Baseline

Distribution of mercury in coastal marine sediments of China: Sources and transport

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ABSTRACT

A total of 220 surface sediments and eight sediment cores were analyzed to study the distribution and transport of Hg in Chinese marginal seas. Spatial distribution showed a general offshore decreasing trend towards the outer continental shelf. Vertical profiles of sediment cores displayed a general increasing trend from bottom to surface layers. Coastal land-based discharges and river-derived inputs are probably the main sources of Hg in coastal sediments of China seas, while TOC, pH, ocean currents and sediment characteristics could play important roles in the transport and spatial distribution of Hg in sediment. The influence of TOC on Hg concentration is more significant than that of pH. The mud deposits on the coastal shelves are main sinks of Hg in the region. The results showed that sedimentary Hg was affected by regional anthropogenic activities and riverine runoffs, and was also influenced by long-range atmospheric transport and ocean current circulations.

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Current global atmospheric deposition of mercury (Hg) has been estimated to be three times higher than preindustrial levels (Selin et al., 2008). The anthropogenic Hg emission of China is the largest in the world owing to its rapid industrialization and urbanization in the last several decades, taking up approximately 30% of the global Hg emission (Pacyna et al., 2010; Wu et al., 2006). In the next few decades, anthropogenic Hg emission in China will still be likely to increase because of the continuously increasing coal consumption and nonferrous metal production (Jiang et al., 2006; Maxson, 2009; Wang et al., 2011). Unraveling the fate of anthropogenic Hg in the environment is an imperative and challenging task.

Coastal regions are under combined impacts of various Hg sources, such as river/sewage discharges and atmospheric deposition. Near-shore regions are often hotspots for the formation of methylmercury (MeHg), which is the most toxic Hg species (Hammerschmidt and Fitzgerald, 2004). Coastal regions thereby act as a dynamic link between the terrestrial and ocean systems, where produced MeHg can be easily transported to the open sea. Elevated Hg concentrations have been reported in mollusks and fish from Chinese coastal regions (Pan and Wang, 2012). Some

coastal residents, fishermen and their families have been found to be at risk of elevated Hg exposure (Cheng et al., 2009; Fok et al., 2007). Therefore, Hg contamination in the coastal environment of China deserves great attention not only for the potential ecological impacts but also for the concern regarding seafood safety to human health.

Sediment plays an important role in the geochemical cycling of Hg, serving not only as a sink but also as a secondary source of Hg in the aquatic environment. Sediment could receive Hg from both natural and anthropogenic sources. Therefore, investigating the distribution of Hg in coastal sediments is an effective way to understand the translocation and cycling of Hg, and to reconstruct the chronology of anthropogenic Hg emissions at the land-ocean interface zone. Some efforts have been made to study the contamination of Hg in sediments from several specific estuaries and coastal regions of China, including Bohai Sea, East China Sea, Xiamen Bay, Pearl River Estuary and Hong Kong (Fang and Chen, 2010; Gu et al., 2012; Luo et al., 2010; Yan et al., 2010; Zhou et al., 2007). However, the behavior of Hg may differ from one estuary to another due to the various inputs, physical, chemical and hydrological conditions. The overall distribution patterns and transport of Hg in coastal sediments of the entire China seas have not been systematically studied and remain unclear until now.

The main objectives of this work were to assess the spatial and vertical distribution characteristics of Hg in coastal sediments from

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four China marginal seas, as well as to identify the possible sources and major control factors for the transport of Hg. The results can provide essential information to better understand the transport and fate of Hg in coastal regions.

As a part of the western Pacific Ocean, the China Sea consists of Bohai Sea (BS), Yellow Sea (YS), East China Sea (ECS) and South China Sea (SCS). The map of the study regions and sampling locations is shown in Fig. 1. A total of 220 surface sediments were collected from the four China seas during 2011–2012, which include 29 samples from the BS, 66 from the YS, 71 from the ECS, and 54 from the SCS. The sampling sites covered most of the marginal seas except for the SCS, where all the sites were distributed within the Pearl River Estuary (PRE). Eight sediment cores were collected in April 2011, including four cores (1–4) from the central mud area of the YS and four (5–8) from the inner and central mud areas of the ECS. These sediment cores were cut into 1 cm intervals along the profile using a stainless steel cutter. All samples were immediately kept in a –4 °C refrigerator after collection and thereafter transported to the laboratory where they were freeze-dried at

–45 °C and homogenized to a fine ground powder through a sieve (80 meshes per inch).

