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Research Article

Assessing Heavy Metals Accumulation in the Leaves and Sediments of Urban Mangroves (*Avicennia marina* (Forsk.) Vierh.) in Bahrain

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Accumulation of 8 heavy metals (HM), Cr, Cu, Fe, Mn, Mo, Ni, Pb, and Zn, was assessed in the leaves and sediments of gray mangrove (*Avicennia marina* (Forsk.) Vierh.) at three sites along Tubli Bay using Inductive Coupled Plasma Analyzer. The results showed no significant differences in HM contents in the leaves of mangroves between the sites except for Mo and Zn. HM concentrations (mg L^{-1}) in leaves were in the following order: Fe > Zn > Mn > Mo > Cu > Ni > Cr. Significant differences existed between sediment content of Tubli site and the other two sites with regard to Cr, Cu, Pb, and Zn due to direct exposure of the site to wastewater outfalls. A general trend of decline in HM concentrations was noticeable from the top towards deeper sediment layers in all the sites of the study area. Concentration of HM in sediment was in the following order: Fe > Mn > Zn > Cu > Cr > Pb > Ni. HM concentration in sediments far exceeded the amount present in the leaves of the mangrove except for Ni. The amounts of HM in mangrove sediments were comparable to regional findings and fell within known sediment quality guidelines.

1. Introduction

Accumulation of heavy metals (HMs) in natural ecosystems poses threats to human health and biodiversity due to their persistence and toxicity [1–4]. Worldwide, coastal and marine ecosystems are subject to HM pollution from municipal wastes and runoffs from agriculture and industrial sources [5–10]. Coastal vegetation plays a major role in trapping and storing these pollutants [11–13]. In this context, mangrove (*Avicennia marina* (Forsk.) Vierh.) grows in the intertidal zones along the eastern coastlines of the Arabian Peninsula, forming discrete communities in several locations [14]. In Bahrain, the species grows naturally in only four localities along the coastlines of Tubli Bay measuring 0.5 Km^2 in the total area [15, 16]. Mangroves are considered as one of the most critical habitats in the country [17, 18]. Recently, the species was classified as critically endangered in Bahrain as its area dwindled by more than 60% in the last three decades [18].

Some researchers have reported the importance of mangrove ecosystems in trapping and storing HM in sediments and plant tissues [19, 20]. Undoubtedly, mangrove trees proportionally contribute to HM sequestration in their system [21]. Mangrove shoots and stems were second to sediment in storing HM [22, 23]. However, organic matter-rich sediments underlying mangroves are the main storage body for HM. Variations in mangrove ecosystems to store HM exist between regions due to physical and chemical properties of sediment, the types of anthropogenic activities involved, and the sampling approach of the area [24–29]. For instance, high salinity affects the sediment capacity to retain HM [30]. Various studies have indicated that the concentrations of HM in the sediment of the Arabian Gulf are comparable to other regions of the world. Nevertheless, hot spots exist in some localities, influenced by urban and industrial centers [31]. On the other hand, higher values of Pb, Cr, and Ni along the Iranian coasts were reported compared to mean world sediment [32].

