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Evaluation of trace metal element concentrations in sediments from sites receiving treated wastewater inputs in the Beni Mellal-Khenifra region, Morocco

Nadia Talbi[1], Abdelali Rouhi[2*], Mohamed Merzouki[1], Hasna Merzouki[1]

[1] Faculty of Sciences and Techniques, University Sultan Moulay Slimane, Beni Mellal, Morocco
[2] Regional Center for Education and Training, Beni Mellal, Morocco
* Corresponding author's e-mail: rouhiabdelali@gmail.com

ABSTRACT

Sediments from wastewater treatment plant sites are significant reservoirs of pollutants that can disrupt ecosystems. This study focused on assessing environmental pollution by trace metals, primarily originating from wastewater, in the Beni Mellal-Khénifra region of Morocco. Sediment samples were collected along the 0–25 cm horizon every two months throughout 2022 at three sites: Beni Mellal (BM), Azilal (AZ) and Khouribga (KH). Analyses were conducted by using inductively coupled plasma atomic emission spectrometry (ICP-AES). Sediment quality was evaluated by calculating several indices: the contamination factor (CF), the enrichment factor (EF), the geo-accumulation index (Igeo), the pollution load index (PLI), and the potential ecological risk index (RI). The results showed that the average abundance of metallic elements in the sediments followed the order: Fe > Zn > Cr > Cu > Pb > Ni > Cd. Significant spatio-temporal variations were also observed for all metallic elements except Zn. The calculation of various pollution indices (CF, EF and Igeo) revealed a strong anthropogenic contribution of Cd et all three sites, of Zn et the KH site, and of Cr at both the KH and AZ sites. Multi-element indices (PLI and RI) qualify the sediments as polluted and may present an ecological risk in the following order: KH > AZ > BM. The strong correlation between most of the trace metallic elements at the BM and AZ site suggests a similar origin. These findings indicate that sediment contamination is influenced by the nature and intensity of anthropogenic activities specific of each province of the region. Agricultural and mining activities were identified as the main sources of these elements.

Keywords: sediment, contamination, wastewater, trace metals, pollution indices.

INTRODUCTION

Similar to some developing countries, Morocco is facing significant anthropogenic pressures due to population growth, urbanization, and escalating demands spurred by economic development. The tremendous need of water, whether for domestic, industrial, or agricultural use, especially in recent drought period, has led authorities to view wastewater as a critical resource. This highlights the necessity of developing a comprehensive national wastewater treatment initiative (Azami, 2017). Consequently, various measures have been implemented and integrated to mitigate the negative effects of wastewater production on the environment. There is a necessity of establishing an extensive sanitation network and constructing treatment facilities to serve urban and industrial areas. As a result, a significant number of wastewater treatment plants have been built across the country (Amir, 2005, El Fels, 2014).

Most wastewater is classified based on its source: domestic, industrial, agricultural, or stormwater. Yet, the water gathered in a sanitation network contains suspended mineral and organic substances of various types and in highly variable concentrations. Despite the substantial volume of urban treated wastewater, it presents significant challenges, particularly due to its pollutant content. Among these contaminants, special attention has been given to trace metal elements due to their tendency to accumulate in different components of the ecosystem. This accumulation leads to contamination of soil and sediments, posing a toxicity risk to both living organisms and humans, primarily through the food chain (Mench and Baize, 2004, Bourrelier and Berthelin, 1998).

Soil and sediment contamination by trace metal elements (TME) in the Beni Mellal-Khénifra region has been widely researched as a significant environmental issue. Several studies have explored this topic, especially, those who focus on the contamination of sugar beet cultivation soils (Aallam et al., 2021), the analysis of open canal water quality (Barakat et al., 2012), the effects of the Beni Mellal public landfill on Oued Sabeq soils (El Baghdadi et al., 2015), the quality of soils irrigated with wastewater (Hilali et al., 2020, Barakat et al., 2019), and the pollution of agricultural soils in the phosphate-rich plateaus of Khouribga (Barakat et al., 2022).

In the Beni Mellal-Khénifra region, encompassing the provinces of Beni Mellal, Azilal, and Khouribga, urban wastewater is collected through a combined sewer system that channels both wastewater and stormwater into treatment plants. Once treated, the effluent is released into the natural environment. However, in addition to the residual pollutants from these plants, contamination also occurs due to drainage water during rainfall. This water carries particles from cultivated soils, as well as nearby public and industrial landfills. The aim of our study was to evaluate the levels of trace metal element contamination (Cd, Zn, Cu, Cr, Fe, Pb and Ni) in sediments sampled near wastewater discharge points from treatment plants.

MATERIALS AND METHODS

Study site description

Sampling was conducted at three polluted sites in the Beni Mellal-Khénifra region in central Morocco. All sites are located near the discharge points of urban wastewater from treatment plants: the Beni Mellal site (BM) (32°21’46”N; 6°22’29”W), the Azilal site (AZ) (31°56’18”N; 6°37’10”W), and the Khouribga site (KH) (32°50’31”N; 6°56’29”W) (Figure 1). The BM and AZ sites are affected by urban and agricultural disturbances, while the KH site is additionally impacted by mining activities (phosphates). The three sites are located approximately 100 kilometres apart.

