Response to Reviewer #2's Comments

Anonymous Referee #2:

The manuscript presents ship-based measurements of atmospheric mercury species and dissolved gaseous mercury in the Pearl River Estuary and the South China Sea. The authors used the measurements to infer the sources and sinks of elemental and reactive mercury in the atmosphere, to estimate the sea to air flux of mercury, and to assess how the mercury concentrations in the South China Sea differ from other areas. The manuscript is well-written, and presents the results in clearly with appropriate tables and figures. The authors provide a detailed description of their measurement methods, analyze their observations systematically, and provide support for their main conclusions.

- I do not have any major concerns about the manuscript, but a few minor comments as follows: We are grateful for the precise, valuable and positive comments. We have made corresponding changes in the revised manuscript as highlighted in red color. Please see the responses to the specific comments below. The reviewer's comments are in black and our replies are in blue.
- (1) Line 43: Here and elsewhere, I would reword "suffered less influence of human activities" to something like "less influence of fresh emissions."

Response:

Thanks for your suggestion. We have changed the term "human activities" as "fresh emissions" throughout the manuscript.

See the revised manuscript at lines 15, 344.

(2) Line 164: The back trajectories were initiated at 500m much higher than the measurement altitude. This needs justification.

Response:

Thanks for your comment. The arrival heights of back trajectories were set at 500 m to represent the approximate height of the mixing marine boundary layer where atmospheric pollutants were well mixed. We have added "the start height was set at 500 m above sea level to represent the approximate height of the mixing marine boundary layer where atmospheric pollutants were well mixed" in the revised manuscript.

See the revised manuscript at lines 141-142.

(3) Line 172: Fig. S1 does not show the sampling unit.

Response:

Thanks for your carefully check on this sentence and Figure S1. The picture of the sampling unit (Figure S1) has been added in the revised supplement of this paper. See the revised supplement at line 58.

(4) Line 200: How were non-detects in the RGM and HgP measurements treated?

Response:

Thanks for your comment. This sentence has been revised as "It should be noted that all the observed RGM and Hg^P values were higher than the corresponding blank values, and the average blank values for RGM and Hg^P were subtracted from the samples." See the revised manuscript at lines 179-181.

(5) Line 331: The bimodal distribution seems less obvious in Fig. 5b.

Response:

We agree with the reviewer that the bimodal distribution seems less obvious in Fig. 5b. As a matter of fact, there were three peaks (< 0.4 μ m, 0.7-1.1 μ m, 5.8-9.0 μ m) although the three-modal distribution was not distinct in Fig. 5. We have corrected the statements in the revised manuscript.

See the revised manuscript at lines 23-24, 307-310, 468.

(6) Line 351-353: I am not convinced that these 1-month observations can be extrapolated to an annual dry deposition flux. I recommend removing that calculation unless there is other evidence supporting its validity.

Response:

Thanks for your comment. Since the South China Sea (SCS) is a tropical sea (4 N-21 N), so we can assume that there is no large variation in the average PAR and air temperature etc. over the SCS during the four seasons. Moreover, since our sampling time spans September 2015, so the average RGM and Hg^P values obtained in this sampling period can be roughly considered as the annual mean values. Additionally, Fu et al. (2010) had used the observations (August 2010) in the northern SCS to estimate the annual emission flux of Hg⁰ over the SCS. Therefore, we think we can use the data obtained in this study to roughly estimate the dry deposition fluxes of RGM and Hg^P in the SCS.

Reference:

Fu, X., Feng, X., Zhang, G., Xu, W., Li, X., Yao, H., Liang, P., Li, J., Sommar, J., Yin, R., and Liu, N.: Mercury in the marine boundary layer and seawater of the South China Sea: Concentrations, sea/air flux, and implication for land outflow, J. Geophys. Res., 115, D06303, https://doi.org/10.1029/2009JD012958, 2010.

(7) Line 365-367: It is not obvious why the small variability in the Hg0 concentrations implies that the evasion of DGM was an important source of Hg0.

Response:

We do agree with your comment that the small variability in Hg^0 concentrations could not imply that the evasion of DGM was an important source of Hg^0 . Therefore, we later realized that this inference lacked sufficient evidence. Therefore, we have deleted this sentence (i.e., and the evasion of DGM in local or regional surface seawater of the SCS and surrounding oceans was probably an important source for the GEM in the marine atmosphere.)

(8) Line 381: It is not clear why are higher RH and lower temperature conducive to Hg2 removal? By gas-particle partitioning?

Response:

Thanks for your comment. Yes, by gas-particle partitioning. Previous studies demonstrated that higher RH and lower air temperature were conductive to the transfer of RGM to Hg^P (Rutter and Schauer, 2007; Lee et al., 2016). Thus, we revised the description as "the transfer of RGM to Hg^P due to higher RH and lower air temperature in nighttime (Rutter and Schauer, 2007; Lee et al., 2016)." in the revised manuscript.

See the revised manuscript at lines 357-359.

Reference:

Rutter, A. P., and Schauer, J. J.: The effect of temperature on the gas-particle partitioning of reactive mercury in atmospheric aerosols, Atmos. Environ., 41, 8647–8657, https://doi.org/10.1016/j.atmosenv.2007.07.024, 2007.

Lee, G.-S., Kim, P.-R., Han, Y.-J., Holsen, T. M., Seo, Y.-S., and Yi, S.-M.: Atmospheric speciated mercury concentrations on an island between China and Korea: sources and transport pathways, Atmos. Chem. Phys., 16, 4119-4133, https://doi.org/10.5194/acp-16-4119-2016, 2016.

- (9) Please mention in the main text that the acronyms are defined in the appendix.
 - Response:

Thanks for your suggestion. The sentence "It should be noted that all of the acronyms in this article have been listed in the Appendix." has been added in the revised manuscript. See the revised manuscript at lines 45-46.

(10) Fig. 5: Using the same scales for the y-axes on panels a and b will be helpful.

Response:

Thanks for the suggestion. We have set the same scales for the Y-axes on panels a and b in Fig.5.

See the revised manuscript at line 776.

(11) Fig. 6: The PAR values can be removed. They do not add much information, but clutter the figure.

Response:

Thanks for your suggestion. There are no PAR values in Fig. 6, and I suspect that the PAR values you mentioned are the one in Fig. 7. First of all, the PAR data was essential for the following discussions. Moreover, we have changed the filled color for the RGM concentrations in nighttime. Thus, we feel the revised Fig. 7 is clearer now. See the revised manuscript at line 782.

(12) Fig. 9: How are the point measurements interpolated for the entire region? Was this interpolation necessary to calculate the sea-air flux? If not may be show the measurements like those in Fig. 3b.

Response:

Thanks for your comments and suggestions. The Fig. 9 was plotted by the software of Ocean Data View based on the data obtained at each the sampling station. It is not necessary for the interpolation to calculate the sea-air flux of Hg^0 . The DGM concentrations in surface seawater and emission fluxes of Hg^0 at all stations have been symbolized in the revised Fig. 9 like those in Fig. 3b.

See the revised manuscript at line 789.

(13) Table 2: The correlation coefficients for HgP(2.5)-RGM and RGM-HgP(2.5) differ. Is that correct?

Response:

We feel very sorry for our carelessness. After recalculation, the correlation coefficients for $Hg_{2.5}^{P}$ -RGM and RGM- $Hg_{2.5}^{P}$ were same (p < 0.01, r = 0.453). We have corrected the p and r values for $Hg_{2.5}^{P}$ -RGM in Table 2.

See the revised manuscript at lines 794-795.