Approximately 0.05–0.1 g of sediment was digested with 2 mL of HNO₃ and 1 mL of HCl at 105 °C for 2 h. The concentrations of total mercury (THg) were determined with a cold vapor atomic fluorescence spectrometer (CVAFS, Beijing Titan Instruments Co., Ltd., China). For the quality control, certified reference materials (CRMs), reagent and method blanks and sample replicates were all included in the analytical procedure. The determined concentrations of Hg in two sediment CRMs were 274 ± 17.8 µg kg⁻¹ (n = 10) in GBW07310 (certified value, 280 ± 40 µg kg⁻¹) and 56.3 ± 4.0 µg kg⁻¹ (n = 10) in GBW07312 (certified value, 56 ± 8 µg kg⁻¹). The relative standard deviation (RSD) for triplicate analysis ranged from 3.7% to 16.9%.

Statistical analysis of the obtained data was accomplished using SPSS 16.0 and Origin 8.0 software. A Kolmogorov–Smirnov test was conducted to check the normal distribution of the obtained data. One-way ANOVA, *t*-test and Pearson's correlation analysis were used to assess the significant differences and potential relationships

Fig. 1. Sampling sites of surface sediments and sediment cores.

Table 1
Concentrations of Hg in surface sediments from four China seas and eight sediment cores (µg kg⁻¹).

Sediment	Location		N	Min	Max	Mean
Surface sediment	BS		29	9.7	160	38.8
	YS		66	7.0	47.4	24.2
	ECS		71	8.2	84.3	32.8
	SCS		54	15.4	398	166.1
	Total		220	7.0	398	63.7
Sediment cores	YS	Core-1	37 cm	31.2	168	58.7
		Core-2	32 cm	26.9	93.1	61.4
		Core-3	35 cm	18.9	85.3	41.2
		Core-4	42 cm	27.4	80.5	45.3
	ECS	Core-5	48 cm	43.5	90.9	68.9
		Core-6	37 cm	48.5	83.6	65.1
		Core-7	56 cm	16.1	44.2	28.1
		Core-8	57 cm	14.3	47.9	26.7
		Total	344	14.3	168	47.4

BS: Bohai Sea; YS: Yellow Sea; ECS: East China Sea; SCS: South China Sea.

among the data. The software ArcGIS 10 was adopted to produce the contour maps by applying the Kriging method for the geospatial interpolation of the Hg data.

The concentrations of Hg in surface sediments are shown in Table 1. For all surface sediments, the concentrations of Hg ranged from 7.0 to 398 $\mu\text{g kg}^{-1}$, with an average value of 63.7 $\mu\text{g kg}^{-1}$. The concentrations of Hg were in the range 9.7–160 $\mu\text{g kg}^{-1}$ (average, 38.8 $\mu\text{g kg}^{-1}$) for the BS, 7.0–47.4 $\mu\text{g kg}^{-1}$ (average, 24.2 $\mu\text{g kg}^{-1}$) for the YS, 8.2–84.3 $\mu\text{g kg}^{-1}$ (average, 32.8 $\mu\text{g kg}^{-1}$) for the ECS,

and 15.4–398 $\mu\text{g kg}^{-1}$ (average, 166 $\mu\text{g kg}^{-1}$) for the SCS. The average concentrations of Hg in surface sediments among the four China seas followed the order: SCS > BS > ECS > YS ($P < 0.01$, one-way ANOVA), with the SCS (PRE) as the most polluted region. This may be because the sampling sites in this region are located at the PRE, which is close to major cities and large industrial hubs in South China.

Fig. 2 shows the spatial distribution of Hg in surface sediments. In the BS, a noticeable hotspot was found at the mouth of the

Fig. 2. The spatial distributions of Hg in surface sediments of China seas ($\mu\text{g kg}^{-1}$). (a) Bohai Sea and Yellow Sea, (b) East China Sea, (c) Yangtze and Qiantang River Estuary, and (d) the Pearl River Estuary.

