Mercury contamination and accumulation in sediments of the East China Sea

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Abstract

Mercury (Hg) concentrations in sediment samples collected from the inner and middle shelves of the East China Sea (ECS) were analyzed to evaluate Hg contamination levels and to calculate Hg sedimentation rates and total accumulation in the ECS. The range of Hg concentrations in surface sediments of the inner shelf was 26.5–47.6 ng/g, and that for the middle shelf was 4.1–13.9 ng/g. Hg concentrations correlated well with organic carbon contents but varied inversely with sediment grain size. Enrichment factors indicated that the whole inner shelf and a small portion of the middle shelf were slightly contaminated by Hg. Hg accumulation rates in the ECS ranged between 0.42–48.7 ng/(cm^2·yr), with higher values observed in the inner shelf. Total Hg accumulation in the calculated area (accounting for 80% of the ECS continental shelf area) ranged from 25 to 30 tons/yr; approximately 51% and 17% of the accumulated Hg mass was deposited in the Yangtze estuarine zone and the inner shelf, respectively.

Key words: Hg; contamination; enrichment factor; accumulation; the East China Sea

Introduction

The Minamata Bay disaster (1950–1970) revealed the harmful effects of mercury (Hg) on human health, and Hg has subsequently become one of the most critical trace metals examined in environmental studies (Tomiyasu et al., 2006). Hg is released and mobilized through natural processes and by anthropogenic activities. Pacyna et al. (2003) comprehensively reviewed global and continental atmospheric Hg emissions and indicated that the anthropogenic contribution of Hg mobilization globally was approximately 1913 tons in 1995. The stationary combustion of fossil fuels, particularly coal, is the major contribution, accounting for over 75% of the total. China is the largest producer and consumer of coal in the world; as of 2003, China consumed nearly 1531Mt of coal, 28% of the world's total consumption (Wong et al., 2006). In addition, with the rapid development of industry during the last two decades, Hg consumption in China is greater than 900 tons, 50% of worldwide production in 2000 (Jiang et al., 2006). China contributes a significant portion of the global anthropogenic Hg emissions to the environment.

China is surrounded from the north to south by the Yellow Sea, the East China Sea (ECS), the Taiwan Strait and the South China Sea. The East China Sea, located between 26°–31°N and 121°–126°E, is one of the largest marginal seas in the Western Pacific, and it is also the main discharge area of the Yangtze River. This is the world's fourth largest river in terms of suspended load. Its annual suspended load transport is approximately 4.78×10^8

tons/yr (Milliman, 1991). There are many developed cities along the coast, such as Shanghai and Ningbo, resulting in a large number of factories in the area. Due to a lack of waste control practices, many studies have indicated that trace metal contamination exists in Hangzhou Bay, extending to the ECS (Huh and Chen, 1999; Yuan et al., 2004). In addition, the Huangpu River, a small tributary near the Yangtze estuarine mouth, is also contaminated by Hg (Ding et al., 2005). However, knowledge about Hg distribution in the ECS is limited (Shi et al., 2005). Therefore, the purpose of this study was to investigate the Hg contents in inner and middle ECS shelf sediments, and to estimate annual Hg accumulation in the ECS.

1 Sampling and methods

1.1 Study area

Thirty-eight sediment cores were collected with a box corer during two cruises with the R/V Ocean Research-I from August, 1997 and May, 1999. The box corer was designed to obtain undisturbed sediment. The core samples were sealed and kept frozen for further processing in the laboratory. The sampling stations (Fig. 1) were located outside of the mouth of the Yangtze River in the inner (station I) and middle (station M) shelf area at < 100 m water depth. After thawing in the laboratory, the cores were extruded vertically using a hydraulic jack and sampled at 1–2 cm intervals. The outer edge (approximately 0.5 cm) of each sediment slab was trimmed off to avoid contamination between layers. Approximately 20–30 g of

