

Mercury in the marine boundary layer and seawater of the South China Sea: Concentrations, sea/air flux, and implication for land outflow

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[1] Using R/V *Shiyan 3* as a sampling platform, measurements of gaseous elemental mercury (GEM), surface seawater total mercury (THg), methyl mercury (MeHg), and dissolved gaseous mercury (DGM) were carried out above and in the South China Sea (SCS). Measurements were collected for 2 weeks (10 to 28 August 2007) during an oceanographic expedition, which circumnavigated the northern SCS from Guangzhou (Canton), Hainan Inland, the Philippines, and back to Guangzhou. GEM concentrations over the northern SCS ranged from 1.04 to 6.75 ng m⁻³ (mean: 2.62 ng m⁻³, median: 2.24 ng m⁻³). The spatial distribution of GEM was characterized by elevated concentrations near the coastal sites adjacent to mainland China and lower concentrations at stations in the open sea. Trajectory analysis revealed that high concentrations of GEM were generally related to air masses from south China and the Indochina peninsula, while lower concentrations of GEM were related to air masses from the open sea area, reflecting great Hg emissions from south China and Indochina peninsula. The mean concentrations of THg, MeHg, and DGM in surface seawater were 1.2 ± 0.3 ng L⁻¹, 0.12 ± 0.05 ng L⁻¹, and 36.5 ± 14.9 pg L⁻¹, respectively. In general, THg and MeHg levels in the northern SCS were higher compared to results reported from most other oceans/seas. Elevated THg levels in the study area were likely attributed to significant Hg delivery from surrounding areas of the SCS primarily via atmospheric deposition and riverine input, whereas other sources like in situ production by various biotic and abiotic processes may be important for MeHg. Average sea/air flux of Hg in the study area was estimated using a gas exchange method (4.5 ± 3.4 ng m⁻² h⁻¹). This value was comparable to those from other coastal areas and generally higher than those from open sea environments, which may be attributed to the reemission of Hg previously transported to this area.

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1. Introduction

[2] Mercury (Hg) is a highly toxic pollutant that poses a serious threat to human health and wildlife [Lindqvist *et al.*, 1991; Wolfe *et al.*, 1998]. In contrast to other metals, which tend to exit in the atmosphere in the particulate phase, Hg

exists mainly (>95%) in the gaseous phase (total gaseous mercury (TGM), TGM = GEM + RGM). GEM, the predominant form of atmospheric Hg (generally constitutes more than 90% the total Hg in atmosphere), is very stable with a residence time between 6 month and 2 years [Schroeder and Munthe, 1998]. This enables Hg to undergo long-range transport and hence, becomes well-mixed on a global scale. Long-range transport followed by wet and dry deposition is the primary pathway for Hg delivery into aquatic ecosystems far from emission sources. Pristine ecosystems may be impacted with Hg contamination and subsequent bioaccumulation in the food web, causing concern about human and wildlife consumption of contaminated fish [Meili, 1991; Watras and Frost, 1989; Lindqvist *et al.*, 1991; Rask and Metsala, 1991].

[3] The ocean plays a vital role in the global cycle of Hg. According to recent global models of Hg biogeochemistry,

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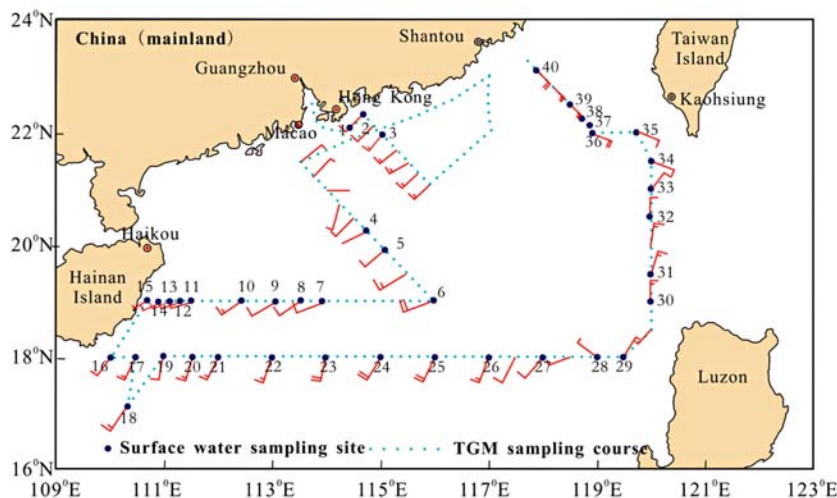


Figure 1. Map showing the sampling stations and 3 h averaged wind directions and wind speed on Beaufort scale 10 m above sea level at the stations.

the ocean releases about 1/3 of the total global Hg emissions to the atmosphere and receives about 30–70% of the global atmospheric deposition [Lamborg *et al.*, 2002; Mason and Sheu, 2002; Strode *et al.*, 2007]. The addition of Hg to oceans is mainly attributed to anthropogenic activities, which have increased since the pre-industrial era [Fitzgerald, 1995; Mason and Sheu, 2002]. Atmospheric emissions of Hg from Asia are much higher than those from other continents in global emission inventories [Pacyna *et al.*, 2006]. In China, anthropogenic Hg emissions are likely to further increase with the expansion of nonferrous production and coal combustion [Streets *et al.*, 2005; Wu *et al.*, 2006]. Higher Hg emissions result in elevated atmospheric Hg concentrations, high deposition levels and reemission fluxes over mainland China [Feng *et al.*, 2004a; Fu *et al.*, 2008a, 2008b, 2008c; Guo *et al.*, 2008], and have the potential to cause Hg pollution in the surrounding regions, even on a global scale [Friedli *et al.*, 2004; Seigneur *et al.*, 2004; Jaffe *et al.*, 1997, 1999, 2005; Travnikov, 2005; Strode *et al.*, 2008; Obrist *et al.*, 2008].

