Air–sea exchange of gaseous mercury in the tropical coast (Luhuitou fringing reef) of the South China Sea, the Hainan Island, China

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Abstract The air–sea exchange of gaseous mercury (mainly Hg(0)) in the tropical ocean is an important part of the global Hg biogeochemical cycle, but the related investigations are limited. In this study, we simultaneously measured Hg(0) concentrations in surface waters and overlaying air in the tropical coast (Luhuitou fringing reef) of the South China Sea (SCS), Hainan Island, China, for 13 days on January–February 2015. The purpose of this study was to explore the temporal variation of Hg(0) concentrations in air and surface waters, estimate the air–sea Hg(0) flux, and reveal their influencing factors in the tropical coastal environment. The mean concentrations ($\pm$ SD) of Hg(0) in air and total Hg (THg) in waters were $2.34 \pm 0.26 \text{ ng m}^{-3}$ and $1.40 \pm 0.48 \text{ ng L}^{-1}$, respectively. Both Hg(0) concentrations in waters ($53.7 \pm 18.8 \text{ pg L}^{-1}$) and Hg(0)/THg ratios (3.8 %) in this study were significantly higher than those of the open water of the SCS in winter. Hg(0) in waters usually exhibited a clear diurnal variation with increased concentrations in daytime and decreased concentrations in nighttime, especially in cloudless days with low wind speed. Linear regression analysis suggested that Hg(0) concentrations in waters were positively and significantly correlated to the photosynthetically active radiation (PAR) ($R^2 = 0.42$, $p < 0.001$). Surface waters were always supersaturated with Hg(0) compared to air (the degree of saturation, 2.46 to 13.87), indicating that the surface water was one of the atmospheric Hg(0) sources. The air–sea Hg(0) fluxes were

estimated to be $1.73 \pm 1.25 \text{ ng m}^{-2} \text{ h}^{-1}$ with a large range between 0.01 and $6.06 \text{ ng m}^{-2} \text{ h}^{-1}$. The high variation of Hg(0) fluxes was mainly attributed to the greatly temporal variation of wind speed.

Keywords Elemental Hg · Air–sea Hg exchange · Emission flux · Tropical coast · South China Sea · China

Introduction

The ocean plays an important role in the global mercury (Hg) cycle because it is both a source and a sink for atmospheric Hg (Mason et al. 2012). Over the past three decades, many studies have been performed to investigate the oceanic Hg biogeochemistry (Fitzgerald et al. 2007 and references therein). Studies suggest that the reduction of divalent Hg (Hg(II)) to elemental Hg (Hg(0)) and oxidation of Hg(0) to Hg(II) (i.e., Hg redox chemistry) in waters can occur simultaneously and is regulated by both photochemical process and microbial process and the photoreduction of Hg(II) is the primary pathway for the production of Hg(0) (Amyot et al. 1997; Lanzillotta et al. 2002; Rolffhus and Fitzgerald 2004; Whalin et al. 2007; Qureshi et al. 2010; Ci et al. 2016). Factors influencing the Hg redox chemistry in the marine environment include the Hg concentration and speciation in water, the strength and spectrum of solar radiation, the concentration and structure of dissolved organic matter and suspended particulate matter, salinity, biological activity, and so on (Qureshi et al. 2012; Ci et al. 2016). Field observations showed that water Hg(0) concentrations in waters of oceans and seas were highly variable both spatially and temporally, and surface waters were supersaturated with Hg(0) compared to air in most study time and regions (Qureshi et al. 2012 and references therein).