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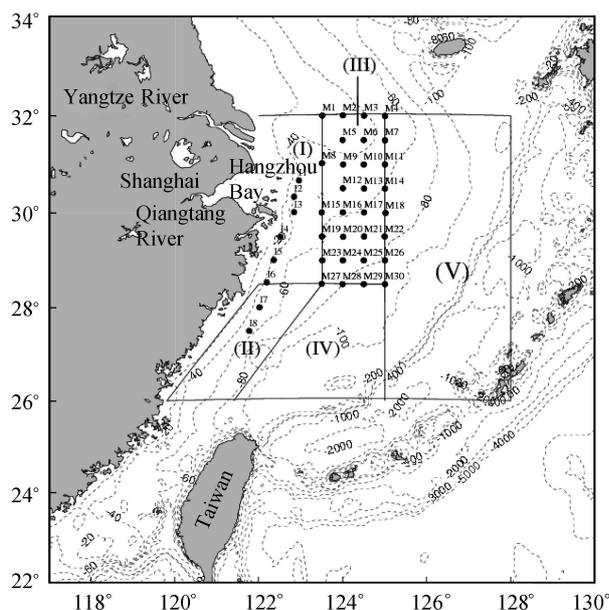
Fig. 1 Map showing sediment sampling stations in the East China Sea where was divided into five boxes for Hg accumulation rate and accumulation calculations. The five boxes representing different mass accumulation rate areas are marked as: (I) estuary; (II) inner shelf; (III) middle shelf; (IV) middle shelf; and (V) outer shelf.

each sample was freeze-dried and ground with a mortar and pestle. The processed sample was stored in an acid-cleaned polypropylene container for further analysis. More detailed descriptions of the sampling and treatment can be found elsewhere (Huh and Su, 1999; Su and Huh, 2002).

1.2 Analytical methods

Concentrations of Hg were determined in surface sediments from all stations. The analytical procedure was modified from USA EPA Method 7474 in which the sample is digested in a microwave unit. In the present study, the samples were digested in a water bath similar to the methods introduced by Shi et al. (2005). The analytical procedure is briefly described below. The total Hg in each freeze-dried sediment sample of approximately 0.5 g was determined after digestion with 10 mL of 50% aqua regia in a Teflon centrifuge vessel (12 hr in a 70°C water bath). After cooling, the digested solution was diluted to 20 mL with Milli-Q water and bromine monochloride (BrCl) solution was added to oxidize the organic-Hg into inorganic-Hg (12 hr in a 50°C water bath). After oxidation, a $NH_2OH\cdot HCl$ solution was used to reduce the excess Br-Cl. This was followed by the addition of stannous chloride ($SnCl_2$) solution to reduce the reactive Hg. The reaction vessel was then purged with Hg-free nitrogen at a flow rate of 360 mL/min for five minutes. Elemental Hg purged from the sample was pre-concentrated on a gold-coated bead trap. This trap was then heated, and the released Hg vapor was measured using a Brooks Rand Model III Cold Vapor Atomic Fluorescence Spectrophotometer, USA.

Hg determinations in reference materials (MESS-2 and PACS-2 from the National Research Council, Canada) were performed using the same analytical procedure. The results were (0.088 ± 0.005) and (2.92 ± 0.18) mg/kg (n

= 9), respectively. The accuracies were (95.7 ± 5.6)% and (96.7 ± 6.2)%, respectively. The detection limit based on three times the reagent blank value was 0.02 ng/g.

1.3 Enrichment factors

Trace metal concentrations in marine sediments can vary widely depending on the natural texture of the sediment and the anthropogenic inputs. For example, the Mn concentration in continental shelf surface sediment is generally less than 1000 μg/g (Goldsmith et al., 2001; Aloupi and Angelidis, 2001). However, an extremely high concentration of Mn, > 5000 μg/g, has been found in the eastern part of the Laptev Sea due to the diagenetic process which provides a large amount of Mn to the upper sediment layer (Nolting et al., 1996). As a result, it is difficult to evaluate whether the observed concentration in marine sediments is influenced by anthropogenic sources or not without a normalizing process. Normalizing elements relative to Al is widely used to compensate for differences in grain size variations, carbonate content and anthropogenic sources (Luoma, 1990). Normalization to the background levels of metals in samples with different characteristics can be accomplished by calculating an enrichment factor (EF) relative to the earth's crust. The equation used is:

$$EF = C_{M,s} \times C_{Al,Crust} \times C_{Al,s}^{-1} \times C_{M,Crust}^{-1}$$

where, $C_{M,s}$, $C_{Al,s}$, $C_{M,Crust}$ and $C_{Al,Crust}$ are metal and Al concentrations in the samples and the crust, respectively (Luoma, 1990). The EF, a useful indicator reflecting the status and the degree of environmental contamination, has been widely used to assess the metals contamination in marine environments (Leivuori, 1998; Pempkowiak et al., 1998; Aloupi and Angelidis, 2001).