[4] Hg in seawater includes dissolved elemental mercury (Hg^0), dissolved and particulate Hg^{2+} complexes (e.g., anions, such as sulfide, thiolate, chloride) [Han *et al.*, 2004], dissolved and particulate methyl mercury (MeHg) complexes, and dimethyl mercury (DMeHg) [Morel *et al.*, 1998; Lamborg *et al.*, 2008; Strode *et al.*, 2007; Kotnik *et al.*, 2007]. In general, atmospheric deposition is identified as the dominant source of THg in open sea regions [Lamborg *et al.*, 2002; Mason and Sheu, 2002; Strode *et al.*, 2007; Sunderland and Mason, 2007]. Also, riverine input, coastal erosion, and lateral and vertical flow may be important on a regional basis [Sunderland and Mason, 2007]. On the other hand, in situ production by various biotic and abiotic processes may be very important for MeHg complexes, DMeHg, and DGM [Compeau and Bartha, 1985; Benoit *et al.*, 1999; Amyot *et al.*, 1994; Siciliano *et al.*, 2002; Sunderland *et al.*, 2009; Gray and Hines, 2009]. DGM is only a small portion of total Hg in seawater, but plays an important role in the oceanic cycle of Hg. Evasion of Hg from the ocean is driven by the formation of DGM by photoreduction and Hg^{2+} reducing bacteria in seawater [Amyot *et al.*, 1994; Siciliano

et al., 2002; Feng *et al.*, 2004b, 2008; Fantozzi *et al.*, 2007].

[5] Numerous studies with regard to the oceanic cycle of Hg have been carried out in many ocean/sea environments [Kim and Fitzgerald, 1986; Leermakers *et al.*, 1997; Wängberg *et al.*, 2001; Gårdfeldt *et al.*, 2003; Temme *et al.*, 2003; Laurier *et al.*, 2003, 2004; Narukawa *et al.*, 2006; Kotnik *et al.*, 2007; Sprovieri and Pirrone, 2008; Andersson *et al.*, 2008a; Lamborg *et al.*, 2008; Sunderland *et al.*, 2009]. However, studies on the distribution, potential sources, and transport of Hg in the coastal seas of China are limited. Previous studies in Bohai Sea and the eastern China Sea have revealed elevated THg concentrations in seawater and TGM concentrations in free and upper troposphere, which may be linked to the riverine discharge and export of Hg enriched air masses from eastern China, respectively [Han *et al.*, 2004; Friedli *et al.*, 2004]. In this study, the concentrations of THg, MeHg and DGM in seawater and GEM in the atmosphere of the northern South China Sea (SCS) are presented for the first time. In addition, based on concurrent measurement of Hg^0 in surface air and water, sea/air exchange fluxes of Hg^0 were estimated using a thin film gas exchange model with the input of ancillary meteorological data. The data set offers a unique opportunity to study the potential outflow of Hg from China, and sea/air exchange of Hg in a low latitude subtropical coastal region.

2. Experimental Setup

2.1. Site Locations

[6] The South China Sea (SCS) is located in the equatorial belt (3°S–25°N) and is the largest semienclosed marginal sea in the western tropical Pacific Ocean. The surrounding areas of the SCS are tropical-subtropical developing countries, including China, Vietnam, Thailand, Malaysia, Indonesia, Philippines, etc. During the period of 10 to 28 August 2007 a sampling campaign was carried out on R/V *Shiyan 3*, which circumnavigated the northern SCS in a zigzag line from Guangzhou to Hainan Island to Luzon (the Philippines) to Taiwan Island, and back to Guangzhou (Figure 1).

2.2. Sampling Techniques and Analysis

[7] GEM concentrations in ambient air were continuously measured during the whole cruise using an automatic Hg vapor analyzer (Tekran 2537A), which was installed in the ship laboratory. Its technique is based on the collection of TGM on gold traps, followed by thermal desorption, and detection of Hg^0 by cold vapor atomic fluorescence spectrometry ($\lambda = 253.7 \text{ nm}$). The instrument features two cartridges which trap gaseous Hg on to gold absorbents. While one cartridge is adsorbing Hg during sampling period, the other is being desorbed thermally and analyzed subsequently for TGM. The functions of each cartridge are then reversed, allowing continuous sampling of ambient air. Particulate mercury (PHg) in ambient air was removed using a 45 mm diameter Teflon filter (pore size $0.2 \mu\text{m}$). In this study, the measured TGM concentration was probably dominated by GEM because GEM was reported at a level at least 2 orders of magnitude higher than RGM in marine boundary layer [Chand *et al.*, 2008]. Moreover, most of the RGM in air was likely removed when passing the sampling tube, which was probably coated by sea salt during the cruise. Therefore, the atmospheric Hg measured in this study was referred to as GEM [Radke *et al.*, 2007]. The sampling interval was 5 min with a sampling flow rate of 1.0 L min^{-1} during the entire campaign. The precision of the analyzer was determined to less than 2%, and detection limit is less than 0.1 ng m^{-3} . Data quality was checked via periodic internal recalibrations every 25 h. To diminish the contamination from exhaust plume of the ship as possible, we installed the sampling inlet at the front deck 15 m above the sea surface using a heated Teflon sampling tube (1/4 inch in diameter and 15 m in length) together with a Teflon tube (1/4 inch in diameter and 20 m in length). Unfortunately, during 14–15 August and at anchor stations, some of the air samples were contaminated by the exhaust plume of the ship. Therefore, GEM data during these periods were all screened out from the data set, and only GEM values observed during the ship traveling were used for the current study.

[8] Sampling and analysis of DGM in seawater were performed using the method described by Gårdfeldt *et al.* [2002], and briefly summarized below. Seawater was sampled at a depth of 10–50 cm below the surface by using a 1 L volume Teflon vial and immediately transferred into a 1.5 L volume borosilicate glass bottle. The DGM in the water sample was extracted by introducing a stream of Hg-free argon at the flow rate of $350\text{--}400 \text{ mL min}^{-1}$ for 60 min [Gårdfeldt *et al.*, 2002]. The extracted gaseous Hg was collected on a gold trap heated to $\sim 50^\circ\text{C}$ during the extraction to avoid condensation of water vapor. The gold trap was analyzed with the standard dual amalgamation and CVAFS detection technique [Brosset, 1987]. The analytical blank was determined on board by extracting MQ water for DGM as described above. A detection limit of 3.1 pg L^{-1} ($n = 6$) was obtained on the basis of 3 times the standard deviation of blank. The borosilicate glass bubble bottle was kept clean by continuously purging it with a small stream of Hg-free Argon gas.