Collection of sediment samples

Sediment samples were collected every two months during the year 2022 from the three study sites. Each sediment sample was a composite of three subsamples taken at a depth between 0 and 25 cm, then air-dried, sieved through a 2 mm mesh, and stored in polyethylene bags in the freezer for physicochemical analysis.

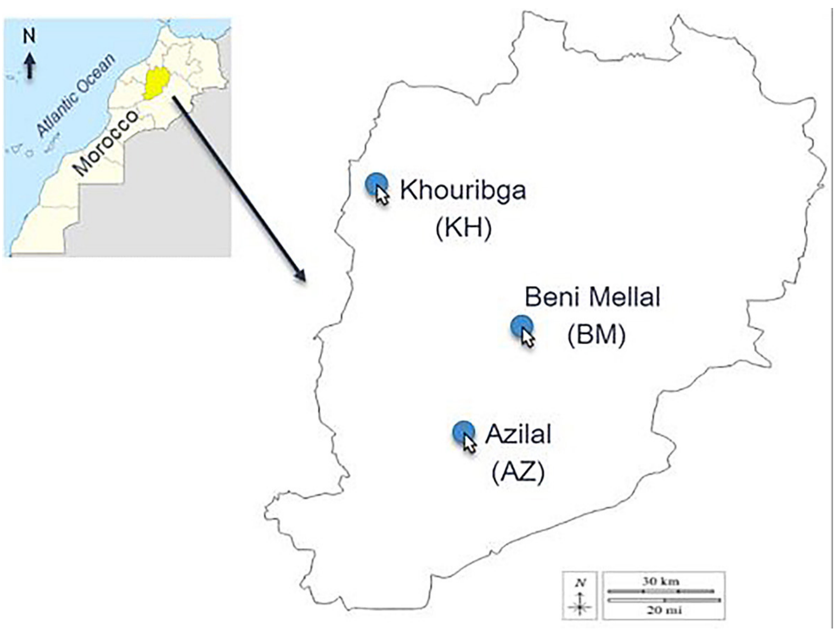

Figure 1. Location of the studied stations (AZ, BM and KH), Morocco

Physicochemical analyses

The pH and conductivity (EC) of the sediment were measured sequentially in an aqueous solution after one hour of agitation, using sediment/solution ratios of 1:2.5 (w:v) and 1:5 (w:v). Organic matter (OM) content was determined by first drying the samples in an oven at 105 °C for 24 hours, then burning them in a furnace at 375 °C for 16 hours. The sediment temperature was measured directly at the sampling site.

For trace metal element analyses, sediment samples were dried in an oven (60 °C) until completely dry. They were then ground and homogenized in a porcelain mortar. The metals were dissolved from approximately 0.3 g of sediment using a mixture of 2 ml hydrochloric acid and 5 ml nitric acid, heated in a sand bath at 110 °C until complete drying. The resulting residue was then suspended in a 1N hydrochloric acid solution (Charlou and Joanny, 1983). Three blank tubes, containing only the acids, were treated in the same manner as the sediment samples. The final solutions were adjusted to a volume of 20 ml with double-distilled water. Metal measurements were performed by ICP (inductively coupled plasma atomic emission spectrometry). The results are expressed in mg/kg of dry weight.

Evaluation of sediment contamination using pollution indices

To assess the sediment quality of the studied sites, we calculated various pollution indices. This analysis allows for the classification of the sites based on the level of contamination by trace metal elements (TME) and helps identify their potential sources. To do this, we calculated the contamination factor (CF), enrichment factor (EF), geo-accumulation index (Igeo), pollution load index (PLI), and potential ecological risk index (RI) (Table 1).

Table 1. Classification of the studied sediments according to pollution indices

Index	Formula	Classification	Sediment quality
CF	$CF = \frac{[M]}{[M]background}$ (1)	CF < 1	Low contamination
		1 ≤ CF < 3	Moderate contamination
		3 ≤ CF < 6	considerable contamination
		CF ≥ 6	Very high contamination
EF	$EF = \frac{[M]/[Fe]sample}{[M]/[Fe]background}$ (2)	EF ≤ 1	Non- enrichment
		1 ≤ EF < 3	Minor enrichment
		3 ≤ EF < 5	Moderate enrichment
		5 ≤ EF < 10	Moderately high enrichment
		10 ≤ EF < 25	High enrichment
		25 ≤ EF	Very high enrichment
Igeo	$Igeo = Log2\left[\frac{[M]}{1{,}5 \times [M]background}\right]$ (3)	Igeo ≤ 0	Unpolluted
		0 < Igeo ≤ 1	Unpolluted to moderate polluted
		1 < Igeo ≤ 2	Moderately polluted
		2 < Igeo ≤ 3	A moderately to highly polluted
		3 < Igeo ≤ 4	Heavily polluted
		4 < Igeo ≤ 5	Heavily polluted to extremely polluted
		5 < Igeo	Extremely polluted
PLI	$PLI = (CF1 \times CF2 \times \ldots CFn)1/n$ (4)	PLI = 0	Unpolluted sediment
		PLI = 1	Pollution is present
		PLI > 1	Deteriorated site quality
RI	$RI = \sum_{i=1}^{i=n} Ti \times CFi$ (5)	RI≤ 90	Low risk
		90 < RI ≤ 180	Moderate risk
		180 < RI ≤ 360	Considerable risk
		360 < RI ≤ 720 RI > 720	High risk Very high risk

The contamination factor (CF) proposed by Tomlinson et al. (1980) expresses the ratio between the concentration of each metal [M] in the sediment and its geochemical background value $[M]_{background}$. It is calculated using the Equation 1, and four contamination classes are determined by this index.

