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Arsenic enrichment in mangroves, and sediments along Karachi coast, Pakistan

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PEER REVIEW

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Comments
The study decribes the quantification of Arsenic in the *A. marina* using hydride generation atomic absorption spectrometry, showing periodic and plant–part wise distribution. The article helps to understand the bioavailibity and cycling of Arsenic in the mangrove system.
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ABSTRACT

Objective: To assess the arsenic (As) concentration in different parts of mangroves *Avicennia marina* and sediments in Karachi coastal area *i.e.* Korangi Creek , Manora, Kakapir and Sandspit.
Methods: Sites are identified for sampling owing to their vicinity to industrial activities. Sandspit is targeted for its being devoid of industries. The hydride generation atomic absorption spectrometry (HG–AAS) were used to analyse the concentration of arsenic in mangrove and sediment.
Results: The high concentration of As was found in roots and middle aerial part as compared to the upper part of mangroves. The concentrations of As was found higher in sediments as compared to the mangroves. There is a seasonal variation of As enrichment in mangrove and sediments as dry seasons showed higher concentration while in rainy season dilution factors may be attributed to the low level of As. The concentration variation of As in sampling sites of mangroves and sediments following the trend *i.e.* Korangi Creek >Manora>Kakapir>Sandspit. The statistical analysis (Two way ANOVA) of data exhibited no significant difference (P>0.05) for trace metals concentrations in mangrove as well as in sediments.
Conclusions: It is obvious to conclude that As should be continuously monitored in different environmental segments. The data must correlate with geographical distribution of As, quantification in different species, their solubility and bioavailability to understand the possible factors responsible for environmental pollution. The present study will be helpful to improve water management resources.

KEYWORDS
Avicennia marina, Sediment, Karachi coast, Arsenic

1. Introduction

The activity of trace metals in aquatic system and their impact on aquatic life vary depending upon the metal species. Many human activities (*e.g.* mining, overuse of chemicals, industrial waste from ports and refineries) have a negative impact on several biological processes[1]. These trace metals are much more likely available to biota than those amounts bound to the soil[2]. Mangrove ecosystem plays an important role in the biogeochemical cycles of the coastal environment, particularly, to provide nutrients and organic carbon to tropical coastal oceans[3,4]. The mangrove forests detain terrestrial–derived nutrients, pollutants and sediments before they reached the coastal ocean[5]. Mangrove and estuarine ecosystems may act as a sink for trace metals because of their variable physical and chemical properties[6]. The waterlogged mangrove sediments are anoxic and reduced in nature due to good supply of sulfide ions and decomposing organic matter in the sediments[7–9]. Trace metals are among the most persistent of pollutants in sediments due to their resistance to decomposition in natural conditions. Most trace elements can be adsorbed by

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clays, complex with organic compounds or co-precipitate with oxides and hydroxides[10].

Sediments prove to be an indicator of the aquatic environment and provide a surface to reserve metals[11-13]. The quality of the marine environment is constantly being monitored by various national authoritative bodies, such as SEPA in Scotland to analyze water, sediment and/or biota[14,15].

Arsenic combines with carbon and hydrogen to form organo-arsenic compounds in animals and plants. The dissolved forms of arsenic in the water include arsenate, arsenite, monomethylarsonic acid and dimethylarsinic acid[16]. Different arsenic species have an affinity for clay mineral surfaces and organic matter, and this can affect their environmental behavior. Therefore, arsenic can be released into the environment from sources such as pesticides applications, wood preservatives, mining activities and petroleum refining. Several studies have been conducted concerning arsenic and associated metals in stream sediments, soils, and crop plants[17-19].

The arsenic contaminated sediment serves as a long-term source of arsenic because its mobility and transport in the environment are strongly influenced to associated solid phase[20]. The distribution and transport of arsenic in sediment is a complex process that depends on water quality, native biota and sediment type. There is a potential for arsenic release when there is fluctuation in Eh, pH, soluble arsenic concentration and sediment organic content[21].

Pakistan is largely arid and semi-arid, receiving less than 250 mm annual rainfall, with the driest regions receiving less than 125 mm of rain annually. It has a diverse landscape, with high mountain systems, fragile watershed areas, alluvial plains, coastal mangroves, and dune deserts. Karachi in Pakistan is located on the northern border of the Arabian Sea and its population is over 18 million. Existing estimates show that mangroves cover approximately 129 000 hectares in the Indus Delta and about 3 000 hectares on the Balochistan coast in the Miani Hor, Kalmat Khor, and Gawadar Bay areas. The Indus Delta therefore supports 97% of the total mangrove forest (37% of the Delta area) while the 3 pockets on the Balochistan coast support the remaining 3% (varying from 8% of the total area in Gawadar Bay to 21% in Kalmat Khor and 25% in Miani Hor)[22]. The heavy metals are the major component of the waste being discharged by the city on the Karachi coast[23].