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The tropical ocean is an important part of the global ocean system. This marine system is characterized by high solar radiation and water temperature, inducing special biogeochemistry of trace elements (Fitzgerald et al. 1984; Christian et al. 2001). Currently, most of investigations on the air–sea $\text{Hg}(0)$ exchange were conducted in the temperate (Mason et al. 1995, 1999; Lanzillotta et al. 2002; Gårdfeldt et al. 2003; Andersson et al. 2007; Fantozzi et al. 2007, 2013; Kuss and Schneider 2007; Ci et al. 2011a, d, 2015; Soerensen et al. 2013) and polar (Mason et al. 1995; Andersson et al. 2008b) marine environments. Only a few measurements focused on tropical oceans and seas (Kim and Fitzgerald 1986; Mason and Fitzgerald 1993; Fu et al. 2010; Kuss et al. 2011; Tseng et al. 2012, 2013; Soerensen et al. 2014). Particularly, the investigation focused on the Hg biogeochemistry in the tropical coastal environment is poor.

Over the last two decades, Hg biogeochemistry in coastal marine environments is receiving increasing attention (Mason et al. 1999; Ferrara et al. 2000; Lanzillotta et al. 2002; Gårdfeldt et al. 2003; Rolfhus and Fitzgerald 2004; Andersson et al. 2007; Fantozzi et al. 2007; Ci et al. 2011a, c). Due to its unique physicochemical properties, the coastal marine ecosystem is considered as an important reactor for rapid, significant, and complicated biogeochemistry of Hg (Fitzgerald et al. 2007). In this study, we simultaneously measured the $\text{Hg}(0)$ concentrations in surface water and overlaying air in a typical tropical coast of the South China Sea (SCS), Hainan Island, China, and further estimated the air–sea $\text{Hg}(0)$ fluxes using a two-layer exchange model. Environmental parameters (e.g., solar radiation, wind speed, and water temperature) affecting the $\text{Hg}(0)$ dynamics are discussed. This study increases the dataset of the global ocean $\text{Hg}(0)$ flux and advances the knowledge of Hg biogeochemistry in the tropical marine environment.

Experimental methods

Site description

The study was performed from 27 January to 8 February 2015 at the Tropical Marine Biological Research Station of the South China Sea Institute of Oceanology, Chinese Academy of Sciences. This station is located at the Luhuitou of Sanya City, the southern tip of Hainan Island, China ($18^{\circ}12.71' \text{ N}$, $109^{\circ}28.39' \text{ E}$; Fig. 1). This coastal region is characterized by a wide distribution of coral reef and mainly receives offshore water from the open water of the SCS (Zhang et al. 2013). The wet and warm southwest monsoon from a low-latitude ocean prevails during April to September, and the dry and cold northeast monsoon from continental East Asia predominates during October to the following March.

Fig. 1 The location of the sampling station (Luhuitou) and the surrounding marine environment

Sampling, analysis, and measurement

The details and quality assurance/quality control for determination of $\text{Hg}(0)$ and total Hg (THg) in water are given in our previous studies (Ci et al. 2011a, c). Briefly, the water samples were manually collected from the sea surface and purged with Hg -free air. The released $\text{Hg}(0)$ was collected on a gold trap and was quantified by a cold vapor atomic fluorescence spectrophotometer (model III, Brooks Rand, USA) using the two-stage gold amalgamation method. After $\text{Hg}(0)$ collections, the subsamples were used to determine the remaining THg, following Gill and Fitzgerald (1987) and the USEPA Method 1631 (USEPA 2002). Briefly, 100-mL samples were oxidized with BrCl and NH_2OH pre-reduction, SnCl_2 reduction, Hg -free air purging, gold trap pre-concentration, thermal desorption, and cold vapor atomic fluorescence spectrophotometry (CVAFS) quantification.

Atmospheric $\text{Hg}(0)$ was measured at the station following the Method IO-5 of USEPA (1999). The principle of this method is also the two-stage gold amalgamation and CVAFS determination. For a typical measurement, the ambient air was delivered to the gold trap with a flow rate of 1.0 L min^{-1} for 2 to 3 h through a Teflon tube. A 47-mm-diameter Teflon pre-filter was placed in the inlet to remove the particle matter (including particulate Hg). This measurement was considered as that of atmospheric $\text{Hg}(0)$ (Ci et al. 2011d; Tseng et al. 2012; Gustin et al. 2015).