The enrichment factor (EF) quantifies anthropogenic inputs of trace metal elements (TME) to the concentrations observed in the sediment. It is obtained by dividing the concentration of the studied element [M] by that of a reference element, whose concentration is considered to be little or not influenced by anthropogenic activities (Mazurek et al., 2016). In this study, Fe was used as the reference. The calculation formula is presented by Equation 2.

The geo-accumulation index (Igeo) proposed by Müller (1969) establishes a relationship between the measured concentration of a trace metal element in the sediment and its local geochemical background concentration, including a lithological correction factor of 1.5 for metal elements. It is calculated using Equation 3.

The pollution load index (PLI) allows for estimating the overall degree of contamination of the sediments at the studied site, based on the total concentration of all the metals studied. Equation 4, developed by Tomlinson et al. (1980), is used to calculate this index.

The potential ecological risk index (RI) is used to quantitatively express the potential ecological risk of trace metal elements (TME) and link their ecological and environmental effects to their toxicological effects. It is expressed as suggested by Håkanson (1980) according to Equation 5.

RESULTS

Physicochemical parameters of the sediments

The results of physicochemical parameters of the sediment proprieties are presented in Table 2. The electrical conductivity (EC) analyses conducted on the three sites show that the values range from 0.31 dS/m to 2.73 dS/m for the BM and KH sites, respectively. According to the SSDS (1993) criteria, all three sediments are classified as non-saline to slightly saline. Regarding pH, according to SSDS (1993), the KH station has a neutral pH. In contrast, the sediment at the BM site is slightly alkaline, while the sediment at the AZ site can be classified as slightly alkaline to alkaline. In the study area, organic matter values range from 1.62% to 15.53% for the BM and KH sites, respectively. The sediments from the KH site are considered highly organic, while those from the BM site are classified as poor to moderately rich in organic matter. The AZ site, on the other hand, is poor in organic matter.

Spatial and temporal variation of trace metal elements (TME) in the sediments

The results related to the concentration of TME in the sediments of the three studied sites are shown in Table 3.

The average concentrations of Cd in the sediments of the three sites ranged from 1.37 ± 0.02 mg/kg (in September at BM) to 10.20 ± 0.15 mg/kg (in July at KH). The analysis of variance (ANOVA) revealed highly significant and significant differences based on the sites ($F = 44.64$; $p < 0.000$) and the sampling dates ($F = 4.31$; $p = 0.0025$). Comparison of the annual average concentrations showed that the sediments at the KH

Table 2. Physicochemical parameters of sediments from the study sites

Parameters		Sites		
		AZ	BM	KH
T (°C)	Mean	16.90	22.38	18.63
	Min–Max	7–25	18–25	13.50–24
pH	Mean	7.42	7.58	6.94
	Min–Max	6.87–8.29	7.40–7.83	6.62–7.13
OM (%)	Mean	4.03	2.88	11.40
	Min–Max	2.77–5.14	1.62–3.77	8,33–15.53
EC (mS/cm)	Mean	0.61	0.54	1.73
	Min–Max	0.34–0.99	0.31–0.73	0.88–2.73

Table 3. basic statistical data of TME measurements at the sediment level of the three study sites and in other local studies

Sampling site	Cd	Pb	Zn	Cu	Cr	Ni	Fe (×10³)	References
(AZ)								Present work
Min	2.81	6	34.49	12.33	43.61	17.18	29.65	
Max	5.75	39.63	116.97	28.62	61.73	26.86	41.31	
Mean	4.31	21.47	77.44	22.23	53.63	21.19	35.13	
SD	(1.22)	(15.07)	(30.28)	(6.79)	(7.48)	(3.67)	(5.14)	
(BM)								
Min	1.37	2.26	28.72	9.05	37.23	11.51	16.94	
Max	4.35	45.64	190.39	66.70	177.66	22.39	33.64	
Mean	3.15	21.4	84.05	24.13	74.09	16.12	27.41	
SD	(1.08)	(15.19)	(56.44)	(21.6)	(52.72)	6.74	(6.15)	
(KH)								
Min	4.90	27.73	211.52	26.76	54.83	18.30	18.39	
Max	10.20	56.53	419.64	74.49	108.89	35.34	41.55	
Mean	7.23	47.83	315.18	53.4	83.13	27.27	33.51	
SD	(1.78)	(10.49)	(85.61)	(18.42)	(17.87)	5.78	(8.32)	
Local studies								
Beet field soils, Beni Mellal	<2	30.5–55	12–26	14-22.5	48.33–60	34.67–45.33	–	Aallam et al. (2021)
Agricultural soils, Beni Amir	2.5–40.08	3.4–134.7	24.5–1272	1.46–191.2	16.07–294	–	–	Oumenskou et al. (2018)
Water from irrigation canals	0–0.8	0.6–5	–	–	2.5–10	–	–	Barakat et al. (2013)
Sludge from the Khouribga WWTP	3.28	107.53	1386.67	147.43	27.82	–	11.213	Aadraoui et al. (2018)
Oued Day, Beni Mellal	4.89	109.66	75.98	87.40	77.77	42.33	26.248	Hilali et al. (2020)
Oued Sabeq, Beni Mellal	3.53	294.36	–	162.98	66.72	–	2.11%	El Beghdadi et al. (2015)
Raw water from the Azilal WWTP	–	0.66–5.62	1.06–6.63	0.13–3.13	–	–	1.23–7.37	Azami Idrissi et al. (2015)
Standards								
*Local Background	0.85	32.45	43.76	31.4	25.21	50	12.14	Oumenskou et al., (2018)
*Earth's crust	0.6	14	75	50	100	58	41	Taylor (1964); Turhan et al., (2020)
*World average	0.41	27	70	38.9	59.5	29	–	Kabata-Pendias (2011)
*FAO/WHO	3	100	300	100	100	50	–	Chiroma et al., (2014)

