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RESEARCH ARTICLE

Bioaccumulation of potentially toxic elements in three mangrove species and human health risk due to their ethnobotanical uses

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Abstract

The aim of this study was to assess probabilistic human health risk due to ethnobotanical usage of *Avicennia officinalis*, *Porteresia coarctata* and *Acanthus ilicifolius*. The study was conducted at the tannery outfall near Sundarban (Ramsar wetland, India) mangrove ecosystem affected by potentially toxic elements (Cd, Cr, Cu, Hg, Mn, Ni, Pb, and Zn). Total metal concentrations (mg kg^{-1}) were considerably higher in the polluted rhizosphere namely, Cd (1.05–1.97), Cu (36.3–38.6), Cr (144–184), Hg (0.04–0.19), Mn (163–184), Ni (37.7–46.4), Pb (20–36.6), and Zn (97–104). Ecological risk index indicated low to moderate ecological risk in this site, whereas the ecological risk factor showed high potential ecological risk due to Cd pollution. BCR Sequential extraction of metals showed more exchangeable fraction of Cd (47–55%), Cr (9–13%), Hg (11–13%), and Pb (11–15%), at the polluted site. Mercury, though present in trace amount in sediment, showed the highest bioaccumulation in all the three plants. Among the toxic trio, Hg showed the highest bioaccumulation in *A. officinalis*, Cd in *P. coarctata* but Pb has the lowest bioaccumulation potential in all the three species. Occasional fruit consumption of *A. officinalis* and dermal application of leaf, bark of *A. officinalis* (antimicrobial), *A. ilicifolius* (anti-inflammatory, pain reliever when applied on wounds) indicated negligible human health risk. However, long-term consumption of *P. coarctata* (wild rice variety) seeds posed health risk (THQ>1) both in adults and children age groups. This study concludes that nature of ethnobotanical use and metal contamination levels of the mangrove rhizosphere can impact human health. The transfer process of potentially toxic elements from rhizosphere to plants to human body should be considered while planing pollution mitigation measures.

Keywords Heavy metal; · Sequential extraction; · Bioaccumulation factor; · Traditional medicine; · Hazard quotient; · Mercury; · Cadmium

Introduction

Mangroves are one of the most resilient ecosystem, playing a vital role in conservation of tropical/sub-tropical region, acting as a buffer between land and sea, as well as protecting the dense population residing along these coasts from natural disasters (Marchand et al. 2006; Maiti and Chowdhury 2013). Despite their role in maintaining the coastal ecosystem, mangroves are threatened and are disappearing worldwide at the rate of 1 to 2% per year, as a result of over-exploitation, pollution, and land conversion (Marchand et al. 2016).

Recent studies have focused on the pollution issues of potentially toxic elements (PTEs) in mangrove habitats spanning across the globe like in China, Japan, Papua New Guinea, Bangladesh, Indonesia Australia, Brazil, USA, and India (Tam and Wong 2000; Akhand et al. 2012; Souza et al. 2015; Rodríguez-Iruretagoiena et al. 2016; Marchand et al. 2016; Anbuselvan and Sridharan 2018). Exposure of PTEs can cause

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deleterious health impacts on human and environment (Marchand et al. 2016; Naz et al. 2021). Sediment being the "source and sink" of PTEs has the potential to transport PTEs to plants, growing on the contaminated sediment along the riverbanks. Recent studies have identified halophytes with high potential of metal accumulation in their roots and shoots growing in saline/hypersaline coastal soil or estuaries (Agoramoorthy et al. 2008; Akhand et al. 2012; Nath et al. 2014; Sarkar et al. 2014;Akhand et al. 2016; Chowdhury and Maiti 2016a, 2016b). Hence, using these plants with bioaccumulated PTEs in a polluted estuarine environment can cause deleterious human health hazards. Researchers tried to predict the probabilistic health risk associated with consumption/ingestion of water or food products and dermal exposure to toxic metal contaminated dust/soil at polluted regions (Zheng et al. 2007; Chotpantarat et al. 2014; Chowdhury et al. 2015; Naz et al. 2016a, 2016b; Naz et al. 2018). But, other exposure routes such as usage of plants with ethnobotanical properties are generally not investigated by environmentalists while assessing human health hazard. India and most of the Asian countries believe on the ethnobotanical usage of the natural plant and mangroves have widely been used in traditional medicine or as food adjuncts (Bandaranayake 1998; Sheng-Ji 2001; Muthu et al. 2006; Dahdouh-Guebas et al. 2006; Qasim et al. 2014). Recent study by Naz et al. 2020 has identified health risks due to the ethnobotanical usage of plants growing at a Chromite mine area of India contaminated by potentially toxic elements. But till date, there no reported research on metal contamination impact due to the ethnobotanical usage of mangroves on human health.

Mangroves are widely used for ethnomedicinal properties in Asiatic nations (Wu et al. 2008). Research by Nabeelah Bibi et al. 2019indicates that India and Bangladesh accounts for about 51% of global use of mangrove as ethnopharmacological supplements. *Avicennia officinalis*, *Porteresia coarctata*, and *Acanthus ilicifolius* have wide ethnobotanical usage in the region. Scientific evidences prove that leaf of *A. officinalis* has alkaloids - terpenoids, phenolics, pentacyclic triterpenoids, and flavonoids (Shanmugapriya et al. 2012; Hossain 2016). Leaves of this plant are widely used as powder or paste for ameliorating joint pain as well as healing of wounds by the local community. *A.ilicifolius* leaf contains terpenoid, phenolic compounds and is widely used across India, Bangladesh as aphrodisiac, to alleviate rheumatism pain, relief for asthma, diabetes, diuretic, dyspepsia, leprosy, hepatitis, blood purifier, cold cure, gangrenous wounds, skin allergies, and snake bites (Bandaranayake et al. 1998; Babu et al. 2001;Hossan et al. 2010; Firdaus et al. 2013; Nabeelah Bibi et al. 2019). *P. coarctata* is a wild rice variety with a mean carbohydrate content of 39% of dry weight and is sometime taken as a good grain by local communities in times of acute food crisis during cyclone or storm surges. (Sengupta and Majumder 2010; Ghosh and Mitra 2015).

Literature review of available academic studies revealed limited information on metal accumulation in different plant parts of the three mangroves (*Avicennia officinalis*, *Porteresia coarctata*, and *Acanthus ilicifolius*), growing naturally in a contaminated habitat (Chakraborty et al. 2013; Shackira and Puthur 2013; Wu et al. 2015; Chakraborty et al. 2018). Hence, understanding the bioaccumulation processes of PTEs can act as a proxy to comprehend the metal uptake potential and eventual human health risk posed by use of these plant parts in traditional medicine around the mangrove habitats.

In this study, we have considered the PTE transport processes, their chemical fractions in sediment and their transfer to different mangrove plant parts and eventual health hazard due to the traditional use. This study was conducted at tannery, municipal out-fall site near the metropolitan city of Kolkata and its suburbs having an approximate population of 14.3 million as per 2011 census, which is India's third populous metropolitan area. This outfall site is at the doorway of the world's largest contiguous mangrove forest of Sundarbans, shared by India (40%) and Bangladesh (60%) and recently declared as a Ramsar Wetland site on February 2019 (site number: 2370). Past researches have shed light into the alarming PTE pollution (Cu, Cd, Cr, Hg, Ni, Mn, Pb, Zn) at Indian Sundarbans due to anthropogenic interventions (Saha et al. 2001;Banerjee et al. 2012; Rodríguez-Iruretagoiena et al. 2016; Akhand et al. 2016; Chowdhury and Maiti 2016a, 2016b).

Thus, the aim of this study was to address the research question, whether PTE accumulation in the rhizosphere and its subsequent bioaccumulation in plant species can be a human health concern due to the ethnobotanical use of the plant parts? The primary objective of this study was, (a) determining the concentration of PTEs (Cd, Cr, Cu, Hg, Ni, Pb and Zn) in the rhizospheric sediment of three halophytes (*Avicennia, officinalis, Acanthus ilicifolius* and *Porteresia coarctata*) growing at Kolkata municipality outfall, (b) sequential extraction of PTEs in the rhizospheric sediments of the targeted halophytes to understand the fractions of metal present in exchangeable (bioavailable), oxidisable, reducible and residual form (c) bioaccumulation potential of the concerned PTEs in different plant parts, and (d) Probabilistic human health risk assessment by the traditional usage of target plant parts growing at the study area.