The Hg analysis system was calibrated using a gas-tight micro-syringe to inject $\text{Hg}(0)$ -saturated air from water bath, and the analytical precision was $<2\%$ (Ci et al. 2011a). The method detection limit (three times the standard deviations of system blanks) for $\text{Hg}(0)$ and THg in waters and $\text{Hg}(0)$ in the atmosphere was 3.0 pg L^{-1} , 0.10 ng L^{-1} , and 0.05 ng m^{-3} , respectively.

The related environmental parameters, including the photosynthetically active radiation (PAR), water temperature, and wind speed, were monitored with the corresponding sensors (Ci et al. 2011a).

Model for air–sea Hg(0) flux

The details about the calculation of air–sea Hg(0) flux and the uncertainties and limitations of this method are given in our previous studies (Ci et al. 2011a, d). Briefly, the air–sea Hg(0) exchange flux (F) is calculated by $F = K(\text{Hg}(0)_{\text{water}} - \text{Hg}(0)_{\text{air}}/H')$, where F ($\text{ng m}^{-2} \text{h}^{-1}$) is the Hg(0) emission flux from (+) or deposition flux into (–) the surface water. The gas transfer coefficient (K) is calculated by Wanninkhof (1992), $K = 0.31u_{10}^2(S_{\text{CHg}}/600)^{0.5}$, where u_{10} is the wind speed (m s^{-1}) at 10 m above the sea surface and the S_{CHg} is the temperature- and salinity-dependent dimensionless Schmidt number of Hg(0) for seawater. The S_{CHg} is defined as ν/D , where ν is the kinematic viscosity ($\text{cm}^2 \text{s}^{-1}$) of seawater and D is the aqueous diffusion coefficient ($\text{cm}^2 \text{s}^{-1}$) of Hg(0). The ν and D are calculated according to Wanninkhof (1992) and Kuss et al. (2009), respectively. H' is the dimensionless Henry's law constant: $H' = \exp(-2404.3/T + 6.92)$ (Andersson et al. 2008a), where T is the surface water temperature (K).

The degree of saturation (S_a) of Hg(0) in surface waters with respect to the atmospheric Hg(0) is defined as $S_a = \text{Hg}(0)_{\text{water}}H'/\text{Hg}(0)_{\text{air}}$. The S_a value greater than 1 indicates the supersaturation of Hg(0) in surface water and subsequent Hg(0) emission from sea surface; otherwise, undersaturation is indicated and the Hg(0) deposition from the atmosphere to ocean occurs.

Results and discussion

THg in water

Table 1 presents the summary of THg in water, Hg(0) in air and surface water, wind speed, water temperature, saturation of degree of Hg(0) in surface water (S_a), and air–sea Hg(0) fluxes. The concentrations of THg (remaining THg + Hg(0)) in waters varied from 0.96 to 2.23 ng L^{-1} with mean concentrations of $1.40 \pm 0.48 \text{ ng L}^{-1}$ ($n=8$). These concentrations were clearly higher than those in the open oceans, such as the North Atlantic Ocean (0.48 ng L^{-1} , Mason et al. 1998) and the North Pacific Ocean ($\sim 0.2 \text{ ng L}^{-1}$, Laurier et al. 2004), and comparable to those of coast, offshore, and estuary, such as the Yellow Sea ($1\text{--}3 \text{ ng L}^{-1}$, Ci et al. 2011b, d) and the Connecticut River Estuary ($\sim 2 \text{ ng L}^{-1}$, Rolffhus et al. 2003).