1 Speciated atmospheric mercury and sea-air exchange of

2 gaseous mercury in the South China Sea

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8 Abstract

The characteristics of the reactive gaseous mercury (RGM) and particulate mercury (Hg^P) in the 9 marine boundary layer (MBL) is poorly understood due in part to sparse data from sea and ocean. 10 Gaseous elemental Hg (GEM), RGM and size-fractioned Hg^P in marine atmosphere, and dissolved 11 gaseous Hg (DGM) in surface seawater were determined in the South China Sea (SCS) during an 12 oceanographic expedition (3-28 September 2015). The mean concentrations of GEM, RGM and 13 $Hg_{2.5}^{P}$ were 1.52 ± 0.32 ng m⁻³, 6.1 ± 5.8 pg m⁻³ and 3.2 ± 1.8 pg m⁻³, respectively. Low GEM 14 level indicated that the SCS suffered less influence from fresh emissions, which could be due to 15 16 the majority of air masses coming from the open oceans as modeled by backward trajectories. Atmospheric reactive Hg (RGM + Hg $_{2.5}^{P}$) represented less than 1 % of total atmospheric Hg, 17 indicating that atmospheric Hg existed mainly as GEM in the MBL. The GEM and RGM 18 concentrations (1.73 \pm 0.40 ng m⁻³ and 7.1 \pm 1.4 pg m⁻³ respectively) in the northern SCS were 19 significantly higher than those $(1.41 \pm 0.26 \text{ ng m}^{-3} \text{ and } 3.8 \pm 0.7 \text{ pg m}^{-3})$ in the western SCS, and 20 the Hg^{P_{2.5} and Hg^{P₁₀} levels (8.3 and 24.4 pg m⁻³) in the Pearl River Estuary (PRE) were 0.5–6.0} 21 times higher than those in the open waters of the SCS, indicating that the PRE was polluted to 22 some extent. The size distribution of Hg^{P} in PM_{10} was observed to be three-modal with peaks 23 around $<0.4 \mu m$, $0.7-1.1 \mu m$ and $5.8-9.0 \mu m$, respectively, but the coarse modal was the dominant 24 size, especially in the open SCS. There was no significant diurnal variation of GEM and Hg^P_{2.5}, 25 but we found the RGM concentrations were significantly higher in daytime $(8.0 \pm 5.5 \text{ pg m}^{-3})$ than 26 in nighttime $(2.2 \pm 2.7 \text{ pg m}^{-3})$ mainly due to the influence of solar radiation. In the northern SCS, 27 the DGM concentrations in nearshore area (40–55 pg l^{-1}) were about twice as high as those in the 28 open sea, but this pattern was not significant in the western SCS. The sea-air exchange fluxes of 29 Hg⁰ in the SCS varied from 0.40 to 12.71 ng m⁻² h⁻¹ with a mean value of 4.99 \pm 3.32 ng m⁻² h⁻¹. 30 The annual emission flux of Hg⁰ from the SCS to the atmosphere was estimated to be 159.6 tons 31 yr^{-1} , accounting for about 5.54 % of the global Hg^0 oceanic evasion though the SCS only 32 represents 1.0 % of the global ocean area. Additionally, the annual dry deposition flux of 33 atmospheric reactive Hg represented more than 18 % of the annual evasion flux of Hg⁰, and 34

therefore the dry deposition of atmospheric reactive Hg was an important pathway for the input ofatmospheric Hg to the SCS.

37 1 Introduction

38 Mercury (Hg) is a naturally occurring metal. Hg is released to the environment through both the 39 natural and anthropogenic pathways (Schroeder and Munthe, 1998). However, since the Industrial Revolution, the anthropogenic emissions of Hg increased drastically. Continued rapid 40 41 industrialization has made Asia the largest source region of Hg emissions to air, with East and 42 Southeast Asia accounting for about 40 % of the global total (UNEP, 2013). Three operationally defined Hg forms are present in the atmosphere: gaseous elemental Hg (GEM or Hg⁰), reactive 43 gaseous Hg (RGM) and particulate Hg (Hg^P) (Schroeder and Munthe, 1998; Landis et al., 2002), 44 while they have different physicochemical characteristics. It should be noted that all of the 45 46 acronyms in this article have been listed in the Appendix. GEM is very stable with a residence time of 0.2-1.0 yr due to its high volatility and low solubility (Radke et al., 2007; Selin et al., 47 48 2007; Horowitz et al., 2017). Therefore, GEM can be transported for a long-range distance in the atmosphere, and this makes it well-mixed on a regional and global scale. Generally, GEM makes 49 up more than 95 % of total atmospheric Hg (TAM), while the RGM and Hg^P concentrations 50 (collectively known as atmospheric reactive mercury) are typically 2-3 orders of magnitude 51 52 smaller than GEM in part because they are easily removed from ambient air by wet and dry 53 deposition (Laurier and Mason, 2007; Holmes et al., 2009; Gustin et al., 2013), and they can also be reduced back to Hg^{0} . 54

Numerous previous studies have shown that Hg⁰ in the marine boundary layer (MBL) can be 55 rapidly oxidized to form RGM in situ (Laurier et al., 2003; Sprovieri et al., 2003, 2010; Laurier 56 57 and Mason, 2007; Soerensen et al., 2010a; Wang et al., 2015; Mao et al., 2016; Ye et al., 2016). Ozone and OH could potentially be important oxidants on aerosols (Ariya et al., 2015; Ye et al., 58 2016), while the reactive halogen species (e.g., Br, Cl and BrO, generating from sea salt aerosols) 59 may be the dominant sources for the oxidation of Hg^0 in the MBL (Holmes et al., 2006, 2010; 60 Auzmendi-Murua et al., 2014; Gratz et al., 2015; Steffen et al., 2015; Shah et al., 2016; Horowitz 61 62 et al., 2017). However, a recent study showed that Br and BrO became dominant GEM oxidants in 63 the marine atmosphere with mixing ratios reaching 0.1 and 1 pptv, respectively, and contributing \sim 64 70 % of the total RGM production during midday, while O_3 dominated GEM oxidation (50–90 % of RGM production) when Br and BrO mixing ratios were diminished (Ye et al., 2016). The wet 65 and dry deposition (direct or uptake by sea-salt aerosol) represents a major input of RGM and Hg^P 66 67 to the sea and ocean due to their special and unique characteristics (i.e., high reactivity and water solubility) (Landis et al., 2002; Holmes et al., 2009). Previous studies also showed that 68 69 atmospheric wet and dry deposition of RGM (mainly HgBr₂, HgCl₂, HgO, Hg-nitrogen and sulfur 70 compounds) was the greatest source of Hg to open oceans (Holmes et al., 2009; Mason et al., 2012; Huang et al., 2017). A recent study suggested that approximately 80 % of atmospheric reactive Hg
sinks into the global oceans, and most of the deposition takes place to the tropical oceans
(Horowitz et al., 2017).

74 The atmospheric reactive Hg deposited to the oceans follows different reaction pathways. 75 One important process is that divalent Hg can be combined with the existing particles followed by sedimentation, or be converted to methylmercury (MeHg), the most bioaccumulative and toxic 76 77 form of Hg in seafood (Ahn et al., 2010; Mason et al., 2017). Another important process is that the divalent Hg can be converted to dissolved gaseous Hg (DGM) through abiotic and biotic 78 79 mechanisms (Strode et al., 2007). It is well known that almost all DGM in the surface seawater is Hg⁰ (Horvat et al., 2003), while the dimethylmercury is extremely rare in the surface seawater 80 (Bowman et al., 2015). It has been found that a majority of the surface seawater was 81 supersaturated with respect to Hg⁰ (Soerensen et al., 2010b, 2013, 2014), and parts of this Hg⁰ 82 may be emitted to the atmosphere. Evasion of Hg⁰ from the oceanic surface into the atmosphere is 83 partly driven by the solar radiation and aquatic Hg pools of natural and anthropogenic origins 84 85 (Andersson, et al., 2011). Sea-air exchange is an important component of the global Hg cycle as it 86 mediates the rate of increase in ocean Hg and therefore the rate of change in level of MeHg. Consequently, Hg⁰ evasion from sea surface not only decreases the amount of Hg available for 87 methylation in waters but also has an important effect on the redistribution of Hg in the global 88 89 environment (Strode et al., 2007).