The knowledge of trace metal concentrations in different compartments of mangrove ecosystem is important to understand the fates of the mangroves and can alert coastal managers of possible impacts upon the detritus driven food web which can potentially lead to the bioaccumulation of contaminants in organisms.

The present work therefore aimed to quantify the concentrations of arsenic in mangrove *Avicenna marina* (including roots, stems and leaves) from four typical mangrove areas of Karachi coast *i.e.*, Korangi Creek, Manora, Kakapir and Sandspit; to determine the distribution of trace metal levels in sediment cores, and to assess the potential risk of trace metals in the mangrove ecosystem along Karachi coast line.

2. Materials and methods

Sites Kakapir, Manora, Korangi Creek and Sandspit were selected owing to their vicinity to industrial activities. Sandspit was targeted for its being devoid of industries (Figure 1).

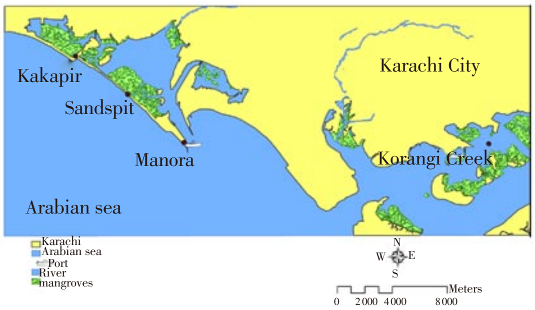

Figure 1. The sites of Karachi coast, Pakistan.

Random selection of sediment and mangroves samples helped to ensure quality data. Sediment and mangrove sample were collected from the sites of low tide zone. Sediments were collected by digging one cubic centimeter of sample with PVC stainless corer to avoid surface contamination through unwarranted sources. Mangrove tissues, including roots, perennial branch (diameter about 5-8 cm), leaves were simultaneously collected. Mangrove samples were washed by deionized water in laboratory to remove the possible dust then oven dried at (70±0.5) °C. The samples of sediment were oven dried at 105 °C, and both the samples grinded by pastel mortar. The sediments were sieved through 63 μm metallic sieve for elemental analysis. Approximately 2 g sample of mangrove samples were digested with a mixture of nitric acid (HNO_3) and perchloric acid ($HClO_4$) (AR Grade Merck-Germany).

About 2 gram sample of sediment was digested in 100 mL Milli-Q water in a 250 mL beaker by adding slowly 10 mL 60% HNO_3 on a hot plate at a temperature of 95 °C until the solution got reduced to 10 to 15 mL. The aliquot was then diluted to 50 mL with deionized water (Milli-Q water) for further chemical analysis by using an instrument varian AA240 hydride generation atomic absorption spectrometry[24].

To assess the precision and accuracy of results, replicate analysis of blank, standard and samples was done. The relative standard deviations were determined to find the precision of the analysis. Quality assurance and quality control procedures were strictly followed throughout the study. The level of accuracy and quality control was maintained by using the certified reference material and the measurements of duplicates for each batch of samples. National Institute of Standards and Technology standard 1573 (Tomato leaves) was used for the evaluation of procedures. Recovery results were calculated for the determination of accuracy. Experiments were repeated till an accuracy of 95%-105% and precision of +/- 5% were obtained. One standard with one set of samples was analyzed routinely.

Two way ANOVA was used to compare the mean concentration of four locations and seasonal variation in sediment and with different parts of mangrove.

3. Results

Table 1 shows the variation of arsenic in sediments at four sites at bimonthly intervals of Karachi coast line Pakistan. Korangi Creek, Manora and Kakapir showed the highest concentration of arsenic as compared to the Sandspit. Seasonal variation was also observed. In the month of June (pre monsoon) the concentration was highest with Korangi Creek at 19.801 μg/g, Manora at 19.335 μg/g, Kakapir at 13.882 μg/g and lowest concentration was in the month of August (rainy season). As dilution factor is involved in rainy season so the accumulation may be lower in these sites. In Sandspit the lowest concentration was also evaluated 3.188 μg/g in August like other sites and high in Febuary at 7.186 μg/g which may be attribute to the anti clock wise movement of wind.