During 2003 to 2007, Tseng et al. (2012, 2013) studied the Hg distribution in air and water in the South East Asia Time-Series Station (SEATS), which is located in the open water of the SCS and about 6.5° ($\sim 800 \text{ km}$) further east than our station

Table 1 Range and mean ($\pm$ SD) value of THg in water, Hg(0) in air and surface water, wind speed, water temperature, saturation of degree of Hg(0) in surface water (S_a), and air–sea Hg(0) flux from 27 January to 8 February 2015 in the Luhuitou fringing reef ($18^\circ 12.71' \text{N}$, $109^\circ 28.39' \text{E}$) of the South China Sea, Sanya City, China

	Min.	Max.	Mean	SD	Number
THg in water (ng L^{-1})	0.96	2.23	1.40	0.48	8
Hg(0) in air (ng m^{-3})	1.88	3.22	2.34	0.26	94
Hg(0) in water (pg L^{-1})	17.9	101.0	53.7	18.8	187
Wind speed at 10 m (m s^{-1})	0.45	5.80	3.26	1.04	187
Water temperature ($^\circ\text{C}$)	19.01	26.10	22.80	1.61	187
S_a	2.46	13.86	6.96	2.50	187
Hg(0) flux ($\text{ng m}^{-2} \text{h}^{-1}$)	0.01	6.06	1.73	1.25	187

(see Fig. 1 for the location of the SEATS). Fu et al. (2010) also investigated the Hg distribution in air and water in the northern SCS in the 2007 summer cruise. Compared to the study of Fu et al. (2010), our concentrations were slightly higher than those in the open water ($\sim 1 \text{ ng L}^{-1}$) of the northern SCS, but very similar to those offshore ($1\text{--}2 \text{ ng L}^{-1}$) near the Hainan Island. Notably, our concentrations were very similar to the SEATS in the winter ($\sim 1.6 \text{ ng L}^{-1}$), but slightly lower than other seasons ($1.8\text{--}2.2 \text{ ng L}^{-1}$, Tseng et al. 2013).

Hg(0) in atmosphere

The mean concentrations of atmospheric Hg(0) ($\pm$ SD) in the study period were $2.34 \pm 0.26 \text{ ng m}^{-3}$ ($n=94$) with a range of 1.88 to 3.22 ng m^{-3} . These concentrations were significantly higher than those in the Northern Hemisphere background region ($1.50\text{--}1.75 \text{ ng m}^{-3}$, Ebinghaus et al. 2002), open oceans of the Southern Hemisphere ($1.27 \pm 0.2 \text{ ng m}^{-3}$, Soerensen et al. 2010), the tropical Pacific Ocean ($1.08 \pm 0.17 \text{ ng m}^{-3}$, Wang et al. 2014), and some coast of North America ($1.41 \pm 0.20 \text{ ng m}^{-3}$, Weiss-Penzias et al. 2013) but were comparable to the coast and open water of East Asia, such as the coast of Korea ($\sim 3\text{--}4 \text{ ng m}^{-3}$, Nguyen et al. 2007, 2010), the island of Okinawa (2.04 ng m^{-3} , Jaffe et al. 2005), and the open water of the Bohai Sea, the Yellow Sea, and the East China Sea ($\sim 2\text{--}3 \text{ ng m}^{-3}$, Ci et al. 2011a, d, 2015; Wang et al. 2016).

Recently, Lei et al. (2015) measured the total gaseous Hg concentrations (mainly Hg(0)) in the Wuzhi Mountain (958 m a.s.l.), which is a forest background region and located in about 80 km further north than our station (see Fig. 1 for the location of the Wuzhi Mountain). Our concentrations were higher than the annual mean concentrations ($1.58 \pm 0.71 \text{ ng m}^{-3}$) of the Wuzhi Mountain but were comparable to its winter concentrations ($2.02 \pm 0.53 \text{ ng m}^{-3}$). Our concentrations were also comparable to the northern SCS in the summer of 2007 (2.62 ng m^{-3} , Fu et al. 2010). Tseng et al. (2012)