1.4 Hg accumulation rate and accumulation

Ocean margins are regions adjacent to the continents, including inland seas, estuaries, and continental shelves and slopes, where the major portion of continental detritus is delivered into the ocean. Information on the sedimentation rate can be obtained using short-lived, naturally occurring, and anthropogenic radionuclides. With a half-life of 22.3 years and a well-defined source function, ^{210}Pb is the most commonly used chronometer for estimating the sedimentation rate in the continental shelves (De Master et al., 1985; Huh and Su, 1999). The ECS is one of the most important continental shelves around the world due to the Yangtze River discharging into the area. Based on ^{210}Pb profiling, De Master et al. (1985) showed accumulation rates up to 5.4 cm/yr near the mouth of the Yangtze River and decreasing seaward to 0.3 cm/yr in a mud patch about 400 km offshore. Huh and his co-workers (Huh and Su, 1999; Su and Huh, 2002) employed multiple radio-nuclides (^{210}Pb, ^{137}Cs and [239,240]Pu) to comprehensively determine sediment mass accumulation rates (MAR) in the ECS. MAR values ranged from 0.05 to > 2 g/(cm[2]·yr), with a maximum value at the mouth of the Yangtze River. Values generally decreased southward along the inner shelf and eastward offshore.

The Hg accumulation rate at the studied ECS stations

was calculated by multiplying the Hg concentration in surficial sediments found in this study by the MAR observed previously (Huh and Su, 1999; Su and Huh, 2002). Based on the Hg accumulation rate, an attempt is made to calculate the total Hg accumulation in the ECS. The study area was divided into five boxes to facilitate the calculation: estuary (box I), inner shelf (box II), middle shelf (box III and IV), and outer shelf (box V) (Fig. 1), and calculations for each box were made using the reported MAR value (Huh and Su, 1999; Su and Huh, 2002). The middle shelf is divided into two boxes because the MAR in the northern middle shelf differed slightly from that in the southern middle shelf. The Hg accumulation rate range in each box is given by the minimum and maximum values among the studied stations within the box. The minimum, average and maximum Hg accumulation values in each box were calculated from the corresponding Hg accumulation rate values multiplied by the box area.

Because the ECS is adjacent to the Yellow Sea to the north and the Northern Pacific Ocean to the east, its total area is difficult to determine. Two widely accepted values are 0.74×10^6 km^2 for the total ECS area and 0.51×10^6 km^2 for the area with a water depth < 200 m (Wong et al., 2000). The area surveyed in this study is approximately 0.405×10^6 km^2, which accounts for about 60% of the whole ECS area provided by Wong et al. (2000) and about 80% of the continental shelf area.

2 Results and discussion

2.1 Spatial distribution of Hg

Mercury concentrations and enrichment factors in surface sediments at each station are listed in Table 1. The Hg spatial distribution is plotted in Fig. 2. Hg concentrations in the surface sediments ranged from 4.1 to 47.6 ng/g. Higher concentrations (26.5–47.6 ng/g) were observed at the inner
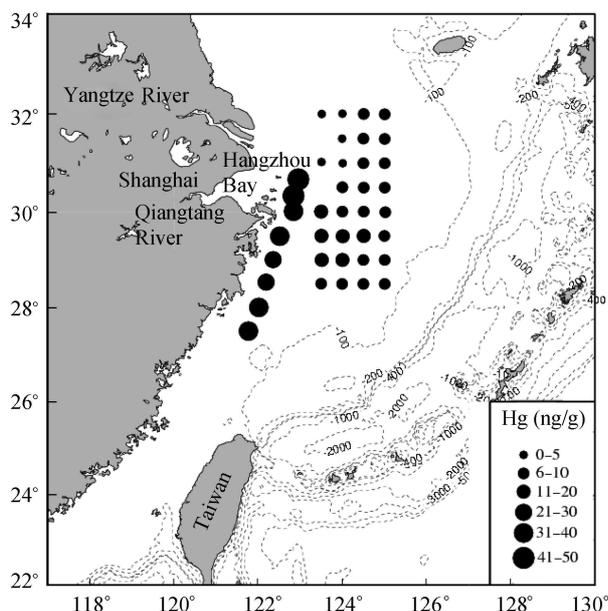
Table 1   Concentration of Hg and Al, enrichment factor (EF) and mass accumulation rate in surface sediments (0–2 cm) in the East China Sea

