	Pb	Fe	Zn	Cu	Cd	Si	F	Al
EC	1										
T	−0.36	1									
pH	0.33	−0.54	1								
Pb	0.32	0.27	0.15	1							
Fe	−0.46	0.19	−0.42	−0.05	1						
Zn	0.20	0.31	−0.29	0.54	−0.08	1					
Cu	0.04	0.12	0.27	0.50	−0.02	0.31	1				
Cd	0.25	0.04	−0.17	0.23	0.37	0.41	0.04	1			
Si	0.43	0.06	0.29	0.26	−0.37	0.06	0.49	−0.03	1		
F	0.78	−0.05	0.40	0.59	−0.49	0.26	0.28	−0.05	0.41	1	
Al	0.72	0.08	0.16	0.44	−0.48	0.28	0.15	0.03	0.36	0.87	1

Table 2. Pearson's correlations matrix for the physicochemical parameters.

Component	Eigen values		
	Total	**% of variance**	**Cumulative %**
1	**3.19**	**39.92**	**39.92**
2	**1.77**	**22.08**	**62.00**
3	**1.10**	**13.70**	**75.69**
4	0.69	8.57	84.26
5	0.63	7.89	92.16
6	0.30	3.73	95.89
7	0.24	2.97	98.87
8	0.09	1.13	100.00

Variables	Component			Communalities
	1	**2**	**3**	
Pb	−0.74	0.42	0.02	0.64
Fe	0.50	0.66	0.15	0.53
Zn	−0.52	0.58	−0.22	0.48
Cu	−0.55	0.28	0.69	0.47
Cd	−0.08	0.78	−0.24	0.39
Si	−0.61	−0.21	0.53	0.44
F	−0.88	−0.22	−0.23	0.85
Al	−0.81	−0.22	−0.38	0.79

Table 3. Factor analysis of groundwater data. The significant factors (>1) are shown in bold.

except sample 10, but none of the groundwater samples contained Al above the specified maximum contaminant level (0.2 mg/l) [12]. The ranges of fluoride are 0.017–0.358 mg/l. Thus, F concentrations are relatively low in the groundwater (<1.5 mg/l). The concentration of the lead in the groundwater samples ranges from 0.017 to 0.292 mg/l. The groundwater quality standard of lead desirable and maximum permissible limit (WHO) is 0.01 mg/l. All of the groundwater samples are exceeding then WHO desirable and maximum permissible limit of Pb. The concentration of Si in the samples varies from 12.2 to 33.3 mg/l with a mean value of 21.6 mg/l.

The second factor exhibits 22.08% of the total variance with positive loading on Cd, Fe and Zn. The concentration of the cadmium in the water samples varies from 0.009 to 0.165 mg/l with a mean of 0.066 mg/l. The groundwater quality standard of cadmium desirable and maximum permissible limit is 0.003 mg/l. All samples are exceeding then desirable limit of Cd. The concentration of iron ranges from 0.055 to 0.499 mg/l. The concentrations of Fe in many of the samples are higher than the WHO permitted limit of 0.3 mg/l [12] and the percent

samples above the limit is 39%. The concentration of the zinc ranges from 0.045 to 0.276 mg/l. The groundwater quality standard of zinc desirable limit is 3 mg/l and maximum permissible limit is 10 mg/l, and all samples are lower than the desirable limit [12].
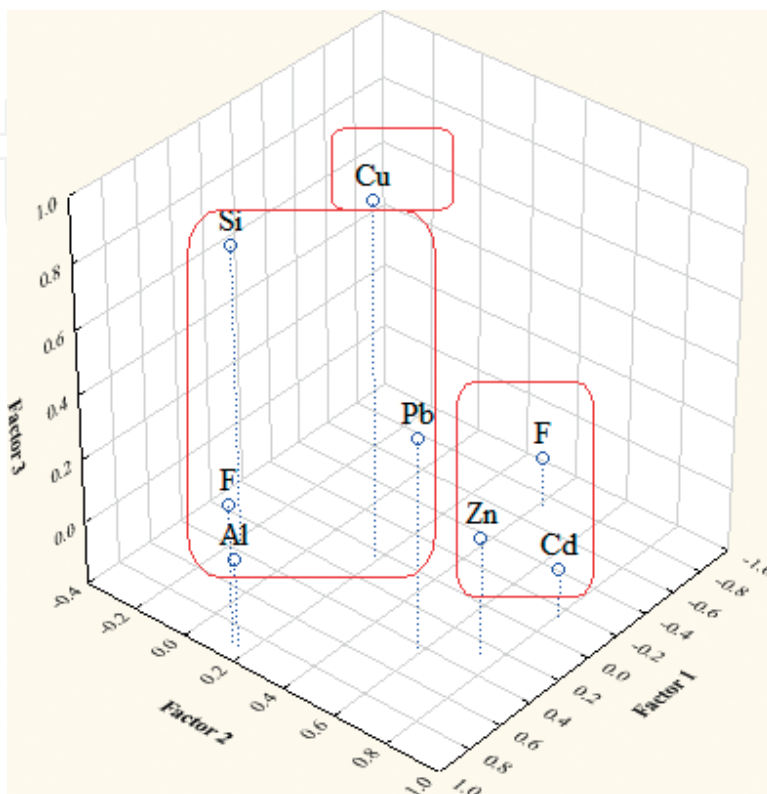