observed a clear seasonality of atmospheric Hg(0) with low concentrations in summer ($2.8 \pm 0.2 \text{ ng m}^{-3}$) and high concentrations in winter ($5.7 \pm 0.2 \text{ ng m}^{-3}$) in the SEATS. Clearly, our concentrations were comparable to the summer but significantly lower than the winter in the SEATS. According to the 72-h back-trajectory analysis, Tseng et al. (2012) suggested that the high atmospheric Hg(0) concentrations in winter were attributed to the northeast monsoon which brought the air masses with enhanced Hg(0) from continental East Asia. In the period of our study, the prevailing wind direction was also northeastern, but the 72-h back-trajectory analysis showed that, due to the low wind speed ($\sim 3 \text{ m s}^{-1}$), the sampled air masses mainly originated from the sea surface near the coast of the Southern China ($22^\circ\text{--}26^\circ \text{ N}$) with little direct anthropogenic Hg emission (data not shown). Therefore, the significant difference of atmospheric Hg(0) in winter between our study and that of Tseng et al. (2012) may be due to the sampled air masses originated from different source regions (clean maritime air vs polluted continental air).

Hg(0) in water

The mean concentrations of Hg(0) in surface water were $53.7 \pm 18.8 \text{ pg L}^{-1}$ ($n=187$) with a range of 17.9 to 101.0 pg L^{-1} . These concentrations were higher than those in the tropical Pacific Ocean ($10\text{--}70 \text{ pg L}^{-1}$, Kim and Fitzgerald 1986; Mason and Fitzgerald 1993; Soerensen et al. 2014) and comparable to the concentrations of Hg(0) in the subtropical and tropical Atlantic Ocean ($\sim 10\text{--}210 \text{ pg L}^{-1}$, Mason and Sullivan 1999; Kuss et al. 2011). Our concentrations were also higher than the annual mean concentrations of some coast (e.g., the coast of the Yellow Sea, $\sim 30 \text{ pg L}^{-1}$, Ci et al. 2011a) and offshore (the Baltic Sea, $10\text{--}32 \text{ pg L}^{-1}$, Kuss and Schneider 2007; the open water of the Yellow Sea, $\sim 35 \text{ pg L}^{-1}$, Ci et al. 2011b) environments in temperate zone but were comparable to some temperate marine environments in warm seasons with similar THg levels, water temperature, and solar radiation, such as the coast and offshore of the Yellow Sea in summer campaigns ($\sim 60 \text{ pg L}^{-1}$, Ci et al. 2011a, d) and the Island of Corsica in the Mediterranean Sea on June 2004 ($\sim 20\text{--}100 \text{ pg L}^{-1}$, Fantozzi et al. 2007).

Tseng et al. (2013) reported a pronounced seasonality of water Hg(0) in the SEATS with high concentrations in warm seasons and low concentrations in cold seasons. Notably, our concentrations of water Hg(0) ($53.7 \pm 18.8 \text{ pg L}^{-1}$) were higher than those in both the summer ($32 \pm 8 \text{ pg L}^{-1}$) and the winter ($12 \pm 6 \text{ pg L}^{-1}$) in the SEATS. Our concentrations were also higher than the mean concentrations ($36.5 \pm 14.9 \text{ pg L}^{-1}$) of the northern SCS in the summer cruise but were very similar to those of stations near the Hainan Island ($\sim 40\text{--}80 \text{ pg L}^{-1}$, Fu et al. 2010). Many field measurements also widely reported that the water Hg(0) concentrations in nearshore environments were generally higher than those in the open water (e.g.,

Mason et al. 2001; Ci et al. 2011d). The field observations and incubation experiments suggest that the special properties of nearshore environment, such as the enhanced THg levels, the estuarine mixing, and the high biological activity, can either promote the production of water Hg(0) and/or increase the Hg(0) input to the coastal water (Amyot et al. 1997; Lanzillotta et al. 2002; Rolfhus et al. 2003; Covelli et al. 2008; Fantozzi et al. 2009; Schartup et al. 2015; Ci et al. 2016).

In this study, the mean value of Hg(0)/THg ratios was 3.8 %. These values were significantly lower than those in open oceans ($\sim 10\text{--}50 \%$, e.g., Mason et al. 2001; Soerensen et al. 2013; Schartup et al. 2015) and comparable to concentrations of many coast and offshore environments ($1\text{--}5 \%$, e.g., Mason et al. 1999; Ci et al. 2011a, d). It should be noted that our values were slightly higher than those in the northern SCS in the summer cruise (3.0 %, Fu et al. 2010) but significantly higher than those in the winter in the SEATS ($\sim 1 \%$) and other seasons ($1\text{--}1.6 \%$) (Tseng et al. 2013).