Station	Location	Hg (ng/g)	Al* (%)	EF	Mass accumulation rate** (g/(cm²·yr))
I1	30°39.96′N, 122°57.06′E	47.6	8.9	6.3	0.40
I2	30°19.95′N, 122°50.13′E	44.7	ND	ND	1.09
I3	30°00.52′N, 122°50.18′E	37.4	9.2	4.8	0.91
I4	29°29.61′N, 122°30.56′E	39.6	7.8	6.0	0.93
I5	29°00.39′N, 122°21.07′E	29.9	7.8	4.5	1.05
I6	28°31.95′N, 122°10.94′E	26.5	8.9	3.5	1.07
I7	28°00.36′N, 122°00.59′E	30.5	8.0	4.5	0.56
I8	27°30.12′N, 121°45.81′E	30.5	8.0	4.5	0.14
M1	31°59.92′N, 123°30.44′E	4.1	5.6	0.9	ND
M2	32°00.20′N, 122°59.88′E	4.2	6.1	0.8	0.19
M3	32°00.13′N, 124°30.20′E	5.9	6.4	1.1	0.24
M4	31°59.78′N, 125°00.30′E	6.2	5.5	1.3	ND
M5	31°30.25′N, 123°59.61′E	4.9	5.4	1.1	0.15
M6	31°29.98′N, 124°29.83′E	8.1	6.5	1.5	0.66
M7	31°29.74′N, 125°00.14′E	5.9	6.8	1.0	0.11
M8	31°01.36′N, 123°30.11′E	4.5	6.1	0.9	ND
M9	30°59.95′N, 124°00.21′E	4.3	ND	ND	0.98
M10	31°00.10′N, 124°29.99′E	8.1	5.9	1.6	0.31
M11	30°59.99′N, 124°59.97′E	9.8	6.8	1.7	0.17
M12	30°30.21′N, 124°00.00′E	6.7	5.6	1.4	0.16
M13	30°29.95′N, 124°30.25′E	9.7	6.3	1.8	0.14
M14	30°30.01′N, 125°00.05′E	9.3	6.2	1.8	0.06
M15	30°00.12′N, 123°29.75′E	11.7	6.5	2.1	0.53
M16	30°00.92′N, 124°00.03′E	8.3	5.9	1.7	1.01
M17	30°00.18′N, 124°30.10′E	5.8	6.0	1.1	0.14
M18	29°59.65′N, 125°01.04′E	5.7	6.0	1.1	0.11
M19	29°30.03′N, 123°30.04′E	13.9	6.7	2.4	0.62
M20	29°29.59′N, 124°00.20′E	13.3	6.1	2.6	0.34
M21	29°29.95′N, 124°30.11′E	11.1	5.7	2.3	0.81
M22	29°30.13′N, 124°59.68′E	5.9	6.3	1.1	0.65
M23	29°00.00′N, 123°30.28′E	10.8	6.7	1.9	0.44
M24	28°59.85′N, 123°59.92′E	11.6	5.4	2.5	0.21
M25	28°59.89′N, 124°29.84′E	7.4	5.8	1.5	0.17
M26	28°59.99′N, 125°00.00′E	5.7	5.9	1.1	0.13
M27	28°30.31′N, 123°29.67′E	8.9	5.8	1.8	0.40
M28	28°29.67′N, 123°59.58′E	9.8	5.7	2.1	0.61
M29	28°29.94′N, 124°30.02′E	7.8	6.8	1.3	0.07
M30	28°29.99′N, 125°00.05′E	5.3	5.9	1.1	0.08

* Data are from Lu, 2001; ** Data are from Huh and Su, 1999 and Su and Huh, 2002.

shelf stations, while lower concentrations (4.1–13.9 ng/g) at the middle shelf stations. The Hg concentration exhibits a fair decrease southwards along the inner shelf and shows a decrease in the southeastern direction along the middle shelf. Shi et al. (2005) studied Hg contents in sediments collected outside of the Yangtze Estuary and Hangzhou Bay. Their results showed that concentrations of total Hg in surficial sediments (0–1 cm) of the surveyed area ranged within < 0.5–62.3 ng/g, with higher concentrations in Hangzhou Bay. The Hg concentrations along the inner shelf found in the present study are in good agreement with those observed by Shi et al. (2005).