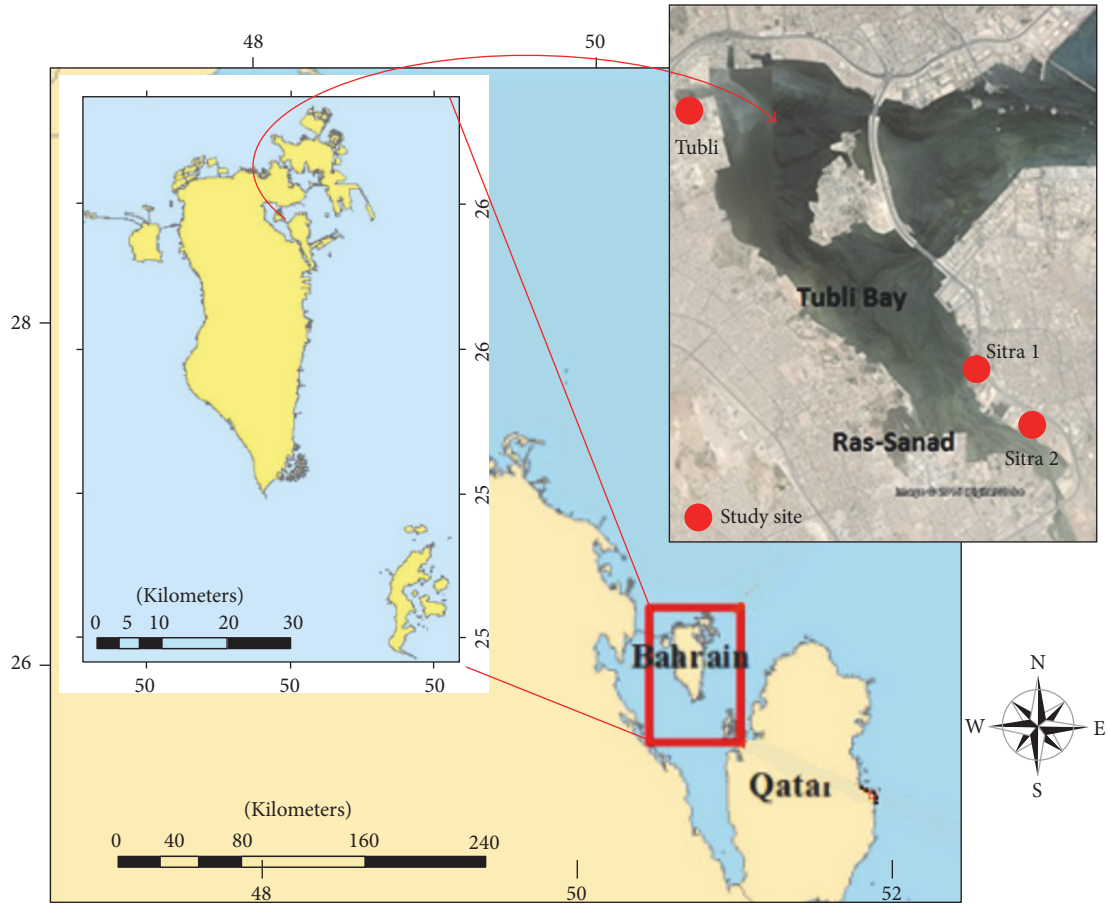

Figure 1: Study site locations.

Mangrove ecosystem in Bahrain is experiencing various forms of pollution, most importantly, wastewater discharge from the Tubli Sewage treatment plant and to some extent from urban and industrial runoff. The magnitude and distribution of HM contaminations in sediment and the role of mangrove in sequestering HM in this part of the world are poorly known. This study aimed to assess accumulations and distribution of HM in the leaves and underlying sediment of mangrove trees as affected by the urban municipal wastewater discharge in three sites of mangrove at the Tubli Bay area.

2. Methodology

2.1. Study Area. Mangrove stands selected for this study are located in northern and western coasts of Tubli Bay. The Bay itself is a shallow body of water located in North West of the main island of Bahrain. The Bay area is 10 Km^2, with an average depth of 5 m. Three mangrove sites were chosen based on ease of accessibility to represent various degrees of exposure to municipal wastewater discharge. Tubli site (26°11′47.25″N, 50°33′39.12″E) is located at the northern end of Tubli Bay where nearly 100,000 m^3 of secondary treated municipal wastewater discharges into the Bay daily [33]. The site is directly affected by wastewater discharge by two canals bounding northern and southern edges of mangrove stand. The other two locations, Sitra (1) (26°9′17.20″N, 50°36′26.64″E) and Sitra (2) (26°8′51.48″N, 50°36′58.79″E), are situated on the western coast and presumed relatively pollution-free. They are 1 km apart and 7 km away from the discharged wastewater outfalls (Figure 1).

2.2. Sampling Procedures. At each study site, six randomly selected trees were chosen, representing six monitoring plots. Leaves from each tree were collected, washed with distilled water, oven-dried at 60°C till constant weight, and then ground and stored. One gram of ground leaves was taken and digested using the dry ashing method [34].