Material and methods

Study site description

Mangrove and halophytes at the Ghushighata outfall (22.52373° N, 88.69492° E), exposed to the municipal sewage and tannery effluent, wasselected as the study area to understand the effect of PTEs contamination on three species, *Avicennia officinalis*, *Acanthus ilicifolius*, and *Porteresia coarctata*. Control site was selected amidst the undisturbed,

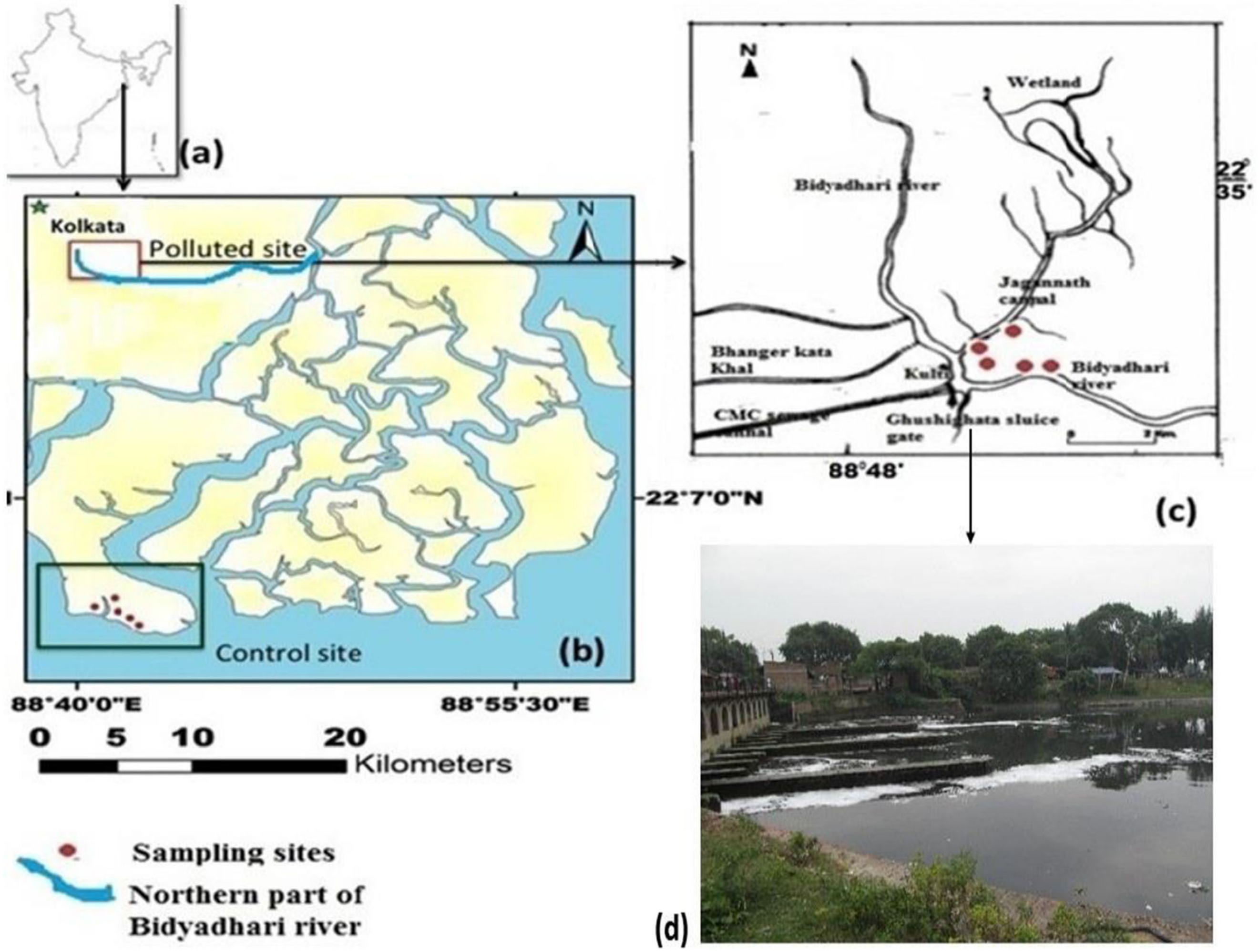


Fig. 1 The study area and the sampling site, (a) study area is marked out from the map of India, (b) the area of Sundarban Ramsar Wetland site, with the location of the metropolitan city of Kolkata and the un-polluted control patch along with polluted sites, (c) the polluted site is zoomed out showing the municipal and tannery effluent canal (CMC canal) and the outfall, (d) the Kulti sluice gate where the salt water from the estuary mixes with the CMC canal

conserved, pristine mangrove forest of Jharkhali (22.00889°N, 88.7134°E) downstream of the same river (Fig. 1).

Sediment sample collection and laboratory treatment

The sediment samples were collected during the monsoon to post monsoon period between August to September of 2015. The time period was selected because of this coincides with fruiting/ seed dispersal time of the three targeted mangroves (Chowdhury et al. 2016b). Considering the spatial distribution and mangrove species composition, the superficial sediment (0–30 cm) was sampled from base of target plant species from sites, near the tannery outfall as well as control site. For each species, seven individual plants were randomly selected from both polluted as well as control site and samples collected from their rhizospheric sediment. All samples were collected during low tide. At each plant rhizosphere, four sub-samples were retrieved manually using acid washed PVC pipes (30 cm length) with help of stainless steel scoop (to aid in removing the PVC pipes from mangrove mud) and mixed into a composite sample. The plant materials/parts were also collected from same plants.

The samples were preserved in ice box till their transport to laboratory. A composite sample was prepared for that particular species from seven replicated sediment samples, sieved (< 62 micron), stored in sterilized zip lock bag and kept at 4 °C in refrigerator for further analysis (APHA 1985; Maiti 2003; Anbuselvan and Sridharan 2018). The pH and electrical conductivity (EC) of the sediment samples were measured in a soil–water suspension (1:1.5; soil: deionised water) using a pH meter (Cyberscan 510) and conductivity meter (EI 601) respectively (Maiti 2013). For soil texture analysis, samples were wet sieved through a 62-μm mesh for 15 min in a mechanical sieve shaker. The sample that retained on the sieve was weighed and expressed as sand. The samples which passed through the sieve were expressed as mud. The finer

Table 1 Soil texture (sand, silt, clay) of the samples collected from the rhizosphere of the plants from polluted and control site in % ± standard deviation (*n*=3)

Polluted rhizosphere			
	Sand	Silt	Clay
Avicennia officinalis L.	37.61±4.6	43.0±2.4	19.3±3.1
Acanthus ilicifolius L.	46.2±3.2	42.2±3.9	11.59±0.9
Porteresia coarctata (Roxb.) Tateoka	39.2± 3.5	37.6 ± 1.5	23.2 ± 2.4
Control rhizosphere			
	Sand	Silt	Clay
A.officinalis	36.8 ± 4	35.7 ± 4.3	27.5± 1.2
A.ilicifolius	30.6± 1.9	57.8± 1.5	11.6± 1.6
P. coarctata	25.1± 4.7	47.4± 2.2	27.5 ± 2.6

fraction, mud, i.e., silts and clays (> 0.063 mm) were determined by pipette method. The soil texture characteristics of sediments are given in Table 1.

The collected sediment samples were air-dried, crushed in a porcelain mortar, sieved through a nylon sieve (Pore size, 0.45 mm), and kept in polypropylene air tight zip bags for further analysis. Organic carbon (OC) of sediment samples was determined by the rapid dichromate oxidation method (Walkley and Black 1934; Anderson and Ingram 1998). While preparing reagents for analysis of OC, 0.1 g Ag_2SO_4 was added before the use of H_2SO_4 to minimize the interference of Cl^- in the saline mangrove soil as the excess Cl^- get precipitated in the form of AgCl (Saha et al. 2001; Schumacher 2002; Wang et al. 2013; Chowdhury et al. 2018). $K_2Cr_2O_7$ solution was used to oxidize the organic carbon in acid medium. The amount of oxidized carbon in the sample was proportional to the amount of chromic ions produced during oxidation. Colorimetric method was employed to determine the chromic ion concentration and the absorbance was recorded by using UV–visible spectrophotometer (Helios Aquamate, Thermo scientific 9423 AQA 151505, made in England) (Schumacher 2002; Perera et al. 2018).Total Kjeldhal Nitrogen (TKN) was determined by alkaline permanganate method by taking 20g of oven dried sediment sample in an 800 mL Kjeldahl flask with 20 mL of water and 100 mL 0.32% $KMnO_4$ and 2.5% NaOH solution and boiled in presence of glass beads. The content was distilled in a Kjeldahl assembly at a steady rate. Ammonia gas released in this process was collected in a 250-mL conical flask containing 20 mL of boric acid indicator solution (pinkish color turns green) and the distillate was titrated against 0.02N H_2SO_4 (Jackson 1973; APHA 1985; Maiti 2003).

Sequential extraction of metal in sediment

The optimized three-stage extraction procedure was used to determine the distribution of PTEs in different chemical fractions using the method Community Bureau of Reference (BCR) metal extraction process first introduced by Ure et al. 1993 and later applied by many researchers (Mossop and Davidson 2003; Sahuquillo et al. 2003; Naz et al. 2020). This method also been used for the sequential extraction of Hg by Sahuquillo et al. 2003 and Ge et al. 2020. The steps of the extraction process have been elaborated below:

Exchangeable fraction: A total of 40 mL of 0.11 M acetic acid was added to 1 g air-dried sediment and shaken overnight. The mixture was centrifuged to separate the extract from the residue.

Reducible fraction: A total of 40 mL of 0.5 M hydroxylammonium chloride, adjusted with nitric acid to the pH of 1.5, was added to the residue from Step 1 and the extraction performed as above.

Oxidisable fraction: The residue from Step 2 (Reducible fraction extraction) was treated twice with 8.8 M hydrogen peroxide, evaporated to near dryness, then 50 mL of 1.0 M ammonium acetate, adjusted to pH 2 with nitric acid, was added and the extraction performed as above.

Residual fraction: The material remaining at the end of the step 3 (Oxidisable fraction extraction) was digested in 20 mL aqua regia, with microwave assistance (Model: ETHOS One, Italy) at 100% power with pressure set at 120 psi for 25 min (two cycles).

The aliquots were filtered through Hg-free filter paper (pore size 2.5 μm), and analyzed for Cd, Cr, Cu, Hg, Ni, Mn, Pb, and Zn using a flame atomic absorption spectrophotometer (FAAS-GBC Avanta, Australia) at the most sensitive resonance wavelength of each element. Hg was measured in cold vapour-atomic absorption spectrophotometer (CV-AAS, GBC Avanta, Australia) (Marrugo-Negrete et al. 2015; Raj et al. 2020). The assessed PTEs concentrations were equated to the weight of dried material used for digestion.