90 In recent years, speciated atmospheric Hg has been monitored in coastal areas (Xu et al., 91 2015; Ye et al., 2016; Howard et al., 2017; Mao et al., 2017) and open seas and oceans (e.g., 92 Chand et al., 2008; Soerensen et al., 2010a; Mao et al., 2016; Wang et al., 2016a, b). However, there exists a dearth of knowledge regarding speciated atmospheric Hg and sea-air exchange of 93 Hg⁰ in tropical seas, such as the South China Sea (SCS). The highly time-resolved ambient GEM 94 concentrations were measured using a Tekran[®] system. Simultaneously, the RGM, Hg^P and DGM 95 were measured using manual methods. The main objectives of this study are to identify the 96 97 spatial-temporal characteristics of speciated atmospheric Hg and to investigate the DGM concentrations in the SCS during the cruise, and then to calculate the Hg⁰ flux based on the 98 99 meteorological parameters as well as the concentrations of GEM in air and DGM in surface 100 seawater. These results will raise our knowledge of the Hg cycle in tropical marine atmosphere 101 and waters.

102 2 Materials and methods

103 2.1 Study area

104 The SCS is located in the downwind of Southeast Asia (Fig. 1a), and it is the largest semi-enclosed 105 marginal sea in the western tropical Pacific Ocean. The SCS is connected with the East China Sea 106 (ECS) to the northeast and the western Pacific Ocean to the east (Fig. 1a). The SCS is surrounded 107 by numerous developing and developed countries (Fig. 1a). An open cruise was organized by the South China Sea Institute of Oceanology (Chinese Academy of Sciences) and conducted during 108 the period of 3-28 September 2015. The sampling campaign was conducted on R/V Shiyan 3, 109 110 which departed from Guangzhou, circumnavigated the northern and western SCS and then 111 returned to Guangzhou. The DGM sampling stations and R/V tracks are plotted in Fig. 1b. In this study, meteorological parameters (including photosynthetically available radiation (PAR) 112 (Li-COR[®], Model: Li-250), wind speed, air temperature and RH) were measured synchronously 113 with atmospheric Hg onboard the R/V. 114

115 2.2 Experimental methods

116 2.2.1 Atmospheric GEM measurements

117 In this study, GEM was measured using an automatic dual channel, single amalgamation cold vapor atomic fluorescence analyzer (Model 2537B, Tekran[®], Inc., Toronto, Canada), which has 118 been reported in our previous studies (Wang et al., 2016a, b, c). In order to reduce the 119 contamination from ship exhaust plume as possible, we installed the Tekran[®] system inside the 120 121 ship laboratory (the internal air temperature was controlled to 25 °C using an air conditioner) on 122 the fifth deck of the R/V and mounted the sampling inlet at the front deck 1.5 m above the top deck (about 16 m above sea level) using a 7 m heated (maintained at 50 °C) 123 polytetrafluoroethylene (PTFE) tube (1/4 inch in outer diameter). The sampling interval was 5 min 124 and the air flow rate was $1.5 \ 1 \ min^{-1}$ in this study. Moreover, two PTFE filters (0.2 μ m pore size, 125 47 mm diameter) were positioned before and after the heated line, and the soda lime before the 126 instrument was changed every 3 days during the cruise. The Tekran[®] instrument was calibrated 127 every 25 h using the internal calibration source and these calibrations were checked by injections 128 of certain volume of saturated Hg⁰ before and after this cruise. The relative percent difference 129 between manual injections and automated calibrations was < 5 %. The precision of the analyzer 130 was determined to > 97 %, and the detection limit was $< 0.1 \text{ ng m}^{-3}$. 131

132 The meteorological and basic seawater parameters were collected onboard the R/V, which 133 was equipped with meteorological and oceanographic instrumentations. To investigate the influence of air masses movements on the GEM levels, 72-h backward trajectories of air masses 134 were calculated using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) 135 136 model (Draxler and Rolph, 2012) and TrajStat software (Wang et al., 2009) based on Geographic Information System. Global Data Assimilation System (GDAS) meteorological dataset 137 (ftp://arlftp.arlhq.noaa.gov/pub/archives/gdas1/) with $1^{\circ} \times 1^{\circ}$ latitude and longitude horizontal 138 139 spatial resolution and 23 vertical levels at 6-h intervals was used as the HYSPLIT model input. It should be noted that the start time of each back trajectory was identical to the GEM sampling time 140 (UTC) and the start height was set at 500 m above sea level to represent the approximate height of 141 142 the mixing marine boundary layer where atmospheric pollutants were well mixed.

143 2.2.2 Sampling and analysis of RGM and Hg^P

The Hg^P_{2.5} (Hg^P in PM_{2.5}) was collected on quartz filter (47 mm in diameter, Whatman), which has 144 been reported in several previous studies (Landis et al., 2002; Liu et al., 2011; Kim et al., 2012;). 145 146 It should be pointed out that the KCl coated denuders were heated at 500 $^{\circ}$ C for 1 h and the quartz 147 filters were pre-cleaned by pyrolysis at 900 $\,$ °C for 3 h to remove the possible pollutant. The RGM and Hg^P_{2.5} were sampled using a manual system (URG-3000M), which has been reported in 148 previous studies (Landis et al., 2002; Liu et al., 2011; Wang et al., 2016b). The sampling unit 149 includes an insulated box (Fig. S1), two quartz annular denuders, two Teflon filter holder (URG 150 Corporation) and a pump etc. The sampling flow rate was $10 \ 1 \ min^{-1}$ (Landis et al., 2002), and the 151 sampling inlet was 1.2 m above the top deck of the R/V. In this study, one Hg_{25}^{P} sample was 152 collected in the daytime (6:00-18:00) and the other in the nighttime (18:00-6:00 (next day)), 153 while two RGM samples were collected in the daytime (6:00-12:00 and 12:00-18:00, local time) 154 and one RGM sample in the nighttime. Quality assurance and quality control for Hg^P and RGM 155 156 were carried out using field blank samples and duplicates. The field blank denuders and quartz filters were treated similarly to the other samples but not sampling. The mean relative differences 157 of duplicated Hg^P_{2.5} and RGM samples (n = 6) were 13 \pm 6% and 9 \pm 7%, respectively. 158

- Meanwhile, we collected different size particles using an Andersen impactor (nine-stage), 159 160 which has been widely used in previous studies (Feddersen et al., 2012; Kim et al., 2012; Zhu et al., 2014; Wang et al., 2016a). The Andersen cascade impactor was installed on the front top deck 161 of the R/V to sample the size-fractioned particles in PM_{10} . In order to diminish the contamination 162 from exhaust plume of the ship as much as possible, we turned off the pump when R/V arrived at 163 164 stations, and then switched back on when the R/V went to next station. The sample collection began in the morning (10:00 am) and continued for 2 days with a sampling flow rate of 28.3 l 165 min⁻¹. Field blanks for Hg^P were collected by placing nine pre-cleaned quartz filters (81 mm in 166 diameter, Whatman) in another impactor for 2 days without turning on the pump. After sampling, 167 168 the quartz filters were placed in cleaned plastic boxes (sealing in Zip Lock plastic bags), and then were immediately preserved at $-20 \$ C until the analysis. 169
- The detailed analysis processes of RGM and Hg^P have been reported in our previous studies 170 171 (Wang et al., 2016a, b). Briefly, the denuder and quartz filter were thermally desorbed at 500 $\,^{\circ}$ C and 900 $^{\circ}$ C, respectively, and then the resulting thermally decomposed Hg⁰ in carrier gas (zero air, 172 i.e., Hg-free air) was quantified. The method detection limit was calculated to be 0.67 pg m^{-3} for 173 174 RGM based on 3 times the standard deviation of the blanks (n = 57) for the whole dataset. The average field blank of denuders was 1.2 ± 0.6 pg (n = 6). The average blank values (n = 6) of 175 $Hg_{2.5}^{P}$ and Hg_{10}^{P} were 1.4 pg (equivalent of < 0.2 pg m⁻³ for a 12 h sampling time) and 3.2 pg 176 (equivalent of < 0.04 pg m⁻³ for a 2-day sampling time) of Hg per filter, respectively. The 177 detection limits of $Hg_{2.5}^{P}$ and Hg_{10}^{P} were all less than 1.5 pg m⁻³ based on 3 times the standard 178 deviation of field blanks. It should be noted that all the observed RGM and Hg^P values were 179

higher than the corresponding blank values, and the average blank values for RGM and Hg^P were
subtracted from the samples.