A transect line running across the mangrove stand on each site in a seaward leeward direction was established. Sediment cores were taken from three randomly established replicates along the same transect at the three depths (0–5, 5–15, and 15–30 cm) using hand-operated soil auger. Once removed, samples were placed in sealed bags and transported to the laboratory for analysis. Samples were air-dried, homogenized using mortar and pestle, and passed through a 2 mm sieve. One gram of air-dried sediment sample was completely digested with a Berghof microwave digestion system in accordance with EPA method 3052 [34, 35].

Table 1: Mean concentrations of heavy metals in mg L^{-1} (±SEM) ($n = 6$) in the leaves of gray mangrove*.

Site	Cr	Cu	Fe	Mn	Mo	Ni	Zn
Tubli	2.45 (0.18)[a]	18.46 (2.29)[a]	659.77 (118.60)[a]	31.51 (1.71)[a]	5.95 (2.04)[b]	6.55 (0.87)[a]	49.00 (6.00)[a]
Sitra (1)	2.22 (0.27)[a]	18.92 (5.86)[a]	638.30 (132.76)[a]	34.38 (5.63)[a]	35.99 (5.81)[a]	6.10 (1.06)[a]	31.38 (2.72)[b]
Sitra (2)	2.09 (0.33)[a]	17.85 (3.46)[a]	625.97 (82.36)[a]	32.97 (3.26)[a]	11.41 (4.43)[b]	6.81 (2.08)[a]	42.09 (3.80)[a,b]
Einollahipeer et al. (2013)	—	37.73	—	—	—	2.44	33.83
Abohassan (2013)	4.53	4.17	39.54	11.14	—	6.74	4.23
Usman et al. (2013)	8.15–14.9	256–356.6	—	—	—	1.37–4.02	29.5–36.8
Almahasheer et al. (2014)	—	107.22	63	5.08	—	0.39	3.62

*Values in the same column not connected with the same letter are significantly different ($P \leq 0.05$). Pb was not detectable.

Concentrations of HM, cadmium (Cd), chromium (Cr), copper (Cu), iron (Fe), manganese (Mn), molybdenum (Mo), nickel (Ni), lead (Pb), and zinc (Zn), in both plant and sediment samples were detected using inductively coupled plasma atomic emission spectrometry (ICP-OES Optima 8000). Analysis of Variance (ANOVA) was performed to assess variations in heavy metals contents in the leaves of mangrove stands among sites and between sites at different sediment depths. Statistically significant differences were evaluated at the 0.05% level of significance using the Student t-test and the Pearson correlation between factors. JMP 10 statistical analysis package was employed in this analysis [36].

3. Results and Discussion

3.1. Heavy Metals in Plant Leaves. Table 1 shows the mean concentrations of HM (mg L^{-1}) in *Avicennia marina* leaves, while Figure 2 shows ranges of HM levels in the stand of the study sites. The results showed no significant differences in HM content in the leaves of mangroves between the study sites, except for Mo which was significantly higher ($P \leq 0.05$) in Sitra (1) compared to Tubli and Sitra (2). However, Zn was significantly higher ($P \leq 0.05$) in Tubli compared to Sitra (1) and Sitra (2). Mean concentrations of HM in mangrove leaves of Tubli Bay were in the following order: Fe (641.1) > Zn (41.4) > Mn (32.95) > Mo (18.6) > Cu (18.4) > Ni (6.5) > Cr (2.2). The leaves contents of Cr and Ni of this study were lower than those found in [20, 21] and higher in Cu, Fe, Zn, and Mn compared to values reported in [37, 38]. Regionally, the following order was reported for mangrove leaves contents: Fe > Mn > Ni > Cr > Zn > Cu [22]. Higher values of Cu in the leaves of mangroves in the region were reported in [21, 23, 39] indicating site-specific HM pollution sources. Although the concentrations of Cr, Fe, and Ni exceeded the values reported in [40–42], respectively, mangrove did not suffer from any visible toxicity symptoms. This result supports scholars' findings that *A. marina* can be considered as a high-efficient plant for bioaccumulation of HM [4, 22, 30, 41]. Special reference to high Cu concentration was made by many authors [21–23, 39]. Variations of the results of our study with other studies [21–23, 43] can be attributed mainly to site specificity and type of anthropogenic pressure.