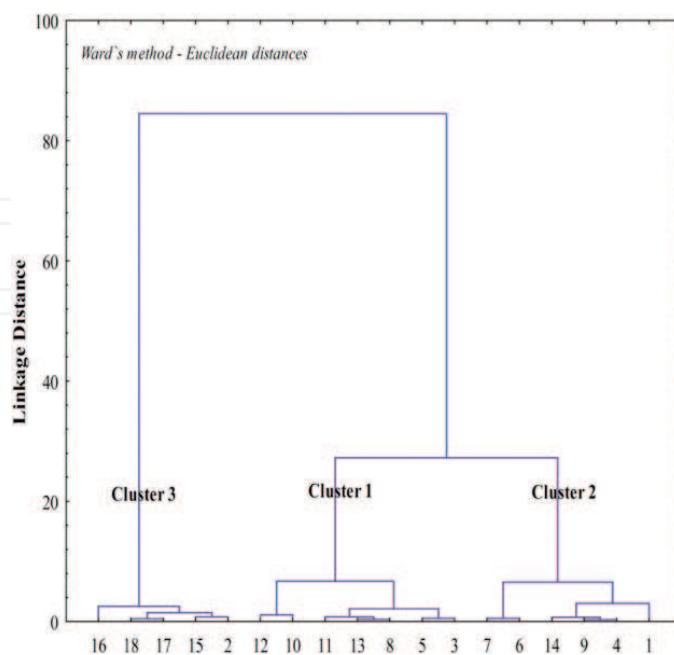**Figure 2.** FA results in the three-dimensional space: plot of loading of the first three factors.

Figure 3. Hierarchical cluster results or dendrogram obtained by CA of the groundwater samples.

	Cluster 1					Cluster 2					Cluster 3				
	Min	Max	Mean	SD	CV	Min	Max	Mean	SD	CV	Min	Max	Mean	SD	CV
EC	950	1540	1164	188	16	830	2730	1463	747	51	1340	2530	1836	489	27
T	14.0	18.0	15.6	1.5	9.7	14.0	17.5	16.6	1.3	7.7	14.0	17.0	15.9	1.3	8.4
pH	7.2	7.8	7.5	0.2	3.1	6.9	7.4	7.2	0.2	2.4	7.3	7.9	7.6	0.2	3.2
Al	0.010	0.060	0.037	0.015	40.278	0.030	0.090	0.057	0.023	41.260	0.040	0.090	0.062	0.023	36.780
Cd	0.016	0.165	0.063	0.051	81.045	0.037	0.125	0.076	0.032	41.771	0.009	0.152	0.060	0.057	94.821
Cu	0.056	0.364	0.219	0.097	44.034	0.089	0.311	0.196	0.086	43.942	0.213	0.430	0.326	0.089	27.271
F	0.031	0.128	0.073	0.033	45.940	0.031	0.358	0.133	0.126	94.271	0.017	0.339	0.203	0.137	67.502
Fe	0.188	0.373	0.278	0.066	23.576	0.182	0.499	0.304	0.124	40.596	0.055	0.378	0.164	0.130	79.412
Pb	0.027	0.101	0.058	0.025	42.274	0.029	0.292	0.102	0.096	94.334	0.017	0.193	0.109	0.073	67.210
Si	12.20	16.70	14.89	1.61	10.80	18.20	23.10	20.85	1.81	8.67	30.60	33.30	31.78	1.02	3.20
Zn	0.045	0.190	0.133	0.059	44.492	0.089	0.238	0.160	0.056	35.082	0.087	0.276	0.154	0.073	47.516

Table 4. Statistical summary of physicochemical parameters in the three clusters.

