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Bioaccumulation and Distribution of Metals in Sediments and *Avicenna marina* Tissues in the Hara Biosphere Reserve, Iran

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Abstract The metal pollution in Sediments and *Avicenna marina* tissues in the Hara Biosphere Reserve was monitored for Lead (Pb), Cadmium (Cd), and Nickel (Ni) with atomic absorption spectrometer. The results showed that the mean concentration of Pb, Cd, and Ni in the water and sediments were much higher than the recommended threshold limits in the most stations, also the highest means of Pb, Cd, and Ni were observed in Avicenna roots and it were 25.26 ± 4.86, 2.17 ± 0.74, and 26.72 ± 6.17 (μg g^{-1}) respectively. Calculating BCF (bioconcentration factor) index illustrates that *A. marina* accumulates Pb, Cd, and Ni 1.62, 1.52 and 0.73 times greater than sediment levels respectively, So it can show that *A. marina* may be employed as a biological indicator exposure of Cd, Pb, and Ni with temporal monitoring, also the factories were main sources of metals contamination in the Hara Biosphere Reserve.

Keywords Contamination · Lead · Cadmium · Nickel · Bioconcentration Factors

Hara Biosphere Reserve in southern Hormozgan province comparing 85,686 hectares areas is located in the middle of the Mehran and Gourzin river deltas between Bandar Khamir and Queshm Island. The Hara Biosphere contains a 100,000 ha international wetland and lies at 26°45′–26°58′N and 55°30′–55°50′E of the Persian Gulf. The variety of Biosphere with its unique mangrove trees provides a divers habitat for birds like egrets, herons, pelicans, and plovers. It also provides a breeding and spawning habitat for fish, shrimp and other crustaceans.

The Persian Gulf is exposed to different sources of marine pollution and is affected by man's activities. The development of urban infrastructure, along the south coast of Iran, and the several industrial complexes was established during the last decades (Pourang and Amini 2001). Industries such as fisheries, maritime cultivation, transport, and tourism, together with deficient wastewater treatments, are a threat to marine life. So, the water quality of the Persian Gulf is influenced by various industries that discharging their wastewater directly to the sea and estuaries. Metals are among the most common pollutants in the Persian Gulf (Agah et al. 2010).

Metals are a common group of marine pollutants, which is not biologically degradable. In seawater, metals appear in two different forms including dissolved cations and metal bound to suspended organic materials. The second form always deposit in the sediment, so that sediment is known as the final destination for discharge metals into the seawater (Chou et al. 2004; Safahieh et al. 2011), then they can transfer into plant tissues from sediments and pose long-term damaging effects on plants (Lindsey et al. 2005). However, all forms of metals in aquatic ecosystem may enter aquatic food chains and accumulate in various types of organisms (Chen et al. 2004; Dórea 2008; Papagiannis et al. 2004) but, the worse impacts of these elements appear when they transfer to the human body through food chains or trophic relations (Sankar et al. 2006). Many metals are essential for plant growth such as Ni, Zn, Mn but they are also toxic for plants at a high concentration (Xu et al. 2006; Jian-Guo et al. 2010), while other metals like Pb and cd are extremely toxic to living organisms and human health even at a low concentration (Peng et al. 2008).

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Avicennia marina (Forsk.) Vierh. var. australasica is dominant in the Hara Biosphere Reserve of Southern Iran, often occurring in mono-specific stands. Avicennia species exhibit greater tolerance and accumulative properties to numerous metals than other mangrove species (Peng et al. 1997; Mac Farlane and Burchett 2002); so it could be a good natural indicator for determining the levels of metals in water and sediments. Hence, the main objectives of this study were: (1) to investigate the metal pollution levels in water, sediment and *A. marina plant* based on human activities in the study area; (2) to determine of this plant ability for removing metals from water bodies.

Materials and Methods

Water and sediment samples were collected from 9 sites in three replicates based on ecological conditions and human activities in the Hara Biosphere Reserve in summer of 2010. Water samples were collected into acid washed 250-mL plastic bottles from approximately 50 cm below the surface water. Sediment samples were collected using grab sampler and the *A. marina* tissue samples were collected at the same sampling sites which water and sediments were taken. All chemicals used were as reagent grade, high purity HNO_3 assay 65 %, HCl assay 37 % and pure $HClO_4$ assay 72 % (Merck, Germany). Prior to any analysis, all equipment and containers were soaked in 10 % HNO_3 and rinsed thoroughly with double distilled water before use.