As mentioned above, the Luhuitou fringing reef is a shallow coast ($\sim 5 \text{ m}$) and widely covered by corals (Zhang et al. 2013). It is well known that coral reef is one of the most productive marine ecosystems and contains a high level of biodiversity (Gattuso et al. 1998). The high productivity of coral reef ecosystem may favor the production of water Hg(0) and subsequently increase the Hg(0)/THg ratio by two pathways. One is that the biological process of the coral reef ecosystem may directly reduce the Hg(II) to Hg(0) via the microbial-mediated Hg(II) reduction (Fantozzi et al. 2009). The other is that the high productivity of the coral ecosystem alters the water chemistry, for example increasing the organic matter in water column, and thus may promote the Hg(0) production since the organic matter generally favors the photoreduction of Hg(II) to Hg(0) (Costa and Liss 1999; Schartup et al. 2015). Therefore, we suppose that the special property of the Luhuitou fringing reef, particularly the large coverage of corals, may greatly promote the production of Hg(0) in coastal water, resulting in higher Hg(0) concentrations and Hg(0)/THg ratio in coastal water than the open water of the SCS. However, to our best knowledge, there is a lack of investigation of Hg biogeochemical cycle under the influence of coral reef ecosystem. More targeted and systematic researches are needed in the future.

Figure 2 shows the time series of Hg(0) data and the measured environmental parameters in the entire study period. As shown in Fig. 2, the Hg(0) concentrations in surface water were highly variable. In many days of the study period (e.g., 30 January and 7 February), the Hg(0) concentrations in waters generally paralleled the solar radiation, presented clearly a diurnal variation with low levels before sunrise, increased to and reached peak levels around noon, and then decreased to low levels in the afternoon and nighttime. This similar correlation has been widely reported (e.g., Fantozzi et al. 2007; Ci

Fig. 2 Time series of Hg(0) in surface water and air, saturation of degree of Hg(0) in surface water (S_a), air-sea Hg(0) flux, and the measured variables from 27 January to 8 February 2015 in the Luhuitou fringing reef (18° 12.71' N, 109° 28.39' E) of the South China Sea, Sanya City, China

et al. 2011a). Linear regression analysis suggested that Hg(0) concentrations in waters were positively and significantly correlated to the PAR (water Hg(0) [pg L^{-1}] $=0.0203 \times \text{PAR}$ [$\mu\text{mol m}^{-2} \text{s}^{-1}$] $+38.655$, $R^2=0.42$, $p<0.001$). This further confirms that the solar radiation is the important controlling factor for the Hg(0) production in coastal water.

The wind force seems also to greatly regulate the variation of Hg(0) concentrations in waters in a short time scale. For example, the daytime of 31 January and that of 1 February were cloudless; however, the diurnal variation of water Hg(0) in these two days was not as pronounced as other cloudless days (e.g., 30 January and 7 February). It is noted that the relatively high wind speed ($\sim 4\text{--}6 \text{ m s}^{-1}$) was recorded in these two days (Fig. 2). Wind force influences the variation of Hg(0) concentrations mainly in the following three ways. First, the high wind speed increases the Hg(0) emission from the sea surface under the condition of supersaturation of water Hg(0) and subsequently reduces the Hg(0) concentration in water column. Second, the high wind speed promotes the vertical mixing of water and thus reduces the exposure time of water to solar radiation which subsequently reduces the photochemical potential of Hg(0) production (O'Driscoll et al. 2003; Ci et al. 2011a). Third, unlike deep open oceans, the shallow coastal environments provide a large ratio of sediment surface-to-water volume and the wind-driven resuspension events induce the elevated vertical mixing of water and then increase the suspended particulate matter of the water column. This process reduces the penetration of solar radiation and also decrease the photochemical potential of Hg(0) production in water column.