182 2.2.3 Determination of DGM in surface seawater

183 In this study, the analysis was carried out according to the trace element clean technique, all containers (borosilicate glass bottles and PTFE tubes, joints and valves) were cleaned prior to use 184 with detergent, followed by trace-metal-grade HNO3 and HCl, and then rinsed with Milli-Q water 185 $(> 18.2 \text{ M}\Omega \text{ cm}^{-1})$, which has been described in our previous study (Wang et al., 2016c). DGM 186 were measured in situ using a manual method (Fu et al., 2010; Ci et al., 2011). The detailed 187 188 sampling and analysis of DGM has been elaborated in our previous study (Wang et al., 2016c). The analytical blanks were conducted onboard the R/V by extracting Milli-Q water for DGM. The 189 mean concentration of DGM blank was 2.3 ± 1.2 pg l⁻¹ (n = 6), accounting for 3–10 % of the raw 190 DGM in seawater samples. The method detection limit was 3.6 pg l^{-1} on the basis of three times 191 the standard deviation of system blanks. The relative standard deviation of duplicate samples 192 generally < 8 % of the mean concentration (n = 6). 193

194 **2.2.4** Estimation of sea-air exchange flux of Hg⁰

The sea-air flux of Hg⁰ was calculated using a thin film gas exchange model developed by Liss 195 and Slater (1974) and Wanninkhof (1992). The detailed calculation processes of Hg⁰ flux have 196 197 been reported in recent studies (Ci et al., 2011; Kuss, 2014; Wang et al., 2016c; Kuss et al., 2018). It should be noted that the Schmidt number for gaseous Hg (ScHg) is defined as the following 198 equation: $Sc_{Hg} = v/D_{Hg}$, where v is the kinematic viscosity (cm² s⁻¹) of seawater calculated using 199 the method of Wanninkhof (1992), D_{Hg} is the Hg⁰ diffusion coefficient (cm² s⁻¹) in seawater, 200 which is calculated according to the recent research (Kuss, 2014). The degree of Hg^0 saturation (S_a) 201 was calculated using the following equation: $S_a = H' DGM_{conc}/GEM_{conc}$, and the calculation of H' 202 203 (the dimensionless Henry's Law constant) has been reported in previous studies (Ci et al., 2011, 204 2015; Kuss, 2014).

205 **3** Results and discussion

206 **3.1** Speciated atmospheric Hg concentrations

Figure 2 shows the time series of speciated atmospheric Hg and meteorological parameters during the cruise in the SCS. The GEM concentration during the whole study period ranged from 0.92 to 4.12 ng m⁻³ with a mean value of 1.52 ± 0.32 ng m⁻³ (n = 4673), which was comparable to the average GEM levels over the global oceans (1.4–1.6 ng m⁻³, Soerensen et al., 2010a, 2013) and Atlantic Ocean (1.52 ± 0.32 ng m⁻³, Laurier and Mason, 2007), and higher than those at background sites in the Southern Hemisphere (0.85–1.05 ng m⁻³, Slemr et al., 2015; Howard et al., 2017), and also higher than those in remote oceans, such as the Cape Verde Observatory

station (1.19 \pm 0.13 ng m⁻³, Read et al., 2017), equatorial Pacific Ocean (1.15–1.05 ng m⁻³, 214 Soerensen et al., 2014) and Indian Ocean $(1.0-1.2 \text{ ng m}^{-3})$, Witt et al., 2010; Angot et al., 2014), 215 216 but lower than those in marginal seas, such as the Bohai Sea (BS), Yellow Sea (YS) and East China Sea (ECS) (Table 1). However, previous studies conducted in the northern SCS showed that 217 the average GEM concentrations in their study period ($2.6-3.5 \text{ ng m}^{-3}$, Fu et al., 2010; Tseng et al., 218 2012) were higher than that in this study. This is due to the fact that the GEM level in the northern 219 220 SCS (Fu et al., 2010; Tseng et al., 2012) were considerably higher than that in the western SCS 221 (this study).

The $Hg_{2,5}^{P}$ concentrations over the SCS ranged from 1.2 to 8.3 pg m⁻³ with a mean value of 222 $3.2 \pm 1.8 \text{ pg m}^{-3}$ (n = 39) (Fig. 2), which was higher than those observed at Nam Co (China) and 223 the Amsterdam Island, and were comparable to those in other coastal areas, such as the Okinawa 224 225 Island, the Nova Scotia, the Adriatic Sea, the Ontario lake and the Weeks Bay (see Table 1), but 226 lower than those in the BS and YS (Wang et al., 2016b), and considerably lower than those in 227 rural and urban sites, such as Xiamen, Seoul (see Table 1), Guiyang and Waliguan (Fu et al., 2011, 228 2012). The results showed that the SCS suffered less influence from human activities. The RGM concentration over the SCS ranged from 0.27 to 27.57 pg m⁻³ with a mean value of 6.1 \pm 5.8 pg 229 m^{-3} (n = 58), which was comparable to those in other seas, such as the North Pacific Ocean, the 230 North Atlantic Ocean and the Mediterranean Sea (including the Adriatic Sea) (Table 1), and higher 231 232 than the global mean RGM concentration in the MBL (Soerensen et al., 2010a), and also higher 233 than those measured at a few rural sites (Valente et al., 2007; Liu et al., 2010; Cheng et al., 2013, 234 2014), but significantly much lower than those polluted urban areas in China and South Korea, such as Guiyang (35.7 \pm 43.9 pg m⁻³, Fu et al., 2011), Xiamen, and Seoul (Table 1). Furthermore, 235 Figure 2 shows that the long-lived GEM has smaller variability compared to the short-lived 236 species like RGM and Hg^P_{2.5}, indicating that atmospheric reactive Hg was easily scavenged from 237 the marine atmosphere due not only to their characteristics (high activity and solubility) but also 238 239 due to their sensitivity to meteorological conditions and chemical environments. This pattern was 240 consistent with our previous observed patterns in the BS and YS (Wang et al., 2016b). Moreover, we found that atmospheric reactive Hg represents less than 1 % of TAM in the atmosphere, which 241 242 was comparable to those measured in other marginal and inner seas, such as the BS and YS (Wang 243 et al., 2016b), Adriatic Sea (Sprovieri and Pirrone, 2008), Okinawa Island (located in the ECS) (Chand et al., 2008), but was significantly lower than those at the urban sites (Table 1). 244