The sediment grain size and total organic carbon (TOC) of the surface sediments at the studied stations were analyzed by Lu (2001). Those results showed that the surface sediments of the inner shelf were mainly composed of silt and clay, with sand content less than 5%, and the TOC concentration ranged from 0.54% to 0.75%. In contrast, sand was the major component (contribution from 46% to 98%), and silt was the secondary component (ranging from 1% to 30%), of the surface samples at the middle shelf stations. TOC concentrations at the middle shelf stations ranged between 0.20%–0.58%, values that were relatively

Fig. 2 Spatial variation of Hg concentration in the surface sediment (0–2 cm) at the studied stations of the East China Sea.

lower than those of the inner shelf stations.

It is well recognized that finer sediments, especially those coated with organic matter, have a larger surface area and a higher adsorption capacity (Stumm and Morgan, 1996). As shown in Fig. 3, Hg concentrations in ECS surface sediments were correlated with clay content ($r^2 = 0.67$; $p < 0.05$) and TOC concentration ($r^2 = 0.57$; $p < 0.05$). These data indicate that higher Hg concentrations in these sediments are associated with finer grain sizes and higher TOC concentrations. These results are in good agreement with previous studies which showed that Hg concentrations in marine sediments correlated well with organic carbon content (Leivuori, 1998; Pempkowiak et al., 1998; Alonso et al., 2000; Shi et al., 2005) and varied inversely with sediment grain size (Pempkowiak et al., 1998; Covelli et al., 2001). Both phenomena may help explain why the Hg concentration found in the middle shelf was lower than that from the inner shelf.

There are two important rivers, the Yangtze and the Qiantang Rivers, discharging into the East China Sea. As the largest river in China, the Yangtze River carries a total of 4.78×10^8 tons of fine sediments to the ECS annually (Milliman, 1991). Lie et al. (2003) studied the freshwater plume of the Yangtze River, showing that the plume tends to move northeastwards along the Chinese coastal area during June to early September when souther-

ly winds prevail. During other seasons, when northerly winds prevail, the plume is confined to the Chinese coast. This observation may also explain why the spatial variation of Hg concentrations in the sediments of the ECS displayed a fair decrease southwards along the inner shelf and a decrease in southeastern direction along the middle shelf.

2.2 Enrichment factor

Mercury concentrations in marine surface sediments can vary widely. A comparison of the Hg concentration in surface sediments and Hg accumulation rates in some marginal seas, such as the Arctic Ocean and the Mediterranean Sea, as well as from disturbed and seriously polluted marine environments around the world, is shown in Table 2. Hg concentrations in sediments of the marginal seas and pristine environments, such as the eastern basin of the Mediterranean Sea and the Arctic Ocean, are generally less than 100 ng/g. Hg concentrations in seriously polluted environments, such as Kaštela Bay (Croatia), the Gulf of Trieste (Italy), and Cartagena Bay (Colombia), can be greater than 10,000 ng/g. It is not immediately clear whether or not Hg concentrations in ECS sediments are at contamination levels, because all Hg concentrations found in this study were lower than 100 ng/g. As described above, the enrichment factor (EF) concept is often used to assess the metal contamination status of a marine environment. This approach is employed here to assess the Hg contamination status of the ECS. For calculating the EF, Hg and Al concentrations in ECS sediments were taken from this study and published results (Lu, 2001), respectively. The Al concentration of the Earth's crust is taken from the literature (Taylor, 1964). However, the Hg concentration in the Earth's crust shown in the literature (Taylor, 1964; Wedepohl, 1995) is relatively high and is possibly incorrect because the instrument sensitivity was lower during those early studies (Chi, 2004). A recent study which investigated the Hg content in the crust, rock and loose sediment around China combined with IGCP 360 Global Geochemical Baseline data recommends that the Hg abundance in the crust is approximately 7 ng/g (Chi, 2004). As a result, this value is adopted here to calculate enrichment factors in ECS sediments. The calculated EF at each station is listed in Table 1. EF values at stations of the inner shelf ranged between 3.5–6.3, suggesting that this area was contaminated by Hg. EF values of the middle shelf stations were generally less than 1.5, and only few stations ranged within 1.5–2.0, indicating that only a small portion of the middle shelf was influenced by anthropogenic sources. An EF value of 1 indicates a predominantly natural origin for the element in the sediment, while values greater than 1.5 indicate enrichment by either natural processes (e.g., biota contributions) or anthropogenic influences (Zhang and Liu, 2002).