Pollution indices

Contamination factor (C^i_f) Contamination factor was initially suggested by Hakanson (1980). Contamination factor was used to assess contaminated sediment by referencing the concentration of metals in the surface layer of bottom sediment to background values of that metal. Contamination factor was calculated as:

$$C_f^i = C_m^i / B_m^i$$

where C_m^i = concentration of ith element in sediment and B_m^i = average shale value of earth's crust for the i^{th} element. Based on the value of C_f^i, the sediments are classified as low ($C_f^i < 1$), moderate ($1 \le C_f^i \le 3$), considerable ($3 \le C_f^i \le 6$) and very high pollution ($C_f^i \ge 6$) (Hakanson 1980).

This index shed light into the potential of pollution and used to assess the tolerance of different plants for different ecological risk. The $C^i{}_f$ is the ratio of heavy metal concentration in the sediment to the average shale value of earth crust (Hakanson 1980; Raj et al. 2017). For background concentration of metals, the average shale values for different metals were used for Pb: 20 mg/kg, Ni: 68 mg/kg, Cu: 45 mg/kg, Cd: 0.3 mg/kg, Cr: 90 mg/kg, Hg: 0.4 mg/kg, Zn: 95 mg/kg, and Mn: 850 mg/kg (Turekian and Wedepohl 1961; Uluturhan et al. 2011).

Ecological risk index (ERI) Hakanson (1980) proposed a method to assess the potential ecological risk for areas under special conservation attention. According to this method, the potential ecological risk factor ($E^i{}_r$) of a single element can be assessed with formulae incorporating the toxic response of each metal as given below,

$$E_r^i = T^i \times C_f^i$$

ERI is the summation of the ecological risk factors as stated below,

$$ERI = \sum_{i=1}^{n} E_r^i$$

$E^i{}_r$ is categorized into five divisions. $E^i{}_r$< 40= Low ecological risk, 40 <$E^i{}_r$< 80= moderate potential ecological risk, 80 <$E^i{}_r$ < 160= considerable potential ecological risk, 160< $E^i{}_r$< 320= high potential ecological risk, $E^i{}_r$> 320= very high ecological risk. The cumulative E_r^i values for all the elements indicate the level of ecological stress to different study area due to metal pollution.

Ecological risk factor ($E^i{}_r$) is the C_f^i multiplied by toxic response factor (T_i), which differs between different metals, as all metals do not possess similar negative foot print in a particular ecosystem. The toxic response factor (Ti) for Cd = 30; Cr =2; Cu = 5; Hg = 40; Mn = 1; Ni = 5; Pb = 5; and Zn = 1, were considered for the ecological risk factor analysis (Hakanson 1980; Raj et al. 2017).

Ecological risk index (ERI) is the summation of the $E^i{}_r$ for all the individual metals. Whereas, 150< ERI= low ecological risk, 150< ERI< 300= moderate ecological risk, 300 < ERI< 600= considerable ecological risk, ERI> 600= very high ecological risk.

Plant sample collection and metal extraction

To determine PTEs in selected halophytes, different parts of the plant were collected (Supplementary materials: Table S1). *Avicennia officinalis* is a tree of Avicenniaceae family. Roots, barks, leaves and fruits of *A.officinalis* were collected separately from seven trees. In case of fruits, only one mature tree growing in the polluted site was selected to collect the fruits as other plants were not in the fruiting age. Root samples were collected in the upper 30 cm of rhizospheric sediment at low tide (*n* = 3 per each type of tree). Bark was collected at breast height from same plants. *Porteresia coarctata*, a Poaceae herb species, was uprooted with seeds. Seven such plants were collected and separated into root, shoot and spikelet. *Acanthus ilicifolius* is a thorny shrub of Acanthaceae family. The leaves, fruit, shoot, and roots were collected from the plants for analysis. As the leaves were thorny, extreme care was taken while separating the leaves and fruits from shoot with help of an acid washed stainless steel scissors. All the plant parts were dried, digested, and analyzed three samples for each plant parts (*n*= 3). That data was used to estimate health risk due to accumulated PTEs. Similar method of sampling was employed by Thành-Nho et al. 2019.

All the collected plant samples were washed several times with distilled water in an ultrasonicator before processing for metal analysis. After that, samples were oven dried at 40°C until a constant weight was achieved and fine particle were made by using mortar and pestle. The fine sized plant samples, (0.5 g) of each parts with three replicates (roots, shoots, fruits, and seeds were digested using microwave-assisted digestion system (Ethos One, Milestone Srl, Italy) HNO_3 (9–71% Nitric acid, 5 mL; and 30% Hydrogen peroxide, 2 mL) at 90°C and made up the volume up to 100-mL with 1% HNO_3 (Chowdhury et al. 2016a; Marrugo-Negrete et al. 2015; Raj et al. 2017; Raj et al. 2020). Blank acid mixtures were digested in the same way. The digested plant samples were passed through Whatman no. 42 filter paper (pore size 2.5 μm), diluted with Hg-free Millipore water and measured in cold vapour-atomic absorption spectrophotometer (CV-AAS, GBC Avanta, Australia) for Hg estimation. Other trace metal concentrations in all the samples were analyzed using flame atomic absorption spectrophotometer (FAAS-GBC Avanta, Australia). Inter batch variations were monitored by repeated analysis of selected samples in various analytical batches (less than 10% relative variation).

Metal accumulation indices

Following formulae was used to determine metal accumulation factors, i.e., bio-accumulation factor (BAF). The mean

Table 2 Highlights the medicinal, pharmacological properties of target mangrove species and dosage used for health risk assessment

Plant species	Family	Ethnobotanical use/economic importance	Active principle/secondary metabolite activity/nutrient content	Parts used#	Dosage#	Reference
Avicennia officinalis L.	Verbenaceae	Root, bark, leaf and fruits having antimicrobial property. The leaf of the plant considered as antiulcer. Bitter fruits are used to eat in local areas. People used to apply leaf and bark extract for the healing of wound.	Contains 3 chlordeoxylapachol, and 20 metabolite obtained was active against K B Human cancer cells in the murine hallow fiber antitumor	Fruit, bark and leaf	Average 30 fruits in a year consumed by local people. Leaf and bark of the plant (powdered/pasted) were used to be applied on wound and joint pain.	Das et al. 2018; Thirunavukkarasu et al. 2010; Jones et al. 2005; Bhimba et al. 2010
Acanthus ilicifolius L.	Acanthacae	Leaf and stem having anti- inflammatory property. The shoot and leaves are applied in wound for pain relief and healing and reportedly have high morphine. Sometime people uses this plants roots and shoots to treat leucorrhea and cloudy urination in women	Contains terpenoid, phenolic compounds, and alkaloid. It shows antiradical efficiency, is potential antioxidant and cytotoxic agents as well as imminent candidate for cancer therapy.	Herbaceous shoot and leaves	Paste of herbaceous shoot and leave applied on wound occasionally, the average body surface area exposed was about 1.5 to 2% of total body surface area. Thus the 1.7% of total body surface area was considered for this study, i.e. 0.028 and 0.014 m^2 for adult and children respectively.	Saranya et al. 2015; Hossan et al. 2010; Firdaus et al. 2013; Singh and Aeri 2013.
Porteresia coarctata (Roxb.) Tateoka	Poaceae	It is also known as ‘wild rice’, people in rural areas used to cook its seed as alternative of rice. The demand of this seed become higher in poor people basically in the condition of food or economic crisis (flood, cyclone/disaster)	Mean carbohydrate content is 39% of dry weight and protein content is 9% of dry weight.	seed	100 and 50 g of seed consumed by adults and children respectively for an average 150 days in a years	Sengupta and Majumder 2010; Ghosh and Mitra 2015

= Data procured through verbal consultation with local population and available literature

metal concentration in the sediments and mangrove plants was considered for calculating the BAF for all three plant species.

$$BAF = \frac{Metal\ concentration\ in\ Plant\ (Root + Fruit\ or\ Seed + Shoot + Leaf)}{Metal\ concentration\ in\ Sediment}$$

Heath risk assessment

The human health risk assessment was carried out by using USEPA model (Wongsasuluk et al. 2014; Naz et al. 2018; Rahman et al. 2019). The exposure of PTEs via ingestion of mangrove plant parts (fruits of *A.officinalis* and seed of *P. coarctata*) and by dermal exposure of leaf and bark of *A. officinalis* and *A. ilicifolius*) grown in polluted and control (non-polluted) sites were analyzed for children and adult population. The average exposure, frequencies, and dosage were established by consultation with local people and available literature as per Naz et al. 2020 (Table 2). The plant parts, i.e., fruit, leaves, bark were considered in terms of their average weight of use for calculating the human health risk, as elucidated in Table 2. Applied formulae for human health risk calculation was

$$ADD_{ing} = \frac{C \times R_{ing} \times EF \times ED \times 10^{-6}}{B_w \times AT}$$

$$ADD_{der} = \frac{C \times SL \times SA \times ABS \times EF \times ED \times 10^{-6}}{B_w \times AT}$$

