



Seasonal assessment of trace element contamination in intertidal sediments of the meso-macrotidal Hooghly (Ganges) River Estuary with a note on mercury speciation

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ABSTRACT

The spatial and seasonal distribution of trace elements (TEs) ($n = 16$) in surficial sediment were examined along the Hooghly River Estuary (~ 175 km), India. A synchronous elevation of majority of TE's concentration (mg kg^{-1}) was encountered during monsoon with the following descending order: Al (67070); Fe (31300); Cd (5.73); Cr (71.17); Cu (29.09); Mn (658.74); Ni (35.89). An overall low and homogeneous concentration of total Hg ($T_{\text{Hg}} = 17.85 \pm 4.98 \text{ ng g}^{-1}$) was recorded in which methyl mercury (MeHg) shared minor fraction (8–31%) of the T_{Hg} . Sediment pollution indices, viz. geo-accumulation index (I_{geo}) and enrichment factor (EF) for Cd ($I_{\text{geo}} = 1.92\text{--}3.67$; $\text{EF} = 13.83\text{--}31.17$) and Ba ($I_{\text{geo}} = 0.79\text{--}5.03$; $\text{EF} = 5.79\text{--}108.94$) suggested high contamination from anthropogenic sources. From factor analysis it was inferred that TE's primarily originated from lithogenic sources. This study would provide the latest benchmark of TE pollution along with the first record of MeHg in this fluvial system which recommends reliable monitoring to safeguard geochemical health of this stressed environment.

1. Introduction

Estuaries are highly productive and dynamic zones that are of great research interest from the aspect of distribution of contaminants (Monteiro et al., 2016). Contamination of trace elements (TEs) in the estuarine environment is of global attention owing to its abundance, stability against degradation, and environmental toxicity (Ali et al., 2016; Hwang et al., 2016). They can accumulate in both suspended particulates and sediments which might be released into the aquatic systems under favourable conditions, enter the food web, and cause health hazards (Ghafari and Yusuf, 2006; Varol, 2011; da Silva et al., 2015; Keshavarzi et al., 2015; Morina et al., 2015). Trace elements originate in the environment through natural processes such as physical and chemical weathering of rocks, erosion and atmospheric deposition and also via anthropogenic activities, i.e. discharge of municipal and industrial wastewaters, runoff from agricultural fields and adjacent coastal lands and riverine fluxes.

Sediments act as source and/or sink for TE's in the environment and contribute to a better understanding of the geochemistry of an ecosystem (Watts et al., 2017). Sediments not only contain valuable information regarding geological and environmental conditions but also manifest the level of TE pollution in the region (Ma and Wang, 2003; Liu et al., 2015). The key factors affecting the rate of sedimentation and distribution characteristics of TE's during riverine transport are mainly attributed to grain size of suspended particles, hydrological regime, organic matter and hydrochemical properties (Dai et al., 2007; Liu et al., 2010). However, due to the change in environmental conditions, sediments may transform from the main sink of TE's to the sources of them for the overlying waters (Gaur et al., 2005; Prica et al., 2008). So the estimation of TE's only in water and in the suspended materials is far from enough due to water discharge fluctuations and low residence time (Varol, 2011). Therefore, the concentrations of TE's in sediments are often monitored to provide basic information for the environmental risk assessment (Long et al., 1995; SEPA, 2002).

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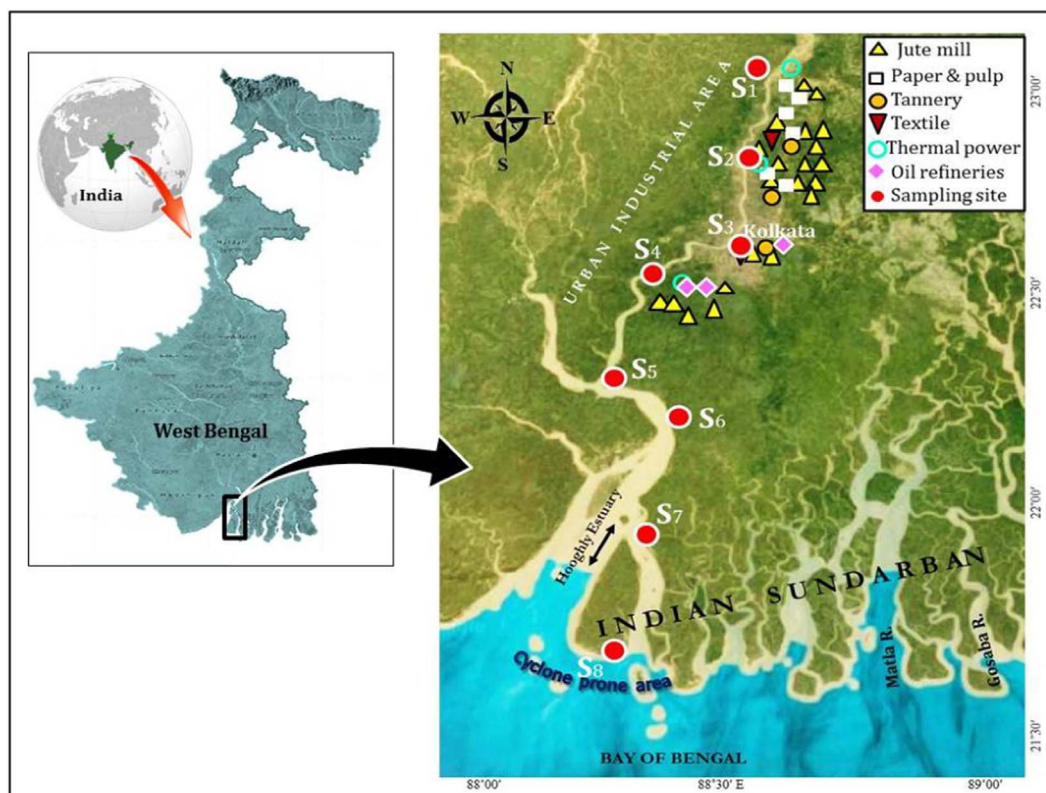