245 3.2 Spatial distribution of atmospheric Hg

246 3.2.1 Spatial distributions of GEM and RGM

The spatial distribution of GEM over the SCS is illustrated in Fig. 3a. The mean GEM concentration in the northern SCS $(1.73 \pm 0.40 \text{ ng m}^{-3} \text{ with a range of } 1.01-4.12 \text{ ng m}^{-3})$ was significantly higher than that in the western SCS $(1.41 \pm 0.26 \text{ ng m}^{-3} \text{ with a range of } 0.92-2.83 \text{ ng})$

 m^{-3}) (t-test, p < 0.01). Additionally, we found that the GEM concentrations in the PRE (the 250 average value > 2.00 ng m^{-3}) were significantly higher than those in the open SCS (see Figs. 2, 3a), 251 252 indicating that this nearshore area suffered from high GEM pollution in our study period probably 253 due to the surrounding human activities. Figure 3a shows that there was large difference in GEM 254 concentration between stations 1–10 and stations 16–31. The 72-h back-trajectories of air masses 255 showed that the air masses with low GEM levels between stations 1 and 10 mainly originated 256 from the SCS (Fig. S2a), while the air masses with high GEM levels at stations 16–31 primarily 257 originated from East China and ECS, and then passed over the southeast coastal regions of China 258 (Fig. S2b). Additionally, Fig. 3a shows that there was small variability of GEM concentrations over the western SCS except the measurements near the station 79. The back-trajectories showed 259 that the air masses with elevated GEM level near the station 79 originated from the south of the 260 261 Taiwan Island, while the other air masses mainly originated from the West Pacific Ocean (Fig. S3a) 262 and the Andaman Sea (Fig. S3b). Therefore, the air masses dominantly originated from sea and 263 ocean in this study period, and this could be the main reason for the low GEM level over the SCS. 264 In conclusion, GEM concentrations showed a conspicuous dependence on the sources and 265 movement patterns of air masses during this cruise.

The spatial distribution of RGM over the SCS is plotted in Fig. 3b. The mean RGM 266 concentration in the northern SCS $(7.1 \pm 1.4 \text{ pg m}^{-3})$ was also obviously higher than that in the 267 western SCS (3.8 \pm 0.7 pg m⁻³) (*t*-test, *p* < 0.05), indicating that a portion of RGM in the northern 268 SCS maybe originated from the anthropogenic emission. We observed elevated RGM 269 270 concentrations in the PRE, and which was consistent with the GEM distribution pattern, indicating 271 that part of the RGM near PRE probably originated from the surrounding human activities. This is confirmed by the following fact: The RGM concentrations in nighttime of the two days in the PRE 272 were 11.3 and 5.2 pg m^{-3} (Figs. 3b and S4), and they were significantly higher than those in the 273 open SCS. Another obvious feature is that the amplitude of RGM concentration is much greater 274 275 than the GEM, and this further indicated that the RGM was easily removed from the atmosphere through both the wet and dry deposition. In addition, we found that the RGM concentrations in the 276 nearshore area were not always higher than those in the open sea except the measurements in the 277 278 PRE, suggesting that the RGM in the remote marine atmosphere presumably not originated from land but from the in situ photo-oxidation of Hg⁰, which had been reported in previous studies (e.g., 279 Hedgecock and Pirrone, 2001; Lindberg et al., 2002; Laurier et al., 2003; Sprovieri et al., 2003, 280 2010; Sheu and Mason, 2004; Laurier and Mason, 2007; Soerensen et al., 2010a; Wang et al., 281 2015). 282

283 **3.2.2** Spatial distributions of $Hg_{2.5}^{P}$ and Hg_{10}^{P}

The concentrations and spatial distribution of $Hg_{2.5}^{P}$ in the MBL are illustrated in Fig. 4a. The highest $Hg_{2.5}^{P}$ value (8.3 pg m⁻³) was observed in the PRE during daytime on 4 September 2015 286 presumably due to the local human activities. The homogeneous distribution and lower level of $Hg_{2.5}^{P}$ in the open SCS indicated that the $Hg_{2.5}^{P}$ not originated from the land and the SCS suffered 287 288 less influence from human activities especially in the open sea. This is due to the fact that the 289 majority of air masses in the SCS during this study period came from the seas and oceans. The spatial distribution pattern of HgP2.5 in this study was different from our previous observed 290 patterns in the BS and YS (Wang et al., 2016b), which showed that Hg^P_{2.5} concentrations in 291 nearshore area were higher than those in the open sea both in spring and fall mainly due to the 292 outflow of atmospheric Hg^P from East China. 293

The concentrations and spatial distributions of Hg_{10}^{P} in the MBL of the SCS are illustrated in 294 Fig. 4b. We found that the Hg_{10}^{P} concentration was considerably (1–6 times) higher in the PRE 295 than those of other regions of the SCS probably due to the large emissions of anthropogenic Hg in 296 surrounding areas of the PRE. Moreover, the highest $Hg_{2,1}^{P}/Hg_{10}^{P}$ ratio (41 %) was observed in the 297 PRE and coastal sea area of Hainan Island, while lowest ratio (22 %) was observed in the open sea 298 (Fig. 4b). The Hg_{10}^{P} concentrations and $Hg_{2,1}^{P}/Hg_{10}^{P}$ ratios were higher in the nearshore area 299 compared to those in the open sea, demonstrating that coastal sea areas are polluted by 300 anthropogenic Hg to a certain extent. Interestingly, we found the mean Hg^P_{2.1} concentration (3.16 301 \pm 2.69 pg m⁻³, n = 10) measured using the Andersen sampler was comparable to the mean Hg^P_{2.5} 302 concentration (3.33 \pm 1.89 pg m⁻³, n = 39) measured using a 47 mm Teflon filter holder (*t*-test, *p* > 303 0.1). This indicated that the fine Hg^P level in the MBL of the SCS was indeed low, and there might 304 be no significant difference in Hg^P concentration in the SCS between 12 h and 48 h sampling time. 305

The concentrations of all size-fractioned Hg^P are summarized in Table S1. The size 306 distribution of Hg^P in the MBL of the SCS is plotted in Fig. 5. One striking feature is that the 307 three-modal pattern with peaks around <0.4 μ m, 0.7–1.1 μ m and 5.8–9.0 μ m was observed for the 308 size distributions of Hg^P in the open sea (Fig. 5a) if we excluded the data in the PRE. The 309 three-modal pattern was more obvious when we consider all the data (Fig. 5b). Generally, the Hg^P 310 concentrations in coarse particles were significantly higher than those in fine particles, and $Hg_{2,1}^{P}$ 311 contributed approximately 32 % (22–41 %, see Fig. 4b) to the Hg_{10}^{P} for the whole data, indicating 312 that the coarse mode was the dominant size during this study period. This might be explained by 313 314 the sources of the air masses. Since air masses dominantly originated from sea and ocean (Figs. S1, S2) and contained high concentrations of sea salts which generally exist in the coarse mode (1-10)315 μ m) (Athanasopoulou et al., 2008; Mamane et al., 2008), the Hg^P₂₁/Hg^P₁₀ ratios were generally 316 lower in the SCS compared to those in the BS, YS and ECS (Wang et al., 2016a). 317