$$HQ = \frac{ADD}{RfD}$$

$$THQ = \sum HQ$$

where ADD_{ing} (mg/kg-d) is average daily dose by ingestion, ADD_{der} (mg/kg-d) is average daily dose by dermal contact, C (mg/kg) is average PTE concentration, EF (Events/day) is exposure frequency, ED (years) is exposure duration, Bw (kg) is average body weight, AT (days) is average time, SL(mg/cm^2/h) is skin adherence factor, ABS is absorbance. RfD (mg/kg-d) is the reference dose. HQ is hazard quotient and THQ is total hazard quotient. THQ is the cumulative sum of all the individual metal's HQs (Liu et al. 2020; Tong et al. 2020). In the present study, the exposure frequency (EF) for fruits of *A. officinalis* was taken 30 days in year, for *P. coarctata* 150 days in year for ingestion; however, the EF for dermal exposure for leaf and bark of *A. officinalis* and *A. ilicifolius* was taken 100 days/year. The exposure duration (ED) for children and adult was taken 12 and 70 years and an average body weight was considered 27 and 70 kg respectively (Naz et al. 2018). The absorbance of each metal was taken 0.001 for people of both age groups (USEPA 2020; Ihedioha et al. 2017). As leaf and bark applied only specific part of the body the skin adherence factor (SL), was considered only 5% of total surface area for both adult and for children (Naz et al. 2018). SA was body surface area (m^2). The average surface area was calculated by using the formulae $SA = 0.015924\ (H \times W)^{0.5}$, where H is average height (cm) and W is average body weight in kg (Redlarski et al. 2016; Yu et al. 2003). Traditional usage of mangrove plant parts donot involves a whole body exposure and dermal applications are restricted to limited part of the body surface. Hence, the maximum exposed body surface area was taken to be 1.7% of total surface (Table 2). Reference dose (RfD) is no-observed adverse effect level (NOVEL) of any exposure of target toxic substance over a long time duration (up to a lifetime) on the human population. Hence, this assessment is based on "chronic" toxicity and not limited to acute responses of the contaminants. RfD is calculated by dose response curve, in which X-axis denotes dose (in mg/kg of body weight/d), and Y axis is response (Barnes et al. 1988; USEPA 2002). The RfD factor used for the calculation in this health risk study was adopted from literature and presented in the supplementary material (Supplementary materials: Table S2).

Quality control and assurance (QA/QC)

All the analysis was carried out at "Ecological Restoration laboratory, department of Environmental Science and Engineering" in Indian Institute of Technology (Indian School of Mines), Dhanbad, India. Analytical grade glassware and reagents were used in whole analysis, and the standard solutions were prepared with millipore deionized water. The accuracy and precision of the PTE analysis were checked against Certified Reference Material, MESS-4 of marine sediment from the National Research Council, Canada for sediment (Cuong and Obbard 2006; Raj et al. 2017; Pal and Maiti 2019). The accuracy of the microwave-assisted acid digestion procedure for total metal determinations was checked using the Certified Reference Material, MESS-4 of marine sediment from the National Research Council, Canada. To check the accuracy of measurement, reference sample was re-checked after every 10 samples. The percentage recovery values for different PTEs were listed in supplementary material (Table S3). An internal check on the results of the microwave extraction procedure, the sequential extraction procedure was performed by a mass balance analysis by comparing the sum of the 4 steps (acid-soluble + reducible + oxidizable + residual) from the sequential extraction procedure with the total metal content from the microwave-assisted acid digestion procedure. The recovery of the sequential extraction procedure was calculated as follows:

$$Recovery = \frac{\text{Fraction 1} + \text{Fraction 2} + \text{Fraction 3} + \text{Residual fraction}}{Total\ Digestion}\ X\ 100$$

The analysis result varies between 74 and 124%, depicted in supplementary material Table S4. Analytical results obtained for reference materials differed by ±5%. The FAAS detection limits of metals (mg/L) were as follows: Cd = 0.0004, Cr=

Table 3 Table elucidates the concentrations of potentially toxic elements (Zn, Cu, Pb, Hg, Cd, Mn, Ni, and Cr) in mg/kg and available nitrogen (Av N) concentrations (mg/kg), electrical conductivity (EC) (μS/cm), and organic carbon (% OC) in the rhizosphearic sediments of three halophytes at two sampling sites (polluted and control)

Plant	Cu	Cd	Ni	Zn	Mn	Cr	Pb	Hg	pH	EC	OC	Av N
Polluted site (*n*=7)												
A. officinalis	37.7 ± 0.6	1.97^{a} ± 0.1	46.4^{a} ± 2.6	97.4 ± 3.9	163^{b} ± 15	184.6^{a} ±11.4	36.6^{a} ± 1.3	0.04 ^{a} ±0.01	5.7^{c} ± 0.5	1434.6^{c} ± 72.3	0.94^{b} ± 0.1	42.5^{b}± 10.1
A. ilicifolius	36.3± 0.6	1.6^{b} ± 0.4	44.8^{a} ± 2.8	100.9 ± 9.9	177.96^{a} ± 2.7	168.7^{b}±10.3	27.3^{b}± 1.3	0.19 ^{b} ±0.02	6.2^{b}± 0.2	1520.3^{b}± 81	0.97^{b}± 0.1	46.7^{b}± 5.8
P. coarctata	38.6± 3.6	1.05^{c} ± 0.1	37.7^{b} ± 2.3	103.9± 11.4	183.96^{a} ± 8.1	143.9^{c}± 2.7	20^{c}± 0.5	0.09 ^{c} ±0.03	6.79^{a}± 0.3	1633.1^{a}± 41.9	1.09^{a}± 0.1	52.5^{a}±2.7
F	2.1	29.5*	23*	0.9	8.23*	36.3*	265.8*	87.9*	14.99*	15.4*	7.16*	3.7*
Control site (*n*=7)												
A. officinalis	26± 1.9	0.25± 0.3	38.75± 4.8	93.75± 2.5	156.25± 2.5	22.88± 1.6	12.38± 1.3	bdl	7.25± 0.1	3640± 325.4	1.7± 0.2	84.1± 9.1
A. ilicifolius	22.9± 0.1	0.49± 0.5	41.1± 3.1	87.45± 5.7	149.45± 7.4	20.375± 4.1	11.175± 0.8	bdl	7.2± 0.1	3614.5± 356.1	1.83± 0.3	80.96± 0.1
P. coarctata	25.18± 3.4	0.7375± 0.3	42.63± 2.8	90.53± 6.1	155.58± 3.2	21.52±0.6	12.55± 1.6	bdl	7.5± 0.2	3627.5± 314.3	1.93± 0.2	86.18±9.1
F	1.998	4.058	1.126	1.566	2.325	0.943	1.368		4.252	0.006	0.602	0.493

The mean data is reported with ±Standard Deviation (SD). *bdl* below detection limit, *n* number of replicates.*Significant difference of mean at significance level <0.05; same letters indicate no significant difference at $p = 0.05$ according to DMRT post hoc test. One-way ANOVA has been conducted separately between the rhizospheres in polluted and control sites

0.003, Cu=0.001, Ni= 0.004, Mn=0.0015, Ni= 0.009, Pb= 0.01, and Zn = 0.0005. The instrumental limit of detection (CV-AAS) for Hg was 0.0005 mg/L.

No experiments had been conducted on humans or any animal subjects and the risk assessment were calculated through empirical model prescribed by USEPA guidelines (USEPA 2005). All experiments have been complied with strict scientific ethics and quality control. The respondents of ethnobotanical survey were all above 18 years in age and have willingly given information about the traditional use of the plants and doses.

Statistical analysis

Statistical analysis was carried out to interpret the result by using Data Analysis Package of MS Excel 2007 and SPSS 16 (SPSS Inc. Chicago, USA). Descriptive statistics like one-way ANOVA was used to access the variance between means of analyzed parameter among the sampling sites and where significant *F* value was observed. Difference between individual means was tested using DMRT (Duncan's Multiple Range Test) at 5% level of significance. Kruskal-Wallis test was only used when normality (Shapiro-Wilk's test) and homogeneity (Leven's test) were violated which was not the case for this study. For soil texture analysis, the sample size was taken as *n*=3, while for all other statistical analysis regarding sediment samples, it was *n*=7. Plant samples and estimation of metals in biological samples were done with a sample size of *n*=3.

Result and discussion

Chemical characteristic of sediments

Sediment samples were collected from the rhizosphere of the target mangrove species from both polluted and control sites. One-way ANOVA revealed that Cd, Cr, Hg, Ni, Mn, and Pb and associated parameters of pH, electrical conductivity (EC), percentage of organic carbon (OC), and available nitrogen (Av N) showed a significant difference in mean ($p < 0.05$) between the rhizospheres of each plant species in the polluted sites except Cu and Zn (Table 3). But in the control site of Jharkhali, one-way ANOVA showed insignificant results ($p > 0.05$) when tested for differences in mean between all these parameters of the rhizosphereic sediment samples. The reason can be that in the polluted sites, the plants tend to modify their rhizospheres to ameliorate metal pollution stress unlike the control site. Study conducted in the same sites showed that metal pollution plays an important role in defining the plant biodiversity and plant community composition (Chowdhury and Maiti 2016a). Study revealed that high metal concentrations in the polluted sites have a selection pressure on the plant species, and only the species that can tolerate metal toxicity