Fig. 1. Map showing the location of eight sampling sites (S_1 to S_8). The intricate river network and position of the multifarious industries on the bank of the Hooghly River Estuary are also shown.

Mercury, a well-known neurotoxic compound, is identified as one of the six most dangerous chemicals in the world by the International Program of Chemical Safety (IPCS) (Integrated Risk Information System, 1993; Bhattacharya et al., 2014). The anthropogenic emission of mercury to the atmosphere in the year 2000 was estimated globally to be 2190 tons (Pacyna et al., 2006) of which 54% was contributed by Asian countries (Li et al., 2009). According to Ullrich et al. (2001), the total Hg (T_{Hg}) concentrations in surface sediments varied from 0.02 to 0.4 $mg\ kg^{-1}$ in uncontaminated or less contaminated rivers, and can be as high as 100 $mg\ kg^{-1}$ in urban, industrial or mining areas. The speciation of mercury provide comprehensive information involving the processes and mechanisms related to the mobility, bioavailability or reactivity of mercury species which cannot be adequately monitored by determining total mercury (T_{Hg}) concentrations (Ullrich et al., 2001; Sunderland et al., 2004; Canário et al., 2007; Du Laing et al., 2009). Inorganic Hg [$Hg(II)$] is the dominant species of Hg in sediments which may be methylated into methyl mercury (MeHg) (Shi et al., 2005; Zhong and Wang, 2008a) which is more mobile than inorganic Hg, 50–100 times more toxic, and more readily bioaccumulated, especially in the aquatic food chain (Compeau and Bartha, 1984; Gabriel and Williamson, 2004; Rimondi et al., 2014). Since the methylation process mainly occurs in sediments, the speciation of mercury (Hg) in the said compartment is crucial to understand the biogeochemistry of this pollutant and to estimate the potential associated exposures of aquatic food webs and ultimately, humans (Hammerschmidt and Fitzgerald, 2001; Carrasco and Vassileva, 2015).

Trace elements in coastal and estuarine sediments have received a great deal of attention in the last several decades, but very few studies have been carried out on mercury pollution along the coastal regions of India (Krishnamoorthy and Nambi, 1999; Ram et al., 2003; Omana and Mahesh, 2008; Chatterjee et al., 2012; Mohan et al., 2014; Ramasamy et al., 2012, 2017). Hence the present work has been undertaken with the following objectives: (i) to investigate TE concentration including

MeHg in surface sediments along Hooghly River Estuary (ii) to evaluate the potential ecological risks of TEs and (iii) to identify the pollution sources and context dependence of these TEs using multivariate analyses.

2. Materials and methods

2.1. Study area and sampling sites

The Hooghly River Estuary (HRE), the first deltaic offshoot of the Ganges River is a coastal plain, funnel-shaped estuary and lie approximately between $87^{\circ}55'01''N$ to $88^{\circ}48'04''N$; $21^{\circ}29'02''E$ to $22^{\circ}09'00''E$. The estuary that forms an important link between the Hooghly–Bhagirathi river system and the Bay of Bengal has evolved as a constituent of the world's largest fluvio-marine delta Ganges-Brahmaputra-Meghna (GBM), within the geographical boundary of India. HRE is well-mixed, positive and mixohaline in nature with a tidal amplitude of 3–6 m. It covers an area of $6 \times 10^4\ km^2$ and is subjected to semi-diurnal tides each of 12 hour duration (Sarkar et al., 2017). It is an estuary of global significance contributing various ecological and economic services and thus supporting the lives of millions of people.

Climate in this region is chiefly influenced by the southwest monsoon (July to October) which is accompanied by heavy rainfall, post-monsoon (November to February) characterized by lower temperature and lower precipitation and pre-monsoon (March to June) dry season with occasionally higher temperature. Almost 70–80% of the precipitation occurs between July and September with an average rainfall of $\sim 1700\ mm$ (Rakshit et al., 2014) resulting in high river discharges ($3000 \pm 1000\ m^3\ s^{-1}$) that gradually diminishes to minimum ($1000 \pm 80\ m^3\ s^{-1}$) during the pre-monsoon (Mukhopadhyay et al., 2006; Ray et al., 2015). A salinity gradient has been induced due to the variation in river discharge which is responsible for the diverse biotopes of plankton communities in the estuary as reported by Rakshit et al.

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