Yellow River Estuary (YRE) (Fig. 2a), suggesting a direct influence of riverine inputs. At the YS, the Hg levels increased from the coastline to the central areas at both the southern and northern YS (Fig. 2a), with the highest Hg levels in the central mud areas. By contrast, a different distribution pattern was found in the ECS, and relatively high levels of Hg were located at the nearshore areas with a general offshore decreasing trend towards the outer shelf (Fig. 2b), indicating a significant influence of both riverine inputs and proximity to terrestrial-based sources along the coast. Besides, Fig. 2c shows more clearly the distribution of Hg along the Zhejiang–Fujian coast, and two hotspots were observed at the Yangtze and Qiantang River estuaries. However, Hg levels at the mouth of these two river estuaries were significantly lower than those at the southward areas. The same distribution pattern has also been found in our previous study in this area in 2002 (Shi et al., 2005). This is possibly caused by the high sedimentation rates at the river mouth (Huh and Su, 1999), and also a result of the nearby land-based Hg discharge and the circulation currents in the estuaries. Similarly, the Hg levels in sediments from the outer part of the PRE were relatively low. Two hotspots were found at the southwestern coastal regions near Zhuhai city (i.e., the key sediment deposition area in the PRE) and the northeastern coastal regions near Shenzhen (e.g., the heavily industrialized city in south China) (Fig. 2d). This also suggests a significant influence of Hg discharges from nearby cities and coastal circulation currents. Compared to our previous survey on the PRE (Shi et al., 2010), the highest and average concentrations of Hg increased from $201 \mu\text{g kg}^{-1}$ to $398 \mu\text{g kg}^{-1}$, and from $54.4 \mu\text{g kg}^{-1}$ to $166 \mu\text{g kg}^{-1}$ during the year 2002–2011, implying a significant increase of the anthropogenic emissions of Hg in the Pearl River Delta (PRD) region. Compared with published data on the background values of sedimentary Hg in the whole SCS (Zhang and Du, 2005), Hg levels in sediments of the PRE is relatively higher, indicating a strong impact of land-based anthropogenic Hg emissions.

Compared with published data on China seas (Table 2), the Hg levels in this study were in good agreement with previous results in magnitude, but slightly higher in absolute values. According to the reported background values of Hg in sediments (Lindqvist et al., 1984), the concentrations of Hg in most surface sediments in this study were still within the background range ($<100 \mu\text{g kg}^{-1}$), except for most sites of the PRE in the SCS and part of the BS (YRE). Compared with other areas listed in Table 2, the concentrations of Hg in sediments collected in this work were lower than those from some countries in Europe, North America, Africa and Asia. This indicates that the Hg contamination level in sediments from the Chinese marginal seas is relatively low, except for some specific regions (the PRE in the SCS and the YRE in the BS).

The concentrations of Hg in the eight sediment cores are shown in Table 1. The four cores (1–4) from the YS are all located in the central mud area of the YS, which is also a site distant from direct input sources and is characterized by cycles of deposition and resuspension under coastal currents interacting with local oceanographic conditions (Yang and Liu, 2007). The Hg levels in these four cores were quite comparable with each other, with Core-1 in the range $31.2\text{--}168 \mu\text{g kg}^{-1}$ (average, $58.7 \mu\text{g kg}^{-1}$), Core-2 in the range $26.9\text{--}93.1 \mu\text{g kg}^{-1}$ (average, $61.4 \mu\text{g kg}^{-1}$), Core-3 in the range $18.9\text{--}85.3 \mu\text{g kg}^{-1}$ (average, $41.2 \mu\text{g kg}^{-1}$) and Core-4 in the range $27.4\text{--}80.5 \mu\text{g kg}^{-1}$ (average, $45.3 \mu\text{g kg}^{-1}$). Generally, the highest Hg values were found in the surface (recent) segments of each profile (depth, 1–5 cm).

The four cores (5–8) from the ECS are located in two widely different areas of the sea. Core-5 and Core-6 were taken from the inner-shelf coastal mud area, a significant sink of sediment-associated pollutants from direct river inputs and land-originated surface runoffs, while Core-7 and Core-8 were from the central-shelf distal mud area, an ideal place for deposition of airborne contaminants transported mainly from China (Zeng et al., 2012). From Table 1, the concentrations of Hg in Core-5 ($43.5\text{--}90.9 \mu\text{g kg}^{-1}$, average: $68.9 \mu\text{g kg}^{-1}$) and Core-6 ($48.5\text{--}83.6 \mu\text{g kg}^{-1}$, average: $65.1 \mu\text{g kg}^{-1}$) were significantly higher than those in Core-7 ($16.1\text{--}44.2 \mu\text{g kg}^{-1}$, average: $28.1 \mu\text{g kg}^{-1}$) and Core-8 ($14.3\text{--}47.9 \mu\text{g kg}^{-1}$, average: $26.7 \mu\text{g kg}^{-1}$) ($P < 0.01$, t -test). These clear differences of Hg concentration suggest that the riverine input and coastal discharge of Hg to the inner-shelf mud area are more significant than atmospheric input to the central-shelf mud area.