[9] To study the THg and MeHg distributions in the north SCS, surface seawater samples were collected using a 1 L volume Teflon vial at 34 anchor stations. Samples were then transferred carefully to precleaned borosilicate glass bottles

(volume: 100 mL) and preserved by adding trace-metal-grade HCl (to 5% of total sample volume). Borosilicate glass bottles with samples were individually sealed into three successive polyethylene bags and stored in a refrigerator (4°C) during the whole cruise. To ensure clean operation, polyethylene gloves were used throughout the field operation. After the cruise, samples were transported soon to the lab and analyzed as soon as possible to prevent the risk of cross contamination and loss of the Hg content (both are analyzed less than 28 days after sampling). THg was analyzed by BrCl oxidation followed by SnCl_2 reduction, and dual amalgamation combined with CVAFS detection [U.S. Environmental Protection Agency (EPA), 1999], while MeHg was determined by using distillation, aqueous phase ethylation and GC separation followed by pyrolysis and GC-CVAFS detection [EPA, 2001]. The detection limits of THg and MeHg were 0.1 ng L^{-1} and 0.009 ng L^{-1} , respectively, which were determined by three times the standard deviation of blanks. Quality assurance and quality control were conducted using duplicates, matrix spikes. The relative percentage difference was $<14\%$ in duplicate samples (6 pairs) and recoveries on matrix spikes of THg and MeHg in samples were in the range of 97–111% and 89–108%, respectively. Moreover, to evaluate the influence of riverine discharge on the distribution of Hg in the SCS, stream water samples were collected at six sites in Zhongshan, Hengli, Nansha, Huangge, Humen, and Huangpu in March 2009.

[10] Meteorological and basic seawater parameters (such as wind speed and surface water temperature) were measured using the ship's monitoring system. Wind direction 10 m above sea surface at each anchor station was determined from Archived Meteorology data available in the Air Resource Laboratory (ARL), National Oceanic and Atmospheric Administration (NOAA) (G. D. Rolph, Real-time Environmental Applications and Display System (READY), 2003, <http://www.arl.noaa.gov/ready/hysplit4.html>). In order to identify the long-range transport of Hg and its precursors, 3 day back trajectories were calculated using Gridded Meteorological Data combined with the Geographic Information System (GIS) based software, Trajstat, from HYSPLIT [Wang *et al.*, 2009]. The Global Data Assimilation System (GDAS) meteorological data archives of the ARL, which are available online at <ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/>, were used as the input. In this study, we calculated 13 back trajectories, which ended at 13 stations with an arrival height of 500 m above the sea surface, and the start time of each back trajectory was matched to the atmospheric GEM sampling time at each station.

2.3. Estimations of Sea/Air Hg Flux

[11] The sea/air flux of Hg was estimated using a thin film gas exchange model developed by Wanninkhof [1992]. This method requires measurements of DGM, GEM, surface water temperature (T), and wind speed normalized to 10 m, and has been described in several studies [e.g., Wängberg *et al.*, 2001; Gårdfeldt *et al.*, 2003]. The degree of DGM supersaturation, SS, was calculated using

$$\text{SS} = \text{DGM} \times H'(T)/\text{GEM}-1 \quad (1)$$

$$H'(T) = \exp(-2404.3/T + 6.92), \quad (2)$$

Table 1. Summary of Atmospheric GEM, Surface Seawater THg, MeHg, DGM Concentrations, and Sea/Air Flux in Northern SCS

Site	Position		Date and Time (Year/Month/Day and Local Time)	THg (ng L ⁻¹)	MeHg (ng L ⁻¹)	DGM (pg L ⁻¹)	GEM (ng m ⁻³)	TW (°C)	Wind Speed U10 (m s ⁻¹)	Supersaturation (%)	Flux (ng m ⁻² h ⁻¹)
	Longitude	Latitude									
1	114.30°	22.06°	2007/8/11 1503			35.0	5.23	27.2	7.4	126	3.3
2	114.75°	22.25°	2007/8/13 1530			26.7	3.32	27.4	8.6	173	3.8
3	114.93°	22.08°	2007/8/13 1820	1.08		29.3	2.44	27.1	10.0	305	6.4
4	114.90°	20.06°	2007/8/13 0923	1.24	0.187						
5	115.10°	19.90°	2007/8/18 0200	1.58	0.082	58.6	4.08	29.0	5.2	409	4.4
6	116.00°	19.00°	2007/8/18 0900	1.01	0.15	22.4	2.11	29.0	14.1	276	9.6
7	113.91°	19.00°	2007/8/18 1845	0.95	0.163	49.4	2.01	29.5	7.7	782	8.3
8	113.5°	19.00°	2007/8/18 2114	2.08	0.134	38.3	1.66	29.4	6.8	726	5.1
9	113.07°	19.00°	2007/8/18 2344	1.33	0.134	33.3	1.63	29.4	7.3	632	4.9
10	112.41°	19.00°	2007/8/19 0313	1.34	0.187	37.4	2.06	29.1	9.4	545	8.6
11	111.50°	19.00°	2007/8/19 0844	1.23		44.6	4.00	28.5	5.9	290	3.8
12	111.30°	19.00°	2007/8/19 1017			34.0	3.33	28.0	6.0	252	2.8
13	111.10°	19.00°	2007/8/19 1200	0.98	0.154	42.7	4.71	27.8	5.3	211	2.7
14	110.90°	19.00°	2007/8/19 1414	1.31		45.9	4.01	28.3	4.9	298	2.8
15	110.70°	19.00°	2007/8/19 1550	2.33	0.117	80.1	3.88	28.2	3.6	616	3.3
16	110.00°	18.00°	2007/8/19 2201	1.25	0.119	77.8	2.26	28.7	8.3	1110	15.3
17	110.50°	18.00°	2007/8/20 0131	1.27	0.101	77.2	1.94	28.7	7.6	1299	13.1
18	110.35°	17.00°	2007/8/20 1638	1.05	0.172	32.2	3.26	29.7	3.8	256	1.2
19	111.00°	18.00°	2007/8/20 2132	1.16	0.089	27.3	1.55	28.6	5.7	5.18	2.5
20	111.51°	18.00°	2007/8/21 0806	1.51	0.114	31.4	1.57	29.4	7.5	616	4.9
21	112.00°	18.00°	2007/8/21 1048	1.21	0.127	24.8	1.82	29.0	8.8	383	4.7
22	113.00°	18.00°	2007/8/21 1819	1.12	0.057	32.2	1.75	29.5	9.1	561	7.1
23	114.00°	18.00°	2007/8/21 0109	1.04	0.096	28.0	1.46	28.9	9.6	578	6.7
24	115.00°	18.00°	2007/8/22 0632	0.94	0.098	46.6	1.43	29.2	8.8	1061	10.3
25	116.00°	18.00°	2007/8/22 1545	0.82	0.063	26.2	1.69	29.3	6.3	454	2.8
26	117.00°	18.00°	2007/8/23 0103	1.09	0.087	32.5	1.35	29.0	3.3	753	1.2
27	118.00°	18.00°	2007/8/23 1102	1.02		43.9	1.62	29.5	3.8	873	2.1
28	119.00°	18.00°	2007/8/23 2150	1.32	0.112	36.7	1.41	29.4	2.4	832	0.8
29	119.50°	18.00°	2007/8/24 0748			30.7	1.73	29.3	1.9	534	0.4
30	120.00°	19.00°	2007/8/24 1530	1.62	0.127	23.8	1.5	29.5	7.7	470	3.7
31	120.00°	19.50°	2007/8/24 2203	1.04	0.045	28.1	1.62	29.1	8.9	516	5.8
32	120.00°	20.50°	2007/8/25 0820	0.88	0.087	22.0	1.57	29.4	5.9	402	2.0
33	120.00°	21.00°	2007/8/25 1132	0.93	0.047	28.1	1.84	29.2	4.5	444	1.6
34	120.00°	21.50°	2007/8/25 1821	1.26	0.055	38.9	1.55	29.4	1.0	799	0.2
35	119.50°	22.00°	2007/8/26 0034	1.99	0.139	29.9	1.5	29.0	4.9	606	2.1
36	119.00°	22.00°	2007/8/26 1000	0.96	0.076	29.5	1.63	28.9	6.6	540	3.5
37	118.75°	22.25°	2007/8/26 1825	1.31	0.199	22.0	1.67	28.7	10.3	363	5.5
38	118.50°	22.50°	2007/8/26 2038	0.75	0.083	21.6	1.54	27.9	9.1	383	4.3
39	118.30°	22.70°	2007/8/26 2231			20.4	2.1	26.6	9.5	223	3.7
40	117.70°	23.30°	2007/8/27 0724	1.15	0.217	35.9	1.58	25.3	4.1	630	1.7