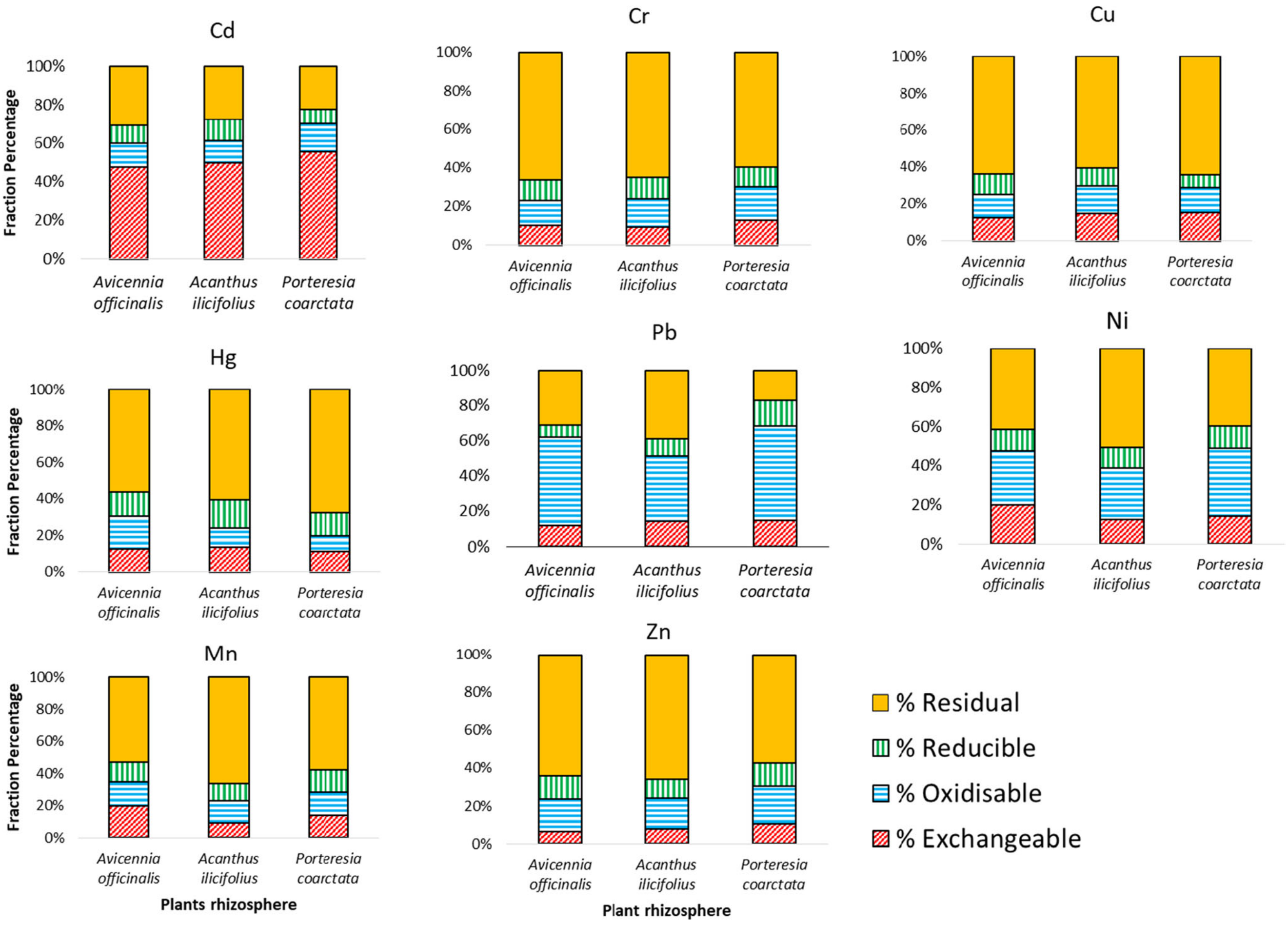


Fig. 2 **a** Fraction of the Potentially Toxic Elements (PTE) found in rhizospheric sediments collected from study area (Polluted site) as determined by the BCR sequential extraction method. **b** Fraction of the potentially toxic elements (PTE) found in rhizospheric sediments collected from study area (control site) as determined by the BCR sequential extraction method. Hg is not indicated in the figure of the control site as that metal has been found to be below detection limit

can survive in the landscape. Mangrove plant community structure also influences the diversity of soil microbiota as evident from works of Li et al. 2011, Chen et al. 2016 and soil microbiota directly influence the nutrient dynamics, as well as overall edaphic environment (Ikenaga et al. 2010).

The concentration (mg/kg) of Cr (143–184), Cd (1.05–1.97), Hg (0.04–0.19), and Pb (20–36.6) were alarmingly high in the polluted site as is evident from current study and past researches in this area (Rodríguez-Iruretagoiena et al. 2016; Chowdhury et al. 2017; Asha et al. 2020). Cr, Cd, and Pb tend to show toxic responses in biota above the level of 90 mg/kg, 0.3 mg/kg, and 20 mg/kg respectively in the sediment (Hakanson 1980; Raj et al. 2017). The average metal concentration in the sediment at the polluted site (*A. officinalis* rhizosphere) was in the order, Cr> Mn> Zn> Ni> Cu>Pb> Cd> Hg. Metal concentrations at *A. ilicifolius* rhizosphere were in the order of Mn> Cr> Zn> Ni> Cu>Pb> Cd> Hg at the polluted site. In the same site, *P. coarctata* rhizosphere was in the order of Mn> Cr> Zn> Ni> Cu>Pb> Cd> Hg.

The sequential extraction of PTEs in polluted and control sediment has been elucidated in Fig. 2a and b respectively. It is evident that the metal partitioning varies with place to place and with mangrove species (polluted vs. control). Studies around the saline estauarine/coastal habitats also shows similar type of metal speciation (Kabata-Pendias and Pendias 2001; Alloway 2013). Here, in the present study, it is evident that polluted site has more exchangeable form of toxic metals like Cr, Pb, Cd, and Hg in comparison with the unpolluted site, proving an extensive risk in the site. But in control site geogenic metals like Zn, Mn has more mobility (exchangeable form) or in form of organic bound fraction in comparison to the polluted site. Microbial activity may be more in the control site, because of less concentration of toxic metals. And Zn, Mn is utilized by microbes owing to its less toxicity. So, both of metals were found to be in higher exchangeable/organic bound fractions in control. Contrary to the prevailing trends, Cd is present in predominant exchangeable form (47–56%) in the polluted rhizospheres. As per similar studies, it is evident

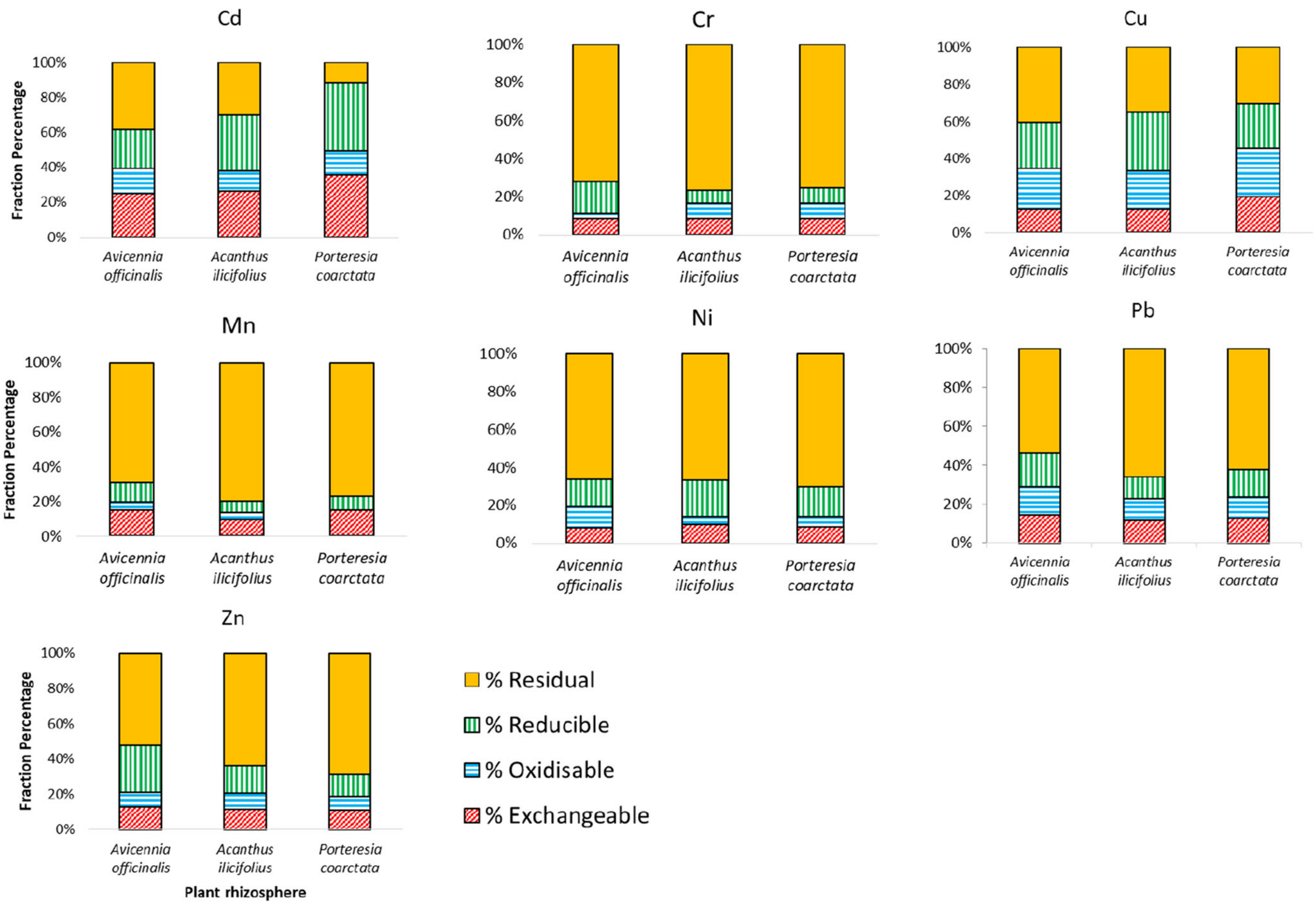


Fig. 2 continued.