Fig. 3 shows the vertical distribution of Hg in the eight sediment cores. For the four cores from the YS, they all showed an obvious increasing trend, with the top layers containing the highest concentrations of Hg. The Hg values in the upper segments (0–10 cm) of each core increased more rapidly than the lower segments. This suggests the significant increase of emissions of Hg in recent years in this region. The four cores from the ECS showed different variations. The Hg values first slightly decreased from the bottom, and then increased to the top in the upper 40 cm. The highest Hg values were also found at the top layers, which could also be attributed to the rapid increasing emission of Hg in recent decades. The relatively high concentrations of Hg at the bottom layers might be resulted from the large historical deposition flux.

In order to identify the key factors influencing the distribution and transport of Hg in coastal sediments of China, the TOC and pH values in all surface sediments were analyzed. The obtained data and results of Pearson's correlation analysis are summarized in Table 3. The TOC contents were in the range 0.08–2.82% with an average of 1.35%. The pH values ranged from 5.96 to 8.68 with

Table 2
Comparison of published concentrations of Hg in different marine sediments ($\times 10^3 \mu\text{g kg}^{-1}$).

Location	Hg	Reference
Toulon Bay, France	1.600 (0.030–27.300)	Tessier et al. (2011)
Ionian Sea, Italy	3.100–7.900	Petronio et al. (2012)
Mersey Estuary, UK	0.840–5.130	Vane et al. (2009)
Guanabara Bay, Brazil	0.870 ($<0.100\text{--}3.220$)	Covelli et al. (2012)
Narragansett Bay, USA	0.555 (0.035–2.629)	Taylor et al. (2012)
San Jorge Bay, Chile	0.259–1.210	Valdés et al. (2010)
Gulf of Mexico	0.100–1.060	Jorge et al. (2009)
Biétri Bay, Ivory Coast	0.790 (0.680–0.880)	Coulibaly et al. (2012)
Mumbai Harbor, India	0.120–0.750	Ram et al. (2009)
Sardinia, Western, Mediterranean	$<0.030\text{--}0.300$	Schintu et al. (2009)
South China Sea, Malaysia	0.100–0.130	Saraee et al. (2011)
East China Sea	0.004–0.048	Fang and Chen (2010)
	0.037 ($<0.001\text{--}0.080$)	Shi et al. (2005)
Pearl River Estuary	0.054 (0.002–0.201)	Shi et al. (2010)
South China Sea	0.061 (0.005–0.148)	Zhang and Du (2005)
China Seas	0.064 (0.007–0.398)	This work

Fig. 3. The vertical distribution of Hg in marine sediment cores.

an average of 8.03. The correlations between Hg concentrations and TOC were significant in all the four China seas, while the correlations between Hg concentrations and pH were significant only in the YS and ECS. When taking all the samples together, the concentrations of Hg showed significant correlations with both TOC

and pH. But the influence of TOC on Hg concentrations was more significant than that of pH.

Fig. 4 shows the sediment particle size distribution and general circulation system of the China seas. Ocean currents and circulation play a key role in sediment transport and deposition in the marginal

Table 3

Correlations of THg, TOC and pH values.

Sampling sites	TOC (%)	pH	$R_{\text{Hg, TOC}}$	$R_{\text{Hg, pH}}$
Bohai Sea ($n = 29$)	1.34 (0.08–2.18)	8.29 (7.91–8.68)	0.489**	–0.135
Yellow Sea ($n = 66$)	1.42 (0.16–2.78)	8.14 (7.70–8.63)	0.640**	–0.623**
East China Sea ($n = 71$)	1.19 (0.51–2.82)	8.21 (7.93–8.60)	0.283*	–0.630**
South China Sea ($n = 54$)	1.47 (0.16–2.63)	7.52 (5.96–8.34)	0.403**	–0.200
Total ($n = 220$)	1.35 (0.08–2.82)	8.03 (5.96–8.68)	0.283**	–0.679**

* Correlation is significant at the 0.05 level.

