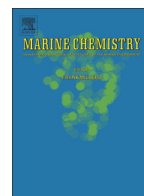




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Organic matter – A key factor in controlling mercury distribution in estuarine sediment

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ABSTRACT

Organic matter (OM) was found to play an important role in controlling mercury (Hg) distribution and speciation in estuarine sediment of the Vembanad Lake. The sedimentary organic carbon (OC) from the northern part of the lake was influenced mainly by marine in situ biological production. However, the sedimentary OC was mainly of terrestrial origin in the southern part of the lake. The average concentrations of Hg in the sediments were relatively higher in the northern part (ranging from ~6.5 to 172.0 $\mu\text{g}/\text{kg}$, with an average concentration of 59.6 $\mu\text{g}/\text{kg}$) than the southern part of the lake (ranging from ~6.8 to 114.0 $\mu\text{g}/\text{kg}$ with an average concentration of 49.7 $\mu\text{g}/\text{kg}$). This study suggests that nature and source of sedimentary OC probably control Hg speciation in the Vembanad Lake system. Speciation study of Hg suggests that a major part (55–90%) of the total Hg was mainly associated with organic phases in the sediment. The variation in concentrations of organic matter bound Hg with the increasing Hg/TOC ratio suggests that more Hg binding sites were available in the sediment from the northern part of the lake but not in the southern part of the lake. Anthropogenic activities were responsible for the higher Hg content in the sediment collected from the northern part of the lake. It is suggested that the strong affinity of sedimentary total nitrogen (TN) for Hg is independent of its source. The sedimentary OM was found to control Hg distribution in different size fraction of the estuarine sediment.

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1. Introduction

Depending upon the physicochemical conditions, sediments which are ecologically important part of aquatic ecosystems can act both as source and sink for contaminants. Hence, sediments are not only considered as carriers but also potential secondary sources of contaminants in an aquatic ecosystem (Burdige, 2011; Chakraborty et al., 2014a,b, 2012; Knee and Paytan, 2011; Nath et al., 2014; Torres et al., 2013). Sediment contamination (by trace/heavy metals) in coastal areas is a major environmental issue because of their potential toxic effects on biological resources and often, indirectly, on human health (Ernst, 2012; Peña-Fernández et al., 2014; Peng et al., 2009; Chakraborty et al., 2010).

Grain size distribution in sediment significantly influences trace/heavy metal concentrations and their bioavailability in sediments. Some studies have shown that highest concentrations of metals are associated with fine grained sediment particles (Chakraborty et al., 2014a, b). Higher specific surface area of clay–silt particles increases the association of trace/heavy metals with these particles (Thorne and Nickless, 1981; Araújo et al., 1988; Cauwet, 1987). However, it has also been reported that metal concentrations in sediments may not increase with decreasing particle size of sediment. High concentration of metal has been reported in the coarser and larger size fractions (Ramesh et al.,

1990; Krumgalz, 1989; Combest, 1991). These exceptions probably demonstrate that metal concentrations in sediment are not controlled exclusively by particle size. There are several other factors (quality and quantity of organic matter, distribution of different mineral phases, metal loading etc.) may also control metal speciation, distribution, accumulation and bioavailability in coastal sediments (Chakraborty and Chakraborti, 2006; Chakraborty et al., 2009). It is, therefore, essential to identify the factors that control toxic metal speciation and their bioavailability in coastal/estuarine sediments.

Mercury (Hg) is considered as one of the most toxic metals found in environment and having no known essential biological function. Moreover, unlike many heavy metals, Hg participates in a variety of biogeochemical processes with a complex cycle characterized by exchanges between different compartments of ecosystem: atmosphere, hydrosphere, and biosphere. In natural environment, Hg occurs in three redox states (0, +I and +II) and can exist in several different chemical forms. All these forms have different chemical properties and are potentially toxic. The organic Hg compounds, of which methylmercury is the most common, are of special concern because of their high toxicity.

Speciation study of Hg in various matrices has attracted much attention and that is reflected by the large number of publications in the recent years (Acquavita et al., 2012; Amos et al., 2014; Bloom and Gill, 1999; Yin et al., 2013; Yu et al., 2012; Lamborg et al., 2014).

A major research and monitoring on mercury (Hg) poisoning have been undertaken mainly in coastal and estuarine sediments in different

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parts of the world. Mercury speciation in sediment depends on several variables such as pH, total organic carbon (TOC), redox condition, sulfide concentration, and other geochemical factors of the system (Gilmour and Henry, 1992).

It has been reported that TOC in sediment can influence Hg distribution and speciation in the sediment by providing enough strong binding sites in sediment surface (Hammerschmidt and Fitzgerald, 2004; Hammerschmidt et al., 2004; Mason and Lawrence, 1999; Stoichev et al., 2004). OC in sediment may increase reduction processes of Hg^{2+} within the system.

Stability of Hg–OC complexes depend on several factors: 1) nature and composition of organic matter; 2) Hg concentrations; 3) pH, ionic strength and redox condition of the medium. The conditional stability constants for Hg–OC complexes vary over 26 orders of magnitude. Recent studies indicate that the conditional stability constants for mercury binding to strong binding sites are of the order of 10^{22} – 10^{28} (Gu et al., 2011; Noh et al., 2013; Randall and Chattopadhyay, 2013).