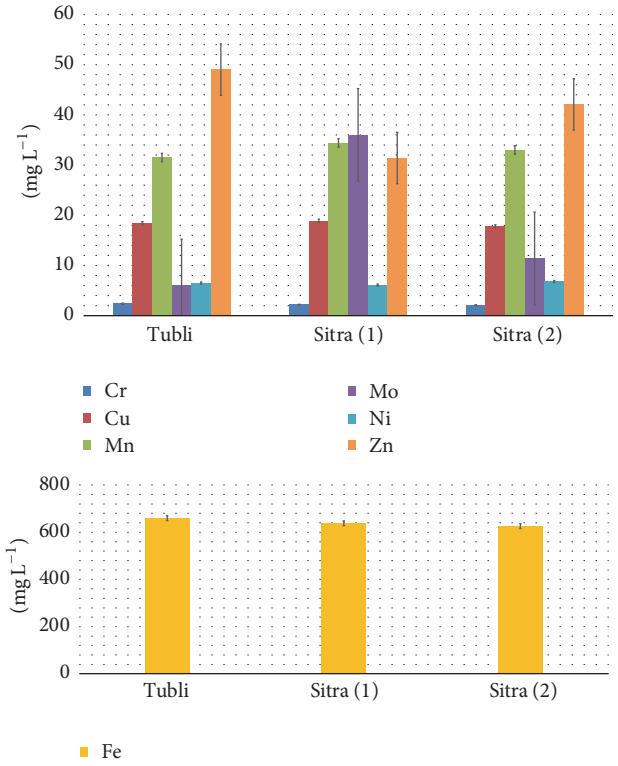

Figure 2: Heavy metals concentrations in the leaves of gray mangrove in the study sites (mg L^{-1}).

There was a high significant correlation between Fe and Cr ($R^2 = 0.8$, $P = 0.0001$) and between Fe and Mn ($R^2 = 0.6$, $P = 0.02$) contents in mangrove leaves. Likewise, a highly significant correlation was found between the Zn and Cu ($R^2 = 0.8$, $P = 0.0002$); however, a negative correlation was observed between Zn and Mo ($R^2 = -0.6$, $P = 0.03$). In the meantime, no significant correlation was found between HM in *A. marina* leaves and the underlying sediment. The same result was obtained in [28], although [41] pointed out that the Cu accumulation in leaves of the plant depends on

Table 2: Mean concentrations of heavy metals in mg L^{-1} (±SEM) ($n = 6$) in the sediment of gray mangrove (mg L^{-1})*.