** Correlation is significant at the 0.01 level.

Fig. 4. Surface sediment particle size distribution and general circulation system in Chinese marginal seas (modified after Hu et al., 2011; Yang et al., 2003; Zhu et al., 2012; Guo et al., 2006). BS: Bohai Sea; YS: Yellow Sea; ECS: East China Sea; BCC: Bohai Coastal Current; YSCC: Yellow Sea Coastal Current; YSWC: Yellow Sea Warm Current; KCC: Korea Coastal Current; ZFCC: Zhejiang–Fujian Coastal Current; TWC: Taiwan Warm Current; KC: Kuroshio Current.

seas. The massive Yellow River-derived sediment flux is transported from the narrow coastal area to the offshore area during winter in the BS, but also moves eastward off the northern Shandong Peninsula driven by the Bohai Coastal Current (BCC) and then southward into the south YS driven by the Yellow Sea Coastal Current (YSCC) (Lu et al., 2011; Yang et al., 2003). The sediments in the BS are therefore mainly deposited as clay in the western part (near the YRE), silt in the central part and sand in the eastern part (Yang et al., 2003). Two general circulation patterns are reported in the YS, including a counterclockwise gyre with a northward inflow of the Yellow Sea Warm Current (YSWC) and a southward inflow of the YSCC, and a clockwise gyre with the YSWC and a southward inflow of the Korea Coastal Current (KCC). As a result, the central part of the YS is dominated by clay, while silt and sand generally blanket the eastern and western parts (Yang et al., 2003). In the ECS, a large proportion of Yangtze River-derived sediments are transported southwards along the coast driven by the Zhejiang–Fujian Coastal Current (ZFCC) and trapped in the inner shelf due to the blocking by the northward offshore Taiwan Warm Current (TWC). An elongated inner-shelf mud wedge is consequently developed from Yangtze mouth to Taiwan Strait, which is broken at the mouth of the Qiantang River (Zhu et al., 2012). Another mud area in the central ECS is formed in the southwest off Cheju Island.

Particle size distribution is another possible factor influencing the distribution of Hg in sediment. Normally, Hg could sorb more strongly to smaller particles because of their greater surface area (Lv and Li, 2006). Ding et al. calculated the regressive equations

between Hg content and particle size, and the results showed that Hg content was most significantly correlated with clay, the next with silt and finally with sand (Ding et al., 2009). In this study, the areas with the highest sedimentary Hg levels are located in muddy sediment areas. The spatial distributions of Hg in surface sediments (Fig. 2) corresponded well with the distribution patterns of particle size (Fig. 4), with the highest Hg concentrations in clay, next higher in silt and the lowest in sand. Obviously, sediment characteristics are also an important factor controlling the distribution of Hg in coastal sediments of China. Therefore, the mud deposits on the coastal shelves of China could be main sinks of Hg in sediments.

In addition, sediment particle size indirectly reflects the hydrodynamic environment. With no direct large river inputs and far away from direct sources, the YS mud area is under convergent hydrodynamic condition and is thereby more characterized by homogeneous conditions. Sedimentary Hg in this area may largely derive from atmospheric deposition and sedimentation. However, different from the YS, the coastal ECS is subject to more heterogeneous conditions, due to the direct continuous riverine inputs from the Yangtze River and other major coastal rivers. Comparatively, the higher Hg levels in the coastal ECS area suggested that riverine inputs and land-based Hg discharge may play more important roles than atmospheric deposition in the distribution of Hg in marine sediments. This indicated that sedimentary Hg was transported under the influence of riverine inputs and sediment characteristics, and further distributed under the influence of ocean currents and circulation.

As a whole, the Hg contamination in coastal sediments of China is relatively low in comparison with some coastal areas in the world. Spatial distribution showed a general offshore decreasing trend of Hg concentrations towards the outer continental shelf. Vertical profiles of sediment cores displayed a general increasing trend from bottom to surface layers. Coastal land-based discharges and river-derived inputs are probably the main sources of Hg in coastal sediments of China seas. Correlation analysis indicated influences of TOC and pH on the Hg concentration in sediments. The good agreement between Hg and particle size distribution suggested that the mud areas in the coastal shelves of China are major sinks of Hg.

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