The third factor exhibits 13.7% of the total variance with positive loading on Cu. The concentration of the copper varies from 0.056 to 0.43 mg/l. The groundwater quality standard of copper maximum permissible limit is 2 mg/l. All groundwater samples are less then maximum permissible level of Cu [12].

3.2. Cluster analysis

Cluster analysis (CA) was applied to group objects (cases) into categories or clusters on the basis of similarities within a cluster and dissimilarities between different clusters with respect to distance between objects [13, 14]. Hierarchical agglomerative cluster analysis was performed on the normalized data set using Euclidean distances as a measure of similarity and Ward's method to obtain dendrograms. Three main clusters can be distinguished in the dendrogram shown in **Figure 3**. **Table 4** shows that the increases of electrical conductivity from the first cluster to the last cluster. Cluster analysis confirmed and completed the results obtained by factor analysis.

The first cluster was composed of the wells 3, 5, 8, 10, 11, 12 and 13, and concerns 39% of the total water samples. The mean of electrical conductivity for this cluster is 1164 μS/cm, which presented low concentrations of all heavy metals compared with others clusters (**Figure 4(a)** and **(h)**).
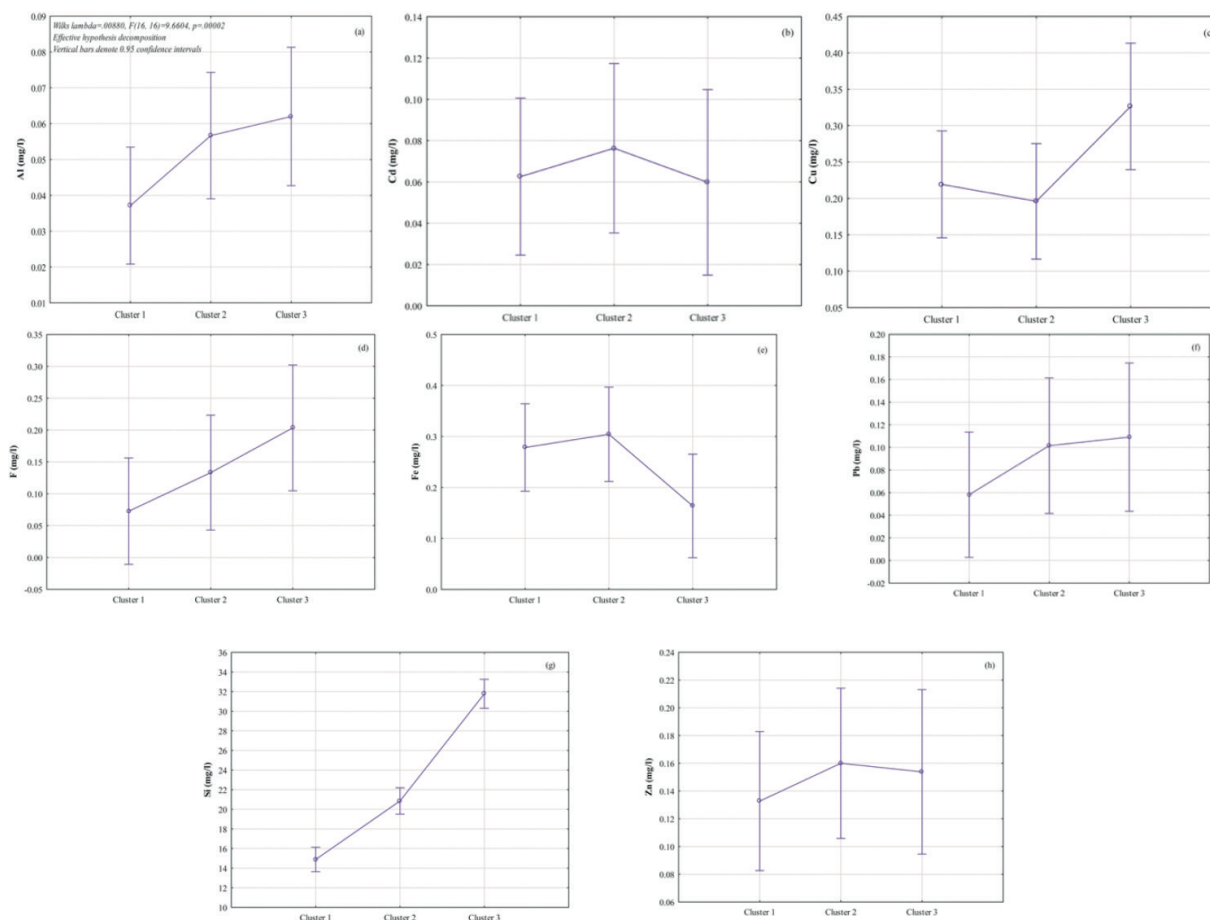