2.3 Hg accumulation rate and accumulation

Based on the Hg concentrations in surface sediments found in this study and the mass accumulation rates reported in the literature (Huh and Su, 1999; Su and Huh, 2002), we calculated Hg accumulation rates at each station
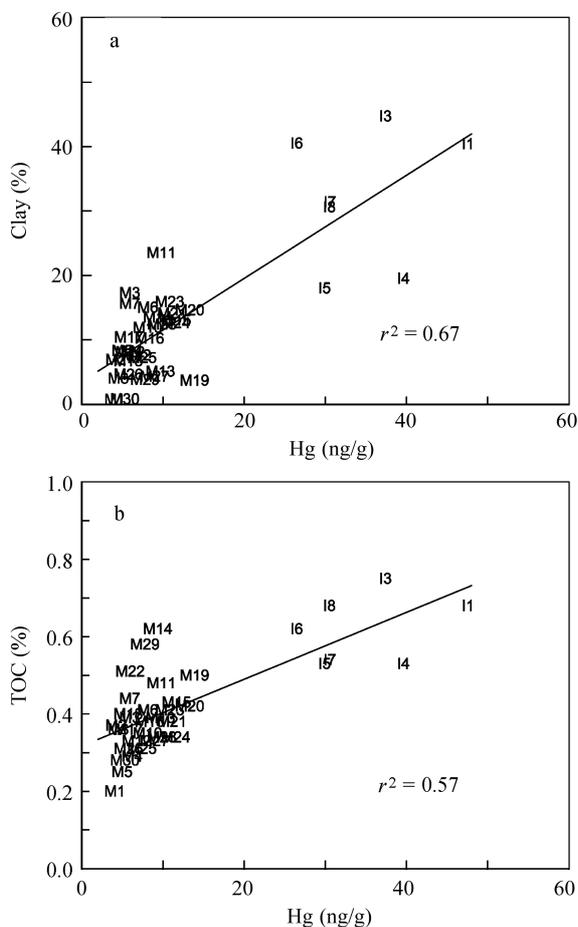
Fig. 3 Scatter plots between Hg concentration and clay content (a), and TOC concentration (b) of the surface sediments of the East China Sea. The symbols is the station number.

Table 2   Comparison of Hg concentration in surface sediments (0–2 cm) and Hg accumulation rate in the East China Sea, the pristine, disturbed and polluted marine environments around the world

Location	Digested chemicals	Total Hg (ng/g)	Hg accumulation rate (ng/(cm²·yr))	Reference
Inner shelf, the East China Sea	Aqua regia	26.5–47.6	4.3–48.7	Present study
Middle shelf, the East China Sea	Aqua regia	4.1–13.9	0.42–9.0	Present study
Pristine marine environments				
Arctic Ocean	Aqua regia/HF	50–100	0.01–0.02	Gobeil et al., 1999
Greenland	Aqua regia	6–257	0.43–39.5	Asmund and Nielsen, 2000
Eastern basin, the Mediterranean Sea	Aqua regia/HF	8.0–40.1	0.09–0.18	Ogrinc et al., 2007
Guaratuba Bay, Brazil	Aqua regia	15.1–44	12–27	Sanders et al., 2006
Disturbed marine environments				
Western basin, the Mediterranean Sea	Aqua regia/HF	78.2–90.3	0.38–1.13	Ogrinc et al., 2007
Strait of Georgia, Canada	HNO_3/H_2SO_4	60–420	5–181	Johannessen et al., 2005
Southern Baltic Sea	$HNO_3/HClO_4/HF$	2–342	ND	Pempkowiak et al., 1998
Gulf of Finland	Aqua regia/HF	50–320	ND	Leivuori, 1998
Lavaca Bay, Texas	–	5–783	ND	Bloom et al., 1999
Polluted marine environments				
Minamata Bay, Japan	$HNO_3/HClO_4/H_2SO_4$	1400–4300	ND	Tomiyasu et al., 2006
Gulf of Trieste, northern Adriatic Sea	HNO_3	100–23300	ND	Covelli et al., 2001
Cartagena Bay, Colombia	–	94–10293	ND	Alonso et al., 2000
Kaštela Bay, Croatia	HNO_3	14280–30400	ND	Kwokal et al., 2002