However, reduction of Hg^{2+} to Hg^0 can be initiated by OC (Alberts et al., 1974; Skogerboe and Wilson, 1981). Mercury reduction by OC is affected by pH, concentrations of dissolved oxygen, and chloride. It has been hypothesized that quinone or semiquinone moieties in organic matter are involved in the transfer of electrons leading to mercury reduction (Alberts et al., 1974).

Significant positive correlation between total Hg and OC contents in estuarine/coastal/open sea sediments has been reported in literature. However, others studies have observed no correlation at all especially in polluted coastal areas (Kehrig et al., 2003; Wu et al., 2013).

The nature of sedimentary OC has been reported to vary from upstream to downstream in an estuary. Particulate and dissolved terrestrial organic carbon has been reported to undergo substantial transformation and biogeochemical processing during fluvial transport as well as during estuarine circulation. Depending on the type of terrestrial

source for the organic matter and the length and speed of the fluvial path as well as residence time in the estuary, the majority of this organic matter is highly modified and/or removed along the way towards the ocean. In addition to that, marine OM from in situ biological production also plays an important role in changing the nature of estuarine sedimentary OC from upstream to downstream. However, the changes in Hg complexing capacity of these substantially transformed and marine derived sedimentary OM in estuaries are not known. Information on fate and transport of Hg in tropical estuarine sediments is limited. The purpose of this study was to determine the impact of TOC and total nitrogen (TN) on the Hg distribution and speciation in surface estuarine sediments, collected from the twenty five geographical locations of the Vembanad Lake, the longest lake in India, from the south west coast of India. The northern part of this lake is well connected with Arabian Sea. Whereas, the southern part of the lake is fluvial dominated and free from brackish water intrusion. An increasing contribution of marine autogenous organic matter towards the seaward side has been reported by Gireeshkumar et al. (2013). Thus, the choice of this area was made in order to understand the variable contribution of marine and terrestrial OM (in sediment) on Hg binding and speciation in different grain size fractions of sediment.

2. Materials and methods

2.1. Study area

The Vembanad Lake, situated in Kerala, is the longest (stretching about 90 km) and largest brackish-water lake of India. It is considered as one of the large tropical estuaries in India. It is permanently well connected to Arabian Sea through two openings (at Kochi and Azhikode). Being connected to Arabian Sea, this lake is regularly influenced by mixed semidiurnal tides (Nath et al., 2000). However 1.5 km long

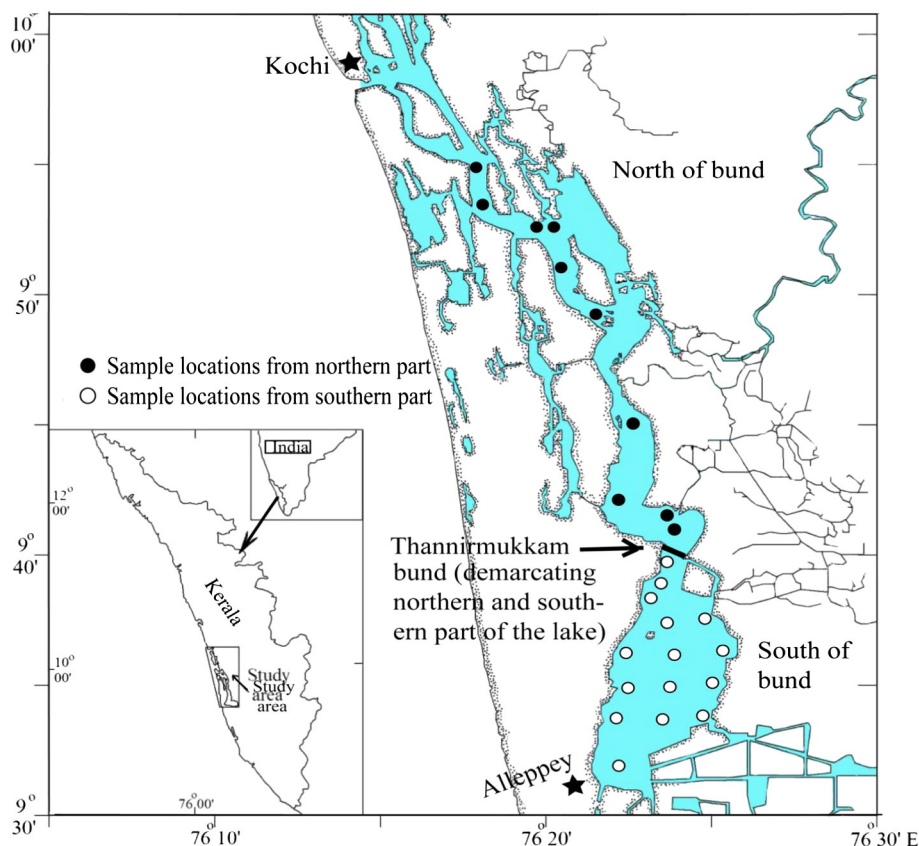


Fig. 1. Map showing the sampling locations from the Vembanad Lake (covering almost entire lake; Kochi in the north and Alleppey in the south). The sampling locations are represented with two different plot symbols; ● (north) and ○ (south).

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