



## Contamination and ecological risk assessment of trace elements in sediments of the rivers of Sundarban mangrove forest, Bangladesh



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### ABSTRACT

In this study, total concentrations of 16 trace elements (Al, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, As, Cd, Sb, Hg, Pb, Th and U) in sediments of the rivers of the Sundarban mangrove forest, after the catastrophic oil spill accident in the Sela river of Sundarban, were determined. The overall mean concentrations of V, Cr, Fe and Cd in surface sediments of the Sundarban are remarkably higher than available literature data of those elements. Trace element contamination assessment, using different environmental contamination indices, reveals that As, Sb, Th and U are low to moderately contaminated while Cd is moderately to severely contaminated in the sediments of this area. The multivariate statistical analyses were applied to reveal the origin and behavior of the elements during their transport in the mangrove ecosystem. High Cr, Ni, Cu and As concentrations suggest the risk of potentially adverse biological effects in the ecosystem.

### 1. Introduction

Trace element contamination in aquatic environments has been intensively studied in recent years due to its toxicity, abundance, and persistence in the environment. Trace element, especially heavy metal, residues can accumulate in aquatic flora and fauna, which may enter into the human food chain and result in health problems (Chabukdhara and Nema, 2012; Ma et al., 2016; Wu et al., 2017). The main reason for element contamination is the increasing complex mixtures of chemicals discharged to the coastal zone from non-point sources. Sediments usually provide useful information on environmental and geochemical pollution status (Larsen and Jensen, 1989; Uluturhan et al., 2011; Tamim et al., 2016) because they are the main sink for various pollutants, including elements discharged into the environment (Dassenakis et al., 1997; N.F. Tam and Wong, 2000; N.F.Y. Tam and Wong, 2000). Recognizing the pollution characteristics of trace elements in river sediments and targeting their potential sources is of key importance for proposing effective strategies to protect watershed ecosystems. Various indices have been developed to assess the contamination and environmental risk of trace elements in surface sediments based on their total content, speciation, bioavailability and toxicity (Yang et al., 2009; Yu et al., 2011). To evaluate the combined risk of several trace elements in

sediments, the pollution load index (PLI) and potential ecological risk index (RI) have also been developed (Yang et al., 2009; Huang et al., 2013).

Mangroves are diverse ecosystems that are found in sheltered estuaries and along river banks and lagoons in the tropics and subtropics. Mangroves act as a fragile link between marine and fresh water ecosystems, pollution sinks, and source of nutrient flux into the marine ecosystem (Maiti and Chowdhury, 2013). In the recent decade, mangrove ecosystem has been increasingly threatened due to the adverse effects of anthropogenic activities such as urbanization, industrialization and aquaculture (Vane et al., 2009; Ahmed et al., 2011; Chaudhuri et al., 2014). Trace element contamination and cycling poses a serious threat to the mangrove ecosystem (Wang et al., 2013; Costa-Boddeker et al., 2017). Adsorption of excessive trace element from sediments to mangrove plants may lead to contamination of the food chain (Ahmed et al., 2011; Álvaro et al., 2015). The measurement of trace element concentrations and characterization of their distribution in the mangrove ecosystem leads to better understanding of the mechanisms controlling the dispersal, accumulation and fate of the elements in the mangrove settings.

The Sundarban mangrove forest, covering about 10,200 km<sup>2</sup> area, 60% of which is in Bangladesh and the rest in India. The progressive

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industrial and agricultural development in Bangladesh and India, and the associated increase of toxic trace element level in the Sundarban mangrove (Ahmed et al., 2011; Silva Filho et al., 2011) demand rigorous control over their concentration and bio-magnification processes in biota. Several studies have been conducted on element pollution in the Indian part of the Sundarban mangroves, (Silva Filho et al., 2011; Banerjee et al., 2012; Bhattacharya et al., 2015; Akhand et al., 2016 etc.) but very few studies have been conducted on the Bangladesh part of Sundarban (Ahmed et al., 2011; Borrell et al., 2016; Kumar et al., 2016). However, these studies are mostly about accumulation of trace elements in aquatic organism of Bangladesh part of Sundarban. In the last few years, several oil, fertilizer and coal fly ash carrying cargo accidents occurred at the Sela and Poshur rivers inside the Sundarban of Bangladesh. Among these incidents, a catastrophic oil carrying cargo accident on December 9, 2014, spilled 350,000 l of fuel oil into the Sela river inside the Sundarban (Phillips, 2014). Since the Sela and Poshur rivers are connected to the small creeks inside the Sundarban, there are concerns over the ecological catastrophe that occurred in the Sundarban due to these accidents. In this study, contamination and ecological risk assessments of trace elements in Sundarban mangrove ecosystem of Bangladesh are conducted using various sediment quality indices and statistical approaches.

## 2. Materials and methods

### 2.1. Study area

Sundarban is the largest mangrove forest in the world and a significant part of it is declared as UNESCO world heritage site (FAO/UNDP, 1998). The Sundarban mangrove forest (88°00'–89°55'E and 21°30'–22°30'N) is located in the estuary of the river Ganges in south-west Bangladesh and in the south-eastern region of the State of West Bengal in India. The forest meets the Bay of Bengal in the south. Sundarban is home to several threatened plant and animal species (including the Royal Bengal Tiger). The forest covers an area of 10,000 km<sup>2</sup>, of which 7000 km<sup>2</sup> (70%) is land, and the remaining 3000 km<sup>2</sup> is under water in the form of rivers, canals and creeks. Most creeks and canals flow into the large rivers, which are interconnected. This delta plain is a tide-influenced depositional system where the tropical south-west monsoon controls the freshwater discharge in the area (Goodbred et al., 2003).