Tables 2-4 reveals that the concentration of arsenic in sediments is higher than that in different parts of mangroves which may be due to acidic condition favor precipitation of orpiment As_2S_3, Reaglar AsS, and other sulphide mineral containing co-precipitated arsenic. As the time passes these precipitate accumulate on the sediment and increase the concentration. Results showed translocation pattern from root to other aerial parts of mangrove from sediments. Overall results revealed that root can accumulate high concentration of arsenic than aerial parts. In comparison of aerial parts anomalous behavior of arsenic concentration was observed. This may be the age of the leaves and stem because old age have more exposure than the new ones. In some cases stem had high level than leaves and vice versa.

The statistical analysis showed no significant difference (P>0.05) for arsenic concentrations in mangrove as well as in sediments.

Table 1
Concentration of arsenic in sediment at bimonthly intervals of different sites of Karachi coast (μg/g, mean±SD).

Months	Sandspit	Kakapir	Manora	Korangi Creek
February	7.751±0.032	7.482±0.032	14.686±0.321	12.551±0.114
	6.552±0.121	8.783±0.037	16.333±0.043	13.782±0.233
	7.255±0.231	10.695±0.223	14.901±0.012	13.222±0.067
Mean	7.186	8.986	15.306	13.185
April	5.350±0.431	12.450±0.012	17.482±0.031	18.360±0.122
	6.721±0.212	11.312±0.043	18.391±0.132	18.962±0.348
	6.511±0.312	11.691±0.032	18.644±0.034	19.020±0.453
Mean	6.194	11.817	18.172	18.780
June	5.211±0.543	13.681±0.322	19.242±0.128	20.022±0.436
	4.952±0.453	14.122±0.564	19.751±0.155	20.181±0.244
	6.113±0.446	13.844±0.732	19.012±0.344	19.200±0.412
Mean	5.425	13.882	19.335	19.801
August	2.321±0.149	5.033±0.654	8.912±0.121	7.252±0.398
	3.033±0.244	5.211±0.321	7.453±0.543	6.911±0.321
	4.211±0.312	4.294±0.322	6.033±0.132	6.335±0.429
Mean	3.188	4.846	7.466	6.832
October	4.752±0.115	6.642±0.054	9.254±0.421	9.451±0.543
	4.212±0.234	6.291±0.043	9.754±0.467	10.210±0.441
	5.341±0.167	6.782±0.013	10.411±0.567	10.985±0.345
Mean	4.759	6.571	9.806	10.215
December	5.995±0.433	7.455±0.677	12.301±0.765	11.254±0.654
	6.102±0.543	8.591±0.543	14.254±0.754	13.452±0.453
	6.311±0.567	8.322±0.453	11.464±0.632	12.091±0.765
Mean	6.136	8.122	12.673	12.265

Table 2
Concentration of arsenic in upper parts of mangroves at bimonthly intervals of different sites of Karachi coast (μg/g, mean±SD).

Months	Parts	Sandspit	Kakapir	Manora	Korangi Creek
February	Leaves	0.812±0.041	0.922±0.009	1.366±0.056	1.695±0.048
	Stem	0.732±0.011	0.951±0.002	1.452±0.001	1.266±0.056
April	Leaves	1.666±0.005	1.766±0.014	1.822±0.002	2.182±0.096
	Stem	0.691±0.020	0.985±0.011	1.331±0.032	1.354±0.058
June	Leaves	2.382±0.023	1.634±0.056	2.325±0.042	3.855±0.023
	Stem	1.322±0.014	2.011±0.008	3.110±0.056	2.266±0.041
August	Leaves	0.594±0.005	0.852±0.024	1.011±0.033	1.312±0.065
	Stem	0.323±0.012	0.311±0.009	0.454±0.055	0.854±0.008
October	Leaves	0.655±0.023	1.201±0.015	1.665±0.021	1.366±0.003
	Stem	0.464±0.007	0.623±0.022	0.744±0.005	0.954±0.020
December	Leaves	0.732±0.0001	1.344±0.032	1.695±0.036	1.522±0.004
	Stem	0.501±0.006	0.791±0.041	1.191±0.078	1.081±0.007

Table 3
Concentration of arsenic in middle parts of mangroves at bimonthly intervals of different sites of Karachi coast (μg/g, mean±SD).