site were the most contaminated (7.23 ± 1.78 mg/kg), followed by those at the AZ site (4.31 ± 1.22 mg/kg) and the BM site (3.15 ± 1.08 mg/kg). The highest (419.64 ± 18.67 mg/kg) and lowest (28.72 ± 3. mg/kg) concentrations of Zn were recorded in November at KH and in September at BM, respectively. The differences in the average concentrations of this metal were found to be highly significant between the sites (F = 93.19; p < 0.000) and not significant between the months (F = 1.19; p = 0.327). The sediments at the KH site had the highest annual average values (315.18 ± 85.61) compared to the BM site (84.05 ± 56.44) and the AZ site (77.44 ± 30.28). A highly significant variation in copper (Cu) concentrations was observed based on the sampling site (F = 21.49; p < 0.000) and sampling date (F = 5.49; p < 0.000). The spatial effect is characterized by the highest level (53.40 ± 18.42 mg/kg) recorded at the KH site, an intermediate level (24.13 ± 21.60 mg/kg) observed at the BM site, and the lowest level (22.23 ± 6.79 mg/kg) recorded at the AZ site.

As for Pb, the highest (56.53 ± 5.18 mg/kg) and lowest (2.26 ± 0.78 mg/kg) concentrations were recorded at the KH and BM sites, respectively. At the KH site, the annual average Pb concentration was 47.83 ± 10.49 mg/Kg, and at the BM site, this value was 21.40 ± 15.19 mg/Kg. A highly significant variation in these concentrations based on the site ($F = 23.41$; $p < 0.000$) and sampling date ($F = 6.04$; $p < 0.000$) was also observed. The highest concentration of Ni (35.34 ± 1.71 mg/Kg) was recorded in the sediments of the KH site in July, while the lowest concentration (11.51 ± 0.66 mg/Kg) was recorded at the BM site in March. The differences in Ni concentrations were found to be highly significant between the sites ($F = 25.88$; $p < 0.000$), and between the months ($F = 4.28$; $p = 0.0027$).

Iron (Fe) also showed a significant variation based on the site ($F = 6.46$; $p = 0.0032$) and sampling date ($F = 9.75$; $p < 0.000$). The maximum concentration (41.55 ± 3.02 mg/Kg) was observed in July at the KH site, while the minimum concentration (16.84 ± 3.41 mg/Kg) was recorded in September at the BM site. The chromium (Cr) concentration in the sediments of the studied sites showed a significant variation according to the site ($F = 4.33$; $p = 0.0184$) and a highly significant variation based on the sampling period ($F = 7.98$; $p < 0.000$). The highest concentration (177.66 ± 3.92 mg/Kg) was recorded in July at the BM site, while the lowest (37.28 ± 8 mg/Kg) was observed at the same site in September. The annual average concentrations were 83.13 ± 17.87 mg/Kg at KH, 74.09 ± 52.72 mg/Kg at BM, and 53.63 ± 7.48 mg/Kg at AZ.

Pollution indices for trace metals

The results of the CF calculations are presented in Figure 2. Cd and Zn showed the highest values for this index (8 and 7.5, respectively) at the KH site, which was found to be highly contaminated. Moderate to considerable contamination of the sediments at the AZ and BM sites by Zn (ranging from 1.77 to 1.99) and Cd (ranging from 3.70 to 5.07) was also observed. However, the sediments at AZ and BM were only weakly contaminated with Cu, Pb, and Ni ($0.32 \leq CF \leq 1.70$). All sites were considered moderately contaminated with Cr and Fe.

The values of the EF for the metal elements are shown in Figure 3. The results revealed that Cd displayed the highest values, while Ni showed the lowest. On average, the enrichment of the sediments in trace metals appears to follow the order: Cd > Cr > Zn > Cu > Pb > Ni. Cd, Zn, and Cr exhibited EF values greater than 1 at certain sampling sites, indicating minor enrichment of the affected soils. This enrichment was observed across all three sites for Cd, at the KH site for Zn, and at the BM and AZ sites for Cr. However, the sediments at the various sites were naturally enriched with Cu, Pb, and Ni.