Site	Depth (cm)/metals	Cr	Cu	Fe	Mn	Ni	Pb	Zn
Tubli	*0–5*	27.3 (3.49)^{a}	85.7 (2.17)^{a}	4521.5 (17.72)^{a}	86.4 (4.71)^{a}	7.7 (1.62)^{a}	26.2 (2.91)^{a}	110.7 (24.69)^{a}
	5–15	24.1 (4.21)^{a,b}	65.7 (20.18)^{a,b}	3597.0 (701.54)^{a,b,c}	81.7 (2.26)^{a}	7.0 (2.16)^{a,b}	22.1 (4.86)^{a,b}	82.4 (45.19)^{a,b}
	15–30	17.2 (1.56)^{b,c}	17.5 (7.41)^{b,c}	2646.1 (724.34)^{b,c}	70.9 (6.78)^{a,b}	3.3 (0.66)^{b,c}	15.5 (2.39)^{b,c}	32.9 (10.66)^{b,c}
Sitra (1)	*0–5*	18.8 (1.13)^{b,c}	21.5 (6.48)^{b,c}	3920.0 (184.81)^{a,b}	71.1 (4.12)^{a,b}	6.00 (0.89)^{a,b}	15.2 (2.07)^{b,c}	35.5 (7.46)^{b,c}
	5–15	18.9 (2.23)^{b,c}	13.1 (3.72)^{c}	3675.7 (655.66)^{a,b}	71.7 (4.90)^{a,b}	5.3 (1.08)^{a,b,c}	13.3 (1.57)^{c}	24.1 (5.22)^{c}
	15–30	18.2 (2.89)^{b,c}	11.9 (3.16)^{c}	3425.4 (844.62)^{a,b,c}	63.4 (8.)^{b}	6.2 (1.77)^{a,b}	12.97 (1.19)^{c}	21.2 (3.04)^{c}
Sitra (2)	*0–5*	16.9 (1.80)^{b,c}	16.6 (3.63)^{c}	3542.5 (445.36)^{a,b,c}	59.7 (7.48)^{b}	4.4 (0.81)^{a,b,c}	16.2 (0.91)^{b,c}	36.3 (3.66)^{b,c}
	5–15	16.2 (2.04)^{c}	13.3 (4.37)^{c}	3336.6 (866.26)^{a,b,c}	57.4 (7.37)^{b}	5.3 (1.45)^{a,b,c}	18.0 (1.70)^{b,c}	37.7 (6.60)^{b,c}
	15–30	11.8 (2.31)^{c}	11.02 (7.34)^{c}	2319.3 (865.06)^{b,c}	35.1 (8.82)^{c}	1.9 (1.04)^{c}	17.2 (2.16)^{b,c}	22.4 (8.34)^{c}
Dehghani and Karbassi (2015)		—	30.11	1894	—	25.08	43.70	—
Einollahipeer et al. (2013)		—	42.13	—	—	54.12	47.90	43.61
NOAA		81	34	—	—	21	46	150
Canadian (ISQGs)		52	18				30	124
EPA								
Nonpolluted		<25	<25	—	—	<20	<40	<90
Slightly polluted		25–75	25–50	—	—	20–50	40–60	90–200
Severely polluted		>76	>50	—	—	>50	>60	>200

*Values in the same column not connected with the same letter are significantly different ($P \leq 0.05$). Mo concentration in mangrove sediment was less than 0.001 mg L^{-1}; NOAA (1997 revision), http://www.esd.ornl.gov/programs/ecorisk/documents/tm95r4.pdf; Canadian (ISQGs), https://www.elaw.org/system/files/sediment_summary_table.pdf; US EPA, 1991. Sediment quality guidelines. Draft report. EPA Region V Chicago IL.

its concentration in the sediment. Various authors pointed out that a large percentage of HM accumulation occurs in stems and roots of mangrove compared to leaf tissues [43]. Nevertheless, very few studies show significant correlations between metal levels in sediment and tissues, and this suggests that mangrove roots may avoid uptake of some metals and that most heavy metals are immobilized in the sediment [12].

3.2. Heavy Metals in Mangrove Sediment. Table 2 illustrates heavy metal concentrations (mg L^{-1}) at various depths of mangrove sediment at the study sites. The concentrations of HM in sediments ranged as Cr: 7.0–43, Cu: <0.001–281, Fe: 925–7203, Mn: 19–108, Ni: <0.001–17, Pb: 7.5–38, and Zn: 7.1–304 mg L^{-1} (Figure 3). These concentrations were within the ranges found in [39, 43, 44]. However, some authors reported higher values for Cu, Ni, and Pb [39, 45] and lower values for Fe in the region [42]. The high concentrations of HM indicate slight to severe pollution in some metals, especially at their upper range values (Cu, Pb, and Zn) according to the EPA sediment quality proposed guidelines, the Canadian interim marine sediment quality guidelines (ISQGs), and NOAA Sediment standards. Mean HM concentrations in the mangrove underlying sediment were distributed in the following order Fe (2319.3), Mn (35.1), Zn (44.8), Cu (28.48), Cr (11.82), Pb (17.2), and Ni (1.9). Clearly, Cu means value in the study sites exceeded the Canadian guidelines and the slightly polluted category of EPA guidelines. The highest concentration of Cu in the sediment is probably due to urban runoff and marine antifouling paints as the Bay is near the Bahrain main harbor.