where DGM is the concentration of Hg⁰ in seawater, GEM is the concentration of Hg⁰ in ambient air above sea surface, H'(T) is the dimensionless partitioning coefficient for Hg⁰ between water and air as known as the Henry's Law constant [Andersson *et al.*, 2008b], and T is the water temperature in K.

[12] The sea/air exchange flux of Hg, F (ng m⁻² h⁻¹), was calculated using

$$F = K_w \times (DGM - GEM/H'(T)), \quad (3)$$

where K_w is the gas transfer velocity of a species in the water/air surface (cm h⁻¹) and was calculated according to [Nightingale *et al.*, 2000]

$$K_w = (0.222 * U_{10}^2 + 0.333 * U_{10}) (Sc_{Hg}/600)^{-0.5}, \quad (4)$$

where U₁₀ is the wind speed at 10 m height, and Sc_{Hg}, the Schmidt number for Hg, is defined as

$$Sc_{Hg} = \nu/D_{Hg}, \quad (5)$$

where ν is the kinematic viscosity (cm² s⁻¹) of seawater calculated using the method of Wanninkhof [1992]. In-tum D_{Hg} is the Hg diffusion coefficient (cm² s⁻¹) in seawater, which was calculated by molecular dynamics simulation, as described by Kuss *et al.* [2009].

3. Results and Discussion

3.1. Atmospheric GEM in the Northern SCS

[13] The summary of atmospheric GEM, surface seawater THg, MeHg, and DGM concentrations and sea/air Hg fluxes in the northern SCS are presented in Table 1. GEM concentrations in the northern SCS ranged from 1.04 to 6.75 ng m⁻³ with a mean value of 2.62 ± 1.13 ng m⁻³ (median: 2.24 ng m⁻³). GEM concentrations in northern SCS are higher compared to the results from other oceans and seas, such as Mediterranean Sea (1.20–2.37 ng m⁻³) [Gårdfeldt *et al.*, 2003], Adriatic Sea (1.6 ng m⁻³) [Sprovieri and Pirrone, 2008], Baltic sea (1.70 ng m⁻³) [Wängberg *et al.*, 2001], North Sea (1.2–1.5 ng m⁻³) [Leermakers *et al.*, 1997], Arctic Ocean (1.7 ng m⁻³) [Andersson *et al.*, 2008a], Atlantic

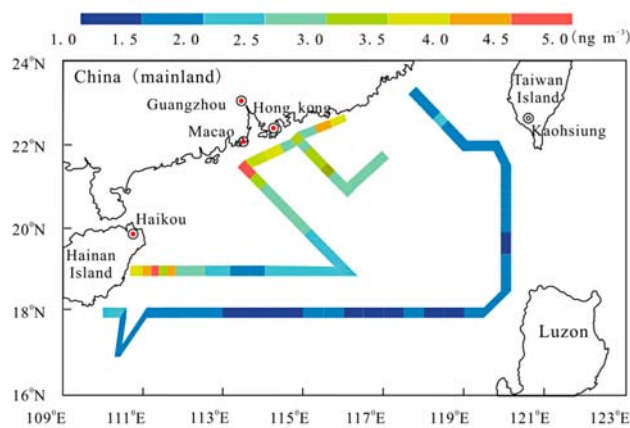


Figure 2. Distribution of average GEM concentrations in ambient air above the northern SCS.

Ocean ($1.00\text{--}2.25\text{ ng m}^{-3}$) [Temme et al., 2003], but they agree well with observations made in the North Pacific Ocean between Japan and Hawaii (2.5 ng m^{-3}) [Laurier et al., 2003].

[14] GEM concentrations showed a clear spatial trend in the northern SCS (Figure 2). Concentrations of GEM were generally higher at sites close to mainland China compared to sites in the open sea. These results are in agreement with recent model studies that showed decreasing TGM concentrations between China and the North Pacific Ocean [Shia et al., 1999; Seigneur et al., 2004; Selin et al., 2007; Strode et al., 2007]. This supports the hypothesis that China constitutes a significant part in the global atmospheric Hg emission inventory. However, in comparison with atmospheric TGM levels reported recently [Feng et al., 2004a; Wang et al., 2007; Fu et al., 2008a; Wan et al., 2009a], these models largely underestimated Hg concentrations in China. This was likely due to the effects of local or regional emissions in those sampling sites, or underestimations of Chinese anthropogenic and natural Hg emissions by model studies.