The Igeo are presented in Figure 4. The average Igeo values at the three sites follow the descending sequence: Cd > Fe > Zn > Cr > Cu > Pb > Ni. The highest average values of this index were recorded at the KH site for Cd, Zn, and Cr, followed by the AZ site for Cd and Fe. According to these results, the KH site is classified as moderately to heavily polluted by Cd and Zn, and moderately polluted by Cr and Fe. In contrast, the AZ and BM sites are moderately polluted by Cd and Fe, and not polluted to moderately polluted by Cr. Additionally, all three sites showed no pollution from Pb and Ni.

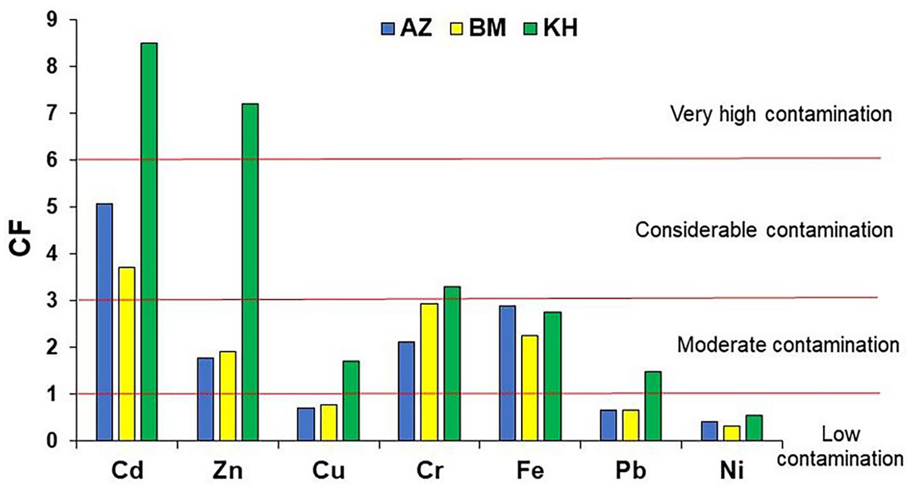

Figure 2. Variations in the Contamination Factor (CF) in the sediments of the three studied sites

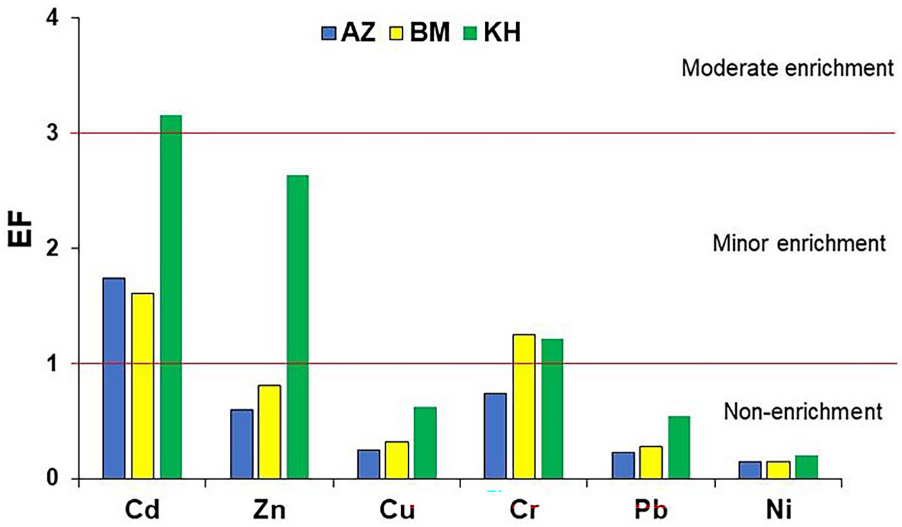

Figure 3. Variations in the Enrichment Factor (EF) in the sediments of the three studied sites

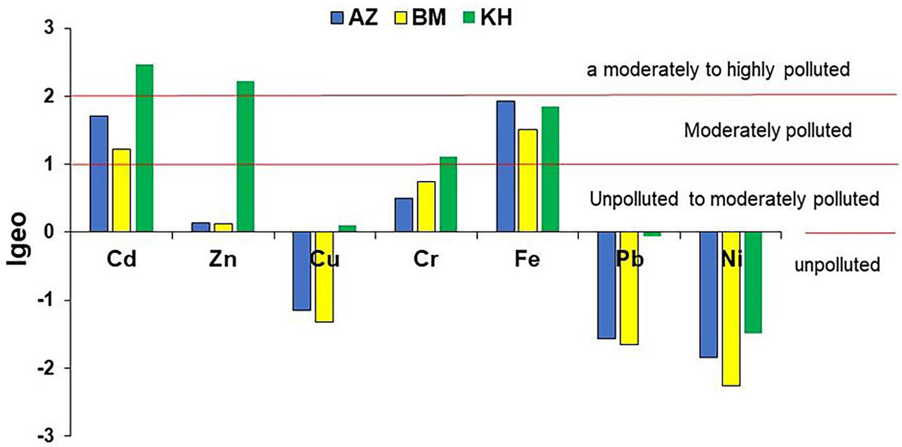

Figure 4. Variations in the Geo-accumulation Indices (Igeo) in the sediments of the three studied sites

The PLI and RI indices for the sediments of the three sites were calculated, and their values are summarized in Table 4. A PLI value greater than 1 indicates a deterioration in the quality of all three environments studied. Among these, the KH site showed a more pronounced deterioration compared to the other two sites, AZ and BM, which did not show a significant difference. Furthermore, the IR index values indicated a moderate risk for the AZ and BM sites, while the KH site presented a considerable risk.