318 **3.3** Dry deposition fluxes of RGM and Hg^P

The dry deposition flux of Hg_{10}^{P} was obtained by summing the dry deposition fluxes of each size-fractionated Hg^{P} in the same set. The dry deposition flux of Hg_{10}^{P} is calculated using the following equation: $F = \sum C \text{Hg}^{P} \times V_{d}$, the *F* is the dry deposition flux of Hg_{10}^{P} (ng m⁻² d⁻¹), $C \text{Hg}^{P}$

is the concentration of Hg^P in each size fraction (pg m⁻³), and V_d is the corresponding dry 322 deposition velocity (cm s⁻¹). In this study, the dry deposition velocities of 0.03, 0.01, 0.06, 0.15 323 and 0.55 cm s⁻¹ (Giorgi, 1988; Pryor et al., 2000; Nho-Kim et al., 2004) were chosen for the 324 following size-fractioned particles: < 0.4, 0.4-1.1, 1.1-2.1, 2.1-5.8 and 5.8-10 µm, respectively 325 (Wang et al., 2016a). The average dry deposition flux of Hg^{P₁₀} was estimated to be 1.08 ng m⁻² d⁻¹ 326 based on the average concentrations of each size-fractionated Hg^P in the SCS (Table S2), which 327 was lower than those in the BS, YS and ECS (Wang et al., 2016a). The dry deposition velocity of 328 RGM was 4.0–7.6 cm s^{-1} because of its characteristics and rapid uptake by sea salt aerosols 329 followed by deposition (Poissant et al., 2004; Selin et al., 2007). The annual dry deposition fluxes 330 of Hg^{P₁₀} and RGM to the SCS were calculated to be 1.42 and 27.39–52.05 tons yr⁻¹ based on the 331 average Hg^{P₁₀} and RGM concentrations and the area of the SCS ($3.56 \times 10^{12} \text{ m}^2$). The result 332 showed that RGM contributed more than 95 % to the total dry deposition of atmospheric reactive 333 Hg. The annual dry deposition flux of RGM was considerably higher than that of the Hg_{10}^{P} due to 334 the higher deposition rate and concentrations of RGM. 335

336 3.4 Temporal variation of atmospheric Hg

337 3.4.1 diurnal variation of GEM

The diurnal variation of GEM concentration during the whole study period is illustrated in Fig. 6. It was notable that there was no significant variability of the mean (\pm SD) GEM concentration in a whole day during this study period, and the GEM concentration dominantly fell in the range of 1.3–1.7 ng m⁻³ (Fig. 6). The statistical result showed that the mean GEM concentration in the daytime (6:00–18:00) (1.49 ± 0.06 pg m⁻³) was comparable to that in the nighttime (1.51 ± 0.06 pg m⁻³) (*t*-test, p > 0.05). The lower GEM concentrations and smaller variability over the SCS further revealed that the SCS suffered less influence of fresh emissions.

345 **3.4.2 Daily variation of RGM**

346 The average RGM concentrations in the daytime and nighttime are illustrated in Fig. 7. Firstly, it 347 could be found that RGM showed a diurnal variation with higher concentrations in the daytime 348 and lower concentrations in the nighttime during the whole study period. The mean RGM concentration in the daytime $(8.0 \pm 5.5 \text{ pg m}^{-3})$ was significantly and considerably higher than that 349 in the nighttime (2.2 \pm 2.7 pg m⁻³) (*t*-test, p < 0.001). This diurnal pattern was in line with the 350 previous multiple sites studies (Laurier and Mason, 2007; Liu et al., 2007; Engle et al., 2008; 351 352 Cheng et al., 2014). This is due to the fact that the oxidation of GEM in the MBL must be 353 photochemical, which have been evidenced by the diurnal cycle of RGM (Laurier and Mason, 354 2007). Another reason is that there was more Br (gas phase) production during daytime (Sander et 355 al., 2003). Figure S3 showed that the RGM concentration in the nighttime was lower than those in 356 corresponding forenoon and afternoon except the measurements in the PRE. This further indicated that (1) the RGM originated from the photo-oxidation of Hg^0 in the atmosphere and (2) the transfer of RGM to Hg^P due to higher RH and lower air temperature in nighttime (Rutter and Schauer, 2007; Lee et al., 2016).

360 In addition, we found that the difference in RGM concentration between day and night in the SCS was higher than those in the BS and YS (Wang et al., 2016b), and one possible reason is that 361 362 the solar radiation and air temperature over the SCS were stronger and higher compared to those 363 over the BS and YS (Wang et al., 2016b) as a result of the specific location of the SCS (tropical sea) and the different sampling season (the SCS: September 2015, the BS and YS: April-May and 364 365 November 2014). Secondly, it could be found that the higher the RGM concentrations in the 366 daytime, and the higher the RGM concentrations in the nighttime, but the concentrations in daytime were higher than that in the corresponding nighttime throughout the sampling period (see 367 368 Figs. 7, S3). This is partly because the higher RH and lower air temperature in nighttime were 369 conductive to the removal of RGM (Rutter and Schauer, 2007; Amos et al., 2012). Thirdly, we 370 found that the difference in RGM concentration between different days was large though there 371 was no significantly difference in PAR values (Fig. 7). However, here again divide two kinds of 372 cases: the first kind of circumstance is that the higher RGM in the PRE (day and night) presumably mainly originated from the surrounding human activities (i.e., 4–5 September 2015); 373 374 the second scenario is that RGM in open waters mainly originated from the in situ oxidation of 375 GEM in the MBL (Soerensen et al., 2010a; Sprovieri et al., 2010). The main reason for the large 376 difference in RGM concentration between different days was that there was large difference in 377 wind speed and RH between different days (see Fig. 2), and the discussion can be found in the 378 following paragraphs.

379 **3.4.3 Daily variation of Hg^P**_{2.5}

Figure 8 shows the Hg^P_{2.5} concentrations in the daytime and nighttime during the entire study 380 period. The Hg^P_{2.5} value in the daytime (3.4 \pm 1.9 pg m⁻³, n = 20) was slightly but not significantly 381 higher than that in the nighttime (2.4 \pm 0.9 pg m⁻³, n = 19) (*t*-test, p > 0.1), and this pattern was 382 383 consistent with the result of our previous study conducted in the open waters of YS (Wang et al., 2016b). The elevated Hg^P_{2.5} concentrations in the PRE and nearshore area of the Hainan Island 384 (Fig. 4 and Fig. 8) indicated that the nearshore areas were readily polluted due to the 385 anthropogenic Hg emissions, while the low $Hg_{2,5}^{P}$ level in the open sea further suggested that the 386 open areas of the SCS suffered less anthropogenic Hg^{P} . Therefore, we postulate that the $Hg^{P}_{2.5}$ 387 388 over the open SCS mainly originated from the in situ formation.

Buring the cruise in the western SCS (16–28 September 2015), we found elevated $Hg_{2.5}^{P}$ concentrations when the RGM concentrations were high at lower wind speed (e.g., 20–22 September 2015, it was sunny all these days) (see Figs. 2, 7, 8). This is probably due to the transferring of RGM from the gas to the particle phase. In contrast, we found that the $Hg_{2.5}^{P}$

- concentrations were elevated when the RGM concentrations were low at higher wind speed (e.g.,
- 394 25–27 September 2015, it was cloudy these days, and there was a transitory drizzly on 26
- September 2015) (see Figs. 2, 7, 8). On the one hand, high wind speed may increase the levels of
- halogen atoms (Br and Cl etc.) and sea salt aerosols in the marine atmosphere, which in turn were
- favorable to the production of RGM and formation of $Hg_{2.5}^{P}$ (Auzmendi-Murua et al., 2014). On
- 398 the other hand, high wind speed was favorable to the removal of RGM and $Hg_{2.5}^{P}$ in the
- atmosphere, this was probably the reason for lower RGM and $Hg_{2,5}^{P}$ concentrations during 25–27
- 400 September as compared to those observed during 20–22 September (see Fig. 2).

401 3.5 Relationship between atmospheric Hg and meteorological parameters

402 Pearson's correlation coefficients were calculated between speciated Hg and meteorological parameters to identify the relationships between them (Table 2). According to the correlation 403 analysis, the Hg^P_{2.5} was significantly positively correlated with RGM. Part of the reason was that 404 RGM could be adsorbed by particulate matter under high RGM concentrations and then enhanced 405 the Hg^P concentrations. Similarly, the Hg^P_{2.5} had a significantly positive correlation with GEM. On 406 the one hand, GEM and Hg^P probably originated from the same sources (including but not limited 407 to anthropogenic and oceanic sources) especially in the PRE and nearshore areas. On the other 408 hand, it was probably due to the fact that GEM could be oxidized to form RGM and then Hg^P, 409 which might be the reason for the positive but not significant correlation between RGM and GEM 410 411 since higher GEM level may result in higher RGM level in daytime.