[15] In the northern SCS, GEM concentrations showed a pronounced dependence on the air mass movement patterns during the whole cruise, which gives insight into the sources influencing the northern SCS. Figure 3 shows 3 day back trajectories arriving at the northern SCS during the cruise. To identify Hg source regions surrounding the SCS, we grouped the air masses into the following three major

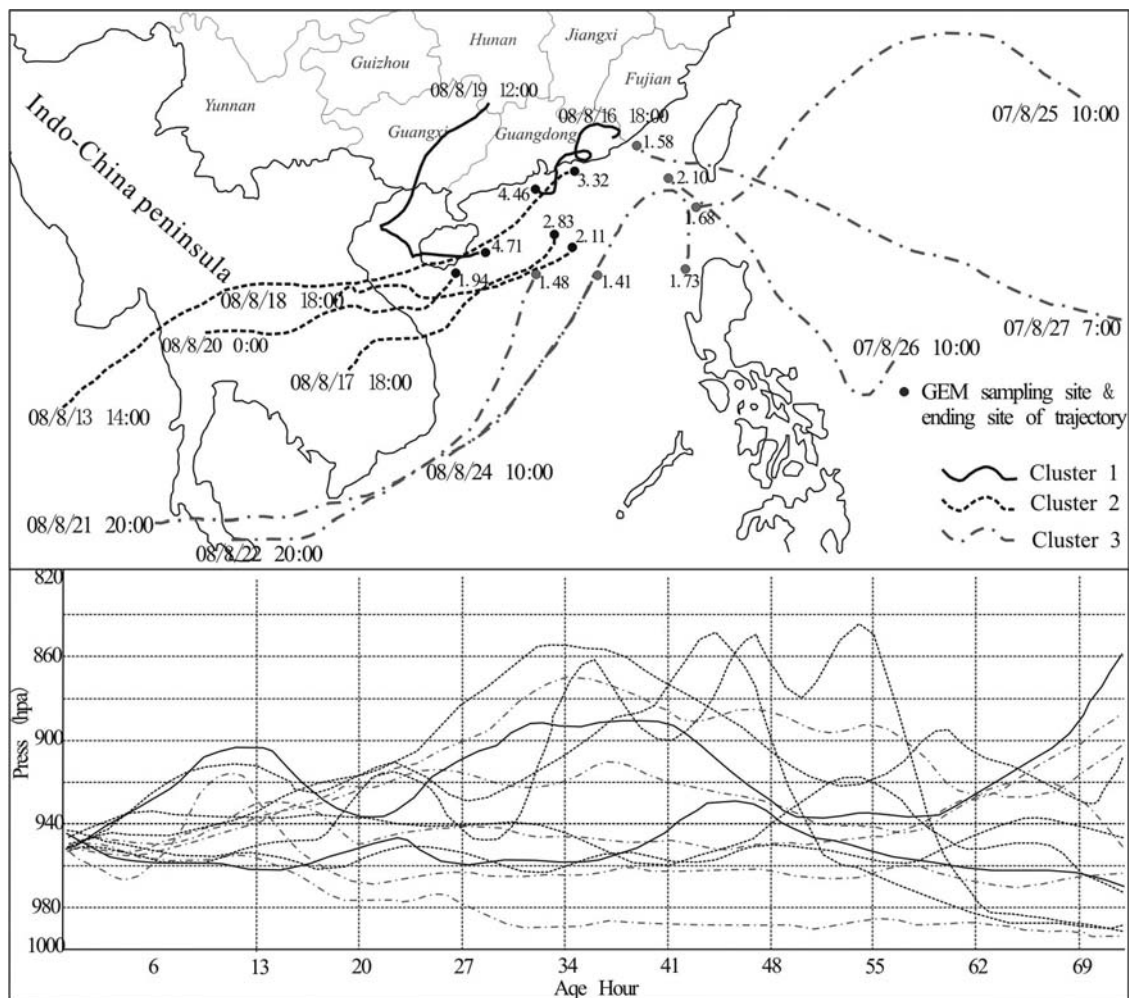


Figure 3. The 72 h back trajectories, clusters, and GEM concentrations during the cruise (time shows the start time of back trajectory and sampling time of atmospheric GEM at each station).

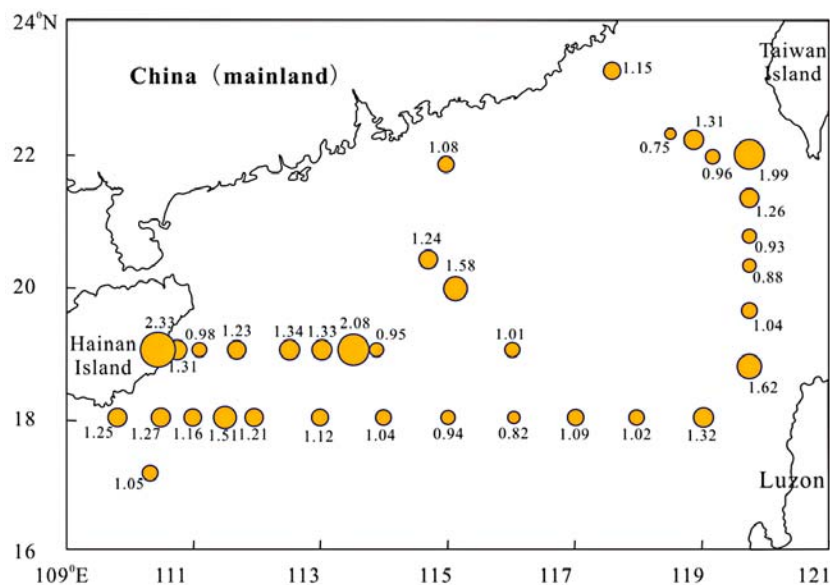


Figure 4. Surface water THg concentrations at each of the station in the northern SCS (unit in ng L^{-1}).

clusters (Figure 3). Cluster 1 consists of air masses originating from the continental inland area, passing over Guangzhou and Guangxi provinces in south China. Cluster 2 shows air masses passing over the Indochina peninsula. Air masses in cluster 3 originated from the open sea.

[16] For the three types of air masses, cluster 1 was related to the highest GEM concentrations. For example, the highest GEM concentration (4.71 ng m^{-3}) was observed in the air mass passing over the Guangxi province and Hainan Island. This reveals that Hg emissions from south China constituted an important source of atmospheric Hg in the northern SCS. This is in agreement with model results, which hypothesized Hg emissions from China seriously impacted the Hg distribution in surrounding regions downwind via long-range transport [Friedli *et al.*, 2004; Seigneur *et al.*, 2004; Jaffe *et al.*, 1997, 1999, 2005; Travnikov, 2005; Strode *et al.*, 2008; Obrist *et al.*, 2008]. Air masses in cluster 2 also exhibited elevated GEM concentrations compared to the global background values ($1.5\text{--}2.0 \text{ ng m}^{-3}$) [Valente *et al.*, 2007], with a range of $1.94\text{--}3.32 \text{ ng m}^{-3}$, indicating the export of atmospheric Hg from the Indochina peninsula. By comparison, Hg emissions in the Indochina peninsula were likely lower than emissions from south China, as air masses passing over this area exhibited much lower GEM concentrations. Last, GEM concentrations in cluster 3 were generally lower than 2.0 ng m^{-3} , which were comparable to global background values ($1.5\text{--}2.0 \text{ ng m}^{-3}$) [Valente *et al.*, 2007]. This is probably because air masses in cluster 3 originated from open sea areas.