The water samples were filtered using a 0.45 μm nitrocellulose membrane filter. Sediment samples were dried at 110°C for 24 h to achieve a constant weight, after cooling the sample was passed through a 0.063 mm plastic sieve and stored in polyethylene bottles until chemical treatment using acid digestion. Acid digestion was performed by placing about 1 g of sample in a beaker and digested with 16 mL mixture of concentrated HCl and HNO_3 with a ratio of (12:4 v/v) for 6 h at 90°C and then 4 mL of concentrated $HClO_4$ was added. The residue was filtered and diluted to 50 mL with double distilled water. The solution was stored in a refrigerator at 4°C for analysis (Tuzen 2003; El-Rjoob et al. 2008). The leaves, stems and roots tissues were separated; oven dried (70°C) and homogenized using a grinder. One gram of the dried sample was placed into 150 mL Erlenmeyer flask, with conc. HNO_3 and conc. $HClO_4$ in a ratio of 5:1.5 v/v. Finally all samples were filtered through a 0.45 mL nitrocellulose membrane filter. Bioconcentration factor (BCF) index were used to quantify the bioconcentration and transfer metals from sediment to plant, it were calculated as metal concentration ratio of plant roots to sediment. The determination of Pb, Cd, and, Ni in water were carried out by graphite furnace atomic absorption spectrometer (Perkin Elmer, AA3030), and in sediments and plants by flame atomic absorption spectrometer (Shimadzu AA 610s). The analytical recovery range of three kinds of metal was 98.7 %–106.4 %, which was measured up to the analytical demand (QA/QC). The detection limits of Pb, Cd, and Ni were 0.002, 0.009, and 0.010 μg g^{-1}, respectively.

Pearson's correlation coefficients (r) were used when calculating correlations between concentration of metals in water, sediments and the plant tissues. To determine the significant differences at different sampling sites, a one-way analysis of variance (ANOVA), Tukey's honest significant difference test was employed. This is because of normalization of data. The paired Student *t test* was used to evaluate the significant differences between the roots and leaves, roots and stems, and stems and leaves for each metal. Data analyses were carried out using the statistical package Minitab (Release 15). All metal analyses in plants are in dry weight (DW). Values are given in means ± standard divisions (SD).

Result and Discussion

Concentrations of Pb, Cd, and Ni in water and sediments from each sampling site are given in Table 1 and the results of correlation analysis for water and sediments in the Hara Biosphere Reserve are shown in Table 3. The highest mean concentration of Pb, Cd, and Ni in water were measured at site 5 (5.33 μg L^{-1}), 3 (2.93 μg L^{-1}), and 9 (9.65 μg L^{-1}) and in sediment were measured at site 7 (42 μg g^{-1}), 2 (4 μg g^{-1}), and 3 (118.90 μg g^{-1}) respectively. Overall the metal concentrations in water and sediments were in descending order of Ni > Pb > Cd. In general, the concentrations of metals in sediment were far higher than those in the water, which showed the accumulated contamination in the Reserve sediments over the years (Namminga and Wilhm 1976). According to the results, the level of Pb, Cd, and Ni in Hara Biosphere Reserve was higher than those in Khor-Mousa, Khor-Jafari, Khor-Zangi (Safahieh et al. 2011) and coastal sediments in the Persian Gulf, UAE and Qatar (De Morsa et al. 2004), as well as Cd, and Ni concentration in this study was higher than those in coastal sediments, Bahrain and Northeast Persian Gulf (Safahieh et al. 2011). The results also indicated that the metal concentrations in water sampling at all stations in the Hara Biosphere Reserve were considerably higher than mean seawater (Table 1). In case of sediment samples Pb concentrations at all stations were higher than mean crust and mean world sediments and Cd concentrations at all stations were higher than mean crust, also Ni concentrations in the Reserve sediments at all stations (except of station 5) was higher than the mean world sediments, but only the stations

Table 1 Means and SD of metal concentrations in water ($\mu g\ L^{-1}$) and sediment ($\mu g\ g^{-1}$) from 9 sites in Hara Biosphere Reserve