The correlation analysis showed that the Hg^P_{2.5} and RGM were all negatively correlated with 412 wind speed and RH (Table 2), and the higher wind speed was favorable to the removal of $Hg_{2.5}^{P}$ 413 over the RGM. This is because the high wind speed might increase the RH levels and then 414 elevated wind speed and RH may accelerate the removal of Hg^P_{2,5} and RGM (Cheng et al., 2014; 415 Wang et al., 2016b). Moreover, both the air temperature and PAR were positively correlated with 416 RGM and Hg^P_{2.5}, and a significantly positive correlation was found between PAR and RGM, 417 418 indicating that the role of solar radiation played on the production of RGM was more obvious than that on the formation of HgP2.5, which were consistent with the previous study at coastal and 419 420 marine sites (Mao et al., 2012).

421 **3.6** Sea-air exchange of Hg⁰ in the SCS

The spatial distributions of DGM and Hg⁰ fluxes in the SCS are illustrated in Fig. 9. The DGM concentrations in nearshore area (40–55 pg l⁻¹) were about twice as high as those in the open sea, and this pattern was similar to our previous study conducted in the ECS (Wang et al., 2016c). The DGM concentration in this study varied from 23.0 to 66.8 pg l⁻¹ with a mean value of 37.1 ± 9.0 pg l⁻¹ (Fig. 9a and Table S3), which was higher than those in other open oceans, such as the Atlantic Ocean (11.6 ± 2.0 pg l⁻¹, Anderson et al., 2011), South Pacific Ocean (9–21 pg l⁻¹,

Soerensen et al., 2014), but considerably lower than that in the Minamata Bay (116 \pm 76 pg l⁻¹, 428 Marumoto et al., 2015). The mean DGM concentration in the northern SCS (41.3 \pm 10.9 pg l⁻¹) 429 was significantly higher than that in the western SCS (33.5 \pm 5.0 pg l⁻¹) (*t*-test, p < 0.01). The 430 431 reason was that DGM concentrations in the nearshore areas of the PRE and Hainan Island were 432 higher than those in the western open sea (see Fig. 9a). The DGM in surface seawater of the SCS 433 was supersaturated with a saturation of 501 % to 1468 % with a mean value of 903 \pm 208 %, which was approximately two thirds of that measured in the ECS (Wang et al., 2016c). The result 434 indicated that (1) the surface seawater in the SCS was supersaturated with gaseous Hg and (2) Hg^0 435 436 evaporated from the surface seawater to the atmosphere during our study period.

The sea-air exchange fluxes of Hg⁰ at each station are presented in Table S3, including GEM, 437 DGM, PAR, surface seawater temperature, wind speed and saturation of Hg⁰. Sea-air exchange 438 fluxes of Hg^0 in the SCS ranged from 0.40 to 12.71 ng m^{-2} h^{-1} with a mean value of 4.99 ± 3.32 439 ng m^{-2} h⁻¹ (Fig. 9b and Table S3), and which was comparable to the previous measurements 440 obtained in the Mediterranean Sea, the northern SCS and West Atlantic Ocean (Andersson et al., 441 442 2007; Fu et al., 2010; Soerensen et al., 2013), but lower than those in polluted marine 443 environments, such as the Minamata Bay, Tokyo Bay and YS (Narukawa et al., 2006; Ci et al., 2011; Marumoto et al., 2015), while higher than those in some open sea environments, such as the 444 Baltic Sea, Atlantic Ocean and South Pacific Ocean (Kuss and Schneider, 2007; Andersson et al., 445 2011; Kuss et al., 2011; Soerensen et al., 2014). Interestingly, we found the Hg⁰ flux near the 446 station 99 were higher than those in open water as a result of higher wind speed (Table S3). 447

In order to better understand the important role of the SCS, we relate the Hg⁰ flux in the SCS 448 to the global estimation, an annual sea-air flux of Hg⁰ was calculated based on the assumption that 449 there was no seasonal variation in Hg⁰ emission flux from the SCS. The annual emission flux of 450 Hg⁰ from the SCS was estimated to be 159.6 tons yr⁻¹ assuming the area of the SCS was $3.56 \times$ 451 10^{12} m² (accounting for about 1.0 % of the global ocean area), which constituted about 5.5 % of 452 the global Hg⁰ oceanic evasion (Strode et al., 2007; Soerensen et al., 2010b; UNEP, 2013). We 453 attributed the higher Hg⁰ flux in the SCS to the specific location of the SCS (tropical sea) and the 454 higher DGM concentrations in the SCS (especially in the northern area). Therefore, the SCS may 455 456 actually play an important role in the global Hg oceanic cycle. Additionally, we found that the percentage of the annual dry deposition flux of atmospheric reactive Hg to the annual evasion flux 457 of Hg⁰ was approximately 18–34 %, indicating that the dry deposition of atmospheric reactive Hg 458 was an important pathway for the atmospheric Hg to the ocean. 459

460 4 Conclusions

461 During the cruise aboard the R/V *Shiyan 3* in September 2015, GEM, RGM and Hg^{P} were 462 determined in the MBL of the SCS. The GEM level in the SCS was comparable to the background 463 level over the global oceans due to the air masses dominantly originated from seas and oceans.

464 GEM concentrations were closely related to the sources and movement patterns of air masses during this cruise. Moreover, the speciated atmospheric Hg level in the PRE was significantly 465 higher than those in the open SCS due to the anthropogenic emissions. The Hg^P concentrations in 466 coarse particles were significantly higher than those in fine particles, and the coarse modal was the 467 dominant size though there were three peaks for the size distribution of Hg^{P} in PM_{10} , indicating 468 that most of the Hg_{10}^{P} originated from in situ production. There was no significant difference in 469 GEM and Hg^P_{2.5} concentrations between day and night, but RGM concentrations were 470 significantly higher in daytime than in nighttime. RGM was positively correlated with PAR and air 471 472 temperature, but negatively correlated with wind speed and RH. The DGM concentrations in nearshore areas of the SCS were higher than those in the open sea, and the surface seawater of the 473 SCS was supersaturated with respect to Hg⁰. The annual flux of Hg⁰ from the SCS accounted for 474 about 5.5 % of the global Hg⁰ oceanic evasion though the area of the SCS just represents 1.0 % of 475 the global ocean area, suggesting that the SCS plays an important role in the global Hg cycle. 476 477 Additionally, the dry deposition of atmospheric reactive Hg was a momentous pathway for the 478 atmospheric Hg to the ocean because it happens all the time.

479 5 Appendix A

480 Table A1 List of acronyms and symbols

Abbreviation	Full name
BS	Bohai Sea
YS	Yellow Sea
ECS	East China Sea
SCS	South China Sea
PRE	Pearl River Estuary
MBL	Marine boundary layer
GEM	Gaseous elemental mercury
RGM	Reactive gaseous mercury
TAM	Total atmospheric mercury
Hg ^P _{2.1}	Particulate mercury in PM _{2.1}
Hg ^P _{2.5}	Particulate mercury in PM _{2.5}
$\mathrm{Hg}^{\mathrm{P}}_{10}$	Particulate mercury in PM ₁₀
DGM	Dissolved gaseous mercury

481 Data are available from the first author Chunjie Wang (888wangchunjie888@163.com).

482 Author contributions. XZ and ZW designed the study. CW and FH organized the mercury

483 measurements. CW performed the data analysis, and wrote the paper. All authors contributed to

the manuscript with discussions and comments.