[17] In general, air masses originated from open sea areas and Indochina peninsula were prevalent during the whole cruise, which was controlled by the southeast summer monsoon. This implies that the northern SCS was less affected by the export of Hg from south China in the summer and might have lower atmospheric GEM concentrations compared to other seasons. However, air masses originating from China will probably increase during the cold season in

the SCS because of prevalent Asian winter monsoon in this period. At the same time, winter elevated TGM concentrations were reported in mainland China [Feng *et al.*, 2004a; Wang *et al.*, 2007; Fu *et al.*, 2008a]. Therefore, we speculate the export of Hg from China to SCS will increase during the cold season.

3.2. THg and MeHg Concentrations in Surface Seawater of SCS

[18] The mean THg and MeHg concentrations in the surface seawater of the northern SCS were $1.2 \pm 0.3 \text{ ng L}^{-1}$ (median = 1.2 ng L^{-1} , ranged from 0.8 to 2.3 ng L^{-1}) and $0.12 \pm 0.05 \text{ ng L}^{-1}$ (median = 0.11 ng L^{-1} , ranged from 0.05 to 0.22 ng L^{-1}), respectively (Figures 4 and 5). The average fraction of Hg present as MeHg was 10%, which was much lower compared to observations from the Mediterranean Sea [Kotnik *et al.*, 2007]. THg and MeHg concentrations in the northern SCS were much higher than those reported in open sea environments, such as the Mediterranean Sea (THg: 0.26 ng L^{-1} , MeHg: $0.03\text{--}0.06 \text{ ng L}^{-1}$) [Kotnik *et al.*, 2007], the North Atlantic (THg: 0.48 ng L^{-1}) [Mason *et al.*, 1998], the west Black Sea (THg: 0.56 ng L^{-1} , MeHg: $< 0.02 \text{ ng L}^{-1}$) [Lamborg *et al.*, 2008], and the North Pacific (THg: 0.23 ng L^{-1}) [Laurier *et al.*, 2004]. However, levels of THg and MeHg in the northern SCS were comparable to values reported in coastal environments, such as the Gulf of Trieste (THg: $0.18\text{--}4.9 \text{ ng L}^{-1}$) [Faganeli *et al.*, 2003], Long Island Sound (THg: $0.46\text{--}3.98 \text{ ng L}^{-1}$) [Rolffhus and Fitzgerald, 2001], Coast of Black Sea (THg: $0.32\text{--}2.0 \text{ ng L}^{-1}$, MeHg: $0.01\text{--}0.2 \text{ ng L}^{-1}$) [Lamborg *et al.*, 2008], and Chesapeake Bay (THg: 1.32 ng L^{-1}) [Lawson *et al.*, 2001]. This suggested the SCS was probably impacted by outflow of Hg pollution originating from terrestrial sources.

[19] Generally, pathways for Hg entering the marine environment include atmospheric deposition, riverine input, as well as lateral and vertical seawater flow [Sunderland and Mason, 2007]. As discussed in section 3.1, south China and

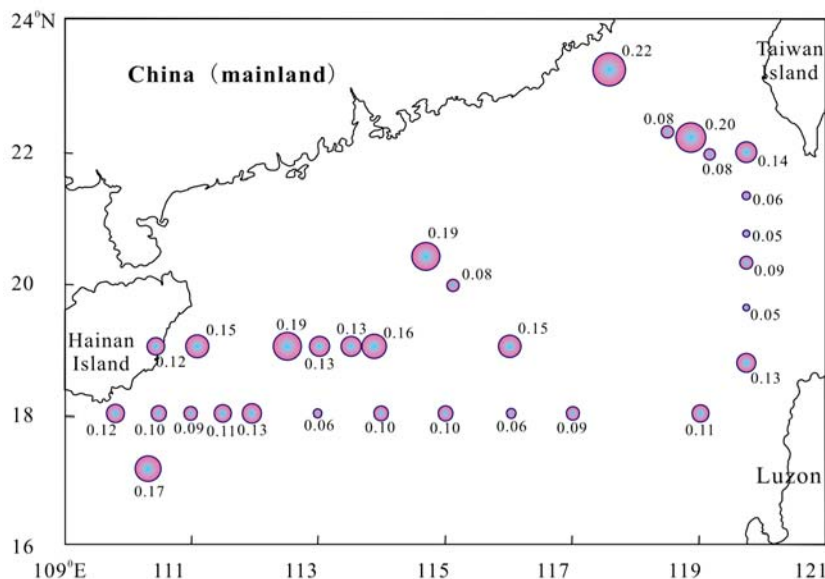


Figure 5. Surface water MeHg concentrations at each of the station in the northern SCS (unit in ng L^{-1}).

the Indochina peninsula were important atmospheric Hg source regions. Elevated emissions from these areas probably resulted in significant Hg delivery into the SCS via long-range atmospheric transport followed by wet and dry deposition. To estimate the wet and dry deposition of Hg in the SCS, mean THg and MeHg concentrations in precipitation of the SCS were assumed to be 10.3 and 0.10 ng L^{-1} , respectively, which were predicted on the basis of mean values of THg (4 – 14.3 ng L^{-1} , $n = 4$) and MeHg (0.04 – 0.16 ng L^{-1} , $n = 3$) recently reported from remote areas of China [Fu et al., 2008b, 2009, 2010; Wan et al., 2009b]. These values are much higher compared to other oceans [Gill and Fitzgerald, 1987], but maybe within the range of possibility. In the Pearl River Delta of China, reported values for wet deposition fluxes of heavy metals (e.g., Pb, Zn, Cr, Cu) were more than fivefold higher compared to those reported in North America and Europe [Wong et al., 2003], and therefore higher estimates for THg and MeHg concentrations may be reasonable due to higher anthropogenic Hg emissions in south China and the Indochina peninsula. Based on the values for THg and MeHg, the annual deposition fluxes of THg and MeHg to the SCS were approximately 71 and 0.7 tons yr^{-1} assuming the mean precipitation depth in the SCS to be 2000 mm [Chen et al., 2005] and area of the SCS to be 3.5×10^{12} m^2 , respectively. Dry deposition of THg in the SCS was calculated using the atmospheric RGM and PHg concentrations (RGM: 4.5 pg m^{-3} , PHg: 3.0 pg m^{-3}) [Jaffe et al., 2005; Chand et al., 2008] observed at Okinawa Island near the SCS and the reported dry deposition velocities for these Hg species. Dry deposition velocity of RGM was set at a relative high value of 4 cm s^{-1} because of rapid uptake by sea salt aerosols followed by deposition [Selin et al., 2007], and dry deposition velocity of PHg was set at 0.25 cm s^{-1} , which was generally accepted in model studies [Shia et al., 1999]. Therefore, the dry deposition flux of THg in the SCS was calculated to be 21 tons yr^{-1} , which was much lower than wet deposition.