that Cd has similar ionic radius like Ca, which is 0.97°A in former and 0.99°A in later. Hence, Cd can co-precipitate with carbonates and favoring its incorporation to the calcite lattice that result in the formation of $Cd_{\alpha}Ca_{1-\alpha}CO_3$ (Ma et al. 2016; Mondal et al. 2020). Pb was mostly present in oxidizable form (37–53%) in the polluted rhizospheres. Literatures indicate that in presence of organic matter and decomposing biological materials, Pb can be found in the oxidizable form as it binds on the organic matter surface (Anastopoulos et al. 2019). Sundarban ecosystem has high organic matter content and organic carbon residues (Chowdhury et al. 2018, 2019). Most of the other metals were in the residual fractions indicating their lower mobility in the mangrove rhizospheres. Metal fractions from Hoogly river (lower part of Ganges) sediment

Table 4 Contamination factor (C_f), for the rhizospheric sediments at polluted study sites

C_f^i								
Plant rhizosphere sediment (Polluted site)	Cd	Cr	Cu	Hg	Mn	Ni	Pb	Zn
A. officinalis	6.56	2.05	0.84	0.10	0.19	0.68	1.83	1.02
A. ilicifolius	5.45	1.87	0.81	0.48	0.21	0.66	1.37	1.06
P. coarctata	3.50	1.60	0.86	0.23	0.22	0.55	1.01	1.09
C_f^i								
Control site	Cd	Cr	Cu	Hg	Mn	Ni	Pb	Zn
A. officinalis	0.83	0.25	0.58	-	0.18	0.57	0.62	0.99
A. ilicifolius	1.63	0.23	0.51	-	0.18	0.60	0.56	0.92
P. coarctata	2.46	0.24	0.56	-	0.18	0.63	0.63	0.95

low($C_f^i < 1$), moderate ($1 \leq C_f^i \leq 3$), considerable ($3 \leq C_f^i \leq 6$), and very high pollution ($C_f^i \geq 6$)

Table 5 Ecological Risk Factor (E^i_r) and Ecological Risk Index (ERI) for the rhizospheric sediments at polluted and control study sites

At polluted site	Cd	Cr	Cu	Hg	Mn	Ni	Pb	Zn	ERI
A. officinalis	196.79	4.10	4.19	4.00	0.19	3.41	9.15	1.02	222.85
A. ilicifolius	163.57	3.75	4.03	19.00	0.21	3.30	6.83	1.06	201.75
P. coarctata	105.00	3.20	4.29	9.00	0.22	2.77	5.00	1.09	130.57
At control site									
A. officinalis	25.00	0.51	2.89	00	0.18	2.85	3.09	0.99	35.51
A. ilicifolius	49.00	0.45	2.54	00	0.18	3.02	2.79	0.92	58.91
P. coarctata	73.75	0.48	2.80	00	0.18	3.13	3.14	0.95	84.43

E^i_r< 40= low ecological risk, 40 <E^i_r< 80= moderate potential ecological risk, 80 <E^i_r< 160= considerable potential ecological risk, 160<E^i_r< 320= high potential ecological risk, E^i_r> 320= very high ecological risk. 150< ERI= low ecological risk, 150< ERI< 300= moderate ecological risk, 300 < ERI< 600= considerable ecological risk, ERI> 600= very high ecological risk

profile also records similar trends (Mondal et al. 2020). Less pH and finer fractions of sediment indicate more exchangeable form of metals. In saline/hypersaline soil, Na^+ and K^+ ions compete with the heavy metals ions and metal tend to stay in immobilized form. But with increase in pH and more organic content in soil results in de-mobilization of these metal ions as evident from similar polluted mangrove sites (Chowdhury and Maiti 2016a;Ma et al. 2016; Marchand et al. 2006, 2016)

Pollution indices

Contamination factor (C_f) To properly understand the pollution status on ecosystem 'contamination factor' (C^i_f), are used, that takes into account the relative toxicity of a particular metal in the environment (Table 4). Contamination factor (C^i_f) compares the pollution severity in a particular site by comparing the metal concentration of a reference value from pre-industrial period. As per this study, C^i_f for Cd was in the category of "very high pollution" for rhizosphearic sediment from *A. officinalis* but it was in the category of "considerable pollution" for *A. ilicifolius* and *P.coarctata*. However, rhizosphearic sediment of all the three target species was "moderately polluted" with Cr, Pb, and Zn pollution. Other studied metals (Cu, Hg, Mn, and Ni) were in the category of "low pollution."

In the control sites, the C_f values of all the metals were in low pollution range except Cd which was showing moderate pollution levels. The Sundarban ecosystem overall has high Cd concentration in sediment phase as reported by previous studies (Akhand et al. 2016; Chowdhury and Maiti 2016b).

Ecological Risk Index (ERI) Ecological Risk Index shows a better outlook level of metal contamination on backdrop of its effect on ecosystem. In this study, all the metals except Cd showed "low ecological risk," whereas Cd pollution was under "high potential ecological risk" category for *A. ilicifolius* and *A. officinalis* sediment samples, and "considerable potential ecological risk" for rhizosphere of *P. coarctata* (Table 5). Ecological risk index showed "considerable potential ecological risk" for *P. coarctata* whereas all other two plant species have "high potential ecological risk." In the control site, the result showed "moderate ecological risk" for Cd whereas all other metals were having "low ecological risk." The ERI results showed "low- moderate potential ecological risk," which indicated minimum pollution levels in the control site and substantiated its choice as control for this study.

Potential toxic elements in mangrove plants

Potential toxic elements in root, shoot/bark, leaves, and fruits/seeds of plants at polluted site are elucidated in Fig. 3. The concentration of PTEs in the plant growing in the control site has been depicted in supplementary material (Figure S1). It is evident that Zn and Mn have the highest concentration in plants root system, which play an important part in several key plant enzyme pathways. The roots of *A. officinalis* showed the highest concentration of Cr (185 mg/kg) followed by the roots of *A. ilicifolius* (105 mg/kg) and *P. coarctata* (86 mg/kg). However, in shoots, Cr was the highest in *A. officinalis* (72 mg/kg) followed by *A. ilicifolius* (63 mg/kg) and *P .coarctata* (24 mg/kg). *A. officinalis* also showed high concentration of Pb and Cd in its roots (Pb—6.63 mg/kg, Cd—2.07 mg/kg), and bark (Pb—2.2 mg/kg, Cd—2.8 mg/kg) . *A. ilicifolius* has the highest Pb concentration in its root (10.6 mg/kg) amongst the three plants. Comparatively, higher concentrations of Pb were recorded in leaf (3.2 mg/kg) and fruit (1.2 mg/kg) of this plant . Mercury in roots of selected plant species was analyzed to be 0.12, 0.27 and 0.05 mg/kg in *A.officinalis*, *A.ilicifolius* and *P. coarctata* respectively. Table 6 showed the PTE concentration in different parts of the mangrove. There is limited study on metal accumulation in different plant parts of the targeted mangrove species growing naturally in a polluted soil.

Table 6 A comparative study on the metal contamination (mg/kg) in mangroves plants

Plant	Plant parts	Study area	Cu	Cd	Ni	Zn	Mn	Cr	Pb	Hg	Reference
A. officinalis	Root	Indian Sundarbans	20.77			49.46		8.44	11.96		Chakraborty et al. 2013
	Leaf		11.88			28.23		3.63	4.57		
	Bark/Shoot		13.77			31.25		4.69	6.22		
	Root	Indian Sundarbans (Polluted site)	32.43	2.07	47.47	85.00	87.27	185.33	6.63	0.12	This study
	Root	Indian Sundarbans (Control)	11.2	0.7	21.6	83.6	86.1	13.3	7.79		
	Bark	Indian Sundarbans (Polluted site)	11.5	2.8	13.8	77.5	60.0	71.7	2.2	0.1	
	Bark	Indian Sundarbans (Control)	11.93	0.6	20.03	86.77	86.77	9.23	1.23		
	Leaf	Indian Sundarbans (Polluted site)	11.20	2.20	11.60	83.6	86.13	50.0	0.12	0.08	
	Leaf	Indian Sundarbans (Control)	9.57	0.11	9.83	81.5	48.2	2.13	0.35		
	Fruit	Indian Sundarbans (Polluted site)	9.6	0.4	5.2	47.7	30.7	7.0	0.0	0.1	
	Fruit	Indian Sundarbans (Control)	6.83		3.33	35.50	14.27	0.27			
A. ilicifolius	Root	Kadalundi-Vallikkunnu Community Reserve (KVCR)	144.8	5.57	8.50	22.70		11.90	35.63		Shackira and Puthur 2013
	Leaf		57.0	2.40	3.77	27.97			17.70		
	Root	Maowei Gulf, China	11.21	0.429	2.86	39.65		18.41	8.23	0.027	Wu et al. 2015
	Stem		5.35	0.071	0.40	23.04		4.96	1.21	0.16	
	Leaf		6.00	0.056	1.15	41.23		6.41	2.38	0.56	
	Root	Indian Sundarbans (Polluted site)	26.53	1.80	40.67	78.60	75.07	105.33	10.60		This study
	Root	Indian Sundarbans (Control)	9.5	0.8	11.1	70.5	95.9	3.8	3.20		
	Shoot	Indian Sundarbans (Polluted site)	11.57	1.87	11.87	73.33	56.73	62.50	1.50	0.39	
	Shoot	Indian Sundarbans (Control)	8.2	0.57	10.87	70.67	86.87	1.13	1.47		
	Leaf	Indian Sundarbans (Polluted site)	9.5	1.0	11.1	43.9	95.9	37.1	3.2	0.5	
	Leaf	Indian Sundarbans (Control)	4.53	0.36	4.83	63.5	55.23	0.79	0.15		
	Fruit	Indian Sundarbans (Polluted site)	2.6	0.0	6.7	23.0	46.0	10.8	1.2	0.3	
	Fruit	Indian Sundarbans (Control)	3.3	0.4	4.1	32.8	14.3	0.1			
P. coarctata	vegetative tissue	Sagarisland,IndianSundarbans	34.9	0.87		124			2.29	0.06	Chakraborty et al. 2018
	Root	Indian Sundarbans (Polluted site)	31.9	1.46	36.73	61.63	71.60	85.67	9.83	0.05	This study
	Root	Indian Sundarbans (Control)	11.2	1.5	46.7	52.2	85.7	16.1	1.2		
	Shoot	Indian Sundarbans (Polluted site)	17.7	1.3	10.8	61.4	58.5	24.0	1.1	0.1	
	Shoot	Indian Sundarbans (Control)	10.37	1.13	10.87	58.97	90.07	17.13	0.21		
	Leaf	Indian Sundarbans (Polluted site)	11.20	2.53	16.67	52.20	85.67	35.10	1.17	0.11	
	Leaf	Indian Sundarbans (Control)	5.20	0.08	2.80	43.00	69.73	5.92			
	Seed	Indian Sundarbans (Polluted site)	5.53	0.44	10.60	27.20	34.49	17.50		0.10	
	Seed	Indian Sundarbans (Control)	4.8		1.4	20.4	7.3	1.0			