Mongla sea port, the second largest sea port of Bangladesh, is situated at the confluence of the Poshur river and the Mongla river. The Port is very close to the Sundarban mangrove forest. The port has trade links with almost all major ports of the world. All ships and cargo boats are carrying goods to the ports through the rivers of the Sundarban. Sundarban mangrove ecosystem is degrading day by day due to different anthropogenic activities like port activities, industrial effluents, ship breaking, fishing, tourism, agriculture, aquaculture etc. (Hussain and Acharya, 1994).

### 2.2. Sample collection and preparation

The superficial sediment (0–5 cm) samples were collected from fifteen sampling points in December 2015, one year after the catastrophic oil spill accident in Sundarban. Three replicate samples were collected from each point and mixed into a composite sample. Geographical locations of the sampling points are presented in Fig. 1. Sampling points started from the freshwater zone near Mongla port (S-1) to saline water zone (S-15). The river bed sediments were collected using an acrylic pipe sampler during low tides. The upper 5 cm of each sample was taken using an acid-washed plastic spatula. All samples were immediately packed in acid-rinsed polyethylene plastic bags and stored at low temperature (4 °C) before sample preparation at the laboratory. The samples were dried in an electric oven at 45 °C for 72 h to gain constant weight. The dried samples were then ground into a small grain

size and homogeneous mixture using a mortar and pestle and sieved through 0.25 mm aperture to remove organic materials, stones and lumps. The homogenous powdered samples were stored in labeled glass bottles until elemental analyses.

### 2.3. Sample analysis

#### 2.3.1. Instrumental neutron activation analysis (INAA)

For trace element determination by INAA, about 50 mg of each dried powder sample was weighed in polyethylene bag and heat sealed. Two certified reference materials (CRMs) from International Atomic Energy Agency (IAEA): Soil-7 and IAEA-SL-1 (Lake Sediment), and one standard reference material: NIST-1633b (Coal Fly Ash), along with the sediment samples, were analyzed in this study. Relative standardization method of INAA was applied for element determination. IAEA-Soil-7 was used as the standard, while IAEA-SL-1 and NIST-1633b were used as the control samples.

The samples and standards were irradiated using pneumatic transfer (rabbit) system at the 3 MW TRIGA Mark-II research reactor of Bangladesh Atomic Energy Commission. Short-term and long-term types of irradiation were performed targeting radioactive nuclei with short and long-half-lives. Short irradiation of each sample/standard was performed separately at a thermal neutron flux of  $1.77 \times 10^{12} \text{ cm}^{-2} \cdot \text{s}^{-1}$  for 1 min at 250 kW, whereas long irradiation was performed simultaneously with all the samples and standards at a thermal neutron flux of  $1.70 \times 10^{13} \text{ cm}^{-2} \cdot \text{s}^{-1}$  for 7 min at 2.4 MW. For the calculation of trace element concentrations, the considered product radionuclides with their half-lives and gamma-ray energies are given in Table 1. In the case of long irradiation, neutron flux gradient within the sample stack was determined by irradiating three IRMM-530RA Al–0.1% Au (0.1 mm foil) monitor foils placing them at the bottom, middle and top of the sample stack. After irradiation, the activities of irradiated samples and standards were measured using a high resolution HPGe detector (resolution at FWHM is 1.88 keV at 1332.5 keV of <sup>60</sup>Co) coupled with a digital gamma spectrometry system. For long irradiated samples, first counting was performed for 1 h after a decay time of 2 days for the elements As and Sb, while the second counting was performed for 2 h after a decay time of 4–5 days for U. Third counting was performed for 3 h after a decay time of 4 weeks for the elements Cr, Fe, Co, Zn and Th. Samples and standards irradiated for a short time, first counting was performed for 300 s after a decay time of about 300 s for the elements Al and V, and second counting for 600 s after a decay time of 2 h for Mn. To determine element concentration, by considering more than one gamma-ray, the average concentration obtained by the concerned gamma-rays is reported. For correcting the <sup>28</sup>Si(n,p)<sup>28</sup>Al and <sup>31</sup>P(n,α)<sup>28</sup>Al interferences in determining the Al concentration using <sup>28</sup>Al radionuclide, high purity Si (SiO<sub>2</sub>) and P (KH<sub>2</sub>PO<sub>4</sub>) chemical reagents (from Spex, USA) were prepared and irradiated with the samples. The specific activity of Si due to <sup>28</sup>Si(n,p)<sup>28</sup>Al reaction was used to subtract the interfering activities where Si concentrations of the studied sediment samples and standards were determined by atomic absorption spectrometry (AAS) (Bernas, 1968). The Al concentration correction, due to <sup>28</sup>Si(n,p)<sup>28</sup>Al reaction was yielded 2–4% for the samples. The P interference, due to <sup>31</sup>P(n,α)<sup>28</sup>Al reaction in Al determination, was calculated in the same way as stated above for IAEA-SL-1 and NIST-1633b which yielded no significant concentration corrections (< 0.01%) for Al. The P contents of the reference materials were used from their certified values for the correction. Since sediment samples contain around same level of P, as in IAEA-SL-1, therefore, no correction was applied for P interference in Al determination by INAA of the sediment samples.

#### 2.3.2. Atomic absorption spectrometry (AAS)

All chemical reagents were of analytical grade or of Suprapure quality (E. Merck, Germany). Double deionized water (Milli-Q System, Millipore) was used for the preparation of all solutions. The element

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