Study of the correlations between trace metal elements

In order to establish potential correlations between the different trace metal elements (Cd, Zn, Cu, Cr, Fe, Pb, and Ni) analysed in the sediments from the three studied sites, the Spearman method was used. The analysis of the correlation matrices of the trace metal elements in the sediments of the AZ, BM, and KH sites reveals several trends. At the AZ site (Table 5), a strong correlation is observed between the metal elements, except for Cr and Ni, which are only significantly related to Fe and to each other. An absence of correlation between Zn and Cd was also observed. For the sediments from the BM site (Table 6), the analysed trace metals show significant positive correlations, except for Ni, which is only correlated with Cr. The trace metals in the sediments from the KH site (Table 7) show a stronger correlation, except for those between Zn and Cu on one hand, and between Zn and Cd on the other. However, Ni only shows a correlation with Cr.

Table 4. Average values of the metal pollution load index (PLI) and the ecological risk index (RI) of the three study sites. Sampling sites with the same letter are not statistically different at the 95% threshold

Indices	Sites		
	AZ[a]	BM[a]	KH[b]
PLI	1.36 ± 0.37	1.28 ± 0.66	2.56 ± 0.57
RI	172.95 ± 46.52	123.19 ± 47.93	292.93 ±70.48

Table 5. Pearson correlation matrix for the trace metal analysed in the sediments of (AZ) site

Elements	Cd	Zn	Cu	Cr	Pb	Ni	Fe
Fe	0.678*	0.803**	0.707*	0.706*	ns	0.818**	1
Ni	ns	ns	ns	0.908**	ns	1	
Pb	0.565*	0.836**	0.863**	ns	1		
Cr	ns	ns	ns	1			
Cu	0.844**	0.872**	1				
Zn	ns	1					
Cd	1						

Note: ns: not significant; * : $p < 0.05$; ** : $p < 0.01$.

Table 6. Pearson correlation matrix for the trace metal analysed in the sediments of (BM) site

Elements	Cd	Zn	Cu	Cr	Pb	Ni	Fe
Fe	0.836***	0.673**	0.578*	0.616**	0.618**	0.648**	1
Ni	ns	0.575*	0.553*	0.632**	ns	1	
Pb	0.800***	0.924***	0.852***	0.864***	1		
Cr	0.685**	0.985***	0.985***	1			
Cu	0.710**	0.973***	1				
Zn	0.769***	1					
Cd	1						

Note: ns: not significant; * : $p < 0.05$; ** : $p < 0.01$.

The results of the dendrogram analysis from a hierarchical clustering analysis (Bray-Curtis coefficient), based on the trace metal concentrations in the sediments, are shown in Figures 5 and 6. Two distinct clusters with a high level of similarity (> 89%) appear in the studied region. The first group consists of the BM site, while the second group includes the AZ and KH sites. The clustering also revealed that the metals are grouped into four distinct clusters: the first includes Fe, the second includes Cd, the third includes Zn, and the fourth includes Cr, Ni, Pb, and Cu. A strong

Table 7. Pearson correlation matrix for the trace metal analysed in the sediments of (KH) site

Elements	Cd	Zn	Cu	Cr	Pb	Ni	Fe
Fe	0.669**	0.870***	0.592**	0.947***	0.811***	0.935***	1
Ni	0.762***	0.927***	0.629**	0.986***	0.714**	1	
Pb	0.752***	0.541*	0.800***	0.777***	1		
Cr	0.799***	0.897***	0.679**	1			
Cu	0.901***	ns	1				
Zn	0.490*	1					
Cd	1						

Note: ns: not significant; * : $p < 0.05$; ** : $p < 0.01$.

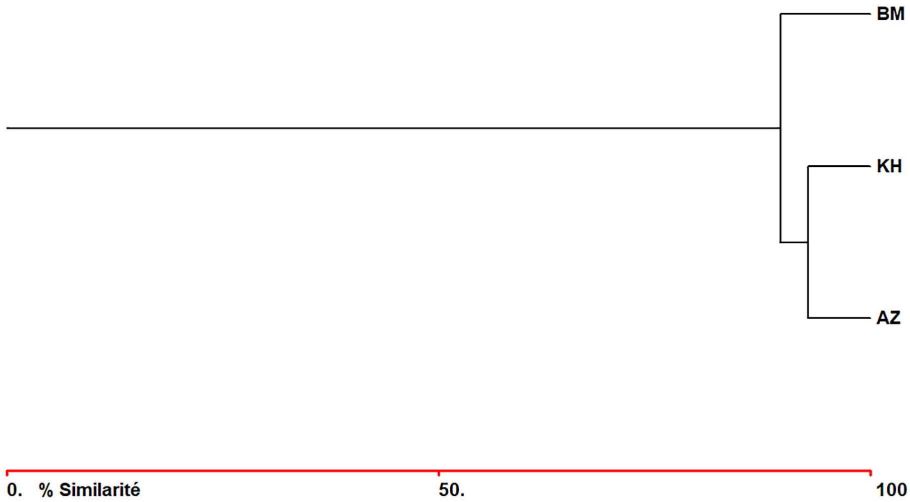

Figure 5. Dendrogram resulting from a hierarchical clustering analysis (Bray Curtis coefficient) of the three sampling sites in the sediments of the three studied sites