Site	Pb		Cd		Ni	
	Water	Sediment	Water	Sediment	Water	Sediment
1	4.49 ± 0.73	35.83 ± 3.18	2.25 ± 0.42	3.91 ± 0.42	3.68 ± 0.35	92.42 ± 6.11
2	3.15 ± 0.62	34.08 ± 2.57	1.46 ± 0.35	4.00 ± 0.65	3.43 ± 0.26	81.26 ± 5.41
3	4.33 ± 0.71	33.00 ± 2.43	2.93 ± 0.74	3.58 ± 0.61	7.72 ± 0.98	118.90 ± 6.35
4	2.83 ± 0.32	39.25 ± 3.07	1.07 ± 0.26	3.50 ± 0.47	2.68 ± 0.31	78.89 ± 5.21
5	5.33 ± 0.63	38.08 ± 4.07	2.35 ± 0.26	3.08 ± 0.45	2.82 ± 0.47	36.81 ± 3.15
6	3.15 ± 0.43	39.50 ± 3.69	0.55 ± 0.09	3.58 ± 0.54	2.46 ± 0.41	83.61 ± 5.56
7	4.16 ± 0.33	42.00 ± 4.12	1.06 ± 0.32	3.50 ± 0.51	3.74 ± 0.73	75.68 ± 5.35
8	2.26 ± 0.24	36.50 ± 3.32	0.75 ± 0.11	3.37 ± 0.45	6.63 ± 1.02	73.62 ± 5.38
9	4.23 ± 0.46	31.66 ± 2.86	1.02 ± 0.21	3.41 ± 0.37	9.65 ± 1.03	77.61 ± 5.69
Average	3.77 ± 0.49	36.65 ± 3.07	1.49 ± 0.30	3.55 ± 0.54	4.64 ± 0.61	79.86 ± 5.72
Mean seawater[a]	0.03	–	0.05	–	2	–
Mean word sediments[b]	–	19	–	–	–	52
Mean crust[b]	–	14	–	0.2	–	80

[a] (Alloway 1995)

[b] (Riley and Chester 1971)

1, 2, 3 and 6 had mean concentration higher than the mean crust. Pearson correlation coefficients shown that there was a positive correlation (r = 0.692, $p < 0.05$) between Cd and Pb in water, we hypothesize that metals with a high positive correlation are possibly from the same pollution source (Üstün 2009), also there was highly negative correlation (r = −0.739, $p < 0.05$) between Pb in sediment and Ni in water. In other cases, there were not found significant relationship between concentrations of metals in water and sediments. Comparison between sampling sites (Table 4) using one-way analysis of variance showed that there were significant differences for Pb, Cd, and Ni concentration in water at the 9 sampling sites. In sediment, comparison between sampling sites showed that there were no significant differences for Pb and Cd; but it was significant differences between 9 sampling sites for Ni.

The highest mean concentration of Pb, Cd, and Ni in water were measured at site 5 (5.33 $\mu g\ L^{-1}$), 3 (2.93 $\mu g\ L^{-1}$), and 9 (9.65 $\mu g\ L^{-1}$) and in sediment were measured at site 7 (42 $\mu g\ g^{-1}$), 2 (4 $\mu g\ g^{-1}$), and 3 (118.90 $\mu g\ g^{-1}$) respectively. Overall the metal concentrations in water and sediments were in descending order of Ni > Pb > Cd. In general, the concentrations of metals in sediment were far higher than those in the water, which showed the accumulated contamination in the Reserve sediments over the years (Namminga and Wilhm 1976). According to the results, the level of Pb, Cd, and Ni in Hara Biosphere Reserve was higher than those in Khor-Mousa, Khor-Jafari, Khor-Zangi (Safahieh et al. 2011) and coastal sediments in the Persian Gulf, UAE and Qatar (De Morsa et al. 2004), as well as Cd, and Ni concentration in this study was higher than those in coastal sediments, Bahrain and Northeast Persian Gulf (Safahieh et al. 2011). The results also indicated that the metal concentrations in water sampling at all stations in the Hara Biosphere Reserve were considerably higher than mean seawater (Table 1). In case of sediment samples Pb concentrations at all stations were higher than mean crust and mean world sediments and Cd concentrations at all stations were higher than mean crust, also Ni concentrations in the Reserve sediments at all stations (except of station 5) was higher than the mean world sediments, but only the stations 1, 2, 3 and 6 had mean concentration higher than the mean crust. Pearson correlation coefficients shown that there was a positive correlation (r = 0.692, $p < 0.05$) between Cd and Pb in water, we hypothesize that metals with a high positive correlation are possibly from the same pollution source (Üstün 2009), also there was highly negative correlation (r = −0.739, $p < 0.05$) between Pb in sediment and Ni in water. In other cases, there were not found significant relationship between concentrations of metals in water and sediments. Comparison between sampling sites (Table 4) using one-way analysis of variance showed that there were significant differences for Pb, Cd, and Ni concentration in water at the 9 sampling sites. In sediment, comparison between sampling sites showed that there were no significant differences for Pb and Cd; but it was significant differences between 9 sampling sites for Ni.