[20] Riverine input is also an important source of THg and MeHg in the SCS. The three largest rivers flowing into the SCS include the Mekong River, Zhujiang River, and Red River, and the annual discharges of these three rivers are 463×10^9 m^3 , 336×10^9 m^3 , and 57×10^9 m^3 , respectively. We collected stream water samples from Pearl River Delta and found THg and MeHg concentrations were highly elevated (mean: THg: 40 ng L^{-1} ; MeHg: 0.21 ng L^{-1} , $n = 6$). Using these data, the riverine input fluxes of THg and MeHg in the SCS were estimated to be 34 and 0.18 tons yr^{-1} , respectively.

[21] Based on these calculations, inputs of THg and MeHg into the SCS via atmospheric deposition and riverine delivery were estimated to be 126 and 0.88 tons per year , respectively. The high inputs were probably caused by anthropogenic Hg emissions and wastewater discharges from the surrounding mainland. For THg, atmospheric deposition and riverine delivery were probably the main sources to the SCS. One of the reasons is that the THg concentrations in the SCS were much higher than those in western Pacific and Indian Ocean which were probably net sinks of THg for the SCS. Besides, upwelling and diffusive transport was suggested to be the sink of THg in Pacific and Indian Ocean [Sunderland and Mason, 2007], which might indicate the upwelling and diffusive of THg from sediment was not the source of THg in the SCS. Atmospheric deposition was considered the major pathway for THg entering the SCS, and this is consistent with previous studies [Mason and Sheu, 2002; Strode et al., 2007; Sunderland et al., 2009]. However, riverine delivery was also an important source of THg to the SCS, which constituted nearly a quarter of total input, and this supports the hypothesis that riverine input can be important on a regional basis [Sunderland and Mason, 2007].

[22] Unlike THg, atmospheric deposition and riverine delivery may be not the primary sources of MeHg to the SCS. The percentage of MeHg input (0.7%) via atmospheric

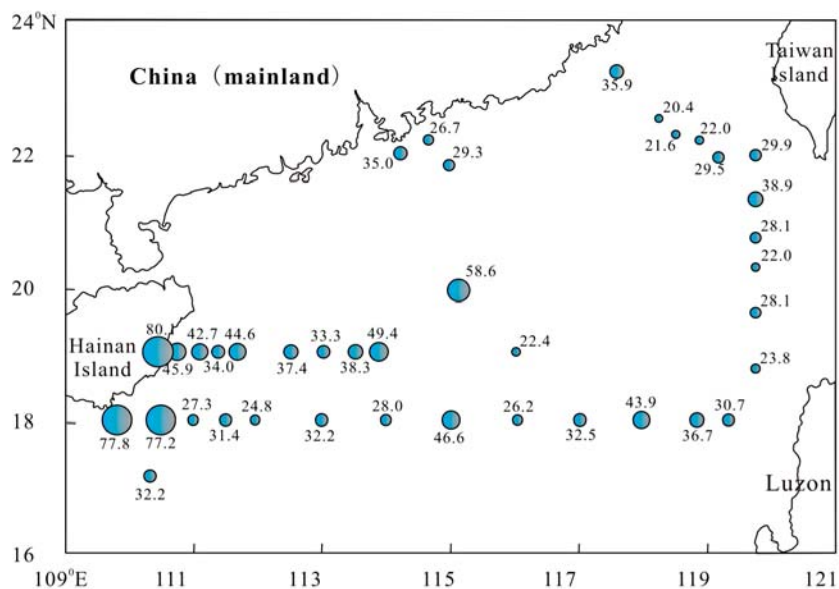


Figure 6. Surface water DGM concentrations at each of the station in the northern SCS (unit in pg L^{-1}).

deposition and riverine delivery was considerably lower compared to the average fraction of Hg present as MeHg (10%) in surface seawater. As suggested by previous studies [Mason and Sullivan, 1999; Sunderland *et al.*, 2009], other sources, such as in situ production of MeHg in deeper and low-oxygen ocean waters followed by upwelling and diffusive transport, might be the dominant source of MeHg to the SCS.

3.3. DGM Concentrations and Sea/Air Hg Fluxes

[23] DGM concentrations in the northern SCS varied from 20.4 to 80.1 pg L^{-1} with an average concentration of $36.5 \pm 14.9 \text{ pg L}^{-1}$ (Table 1). Unlike elevated THg concentrations, DGM concentrations in the study area were comparable to those observed from open sea environments, such as the Arctic ocean ($44 \pm 22 \text{ pg L}^{-1}$) [Andersson *et al.*, 2008a]; the east Mediterranean Sea (22.0–54.2 pg L^{-1}) [Gårdfeldt *et al.*, 2003], and slightly higher than those observed in the Baltic Sea (14–22 pg L^{-1}) [Wängberg *et al.*, 2001], the equatorial Pacific Ocean (6–45 pg L^{-1}) [Kim and Fitzgerald, 1986], the Tyrrhenian Sea (17–26 pg L^{-1}) [Gårdfeldt *et al.*, 2003], and the Irish west coast (13–40 pg L^{-1}) [Gårdfeldt *et al.*, 2003]. However, the spatial variability of DGM concentra-

tions in the northern SCS was similar to THg, with higher DGM concentrations observed close to Hainan Island and lower concentrations observed in the open sea (Figure 6). Accordingly, we found that DGM concentrations correlated well with THg concentrations ($R = 0.38$, $P = 0.03$). Therefore, we speculate that DGM in the northern SCS was not directly driven from mainland output like THg, but elevated THg concentrations in the northern SCS might be favor DGM production.

