



## Short communication

Sediment accumulation and mercury (Hg) flux in *Avicennia marina* forest of Deep Bay, ChinaRuili Li<sup>a</sup>, Minwei Chai<sup>a</sup>, Meixian Guo<sup>b</sup>, Guo Yu Qiu<sup>a,\*</sup><sup>a</sup> Shenzhen Key Laboratory for Heavy Metal Pollution Control and Reutilization, School of Environment and Energy, Shenzhen Graduate School of Peking University, Shenzhen 518055, China<sup>b</sup> Nanshan Second Experimental School, Shenzhen 518053, China

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## ABSTRACT

To investigate the rate of sediment accumulation and mercury (Hg) flux in *Avicennia marina* forest of Deep Bay, China, sediment cores were analyzed. The results showed that Hg concentrations were much higher at all depths compared to the background level. A high correlation between Hg and total organic carbon (TOC) indicated their similar anthropogenic origin. Sedimentation rate was estimated to be  $1.38 \text{ cm a}^{-1}$  by <sup>210</sup>Pb geochronology. The increase in the mass sediment accumulation rates was rapid (range:  $0.5\text{--}0.94 \text{ g cm}^{-2} \text{ a}^{-1}$ ), and the Hg fluxes ranged between 76 and  $116 \text{ ng cm}^{-2} \text{ a}^{-1}$  during the last three decades. The reduction in both Hg concentrations and flux during the last decade may be due to the adoption of contamination control policies. Our results support the notion that the Hg fluxes determined from the sediment cores reveal the effects of anthropogenic influences from the areas around Deep Bay.

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## 1. Introduction

Mangrove wetlands, the intertidal wetlands of the tropics and subtropics, are key ecological habitats that link terrestrial and marine environments. Mangrove wetlands and their adjacent mudflats, are increasingly impacted by urban/industrial development in the tropical coastal zone and are subject to pollution from multiple sources, including, but not limited to, municipal waste, aquaculture, mariculture and shipping, as well as onshore industries and runoff from urban centres (Huang et al., 2003; Cuong et al., 2005; Sanders et al., 2006; Vane et al., 2009; Li et al., 2015). The cycling of heavy metals, because of their toxicity, bioaccumulation capacity and persistence, is a serious question which has been addressed by many studies of mangrove environments (Tam and Wong, 2000; Marchand et al., 2006; Chaudhuri et al., 2014). In contrast with organic pollutants, heavy metals cannot be biologically or chemically degraded and therefore they either accumulate locally or are transported over long distances. The interactions between terrestrial and marine environments play a significant role in regulating the input of heavy metals and

suspended matter into coastal areas and oceans. The biogeochemical cycling of heavy metals in the coastal zone has been altered by man's activities, especially with respect to the human encroachment of rivers and estuaries and the resultant enhanced input of trace metals on these locations from point and non-point sources (Paraquettia et al., 2004; Bodin et al., 2013).

Mangrove sediments are potential repositories of anthropogenic pollution because of their high total organic carbon content (TOC), anaerobic properties and rapid turnover and burial (Tam and Yao, 2002; Marchand et al., 2005; Ding et al., 2009). Sediments have the ability to preserve the environmental history of a drainage basin and can be useful for making predictions about how future changes may alter a certain environment. Alterations to sediment composition may result from anthropogenic disturbances such as urban development, road construction, agriculture, and hydrological changes (Jha et al., 2003; Smoak and Swarzenski, 2004; Sanders et al., 2006). Estuaries are preferential environments for sediment deposition because they are areas of low gradient and may receive large sediment loads. This makes estuaries important sites for studying particle-reactive elements, including mercury (Hg) and atmospherically derived radioactive tracers (Jha et al., 2003; Bergamaschi et al., 2012).