The results of this study showed that the wastewater of most factories which located around the Hara Biosphere Reserve such as Al-Mahdi aluminum factory, lead and zinc Queshm factory and Hormozgan Cement factory were discharged to the Persian Gulf and the Hara Biosphere Reserve directly, without any remediation; only a simple

Table 2 Means and SD of metal concentrations in leaves, stems and roots (μg g^{-1}) from 9 sites in Hara Biosphere Reserve

Site	Pb			Cd			Ni		
	Leaf	Stem	Root	Leaf	Stem	Root	Leaf	Stem	Root
1	14.25 ± 2.76	29.83 ± 2.80	37.00 ± 4.37	2.33 ± 0.57	3.03 ± 0.52	3.08 ± 0.38	14.00 ± 2.29	29.41 ± 5.19	29.33 ± 3.50
2	12.41 ± 1.70	22.83 ± 3.17	13.13 ± 3.37	2.50 ± 0.00	1.33 ± 0.38	1.50 ± 0.25	12.91 ± 2.18	19.33 ± 1.60	18.00 ± 1.14
3	12.91 ± 1.62	20.66 ± 2.70	11.66 ± 4.34	2.08 ± 0.28	0.91 ± 0.14	1.75 ± 0.25	10.16 ± 1.46	22.00 ± 3.78	16.83 ± 0.57
4	7.08 ± 1.52	16.08 ± 1.62	14.66 ± 1.44	1.42 ± 0.14	0.91 ± 0.16	1.25 ± 0.25	10.75 ± 0.25	15.25 ± 2.17	19.41 ± 0.80
5	4.41 ± 1.15	17.45 ± 2.95	19.75 ± 4.50	1.16 ± 0.28	2.33 ± 0.52	1.58 ± 0.38	7.58 ± 1.75	17.41 ± 2.03	19.58 ± 1.42
6	18.75 ± 0.86	14.16 ± 1.44	32.50 ± 3.60	1.66 ± 0.28	0.83 ± 0.28	2.75 ± 0.25	11.16 ± 1.01	20.58 ± 0.14	33.00 ± 5.88
7	22.00 ± 1.56	20.41 ± 3.76	37.83 ± 2.12	1.41 ± 0.80	1.08 ± 0.14	2.91 ± 0.38	13.58 ± 1.01	20.41 ± 2.55	42.16 ± 7.33
8	20.75 ± 2.25	20.33 ± 2.45	23.25 ± 5.25	1.25 ± 0.25	1.33 ± 0.38	1.83 ± 0.38	11.91 ± 2.88	18.58 ± 3.32	23.16 ± 4.41
9	12.33 ± 3.82	23.00 ± 3.63	37.58 ± 0.62	1.50 ± 0.66	1.58 ± 0.52	2.91 ± 0.38	13.25 ± 2.38	17.08 ± 1.18	39.00 ± 5.93
Average	13.87 ± 2.02	20.53 ± 3.58	25.26 ± 4.86	1.70 ± 0.59	1.55 ± 0.44	2.17 ± 0.74	11.70 ± 2.50	20.00 ± 4.57	26.72 ± 6.17

Table 3 Statistically significant relations (pearson correlations, r) between metal concentrations of water, sediment and *A. marina* tissues (n = 9)

Relations	R	*p*
Cd and Pb in water	0.692	<0.05
Pb in sediment and Ni in water	−0.739	<0.05
Cd in sediments and Cd in leaves	0.922	<0.01
Pb and Cd in stems	0.722	<0.05
Pb and Ni in stems	0.700	<0.05
Pb and Cd in roots	0.950	<0.01
Pb and Ni in roots	0.932	<0.01
Cd and Ni in roots	0.883	<0.01

p significance level

physical screening was being performed; It consisted of lots of toxic metals likes Pb, Cd, Ni, Vn and Cr, as well as Oil tankers, commercial ships and recreational boats traffic within the study area released large amounts of Pb, Cd, and Ni-containing compounds into the water and sediments.