Figure 4. Plot of heavy metals in the three clusters.

The second cluster was represented by the wells 1, 4, 7, 9 and 14, and it occupies 33% of the total water samples (mean EC = 1463 μS/cm). This cluster included samples with the highest concentrations of Cd (0.076 mg/l), Fe (0.304 mg/l) and Zn (0.160 mg/l) (**Figure 4(b)**, **(e)**, and **(h)**).

The third cluster was included samples 2, 15, 16, 17 and 18 (28%), where the mean of EC is 1836 μS/cm. In this cluster, the samples were presented the highest concentrations of Al (0.062 mg/l), Cu (0.326 mg/l), F (0.203 mg/l), Pb (0.109 mg/l) and Si (31.78 mg/l) (**Figure 4(a)**, **(c)**, **(d)**, **(f)**, and **(g)**).

4. Pollution evaluation indices

The degree of pollution in groundwater samples were assessed employing two methods; degree of contamination (C_{deg}) and heavy metal evaluation index (HEI) as reported in the literature [15, 16].

The quality of groundwater was evaluated by calculating contamination index (C_{deg}). The degree of contamination is used as a reference of estimating the extent of metal pollution [17]. This index may be classified into three categories as follows: low ($C_{deg} < 1$), medium ($C_{deg} = 1–3$) and high ($C_{deg} > 3$) [15, 18, 19]. The contamination index was computed from the following equation:

$$C_{deg} = \sum_{i=1}^{n} C_{fi} \tag{1}$$

$$C_{fi} = \frac{C_{Ai}}{C_{Ni}} - 1 \tag{2}$$

where, "C_{fi}" is contamination factor for the ith component, "C_{Ai}" is analytical value for the ith component, and "C_{Ni}" is upper permissible concentration of the ith component (N denotes the "normative value").

The heavy metal evaluation index gives an overall quality of groundwater with respect to heavy metals [19]. This index was computed using the relationship:

$$HEI = \sum_{i=1}^{n} \frac{H_C}{H_{MAC}} \tag{3}$$

where, "H_C" and "H_{MAC}" are the measured value and maximum admissible concentration (MAC) of the ith parameter, respectively.

The estimated pollution evaluation indices for the selected heavy metals in the three clusters are shown in **Table 5**. In the first cluster, mean values of HEI and C_{deg} indices were observed to be 29 and 22 (**Table 5**), respectively, which indicated that the water samples of this cluster were contaminated with low degree of pollution by heavy metals, especially Cd and Pb [19]. The mean values of HEI and C_{deg} of the second cluster and the last cluster were respectively

		Cluster 1			Cluster 2			Cluster 3		
	MAC	Mean	HEI	C_{deg}	Mean	HEI	C_{deg}	Mean	HEI	C_{deg}
Al	30	37	1.2	0.2	57	1.9	0.9	62	2.1	1.1
Cd	3	63	21	20	76	25.3	24.3	60	20	19
Cu	2000	219	0.1	−0.9	196	0.1	−0.9	326	0.2	−0.8
F	1500	73	0.05	−0.95	133	0.1	−0.9	203	0.1	−0.9
Fe	300	278	0.9	−0.1	304	1	0	164	0.5	−0.5
Pb	10	58	5.8	4.8	102	10.2	9.2	109	10.9	9.9
Si	—	14,890	—	—	20,850	—	—	31,780	—	—
Zn	3000	133	0.04	−0.96	160	0.05	−0.95	154	0.05	−0.95
∑ HEI/C_{deg}		29	22		39	32		34	27	

MAC: maximum admissible concentrations.

Table 5. Description of pollution evaluation indices for selected heavy metals (μg/l) in groundwater samples.