Mercury (Hg) is one of the most important heavy metals in

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estuarine and coastal sediments. Previous studies have identified wetlands as a sink for total Hg and as a net source of methylmercury (Louis et al., 1994, 1996; Galloway and Branfireun, 2004; Ding et al., 2009; Navya et al., 2015). The main transport routes into wetlands include runoff from the catchment area and atmospheric deposition onto the wetland surface (Lindstrom, 2001). Wetlands can also significantly affect the concentration, speciation, and transport of Hg (Liu and Ding, 2007; Ding et al., 2009; Navya et al., 2015). Many studies have shown that industry, agricultural activities, wastewater disposal, gold mining and the use of fossil fuels are major anthropogenic inputs of Hg into the environment (Azevedo et al., 2011; Liang et al., 2011). Its high mobility, organic matter affinity and biomagnification capacity make this element one of the most environmentally harmful of all the metals (Gorski et al., 2003; Ding et al., 2011; Dhivert et al., 2016). Furthermore, elevated Hg levels in the human body can lead to serious damage, including neurological, immunological, and nephrological disorders, etc (Cornelis et al., 2005; Zahir et al., 2005).

Prominent development in Shenzhen only began in the 1980s. Deep Bay is located next to Shenzhen City, China, which has been the world's most rapidly urbanizing city during the last three decades. Many economic activities were rapidly introduced during this time period, of which industry has been the pre-eminent economic activity. Shenzhen is highly active in industrial production and because of the substantial rise in industrial development during the last three decades, studies investigating local pollutants, as well as sedimentation processes, are needed in this region. Previous studies have preliminarily investigated the total Hg and methylmercury, as well as speciation of mercury in Futian mangrove forest (Ding et al., 2009, 2011), with no systematic research on sediment accumulation and mercury flux accumulation. In this study, the  $^{210}\text{Pb}$  sedimentation rate and the Hg flux in the sediment were calculated using samples collected from the landward region of Deep Bay, near the location of the main freshwater input into the estuary. Based on the discussion above, it was hypothesized that industrial development during the past three decades may be impacting the estuarine environment. To this end, this study investigated (1) mass sediment accumulation rates, (2) the vertical distribution of Hg contamination to examine historic pollution trends and (3) the Hg flux in Deep Bay.

## 2. Materials and methods

The Futian National Nature Reserve in Shenzhen, China, covers an area of 369 ha (Fig. 1). The vegetation includes mangroves, semi-mangroves and seashore plants with *Aegiceras corniculatum*, *Kandelia candel* and *Avicennia marina* being the dominant native mangrove species in the Reserve. The mean annual temperature is 22.6 °C with the lowest mean monthly temperature occurring in January (15.0 °C) and highest in July (28.7 °C). The mean annual precipitation is  $1.8 \times 10^3$  mm and annual sunshine is  $2.2 \times 10^3$  h with alternate dry and wet seasons (Yang et al., 2008).

Three sediment cores, 7.5 cm in diameter and 60 cm in length, were collected in Deep Bay (Fig. 1) using an acrylic tube. The cores were sliced at 2 cm intervals down to 20 cm depth. Then from 20 cm to 60 cm depth the cores were sliced at 4 cm intervals. Sampling was undertaken in July 2011 at low tide. Immediately after extraction, the sediment showed a dark, uniform color throughout the core. Gamma-ray measurements were conducted using a 40% efficient semi-planar intrinsic germanium high purity coaxial detector housed in a lead shield and coupled to a multi-channel analyzer. Activity was calculated according to well-established methods (Alongi et al., 2005). The sedimentation rate was obtained by dividing the  $^{210}\text{Pb}$  decay constant by the slope of the log-linear plot of unsupported  $^{210}\text{Pb}$  versus depth. After

gamma-counting was completed, the samples were dried at 50 °C. A low temperature was used in order to avoid Hg volatilization. The sediment slices were then ground and processed for TOC on a Perkin-Elmer 2400 CHNS Analyzer (Alongi et al., 2005). Total Hg was measured with a ZYG-II cold vapor atomic fluorescence spectrometer (Ding et al., 2009).