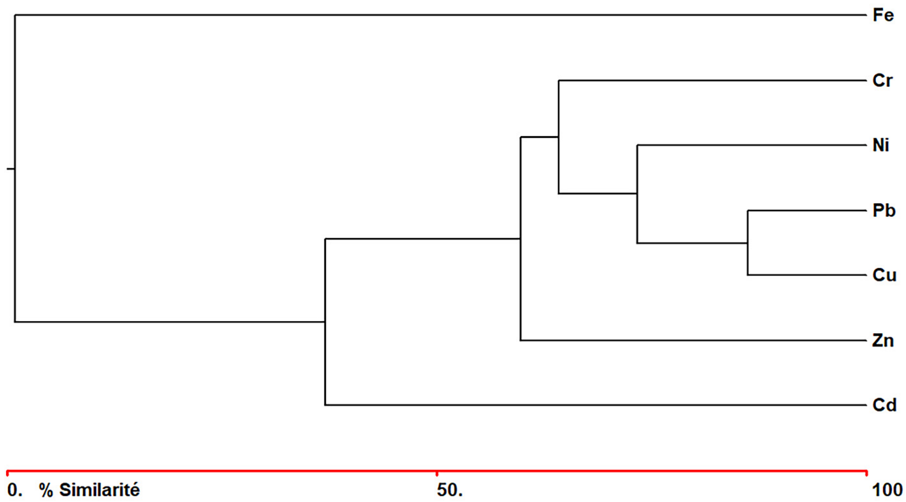

Figure 6. Dendrogram resulting from a hierarchical clustering analysis (Bray Curtis coefficient) of trace metal elements analyzed in the sediments of the three studied sites

and clear similarity (85.81%) between Pb and Cu suggests that these two metals originate from the same source, while Zn seems to have a different origin. The observed similarity between Ni and Pb (70.85%) as well as between Ni and Cu (73.17%) also indicates that these metals share a common origin, distinct from that of Fe and Cd.

DISCUSSION

The average values of Cd, Zn, Cu, Cr, Ni, Pb, and Fe in sediment samples collected from the three studied sites vary depending on the specific metal. Their quantitative distribution in the sediments of the three sites follows the decreasing hierarchy: Fe > Zn > Cr > Cu > Pb > Ni > Cd. This order of abundance is not only linked to the natural abundance of these elements but also suggests an anthropogenic contribution. However, this order differs from that observed for the local geochemical background (Oumenskou et al., 2018), which shows the following hierarchy: Fe > Ni > Zn > Pb > Cu > Cr > Cd. Similarly, the sequence observed by Hilali et al. (2020) in samples from sediments irrigated by wastewater from the Oued Day is as follows: Fe > Pb > Cu > As > Zn > Cr > Ni > Cd. Several authors have shown that total trace metal concentrations in sediments and soils vary depending on the soil type, the metal element, and the sources of contamination (Baize, 2009, Kao et al., 2007, Smouni et al., 2010, Lambiénou et al., 2020, Barakat et al., 2022).

Significant spatio-temporal variations were also observed for all metal elements except for Zn (which showed no significant temporal variation). The spatial distribution showed that the highest annual average concentrations were found in the sediments of the KH site, except for Fe, while the BM site ranked second for Zn, Cu, and Cr. On the other hand, the lowest concentrations of trace metals were detected at the AZ site.

The lack of temporal variation of Zn in the sediments of the three study sites can be explained by its speciation. Indeed, Zn is immobilized by adsorption on minerals such as carbonates, iron oxides and clays, which contributes to the stability of its concentration in the sediments. In addition, organic matter can form complexes with Zn, thus reinforcing this stability (Damme et al., 2010; Hao-Qin et al., 2024). These conditions are met in the treated wastewater spreading areas.

The comparison of the concentrations of the trace metals analysed in the sediments from different sites in the study area with certain standards and some data from the literature is presented in Table 4. In general, the Cd concentrations at the three sites significantly exceed the pollution thresholds set by WHO, AFNOR, and FAO standards, as well as the values for the Earth's crust and the geochemical background. Similarly, the sediments are considered contaminated by Zn compared to the geochemical background values, Earth's crust, and global average. Additionally, at the Khouribga site, the concentrations of Pb and Cu also exceed the average values of the geochemical background, Earth's crust, and the global average. The concentrations of Cd, Cr, and Fe measured by Aadraoui et al. (2018) in the Khouribga wastewater treatment plant (WWTP) sludge is lower than those obtained in our study on the sediments of the treated wastewater application area at the same station (Figure 4). These results can be explained by the fact that pollution from these three elements comes from a source other than treated wastewater. Indeed, Cd contamination varies according to the geographical specificities and anthropogenic activities specific to each region. In the province of Khouribga (KH), which is home to one of the largest phosphate deposits in the world, the phosphate industry is a major source of pollution, contributing to the contamination of surrounding soils and sediments. On the other hand, in the provinces of Béni Mellal and Azilal, which are heavily agricultural, the intensive use of phosphate fertilizers and pesticides is the main factor of contamination. These substances, applied to cultivated soils, are likely to be carried to neighboring ecosystems during rainfall, thus increasing the dispersion of Cd in the environment.