39, 32, 34 and 27 (**Table 5**), revealing high level of pollution with Al, Cd and Pb. Overall, relatively higher heavy metals pollution is observed in the water samples of the second and third cluster than the first cluster.

5. Human health risk assessment

Human health risk assessment was defined as the processes of estimating the probability of occurrence of an event and the probable magnitude of adverse health effects over a specified time period [20, 21]. Exposure of human beings to the metals could occur via three main pathways including direct ingestion, inhalation and dermal absorption through skin; however, ingestion and dermal absorption are common routes for water exposure [22–25].

The numeric expressions for risk assessment have been obtained from USEPA Risk Assessment Guidance for Superfund (RAGS) methodology [22].

$$Exp_{ing} = \frac{C_{water} \times IR \times EF \times ED}{BW \times AT} \tag{4}$$

$$Exp_{derm} = \frac{C_{water} \times SA \times K_p \times ET \times EF \times ED \times CF}{BW \times AT} \tag{5}$$

where, Exp_{ing}: exposure dose through ingestion of water (μg/(kg day)); Exp_{derm}: exposure dose through dermal absorption (μg/(kg day)); C_{water}: concentration of metals estimated in groundwater (μg/l); IR: ingestion rate (2.2 l/day); EF: exposure frequency (365 days/year); ED: exposure duration (30 years); BW: average body weight (70 kg); AT: averaging time (25,550 days); SA: exposed skin area (18,000 cm^2); ET: exposure time (0.58 h/day); CF: unit conversion factor

(0.001 l/cm^3); and K$_p$: dermal permeability coefficient. These parameter values are taken from reference values or pooled from the statistical data of local population [23, 24].

The characterization of non-carcinogenic risks such as hazard quotients (HQ) and hazard index (HI) is carried out using USEPA guidelines [22, 23]:

$$HQ_{ing/derm} = \frac{Exp_{ing/derm}}{RfD_{ing/derm}} \tag{6}$$

$$HI_{ing/derm} = \sum_{i=1}^{n} HQ_{ing/derm} \tag{7}$$

where, HQ$_{ing/derm}$: Hazard quotient via ingestion/dermal route (unitless); HI$_{ing/derm}$: Hazard index via ingestion/dermal route (unitless); and RfD$_{ing/derm}$: ingestion/dermal reference dose (μg/kg/day).

It is generally accepted that HI below 1 is considered to mean no significant risk of non-carcinogenic effects, and if the value of cancer risk is between 10^{-4} and 10^{-6}, it is believed that the carcinogenic risk is acceptable [26, 27].

			Cluster 1		Cluster 2		Cluster 3	
	RfD$_{ing}$	RfD$_{derm}$	Exp$_{ing}$	Exp$_{derm}$	Exp$_{ing}$	Exp$_{derm}$	Exp$_{ing}$	Exp$_{derm}$
Al	1000	200	1.163	2.36E−03	1.791	3.64E−03	1.949	3.96E−03
Cd	0.5	0.025	1.980	4.03E−03	2.389	4.86E−03	1.886	3.84E−03
Cu	40	8	6.883	1.40E−02	6.160	1.25E−02	10.246	2.08E−02
F	60	60	2.294	4.67E−03	4.180	8.50E−03	6.380	1.30E−02
Fe	700	140	8.737	1.78E−02	9.554	1.94E−02	5.154	1.05E−02
Pb	1.4	0.42	1.823	1.48E−03	3.206	2.61E−03	3.426	2.79E−03
Zn	300	60	4.180	5.10E−03	5.029	6.14E−03	4.840	5.91E−03
			Cluster 1		Cluster 2		Cluster 3	
			HQ$_{ing}$	HQ$_{derm}$	HQ$_{ing}$	HQ$_{derm}$	HQ$_{ing}$	HQ$_{derm}$
Al			1.16E−03	1.18E−05	1.79E−03	1.82E−05	1.95E−03	1.98E−05
Cd			3.96E+00	1.61E−01	4.78E+00	1.94E−01	3.77E+00	1.53E−01
Cu			1.72E−01	1.75E−03	1.54E−01	1.57E−03	2.56E−01	2.60E−03
F			3.82E−02	7.78E−05	6.97E−02	1.42E−04	1.06E−01	2.16E−04
Fe			1.25E−02	1.27E−04	1.36E−02	1.39E−04	7.36E−03	7.49E−05
Pb			1.30E+00	3.53E−03	2.29E+00	6.21E−03	2.45E+00	6.64E−03
Zn			1.39E−02	8.50E−05	1.68E−02	1.02E−04	1.61E−02	9.84E−05
∑HI$_{ing/derm}$			5.50	0.17	7.32	0.2	6.61	0.16

Table 6. Summary of the health risk assessment for selected metals through ingestion pathway and dermal absorption in water samples.