Months	Parts	Sandspit	Kakapir	Manora	Korangi Creek
February	Leaves	0.912±0.005	1.323±0.001	1.494±0.004	1.752±0.014
	Stem	0.722±0.021	0.982±0.002	1.221±0.015	1.355±0.052
April	Leaves	1.294±0.054	1.411±0.036	1.864±0.025	3.543±0.004
	Stem	1.121±0.084	1.382±0.005	1.710±0.035	2.251±0.062
June	Leaves	1.474±0.001	3.625±0.009	1.323±0.066	4.565±0.001
	Stem	7.101±0.078	4.912±0.002	4.684±0.047	7.121±0.017
August	Leaves	0.322±0.002	0.903±0.006	1.312±0.012	1.565±0.002
	Stem	3.415±0.005	2.911±0.001	1.440±0.010	1.712±0.023
October	Leaves	0.532±0.003	0.711±0.005	0.796±0.033	0.854±0.054
	Stem	0.611±0.013	0.696±0.007	1.165±0.085	1.222±0.006
December	Leaves	0.624±0.065	0.955±0.008	1.082±0.065	1.154±0.025
	Stem	0.712±0.077	0.621±0.009	0.833±0.005	1.276±0.036

Table 4
Concentration of arsenic in bottom parts of mangroves at bimonthly intervals of different sites of Karachi coast (μg/g, mean±SD).

Months	Parts	Sandspit	Kakapir	Manora	Korangi Creek
February	Leaves	1.232±0.012	1.381±0.014	2.152±0.014	2.544±0.014
	Stem	1.464±0.052	1.712±0.052	2.291±0.058	2.851±0.017
	Roots	1.992±0.033	2.491±0.036	2.812±0.065	3.255±0.058
April	Leaves	1.352±0.011	1.212±0.014	2.281±0.004	8.762±0.045
	Stem	1.523±0.052	1.151±0.025	2.303±0.005	9.424±0.078
	Roots	2.311±0.004	2.665±0.036	3.644±0.001	9.975±0.045
June	Leaves	1.584±0.015	4.011±0.041	3.212±0.007	5.621±0.088
	Stem	2.355±0.006	5.900±0.021	2.133±0.026	3.412±0.075
	Roots	5.212±0.069	5.252±0.035	4.134±0.089	7.023±0.098
August	Leaves	0.482±0.033	1.133±0.021	1.705±0.075	1.812±0.012
	Stem	0.754±0.085	1.454±0.005	1.833±0.004	1.955±0.022
	Roots	1.353±0.001	1.612±0.035	2.635±0.004	2.834±0.036
October	Leaves	0.784±0.009	1.321±0.014	1.844±0.098	1.232±0.005
	Stem	0.633±0.025	1.388±0.025	2.122±0.078	1.651±0.012
	Roots	1.535±0.053	1.764±0.069	2.654±0.045	2.454±0.014
December	Leaves	1.081±0.062	1.922±0.058	1.965±0.065	1.855±0.022
	Stem	1.044±0.015	1.966±0.065	2.354±0.048	2.772±0.047
	Roots	1.565±0.021	2.212±0.003	2.722±0.004	2.841±0.088

4. Discussions

Along the entire coast of Pakistan, most affected areas by pollution are in the region of Karachi and its adjacent networks of Creeks[25]. Industries of Karachi discharge untreated municipal and industrial effluents through its two main rivulets, Malir and Lyari[20]. The study aimed to assess the arsenic level in sediments and different parts of mangroves in four coastal sites, Manora channel, Korangi Creek, Kakapir and Sandspit.The variation in the concentration of arsenic with respect to the location in sediments was found in decreasing order as: Korangi Creek> Manora > Kakapir > Sandspit.

In four stations this variation may be related to the dynamic conditions of sea water as metal deposition depends on water flow rate, height of tides, weathering conditions, sampling times, sediments type, and its affinity towards different elements.

The less concentration of arsenic 3.188-7.186 μg/g at Sandspit is because of no major industrial unit this region except domestic waste. In the west of Sandspit the beaches are mixed *i.e.* rocky and sandy of variable magnitude[26]. Korangi Creek had highest concentration (6.832-19.801 μg/g) because it receives waste water from domestic, Malir River, Korangi fisheries jetty, slaughter house and several villages such as Ibrahim Hyderi, many oil refineries.

Manora also shows high level of arsenic (7.466-19.335 μg/g). It is contaminated by Lyari and Malir River[27]. The channel receives domestic wastes from large size fishing villages on the islands of Manora, Baba and Bhitt, oil pollutants from ship traffic and oil refineries, and the wastes from shipyard and SITE area. SITE area had many small and large industries of food, beverages, paper and paper products, tobacco, textile, rubber products, chemicals, non-metallic minerals, basic metal, metal products, machinery, petroleum products, coal etc[25]. The wastes brought by the Lyari River (black water) are mostly untreated and are rich in total dissolved solids of 30 tons/day, creating many problems for the channel. The surface sediment here has changed from black into shades of greenish black and finally brown with the odor of sewer sludge and hydrogen sulphide[27]. The Kakapir was contaminated due to the domestically planted small industries and the arsenic level approached to 4.846-13.882 μg/g. The effluents from these industries carry many toxic compounds including arsenic. Arsenic has been used in various fields *i.e.* agriculture, livestock, electronics, industry and metallurgy.