The values of S_a ranged from 2.46 to 13.87 with a mean value of 6.96 ± 2.50 . Clearly, the S_a values were always greater than 1, indicating that the surface water was always the source

of atmospheric Hg(0). Our values were comparable to those of the tropical and subtropical oceans, such as the Equatorial Pacific Ocean (0.79–40, Kim and Fitzgerald 1986; Mason and Fitzgerald 1993), the Atlantic Ocean ($\sim 4\text{--}20$, Mason and Sullivan 1999; Kuss et al. 2011), and many coasts and off-shore of the temperate marine environments in warmer seasons, such as the Yellow Sea (4.50–7.80, Ci et al. 2011a, d) and the Mediterranean Sea (6–11, Andersson et al. 2007) in summer. Our values were also comparable to those in the northern SCS in summer (5.34, Fu et al. 2010). However, Tseng et al. (2013) found that the water Hg(0) in the SEATS in winter was undersaturation ($S_a=0.6$) due to the combination of low water Hg(0) and high atmospheric Hg(0).

Air-sea Hg(0) flux

The air-sea Hg(0) fluxes in this study were estimated to be $1.73 \pm 1.25 \text{ ng m}^{-2} \text{ h}^{-1}$ with a large range from 0.01 to $6.06 \text{ ng m}^{-2} \text{ h}^{-1}$. As described above, the two-layer gas exchange model is very sensitive to the wind speed. The high variation of Hg(0) fluxes mainly attributed to the high variation of wind speed. Our fluxes were comparable to the Pacific Ocean ($1\text{--}3 \text{ ng m}^{-2} \text{ h}^{-1}$, Kim and Fitzgerald 1986; Mason and Fitzgerald 1993; Soerensen et al. 2014) and comparable to the subtropical and tropical Atlantic Ocean ($0.5\text{--}20 \text{ ng m}^{-2} \text{ h}^{-1}$, Mason and Sullivan 1999; Kuss et al. 2011) and many temperate marine environments, such as the Yellow Sea ($0.51\text{--}18.3 \text{ ng m}^{-2} \text{ h}^{-1}$, Ci et al. 2011a, d, 2015), the Mediterranean Sea ($0.84\text{--}3.40 \text{ ng m}^{-2} \text{ h}^{-1}$, Andersson et al. 2007), and the Chesapeake Bay ($1.1 \text{ ng m}^{-2} \text{ h}^{-1}$, Mason et al. 1999).

Our fluxes were lower than those of the northern SCS in the summer cruise ($4.5 \pm 3.4 \text{ ng m}^{-2} \text{ h}^{-1}$) since high wind speeds were recorded (Fu et al. 2010). Notably, Tseng et al. (2013)

observed the undersaturation of seawater Hg(0) in winter since the high concentrations of atmospheric Hg(0) and low concentrations of water Hg(0), inducing the net deposition Hg(0) ($-1.5 \text{ ng m}^{-2} \text{ h}^{-1}$) from the atmosphere to the sea surface. This discrepancy further shows the significantly different Hg(0) dynamics between the coast and open water of the SCS.

Conclusions

The measurements of Hg(0) concentrations in the surface water and air in a typical tropical coast of the SCS showed that the behavior of Hg(0) was highly dynamic in a short time scale, depending on many environmental parameters. Measurements of Hg(0) in surface waters showed the pronounced diurnal variation with high concentrations in daytime and low concentrations in nighttime, indicating that the photochemical reduction of Hg(II) is a fundamental process for the production of Hg(0). Compared to the previous study, we found the significant difference of Hg(0) dynamics between the coast and open water of the SCS in winter. We suppose that the unique coastal environment setting in the Luhuitou fringing reef, such as the large coverage of corals, may have important impact on the Hg(0) production in water, inducing the elevated Hg(0) concentrations and the Hg(0)/THg ratio in coastal water than in the open water of the SCS. More focused and comprehensive researches are needed to explore the effect of coral ecosystem on the Hg biogeochemical cycle in the future.

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