High significant differences were observed between the concentrations of Cr, Cu, Mn, Pb, and Zn in top layer sediment at Tubli site ($P \leq 0.05$) compared to the other two sites due to the effect of plant sewage effluent. It was also noted that the concentration of Fe and Mn significantly varied between the top sediment layers (0–5 cm) and the deep ones (15–30 cm) at all sites. HMs were concentrated in a top surface sediment layer, which agreed with [29].

Pearson correlation analysis showed that strong correlation coefficients existed between Cu and Cr (0.788), Fe and Cr (0.8437), Mn and Cr (0.8028), Mn and Fe (0.7521), Ni and

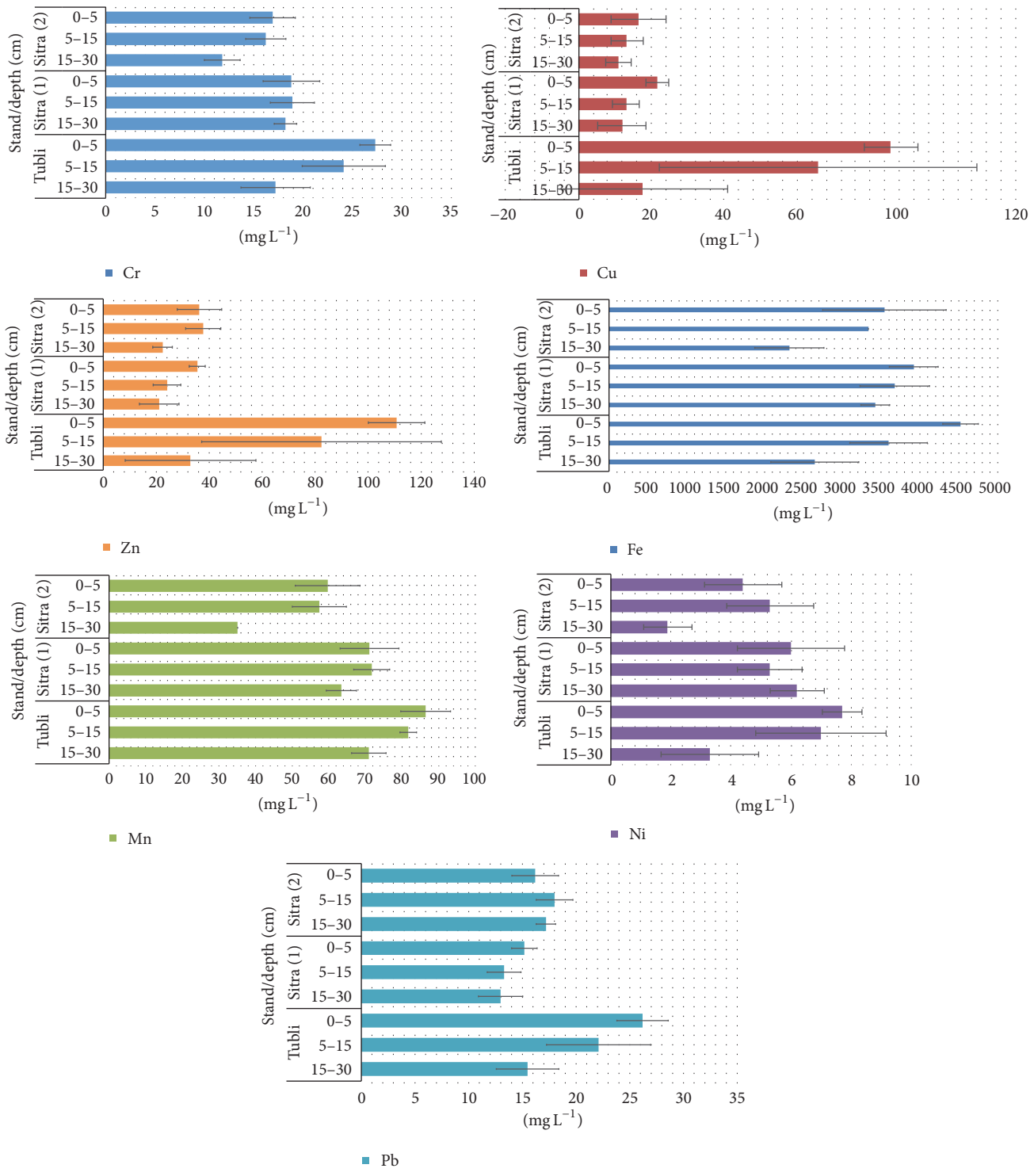

Figure 3: Heavy metals content of mangrove sediment in the study sites (mg L^{-1}).

Cr (0.8311), Ni and Fe (0.8138), Pb and Cu (0.7530), Zn and Cr (0.8111), Zn and Cu (0.9702), and Zn and Pb (0.8105) at ($P < 0.0001$). Also, likely correlations existed between Fe and Cu (0.6254), Ni and Cu (0.6660), Ni and Mn (0.6624), Pb and Cr (0.6359), Pb and Fe (0.5117), Zn and Fe (0.6446), and Zn and Ni (0.6696) ($P < 0.0001$). On the other hand, weak correlation coefficients were found between Mn and Cu ($P = 0.0014$, $R = 0.4248$), between Pb and Mn ($P = 0.0176$, $R = 0.0176$), between Pb and Ni ($P = 0.0030$, $R = 0.3966$), and between Zn and Mn ($P = 0.0007$, $R = 0.4461$). Although significant correlations were observed between a number of metals in this study, these figures must be correlated with

organic matter and sediment texture to reveal the whole picture of HM accumulation in the sediment Bay.

The different distribution of heavy metals in sediments over large spatial scales can be attributed to varying levels of contaminant input and changed sediment deposition patterns, especially for fine sediments and geochemical conditions [26, 29]. Further on, HM concentration in sediments of the study sites far exceeded the amount present in the leaves of mangroves except for nickel which was higher in the leaves. It was reported that significant relationships between *A. marina* leaf tissue and sediment might occur but were usually weak and were not maintained for a very long time [12]. Variation in HM concentrations can be attributed to the physical variables which, essentially, modify biological activities. The combinations of biophysical factors affect the concentration