[24] The calculated sea/air Hg fluxes are presented in Table 1, including GEM concentrations, water temperature, wind speed, and degree of DGM supersaturation at each site. Sea/air exchange fluxes of Hg in the northern SCS varied from 0.2 to 15.3 $\text{ng m}^{-2} \text{ h}^{-1}$ with a mean value of $4.5 \pm 3.4 \text{ ng m}^{-2} \text{ h}^{-1}$. Table 2 shows the comparison of sea/air Hg exchange fluxes estimated using gas exchange method between the study area and some other regions. Generally, the evasion flux of Hg in the northern SCS was lower compared to Tokyo Bay and the east Mediterranean, but higher than results from other oceans. However, it should be noted that large uncertainties are associated with this comparison. In this study, the calculation of Hg flux was based on more recent determinations of Henry's Law constant and

Table 2. Sea/Air Hg Fluxes Estimated Using Gas Exchange Model in Northern SCS and Values Reported in the Literature

Location	Time	Latitude	Longitude	Flux ($\text{ng m}^{-2} \text{ h}^{-1}$)	Reference
Baltic Sea	2–15 Jul 1997	54°N–57°N	14°W–21°W	1.6	Wängberg <i>et al.</i> [2001]
	2–15 Mar 1998	54°W–57°W	14°W–21°W	0.8	Wängberg <i>et al.</i> [2001]
Irish west coast	31 Aug to 7 Sep 1999	59.2°N	9.5°W	2.7	Gårdfeldt <i>et al.</i> [2003]
East Mediterranean	17–23 Jul 2000	32°N–37°N	17°E–29°E	7.9	Gårdfeldt <i>et al.</i> [2003]
West Mediterranean	2–3 Aug 2000	38°N–42°N	6°E–9°E	2.5	Gårdfeldt <i>et al.</i> [2003]
Tyrrhenian Sea	29 Jul to 8 Aug 2000	29°N–42°N	8°E–14°E	4.2	Gårdfeldt <i>et al.</i> [2003]
Arctic Ocean	13 Jul to 25 Sep 2005	63°N–90°N	60°E–180°W	2.4	Andersson <i>et al.</i> [2008a]
Tokyo Bay	Dec 2003; Oct 2004; Jan 2005	35°N–36°N	139°E–140°E	5.8	Narukawa <i>et al.</i> [2006]
North Sea	May and Oct 1996	54.5°N	4.8°E	0.2–4.6	Baeyens and Leermakers, 1998
Equatorial Pacific Ocean	8 Jun to 3 Jul 1984	4.5°S–4.5° N	93°W–155°W	3.3	Kim and Fitzgerald [1986]
Northern SCS	Aug 2007	16°N–24°N	109°E–123°E	4.5	This work

the diffusion coefficient of Hg [Andersson *et al.*, 2008b, Kuss *et al.*, 2009], which decreased the estimated flux by 40% [Narukawa *et al.*, 2006; Gårdfeldt *et al.*, 2003]. In this case, our result is comparable or higher compared to Hg fluxes estimated from other coastal areas and the open sea environment, and this was probably driven by the great delivery of Hg into the SCS from surrounding areas, which caused elevated Hg reemissions.

[25] Using the data obtained from this study, annual Hg evasion from the SCS was calculated based on the assumption there was no seasonal variation in Hg emission fluxes from tropical oceans [Strode *et al.*, 2007]. The annual emission flux of Hg from the SCS was 140 tons yr⁻¹ assuming the area of the SCS was 3.5×10^{12} m², which represented about 25% of Hg emissions from anthropogenic activities in China [Streets *et al.*, 2005]. In general, it is believed that reemission of previously deposited Hg is an important component of ocean Hg emissions [Mason and Sheu, 2002; Seigneur *et al.*, 2004; Selin *et al.*, 2007; Strode *et al.*, 2007]. As discussed in section 3.2, approximately 126 tons of THg entered the SCS annually via atmospheric deposition and riverine delivery, and this was probably the most important reason for the high Hg emission flux in the northern SCS. However, it appears that ocean emission of Hg was larger than the total THg input via atmospheric deposition and riverine input, and this may be attributed to uncertainty in these estimations. First, since there are no data on atmospheric deposition flux and riverine output flux in the study area, the input of Hg to the SCS may be overestimated or underestimated. In addition, there are substantial uncertainties in the estimation of sea/air flux of Hg in various models. Seasonal cycling and spatial distribution of Hg fluxes were not considered in this study, and this probably led to an overestimation of the Hg emission flux. The sampling campaign was carried out during the summer when water temperature was higher, which likely led to overestimation of Hg evasion. Moreover, the thin film Hg exchange model should be further developed, since there are still large discrepancies between model results and measured fluxes [O'Driscoll *et al.*, 2003]. It is obvious that more work is needed to evaluate the Hg dynamics in the SCS and its influence in the Hg cycling on the global scale. Future research should include: (1) long-term measurements of atmospheric Hg species in the SCS, which will be helpful for the study of long-range atmospheric transport, atmospheric physical-chemical process in the marine boundary layer, and regional Hg emission inventories; (2) sea/air exchange of Hg in the SCS; (3) distribution of Hg species, dynamics of Hg in the SCS, and (4) the SCS as a source of Hg to other regions (e.g., western Pacific Ocean).

4. Conclusion

[26] For the first time, the concentrations of atmospheric GEM, THg, MeHg, and DGM in surface seawater were investigated in the northern SCS. In addition, sea/air Hg fluxes were estimated using atmospheric GEM and surface seawater DGM concentrations and the gas exchange method. Elevated GEM concentrations (mean: 2.62 ng m⁻³) and the distinct spatial distribution of GEM suggested export of Hg from China and the Indochina peninsula was an important source of atmospheric Hg to the northern SCS.

THg and MeHg concentrations in the SCS were higher compared to other oceans, with mean values of 1.2 ng L⁻¹ and 0.12 ng L⁻¹, respectively. Higher concentrations of THg and MeHg were observed at sites close to Hainan Island and Taiwan Straits, respectively. Our preliminary assessment suggested great delivery of THg from surrounding mainland via atmospheric deposition and riverine input played an important role in the SCS. Hg enriched waters in the SCS might act as an important source of Hg to the western Pacific Ocean and Indian Ocean via lateral exchange of water masses. The concentrations of DGM in the study area ranged from 20.4 to 80.1 pg L⁻¹ with an overall average concentration of 36.5 pg L⁻¹. Estimated sea/air Hg fluxes showed a mean emission flux of 4.5 ng m⁻² h⁻¹, which was comparable or higher than those reported from other Ocean/Sea environments. High Hg emission in the SCS might be attributed to reemission of Hg previously transported to this area.

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