## 3. Results and discussion

The bulk density of the sediment increased with depth (Fig. 2a,  $R^2 = 0.65$ ,  $P < 0.01$ ) whereas levels of TOC significantly decreased with increasing depth (Fig. 2b,  $R^2 = 0.93$ ,  $P < 0.01$ ). Hg concentrations ranged between 92 and 196 ng g<sup>-1</sup> and declined with increasing depth (Fig. 2c,  $R^2 = 0.74$ ,  $P < 0.01$ ).

The  $^{210}\text{Pb}$  dating method ( $^{210}\text{Pbex}$ ) is a widely used radionuclide tracer to estimate soil redistribution rates based on its fallout radionuclides (Rabesiranana et al., 2016). In the present study, the  $^{210}\text{Pbex}$  was fitted to the least square procedure ( $R^2 = 0.41$ ;  $n = 15$ ;  $P < 0.05$ ), thus allowing calculation of the sedimentation rate (1.38 cm a<sup>-1</sup>) (Fig. 3; Alongi et al., 2005). The results of this calculation estimated the bottom of the sediment core (60 cm) to be nearly 45 years old. Only one study has calculated the sedimentation rates for coastal mangrove wetland in China. In the Jiulongjiang Estuary of Fujian Province, China, the sediment rates were 6–10 cm a<sup>-1</sup> in the low-intertidal zone and 1.3–1.4 cm a<sup>-1</sup> in the high-intertidal zone (Alongi et al., 2005).

Several studies have also determined sedimentation rates in mangrove environments in other countries. In a tropical mangrove forest in Brazil, sediment accumulation rates were estimated to be 0.28 cm a<sup>-1</sup> (Sanders et al., 2010). In the Cananeia-Iguape Estuary, which receives the drainage from a mountainous crystalline complex, the  $^{210}\text{Pb}$ -derived sedimentation rates ranged between 0.53 and 0.98 cm a<sup>-1</sup> (Saito et al., 2001). In contrast, the sedimentation rates in the highly eutrophicated Guanabara Bay (Rio de Janeiro) were as high as 2 cm a<sup>-1</sup> (Wilken et al., 1986). Saenger (2002) estimated that the vertical accretion in mangroves commonly approached 0.5 cm a<sup>-1</sup>.

In this study, mass sediment accumulation rates ranged between 0.50 and 0.94 g cm<sup>-2</sup> a<sup>-1</sup> (Fig. 4). These results are similar to the mass accumulation rates from the Ajkwa River estuarine mangroves of Irian Jaya (West Papua), Indonesia, which ranged between 0.45 and 1.3 g cm<sup>-2</sup> a<sup>-1</sup> (Brunskill et al., 2004). In southern Thailand, sediment accumulation rates in mangrove forest ranged between 0.29 and 0.76 g cm<sup>-2</sup> a<sup>-1</sup> (Alongi et al., 2001). In Malaysia, sediment mass accumulation (range: 0.22–1.14 g cm<sup>-2</sup> a<sup>-1</sup>) was rapid compared to previous results from mangroves and in the Jiulongjiang Estuary of China, mass accumulation rates were also rapid compared with above-cited measurements in mangroves. Accumulation rates decreased between the low-intertidal zone and the high-intertidal forests, with average rates being 4.90–7.50 g cm<sup>-2</sup> a<sup>-1</sup> in the low intertidal stand to 0.80–1.10 g cm<sup>-2</sup> a<sup>-1</sup> in the high-intertidal forest (Alongi et al., 2005).

In Deep Bay, China, the background concentration of Hg in sediments has been established as 71 ng g<sup>-1</sup> (Wang and Wei, 1995). This was the same as the average background concentration found generally in coastal China (Xia et al., 1984). In the present study, Hg concentrations (ranging between 92 and 196 ng g<sup>-1</sup>) surpassed the background levels of both Deep Bay and coastal China in general. However, Hg concentrations were much lower than in environments strongly affected by urban and industrial inputs, such as Guanabara Bay, where the Hg in mangrove sediments can reach 3000 ng g<sup>-1</sup> (Wasserman et al., 2002). Hg distribution may be related to TOC (Silva et al., 2003; Sanders et al., 2006; Chakraborty et al., 2015). This was confirmed by the high correlation observed

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