The concentrations of Pb, Cd, and Ni in the leaves, stems and roots of *A. marina* from each sampling site are given in Table 2. In *A. Marinna* the highest mean concentration of Pb, Cd, and Ni observed in roots and it were measured at site 7 (37.83 μg g^{-1}), 1 (3.08 μg g^{-1}), and 7 (42.16 μg g^{-1}) respectively. Metal concentrations in leaves and stems tissue were in descending order of Pb > Ni > Cd while for root tissue were in descending order of Ni > Pb > Cd and the general trend of bioaccumulation was Sediments > Roots > Stems > Leaves > Water. According to the results of this study, the level of Pb, Cd, and Ni in the Hara Biosphere Reserve were higher than those in Shenzhan of China, reported by Peng et al. (1997); The mean concentrations of Pb in leaves and roots of *A. marina* were 5 and 164 μg g^{-1} respectively at Homebush Bay, Sydney (Mac Farlane et al. 2003). Samecka-Cymerman and Kempers (2004) found Cd concentrations ranged from 0.002 to 5.2 μg g^{-1} in 18 aquatic plant species. Ni concentration in this study was higher than the concentration reported by Jian-Guo et al. (2010). Base on Table 3, highly positive correlation between different metals in the roots and stems were observed and there were no significant relationships between metal concentrations in *A. marina* tissues and those in sediments, except of Cd in leaves and sediments. The absence of the significant correlations between the metal concentrations in the plants and in the sediments may be partly attributed to the fact that the correlation was developed based on the total metal concentrations in sediment instead of the bioavailable fractions, which are the dominant form for metal uptake by plant roots (Cardwell et al. 2002).

Comparison between sampling sites using one-way analysis of variance showed that there were significant differences for Pb, Cd, and Ni concentration in the all tissues of *A. marina* at 9 sampling sites of the Hara Biosphere Reserve (Table 4). The paired Student *t* test (Table 5) showed significant differences between the roots and leaves and between the roots and stems for all the three metals. There were also statistical significant differences between the stems and leaves, for Pb and Ni while there were no significant differences for Cd.

Results of bioconcentration factors (BCF) showed that *A. marina* accumulates Pb, Cd and Ni 1.62, 1.52 and 0.73 times greater than sediment levels respectively. However, plants with a high bioconcentration factor have the potential for phytostabilization (Yoon et al. 2006). Bioaccumulation of Pb, Cd, and Ni in the *A. marina* were in descending order of roots > stems > leaves, so it can show that *A. marina* have high ability for absorption and accumulation of metals in the water ecosystem; these results were corresponding to study of Shete et al. (2007). Mac Farlane et al. (2003) found that *A. marina* root may be employed as a bio-indicator of metals, as metals in roots were reflective of environmental levels.

Table 4 Analysis of variance (ANOVA) of Cd, Ni, and Pb concentration in water, sediment and *A. marina* tissues

	Pb		Cd		Ni	
	F value	*p* value	F value	*p* value	F value	*p* value
Water	5.54	<0.001	11.73	<0.001	63.80	<0.001
Sediment	1.76	ns	2.07	ns	26.79	<0.001
Leaf	17.13	<0.001	3.61	<0.05	3.57	<0.05
Stem	3.66	<0.05	15.12	<0.001	6.26	<0.001
Root	23.78	<0.001	14.52	<0.001	7.87	<0.001

p significance level, *ns* not significant

Table 5 Significant difference of Cd, Cu, and Pb, between the root and leaf, root and stem, and stem and leaf in *A. marina* according to the paired Student *t* test (t)

	Roots and leaves		Roots and stems		Stems and leaves	
	t	p	t	p	t	p
Pb	−5.92	<0.001	−2.26	<0.05	4.37	<0.001
Cd	−2.60	<0.05	3.11	<0.05	0.76	ns
Ni	8.05	<0.001	−3.19	<0.05	−8.93	<0.001

ns not significant

The concentrations of Cd, Pb, and Ni in water and sediments in the Hara Biosphere Reserve significantly exceeded the recommended threshold limits for unpolluted water body. The highest accumulation of Cd, Pb, and Ni observed in *A. marina* root so it has higher potential as an environmental bioindicator than leaf and stem, hence *A. marina* species could be used for removing metals from contaminated water in mangrove ecosystem.

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