In consideration of the ethnobotanical usage of the mangrove plants, this study also explain the PTE contents in respective usable parts. i.e., the leaf, fruit, and seeds. Results showed the absence of Pb in the fruits and seed of the *A. officinalis* and *P. coarctata*, whereas trace of Cd and Hg was present in fruits and seeds of the plants growing in the polluted sites. In *A. officinalis* and *A. ilicifolius*, significantly high metal concentration was found in its shoot/ bark and leaves.

Bioaccumulation factor (BAF) shows the potential of each species to accumulate PTEs in their body. *A. officinalis* (12) showed the highest bioaccumulation of Hg in its areal parts followed by *A. ilicifolius* (8.3) and *P. coarctata* (5.7) (Fig. 4). Whereas, *P. coarctata* has the highest capacity to bioaccumulate Cd (7.9) in its tissues followed by *A. officinalis* (5.2) and *A. ilicifolius* (4.4). Cd is readily taken by the plants through apoplastic and symplastic transport via xylem-phloem and have low toxicity to macrophytes than other pollutants but have potentials to be bioaccumulated in higher tissues (Alloway 2013; Clemens et al. 2002). Cr also showed a similar trend with *P. coarctata* having highest bioaccumulation potential (2.0) followed by *A. officinalis* (1.6) and *A. ilicifolius* (1.5). Pb was negligible in the fruits of *A. officinalis* and seeds of *P. coarctata*.

As most of the toxic metals (Cd, Hg, Cr) had showed a high concentration and bioaccumulation in plant tissues, it can be concluded that these species can cause health hazard if it is regularly ingested/usage by the human. But for health risk assessment, all the metals were considered for THQ

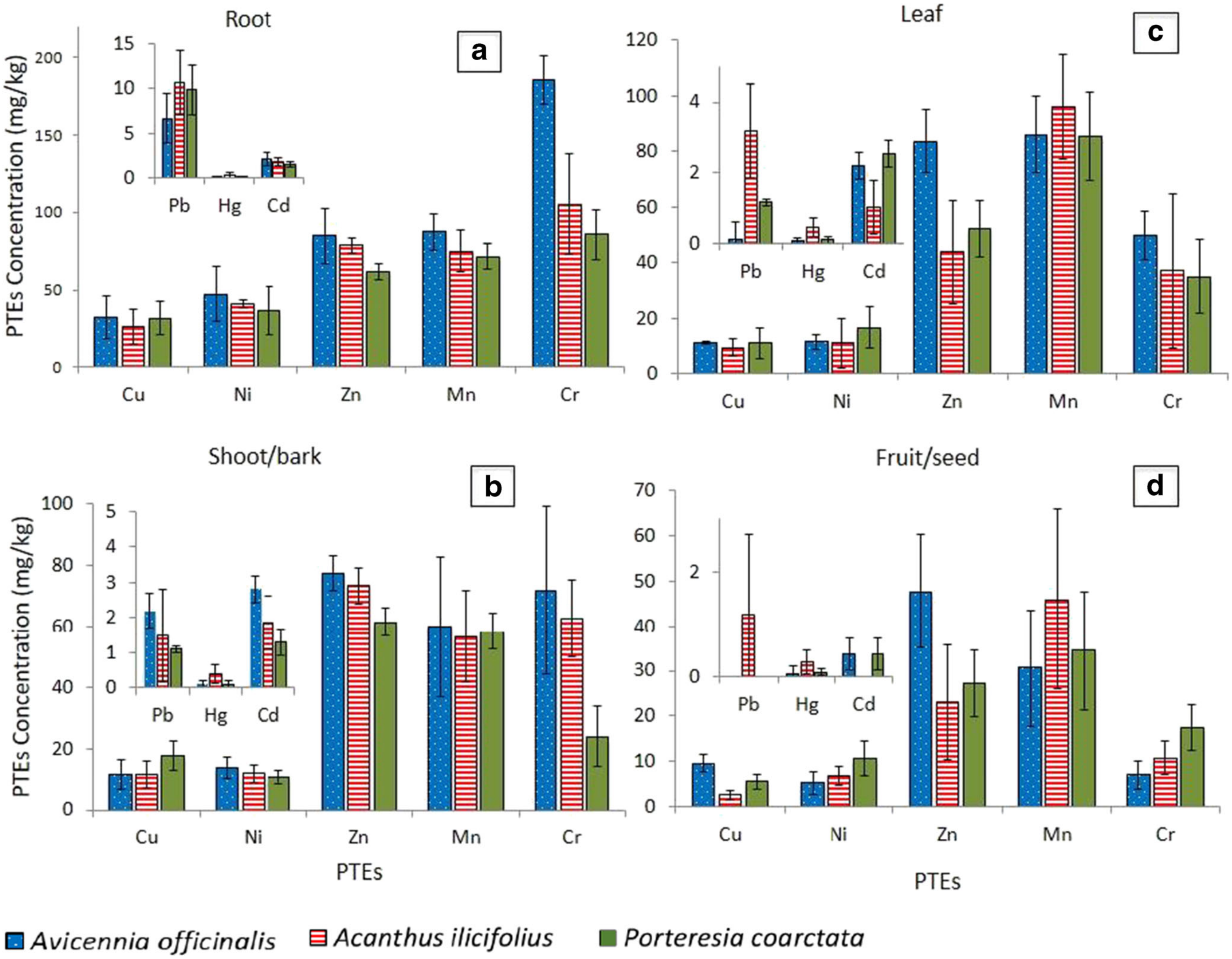
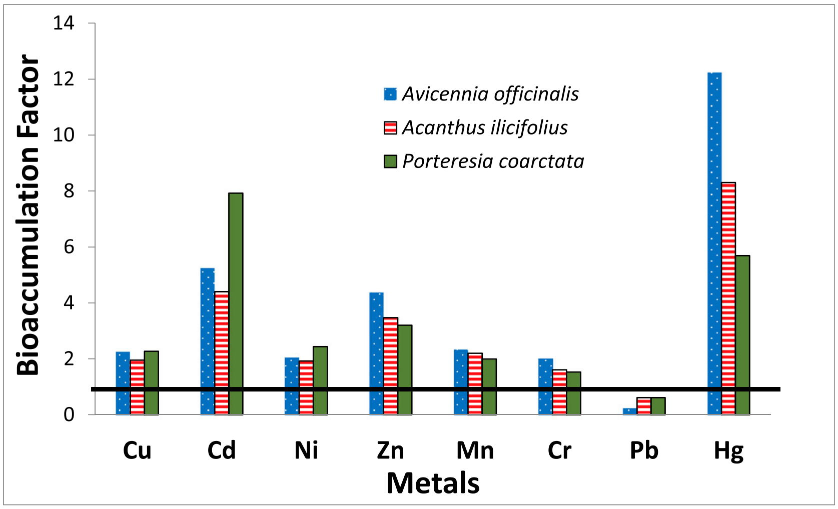


Fig. 3 Concentrations of potentially toxic elements in the root and shoot/bark, leaf, fruit/seed of the three halophytes collected from polluted site. Bioconcentration of PTEs in (A) root, (B) shoot/bark, and (C) leaf (D) Fruit/ seeds of *A. officinalis*, *A. ilicifolius* and *P.coarctata*

calculation. Because even if other metals were showing relatively lower bioaccumulation in the plant parts cumulatively they may impart health impact. In this study, THQ was used to understand the relative human health impact of each metal and

Fig. 4 Bioaccumulation factors (BAF) potential toxic metals (PTEs) in three investigated halophytes at polluted sites (the line designates the limit of 1, to show values with > 1 BAF values)