In contrast, the concentrations of Zn, Cu, and Pb are higher in the sludge than at our sampling site. Indeed, during treatment and decantation, the concentration of chemical contaminants increases in the sludge compared to that observed in raw wastewater (El Fels, 2014, Milik et al., 2017). Furthermore, Aadraoui (2020), through a leaching test carried out on sludge from the same plant, revealed that the concentrations of several trace metal elements (TME) in the eluate, such as copper, lead and zinc, remain below 1 mg/kg. This could explain the lower levels of these metals in the sediments of the studied site, compared to those measured in the sludge from the Khouribga wastewater treatment plant.

The calculation of various pollution indices (FC, FE, and Igeo) revealed a strong anthropogenic contribution for Cd in the sediments of the three sites, for Zn in the sediments of the KH site, and for Cr in the sediments of the KH and AZ sites. However, the multi-element indices (PLI and IR) show that all the soils studied are polluted (PLI > 1) and may pose an ecological risk, with the magnitude of the risk following the order: KH > AZ > BM. This result suggests that sediment contamination depends on the nature and extent of the anthropogenic activities characteristic of each province in the region. Indeed, Khouribga (KH) is a major mining city, suggesting that the phosphate industry, which is the primary economic activity, is the main source of metal pollution in the soil. Several authors who have worked on the discharges from the phosphate industry in Morocco agree that the source of sediment and living organism contamination by trace metals is phosphate gypsum, especially rich in Cd, Zn, and Cr (Fersiwi, 2007, Rouhi, 2013, Merzouki, 2009, Sif, 2002). Consequently, these elements may have harmful effects on organisms living in these sites. In addition to the trace metals from treated urban wastewater, the sediments at the KH site may also be contaminated by leachate from the city's municipal landfill, located a few hundred meters from the study site.

The sampling point for the BM site is located at the Day River. In addition to treated wastewater from the treatment plant, this location also receives a significant amount of untreated wastewater, as well as washing water and mineral and organic waste from the city's scrap, not to mention the garbage from the wild dumps in the surrounding neighborhoods. Similarly, the sediment sampling station at the AZ site is located in the bed of the Oued Alili, which runs through the city of Azilal. This river receives solid waste as well as raw wastewater, often mixed with rainwater during heavy precipitation. This explains the significant peak in trace metals observed in July 2022, a period marked by rainfall. Beni Mellal (BM) and Azilal (AZ) are both urban and agricultural areas, suggesting that these elements could be introduced into the soil due to agricultural activities such as the excessive use of chemical fertilizers, herbicides, and insecticides. These elements would eventually end up in the natural environment during rainfall. Oumenskou (2018) had pointed out this close relationship between agricultural activities in the region and the excessive inputs of trace metals.

The strong correlation between most of the trace metals in BM and AZ indicates their similar origin. This observation was also made by El Hamzaoui et al. (2020) on soil samples collected from the Beni-Moussa area, cultivated with beets and other agricultural products. The trace metals present in the sediments of the studied sites also come from wastewater collected by the sewage systems. Indeed, Barakat et al. (2019) reported that soil irrigation with wastewater in the Beni-Mellal region contributed to their enrichment with Cd, Cr, Cu, Zn, and Fe (moderate to considerable enrichment).

It should also be noted that the presence of toxic elements such as trace metals in the sediments of treated wastewater disposal sites by wastewater treatment plants was likely caused by materials brought in by stormwater, street cleaning, and erosion of the piping system, due to domestic detergents or reagents added during wastewater treatment (Tantawy et al., 2012, Cusido and Soriano, 2011).

CONCLUSIONS

This study aimed to assess the contamination status of trace metal elements (Cd, Zn, Cu, Cr, Pb, Fe and Ni) in sediments from three wastewater spreading areas associated with treatment plants in the Beni Mellal-Khenifra region. The results obtained show that the values of trace metal element (TME) concentrations vary depending on the metal element, the site and the sampling date. The highest average annual concentrations were recorded at the Khouribga site (KH), except for Fe. The Beni Mellal site (BM) is stood out by relatively high concentrations of Zn, Cu and Cr, thus occupying the second position. In contrast, the lowest concentrations were recorded at the Azilal site (AZ). Our study revealed that some elements exceeded the standard levels, especially for Cd and Zn. The pollution indices (CF, EF and Igeo) showed anthropogenic inputs of Cd in the sediments of the three sites, of Zn in the KH site and of Cr in the KH and AZ sites. The calculation of the PLI and RI indices shows that all the sediments analyzed are polluted (PLI > 1) and may present a risk to the natural environment. Treated wastewater is not the only source of contamination by TME, agricultural and mining activities and leachates from the public landfill also contribute to this enrichment. This sediment pollution represents a significant risk of contamination of water resources, human health, and the ecosystem, hence the need to implement a sediment contamination monitoring plan and to consider decontamination.

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