of the HM in surface sediment through modification of the sedimentation and erosion processes [46]. The high density of trees which averaged "4577 tree ha^{-1}" on the study sites coupled with a massive number of pneumatophores (150 root m^{-2}) has an influence on wave attenuation along the coasts of the Bay which, in turn, modify sediment movements through trapping processes that alter HM distribution. Besides, some metals might be affected by a redox condition of sediments [45] and the presence of dense growth of algae in the Bay [47] which may modify HM settlements and concentration thereby [48].

4. Conclusion

The results of this study indicate that the leaves of the mangrove are sequestering a substantial amount of Cu and can serve as a bioindicator of Cu contamination. On the other hand, HM concentration in leaves of mangrove does not reflect the complete picture of HM status in mangrove or its temporal sequestration of these metals. Thus the HM contents of roots and branches have to be taken into consideration in future studies. HM content in the sediment of mangroves was comparable with values for Arabian Gulf coastlines and mostly fall within international standards except some hot spots (Tubli site in this study) where values are much higher for some elements (Cu) due to urban wastewater discharge and Bay proximity to Bahrain main harbor. The different distribution of HM in sediments over spatial scales (sites) reflected varying levels of contaminant input and changed sediment deposition patterns by anthropogenic factors. In this regard, Tubli site had the most concentrations of HM compared to the other two sites as its mangrove stand is directly affected by wastewater outfalls. On the other hand, the concentrations of Cr, Cu, Fe, Mn, and Ni were greater in Sitra (1) than in Sitra (2) while the latter had a higher concentration of Pb and Zn. This result reflects the presence of a point source pollution at Sitra (2) other than wastewater. Finally, the results of this study can serve as an indication of contamination presence in a particular setting and care must be practiced when comparing these results with other studies due to differences in site conditions, analysis methods, and types of pollution involved.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this work.

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