ND: not determined.

(Table 1 and Fig. 4). Relatively high Hg accumulation rates, ranging from 4.3 to 48.7 ng/(cm²·yr), were observed along the inner shelf due to the higher Hg concentrations and sediment mass accumulation rates there compared to the other sites. Mercury accumulation rates in the middle shelf were lower (0.42–9.0 ng/(cm²·yr)). Hg accumulation rate data for marine environments are limited. A comparison of Hg accumulation rates in marine environments worldwide is shown in Table 2. Hg accumulation rates in pristine environments, such as the Arctic Ocean, are as low as < 0.02 ng/(cm²·yr) (Gobeil et al., 1999). Accumulation rates increase to 0.1–1.0 ng/(cm²·yr) in the Mediterranean Sea (Ogrinc et al., 2007) and to 0.43–39.5 ng/(cm²·yr) along the Greenland continental shelf (Asmund and Nielson, 2000). A much higher Hg accumulation rate, 181 ng/(cm²·yr), can be seen in a Hg polluted environment,

such as in the Strait of Georgia, because of the relatively high sediment accumulation rate, (2.01 ± 0.4) ng/(cm²·yr) (Johannessen et al., 2005). These results indicate that Hg accumulation rates in marine environments can vary by four orders of magnitude. With the exception of seriously polluted systems, the range of Hg concentrations in marine environments worldwide is generally less than one order of magnitude. However, the sedimentation rate in different marine environments can vary by up to three orders of magnitude (Gobeil et al, 1999; Huh and Su, 1999; Johannessen et al., 2005; Ogrinc et al., 2007). Thus, the sedimentation rate seems to play a more important role than the Hg concentration in determining the Hg accumulation rate in marine environments.

The calculated results for Hg accumulation and the parameters used for the calculations in each box of the ECS are summarized in Table 3. Hg accumulation ranged between 4.25–44.08 tons/yr with average (18.05 ± 15.48) tons/yr for the entire calculated area. Approximately 51% of this accumulated Hg mass is deposited in the Yangtze estuarine zone (box I), and 17% accumulates on the inner shelf (box II). The accumulated Hg mass on the middle shelf (box III and IV) and the outer shelf (box V) account for only 19% and 13%, respectively, of the total amount. If the average value found in the inner and middle shelves is used to calculate the accumulation on the outer shelf, the Hg accumulation for all shelves together in the ECS is approximately 30 tons/yr. Alternatively, if the value of the outer shelf area is considered as less than 20% of the total value above for the inner and middle shelves, then the value should be reduced by 5 to 25 tons/yr. The main water discharge into the ECS is from the Yangtze River. Ding et al. (2005) found that the annual riverine inputs of dissolved and particulate Hg from the Yangtze River into the ECS are 12.1 tons and 15.8 tons, respectively. The combined value is 27.9 tons which is within the estimated range of the present study. Our result showing about 50% of the Hg accumulation occurring near shore in the ECS compares
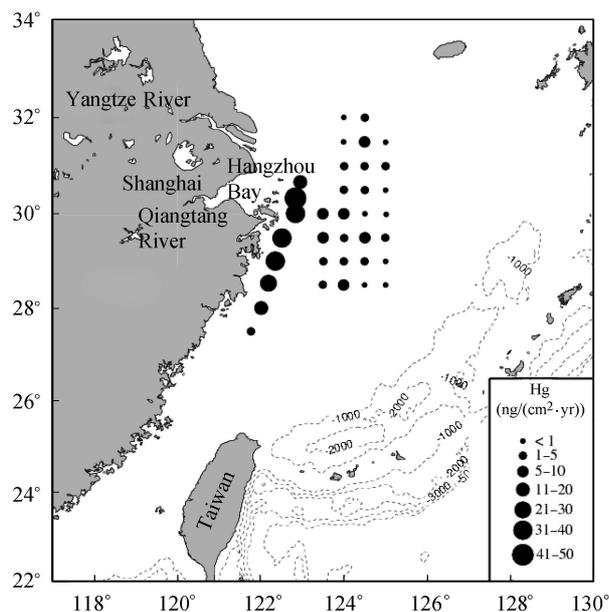
Fig. 4   Spatial variation of Hg accumulation rate at the studied stations of the East China Sea.