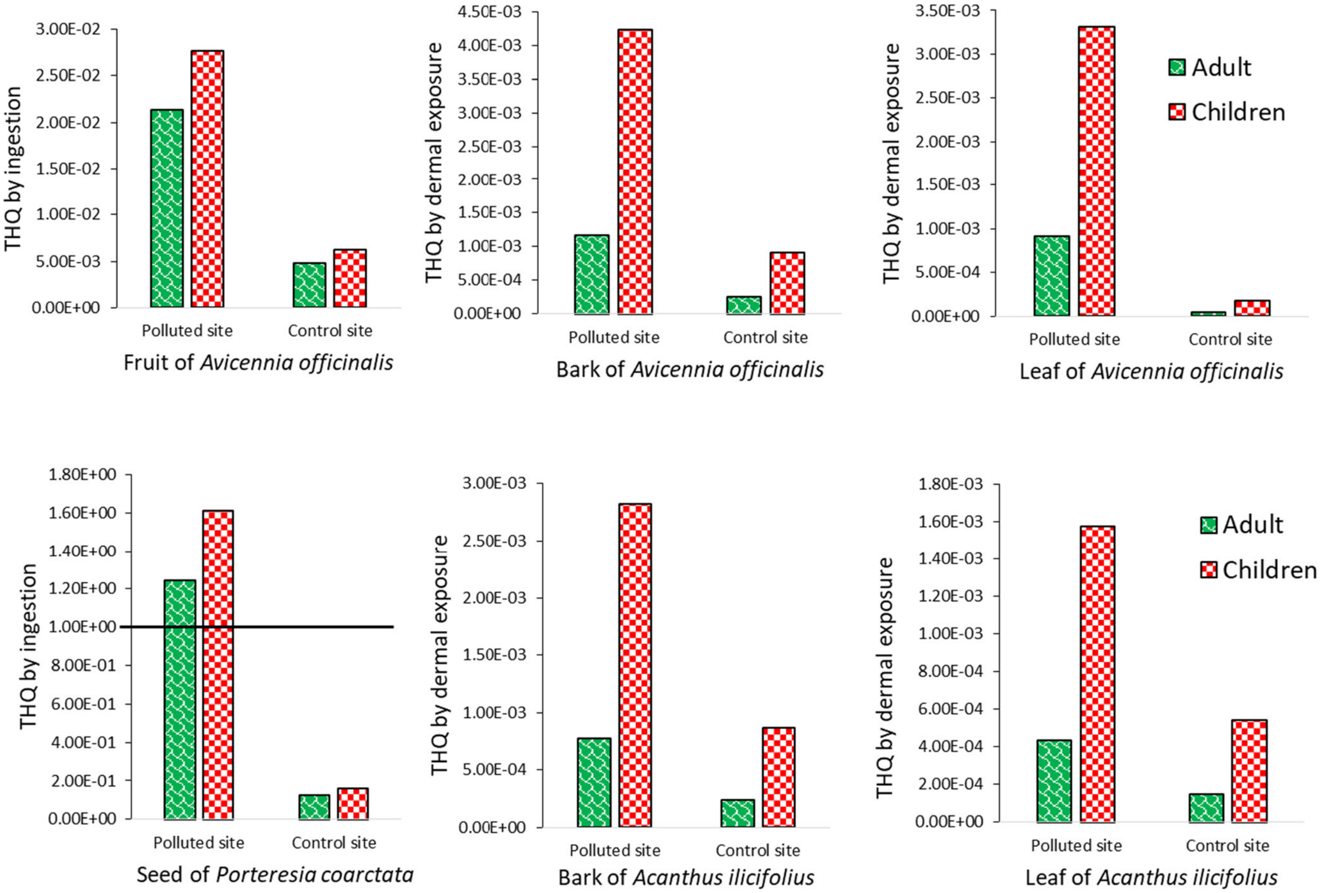


Fig. 5 Comparison total hazard quotient (THQ) values between polluted and control sites between adult and children for the ingestion and dermal uses different plant parts of three selected mangroves species (fruits, seeds, leaf and bark). The straight line indicating the safe limit of THQ (THQ=1)

to segregate out the PTE's having highest probable human health risk. Similar literatures on human health risk assessment due to PTE exposures also considered all the investigated metal concentrations to evaluate cumulative human health hazard (Kumari et al. 2018; Naz et al. 2020). THQ empirical model indicates which metal has the highest propensity to cause health impact in respect to all the studies PTE's.

Human health risk assessment Results indicated that plants growing in metal polluted areas are prone to cause health risk in adult and children population via regular consumption of the ethnobotanical usage (Fig. 5). However, dermal exposure of bark/stem and leaves of *A. officinalis* and *A. ilicifolius* growing in both polluted and non-polluted sites was not posing any potential health risk in both adult and children population but the consumption of *P. coarctata* seeds harvested from the polluted sites showed high total hazard quotient (THQ>1) 1.06 and 1.37 in adult and children respectively indicating health risk. Although, the fruits of *A. officinalis* showed THQ 2.14E-02 (adult) and 2.77E-02 (children) respectively, this was indicating negligible health risk (Fig. 5). Among all metals, Cd was responsible for the maximum contribution in the health risk by ingestion of *P. coarctata* seeds and fruits of *A. officinalis*, followed by Ni, Mn, Zn, Cr, and Cu (Supplementary material Table S5 a,b).

Total hazard quotient through dermal exposure by leaf of *A. officinalis* was 9.10E-04 and 3.31E-03 in adult and children respectively. And health risk via dermal exposure by bark of same plant was 1.17E-03 and 4.24E-03 in adult and children respectively (Fig. 5). The trend of the responsible metals for this health risk via usage of leaf of *A. officinalis* was Cd>Mn>Cr>Zn>Cu>Hg>Pb>Ni and via usage of bark of same plant was Cd>Cr> Mn>Hg>Pb>Zn >Cu>Ni (Supplementary material Table S5a). The THQ by uses of leaf and bark of *A. ilicifolius* was 4.33E-04 (Adult), 1.58E-03 (children) and 7.75E-04(Adult), 2.82E-03 (children) respectively (Fig. 5).The maximum and minimum health risk posing metal via both pathways (leaf and bark application) was Cd and Mn respectively. Although the overall posed health risk by external usage (dermal) of leaves and bark of *A. officinalis* and *A. ilicifolius* was negligible, but the regular exposure can cause threat to local populace.

In this study, Cd was responsible for high THQ in comparison to other exposed metals. Cadmium has been regarded as highly toxic as it does not play any important role in biological functions. Cd^{+2} form of cadmium can replace Zn^{2+} from key enzyme motifs due to their similar electronic configuration (Sigel et al. 2013; Chowdhury and Maiti 2016a, 2016b). This replacement occurs often because both metals are present in a same group in the periodic table having similar d10 electronic configuration and coordination numbers. This can be attributed to be the main reason for the high bioaccumulation of Cd in biological systems and Cd has been found to be biomagnified in higher trophic levels. In animals, Cd can bind with the red blood cells which results in accumulation of cadmium in the kidneys and liver tissues. It has a very long biological half life in any tissues (Alloway et al. 2012). Because of its high toxicity, bioaccumulation potential and almost no biological function, Cd is one of the "Toxic Trio," the most deleterious metal pollutants (Wu et al. 2011).

The quantitative estimation of human health risk has by high degree of uncertainties, which arises from the input parameters and toxicology data used for the study (De Miguel et al. 2007; Yang et al. 2019). Different input parameters, such as the average exposure age, body weight, height, skin surface area, exposure time, and exposure frequency can vary from person to person according to living style, economic, and environmental conditions. Uncertainty in the inputs is reflected in uncertainty in model results and predictions (Borgonovo 2007). Thus, in case of oral and dermal application of mangrove plant parts can vary from person to person and from time to time, further the doses can differ from one person to another. The maximum values of input parameters provide the maximum risk factor, which delivers the maximum extend of risk caused in certain condition (Sahuquillo et al. 2003). Thus, to draw out the maximum extend of health risk and to reduce the uncertainty a certain level, the maximum values of input parameters have been considered except the PTE concentrations (which is taken as average) to identify the health risk in this study. The obtained results may have uncertainties as evident from similar studies, and these results can vary with the variations of input parameters (Yang et al. 2019; Kohzadi et al. 2019).

Conclusion

This study concludes that polluted site had more exchangeable form of toxic metals like Cr, Pb, Cd, and Hg in comparison to the unpolluted site, proving probable ecological and health risk in the region. Ecological risk assessment suggested considerable pollution pressure in the study site. Cd had the highest exchangeable fraction in the polluted soil resulting in high uptake of this PTE by mangroves. *A. officinalis* had the potential to uptake Cd, Cr, Zn, and Mn as indicated by bioaccumulation factor. Mercury was present in trace amounts at the polluted rhizosphere. This study also indicates that Hg was highly accumulated in the three mangroves showing highest bioaccumulation factor. But this toxic metal poses minimum health risk as evident from this health risk assessment study. Among all three halophytes (*A. officinalis*, *A. ilicifolius*, and *P. coarctata*), human health risk due to bioconcentration of Cd was observed highest due to the ingestion of seeds of *P. coarctata* seeds. Low health risk was observed by the oral intake of plants grown in the polluted sites. However, the dermal application of plant parts did not show any significant health risk, which indicated that the plants grown in the metal contaminated sites can be used for external applications. Regular assessment and monitoring studies need to be conducted across coastal mangrove areas impacted by PTE contamination along with accounting bioaccumulation potentials of these toxic elements in the native plants. Ethnobotanical usage of these plants needs to be considered while implementing policy level pollution mitigating interventions.

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Author contribution A.C. Responsible for designing of research question, sampling, analysis, interpretation of data, and drafting of the manuscript. A.N. Taken part in sequential extraction of PTEs from sediment samples and using human health risk assessment tools to interpret the results. A.N. has equal contribution as the first author. S.K.M. Is responsible for guidance, providing laboratory facilities, supervision of analysis and interpretation of results. The final manuscript and authorship sequence have been approved by all the authors before submission.

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Data availability The raw data is available with authors and rest are presented in the manuscript and supplementary materials.

Compliance with ethical standards

Ethics approval and consent to participate The research has been conducted following ethical guidelines. Participants of the survey were adult and have given permission to participate in the interview.

Consent for publication We give permission to publish the manuscript upon acceptance and declare there is no use of any copy righted materials in this manuscript for which any consent from third party is required.

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