The health risk assessment parameters for the selected heavy metals in the groundwater samples of the three clusters via oral and dermal routes were described in **Table 6**. In the three clusters, the estimated mean levels of Exp_{ing} and Exp_{derm} in the water samples are observed in the order of Fe > Cu > Zn > F > Pb > Cd > Al and Fe > Cu > F > Zn > Cd > Al > Pb, respectively. The results indicated that Fe, Cu, Zn and F are the major contributors to the ingestion and dermal exposures to the inhabitants, while Cd, Al and Pb are the least participants. Among the selected metals, Cd and Pb (HQ_{ing} > 1) posed adverse health risks and potential non-carcinogenic health risks to the inhabitants, while rest of the metals caused little or no adverse effects to the residents via ingestion route. However, the mean levels of HQ_{derm} for the selected metals are found to be lower than unity, indicating that the metals would not pose any adverse effect and non-carcinogenic health risk to the consumers via dermal contact.

Hazard index via ingestion intake (HI_{ing}) and dermal contact (HI_{derm}) are computed to assess the overall non-carcinogenic risk posed by selected metals via ingestion and dermal contact of water as a whole. Among the selected metals, Cd and Pb contributed the most to the mean value of HI_{ing} (6.48), suggesting that these metals deserved serious health concern via ingestion path. However, the mean value of HI_{derm} (0.18) is found to be less than unity, demonstrating that the selected metals posed little or no hazard to residents through dermal contact. Since the largest contributors to chronic non-carcinogenic risks were Cd and Pb in the present investigation, therefore, special attention should be paid to Cd and Pb management in the studied area.

6. Conclusion

In this study, the mean concentrations of heavy metals in groundwater sources in decreasing order was as follows: Si > Fe > Cu > Zn > F > Pb > Cd > Al. Factor analysis method identified three factors responsible for data structure explaining 75.69% of total variance in groundwater. Three major water clusters resulted from the cluster analysis. CA confirmed and completed the results obtained by FA. The mean values of HEI and C_{deg} indices indicated that the water samples of the first cluster were contaminated with low degree of pollution by heavy metals, especially Cd and Pb. The mean values of HEI and C_{deg} of the second and the third cluster revealing high level of contamination with Al, Cd and Pb. Non-carcinogenic health risk assessment was computed to assess the adverse health effects on the population. The hazard quotients (via ingestion) of Cd and Pb were found to be higher than the safe limits, posing threat to the consumers. However, no risk related to the dermal contact was associated with the measured metal levels. In the face of this type of pollution, which has an adverse effect on human health, a number of recommendations and guidelines have been drawn from this study to support the rational use of polluted areas, which are listed as follows:

Recommendations

1. Broad dissemination of the water code, in a vulgarized way, to sensitize citizens who act out of ignorance.

2. A research study and advocate in the field of pollution, because it presents a means of prevention.

3. Landfill and industrial waste discharge in these areas

4. Use of fertilizers in an abusive manner

5. The hydrological process and functions within the basins as well as in the larger terrestrial landscape where they occur must be taken in management.

Orientations

1. At the international level, there are guidelines to promote the inclusion of polluted areas in the management of shared watersheds.

2. At the national level, put in place processes of tight control and cross-sectoral harmonization of policy objectives and to raise awareness of the role and value of polluted areas.

3. The water sector must establish a dynamic political, legislative and institutional environment that takes due account of polluted areas to ensure that the sector has the capacity and information to participate constructively in the protection of polluted areas.

Author details

Lazhar Belkhiri[1]*, Ammar Tiri[1] and Lotfi Mouni[2]

*Address all correspondence to: belkhiri.la@gmail.com; belkhiri_laz@yahoo.fr

1 Laboratory of Applied Research in Hydraulics, University of Mustapha Ben Boulaid Batna, Algeria

2 Laboratoire de Gestion et Valorisation des Ressources Naturelles et Assurance Qualité, Faculté des Sciences de la Nature et de la Vie et Sciences de la Terre, Université de Bouira, Algeria

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