Table 3 Area, mass accumulation rate, Hg concentration, Hg accumulation rate and accumulation for each box of the East China Sea

Box	Area (km^2)	Mass accumulation rate* (g/(cm^2·yr))	Hg concentration (ng/g)	Hg accumulation rate (ng/(cm^2·yr))	Hg sedimentation flux (tons/yr)
I	42400	0.40–1.09 (0.74 ± 0.29)	4.05–47.58 (23.28 ± 16.2)	3.58–48.67 (22.14 ± 15.92)	1.51–20.64 (9.39 ± 6.75)
II	38200	0.14–0.56 (0.37 ± 0.21)	8.94–30.53 (23.32 ± 12.46)	3.57–17.10 (8.31 ± 7.61)	1.37–6.53 (3.18 ± 2.91)
III	57400	0.06–1.01 (0.35 ± 0.28)	2.42–13.87 (7.50 ± 2.88)	0.42– 8.99 (2.97 ± 2.71)	0.24–5.16 (1.70 ± 1.56)
IV	69700	0.07–0.61 (0.29 ± 0.26)	5.28–9.84 (7.96 ± 1.98)	0.42–6.00 (2.64 ± 2.67)	0.29–4.18 (1.84 ± 1.86)
V	197000	0.06–0.65 (0.19 ± 0.21)	5.28–9.79 (6.80 ± 1.90)	0.42–3.84 (1.22 ± 1.22)	0.83–7.57 (2.39 ± 2.40)
Total	404700				4.25–44.08 (18.05 ± 15.48)

Values in parentheses are the average and one standard deviation.

* Data are from Huh and Su, 1999 and Su and Huh, 2002.

well with the results of Cossa et al. (1997), who found that Hg accumulation in the Mediterranean Sea ranged between 6.22–13.44 tons/yr, and 50% of this amount was deposited near shore.

It has been reported that Hg partitioning between the particulate and dissolved phases varies significantly from estuarine to oceanic seawater. Particulate Hg dominates in estuarine seawater but decreases in the seaward direction, and is not important in oceanic seawater (Mason et al., 1998; Mason and Suillivan, 1999; Leermakers et al., 2001). As a consequence, these studies concluded that only a small fraction of the riverine transport of Hg is exported into the ocean due to the high retention of Hg in coastal environments. The mechanisms responsible for these changes in Hg partitioning may involve the increased prevalence of chloro-complex-Hg compounds with increasing salinity (Stumm and Morgan, 1996).

3 Conclusions

China launched its modern industrialization in the 1980s and became the largest consumer of coal and Hg worldwide in the last decade. It is expected that anthropogenic Hg emissions from China will strongly influence the adjacent marine environment. However, this study found that Hg concentrations in the East China Sea sediments generally ranged between 4.1–70 ng/g, values that are comparable to those of pristine environments, such as the Arctic Ocean and the continental shelf off Greenland. In addition, in comparison with continental shelves around the world, Hg concentrations in surface sediments of the ECS are relatively low. As the spatial distribution of Hg in ECS sediments exhibits a fair decrease southwards along the inner shelf, and decreases in the southeastern direction along the middle shelf, the present study suggests that only the inner shelf of ESC, especially Hangzhou Bay, is contaminated by Hg. It is estimated that Hg accumulation in the continental shelf area of the ECS ranged between 25–30 tons/yr, and approximately 70% of the total sedimentation flux was deposited in the inner shelf. Consequently, the middle shelf of the ECS is still free from Hg contamination.

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