The seasonal variations in sediment and mangroves were also assessed. The highest concentration in sediment is observed in Korangi Creek 19.801 μg/g and Monora 19.335 μg/g in the month of June which is dry season in Karachi. As the monsoon starts from the month of July to September this concentration declines and approaches to the lowest varying with locations. The tropical climate of this region is encompassing mild winters and warm summer. It follows high humidity levels from March to November, which fells down in winter as the wind direction is north easterly. In summer temperatures (From the end of April till the end of August) ranges from 30 °C to 42 °C. The annual average rain fall is about 2000 mm and it is highly intervallic. Most of the rain falls occur in less than three months, which bring about seasonally variable river flow between dry March to September and wet November to January[28]. The same pattern of arsenic level is shown in the present study according to season.

The fine texture of sediments has the high capacity to hold water so the dissolved arsenic in water bound with organic substances present in sediments. Sandy type sediments, organically poor have little ability to retain metal ions[29].

Results of the present study revealed that fine texture sediments of Manora and Korangi Creek have higher accumulation pattern than that of Sandspit and Kakapir.

Translocation of arsenic from sediment to different parts of mangrove is observed. The lower part of mangrove had higher concentrations as compared to the middle partand upper part. The high concentration in root as compared to the stem and leaves may be due to roots penetrates in the sediment and is in direct contact with the wastes. Generally the concentration of arsenic is more in stems than in leaves, but in some cases in December leaves of middle parts showed 1.082 μg/g and stem 0.833 μg/g. This may be due to old age mangroves accumulate much more arsenic than the new ones.

The accumulation and toxic effects are critically depends on form of arsenic. Inorganic arsenicals and monomethylarsonic acid are mainly accumulated in the roots, while dimethylarsinic acid is more readily translocated to the stem[30]. In hydrophobic condition regardless of the chemical form of arsenic, the concentrations in both root and stem significantly increased with increased levels of arsenic in the medium. The lowest level was found after monsoon because of the dilution effect of rainy season.

The important aspect is bioavailability of arsenic to mangroves and sediments. Its concentration varies from less than 0.01 to about 5.00 μg/g (dry weight basis)[31]. The mangrove sediment is anaerobic reduced, containing high organic matter due to microbial degradation. All molecular oxygens are removed beneath the surface layer, creating ideal conditions for the reduction of sulphate into sulphide. Thus the redox potential is an important factor controlling the retention of trace metals in sediments and along with the interstitial water pH, may change the concentration of dissolved metals and increase its availability. The bioavailability, mobility properties are depending on the different types of binding between trace elements and solid phases of the sample. Therefore, elements in the exchangeable and acid soluble fractions are considered as readily and potentially bioavailable to plant and aquatic organisms. Arsenic is naturally occurring in all soils and natural waters. It is chemically similar to the phosphorous, an essential plant nutrient. Arsenate can compete with phosphate for transport PO_4^{3-} as the substitute for plant nutrition; however the increased application of phosphate fertilizers increase the toxicity, through the release of fixed arsenic.

Conflict of interest statement

We declare that we have no conflict of interest.

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Comments

Background

Arsenic contamination can be devastating to plant growth and development. Mangrove tree *Avicennia marina* often confronts with man–made metal contamination, which not only impact the tree but also other floral resources. Assessment of such important issues is essential to devise mitigation strategies.

Research frontiers

The study assessed arsenic concentrations in various parts of the mangrove and sediments during different period of time using hydride generation atomic absorption spectrophotometry.

Related reports

Arsenic contamination is lethal for plant growth and development. It is essential to analyze the heavy metals uptake and regulations in economically important ecosystems.

Innovations and breakthroughs

The periodic assessment of arsenic in various parts of the plants are helpful to devise strategies to remediate contaminated systems.

Applications

The study could be helpful in formulation of conservation methods both at source and sink levels.

Peer review

The study decribes quantity of arsenic in *Avicennia marina* using hydride generation atomic absorption spectrometry, showing periodic and plant–part wise distribution. The article helps to understand the bioavailibity and cycling of arsenic in the mangrove system.

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