485 Competing interests. The authors declare that they have no conflict of interest.

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492 References

- 493 Ahn, M. C., Kim, B., Holsen, T. M., Yi, S. M., and Han, Y. J.: Factors influencing concentrations of dissolved 494 gaseous mercury (DGM) and total mercury (TM) in an artificial reservoir, Environ. Pollut., 158, 347-355, 495 https://doi.org/10.1016/j.envpol.2009.08.036, 2010.
- 496 Amos, H. M., Jacob, D. J., Holmes, C. D., Fisher, J. A., Wang, Q., Yantosca, R. M., Corbitt, E. S., Galarneau, E.,
- 497 Rutter, A. P., Gustin, M. S., Steffen, A., Schauer, J. J., Graydon, J. A., St. Louis, V. L., Talbot, R. W., Edgerton, E.
- 498 S., Zhang, Y., and Sunderland, E. M.: Gas-particle partitioning of atmospheric Hg(II) and its effect on global
- 499 mercury deposition, Atmos. Chem. Phys., 12, 591-603, https://doi.org/10.5194/acp-12-591-2012, 2012.
- 500 Andersson, M. E., Gårdfeldt, K., Wängberg, I., Sprovieri, F., Pirrone, N., and Lindqvist, O.: Seasonal and daily 501 variation of mercury evasion at coastal and off shore sites from the Mediterranean Sea, Mar. Chem., 104, 502 214-226, https://doi.org/10.1016/j.marchem.2006.11.003, 2007.
- 503 Andersson, M. E., Sommar, J., Gårdfeldt, K., and Jutterström, S.: Air-sea exchange of volatile mercury in the 504 North Atlantic Ocean, Mar. Chem., 125, 1-7, https://doi.org/10.1016/j.marchem.2011.01.005, 2011.
- 505 Angot, H., Barret, M., Magand, O., Ramonet, M., Dommergue, A.: A 2-year record of atmospheric mercury
- 506 species at a background Southern Hemisphere station on Amsterdam Island, Atmos. Chem. Phys., 14, 507 11461-11473, https://doi.org/10.5194/acp-14-11461-2014, 2014
- 508 Ariya, P. A., Amyot, M., Dastoor, A., Deeds, D., Feinberg, A., Kos, G., Poulain, A., Ryjkov, A., Semeniuk, K.,
- Subir, M., and Toyota, K.: Mercury Physicochemical and Biogeochemical Transformation in the Atmosphere 510 and at Atmospheric Interfaces: A Review and Future Directions, Chem. Rev., 115, 3760-3802, 511 https://doi.org/10.1021/cr500667e, 2015.
- 512 Athanasopoulou, E., Tombrou, M., Pandis, S. N., and Russell, A. G.: The role of sea-salt emissions and
- 513 heterogeneous chemistry in the air quality of polluted coastal areas, Atmos. Chem. Phys., 8, 5755-5769,
- 514 https://doi.org/10.5194/acp-8-5755-2008, 2008.
- 515 Auzmendi-Murua, I., Castillo, Á., and Bozzelli, J. W.: Mercury oxidation via chlorine, bromine, and iodine under
- 516 atmospheric conditions: Thermochemistry and kinetics, J. Phys. Chem. A, 118, 2959-2975, 517 https://doi.org/10.1021/jp412654s, 2014.
- 518 Bowman, K. L., Hammerschmidt, C. R., Lamborg, C. H., and Swarr, G.: Mercury in the North Atlantic Ocean: the 519 U.S. Geotraces zonal and Meridional sections, Deep Sea Res. Part II, 116, 251-261, 520 https://doi.org/10.1016/j.dsr2.2014.07.004, 2015.

- 521 Chand, D., Jaffe, D., Prestbo, E., Swartzendruber, P. C., Hafner, W., Weiss-Penzias, P., Kato, S., Takami, A.,
- Hatakeyama, S., and Kajii, Y.: Reactive and particulate mercury in the Asian marine boundary layer, Atmos.
 Environ., 42, 7988–7996, https://doi.org/10.1016/j.atmosenv.2008.06.048, 2008.
- 524 Cheng, I., Zhang, L., Blanchard, P., Graydon, J. A., and St. Louis, V. L.: Source-receptor relationships for speciated
- 525 atmospheric mercury at the remote Experimental Lakes Area, northwestern Ontario, Canada, Atmos. Chem.
- 526 Phys., 12, 1903–1922, https://doi.org/10.5194/acp-12-1903-2012, 2012.
- 527 Cheng, I., Zhang, L., Blanchard, P., Dalziel, J., Tordon, R., Huang, J., and Holsen, T. M.: Comparisons of mercury
- sources and atmospheric mercury processes between a coastal and inland site, J. Geophys. Res., 118, 2434–2443,
- 529 https://doi.org/10.1002/jgrd.50169, 2013.
- 530 Cheng, I., Zhang, L., Mao, H., Blanchard, P., Tordon, R., and Dalziel, J.: Seasonal and diurnal patterns of speciated
- atmospheric mercury at a coastal-rural and a coastal-urban site, Atmos. Environ., 82, 193–205,
 https://doi.org/10.1016/j.atmosenv.2013.10.016, 2014.
- 533 Choi, H. D., Holsen, T. M., and Hopke, P. K.: Atmospheric mercury (Hg) in the Adirondacks: Concentrations and
 534 sources, Environ. Sci. Technol., 42, 5644–5653, https://doi.org/10.1021/es7028137, 2008.
- 535 Ci, Z., Zhang, X., Wang, Z., Niu, Z., Diao, X., and Wang, S.: Distribution and air–sea exchange of mercury (Hg) in
- the Yellow Sea, Atmos. Chem. Phys., 11, 2881–2892, https://doi.org/10.5194/acp-11-2881-2011, 2011.
- 537 Ci, Z., Wang, C., Wang, Z., and Zhang, X.: Elemental mercury (Hg(0)) in air and surface waters of the Yellow Sea
 538 during late spring and late fall 2012: Concentration, spatial-temporal distribution and air/sea flux, Chemosphere,
- 539 119, 199–208, https://doi.org/10.1016/j.chemosphere.2014.05.064, 2015.
- de Foy, B., Tong, Y., Yin, X., Zhang, W., Kang, S., Zhang, Q., Zhang, G., Wang, X., Schauer, J. J.: First field-based
- atmospheric observation of the reduction of reactive mercury driven by sunlight, Atmos. Environ., 134, 27-39,
- 542 https://doi.org/10.1016/j.atmosenv.2016.03.028, 2016.
- 543 Draxler, R. R., and Rolph, G. D.: HYSPLITModel access via NOAA ARL READY Website
 544 (http://www.arl.noaa.gov/ready/hysplit4.html), NOAA Air Resources Laboratory, Silver Spring, MD, 2012.
- 545 Engle, M. A., Tate, M. T., Krabbenhoft, D. P., Kolker, A., Olson, M. L., Edgerton, E. S., DeWild, J. F., and
- 546 McPherson, A. K.: Characterization and cycling of atmospheric mercury along the central US Gulf Coast, Appl.
- 547 Geochem., 23, 419–437, https://doi.org/10.1016/j.apgeochem.2007.12.024, 2008.
- Feddersen, D. M., Talbot, R., Mao, H., and Sive, B. C.: Size distribution of particulate mercury in marine and
 coastal atmospheres, Atmos. Chem. Phys., 12, 10899–10909, https://doi.org/10.5194/acp-12-10899-2012, 2012.
- 550 Fu, X., Feng, X., Zhang, G., Xu, W., Li, X., Yao, H., Liang, P., Li, J., Sommar, J., Yin, R., and Liu, N.: Mercury in
- the marine boundary layer and seawater of the South China Sea: Concentrations, sea/air flux, and implication for
- 552 land outflow, J. Geophys. Res., 115, D06303, https://doi.org/10.1029/2009JD012958, 2010.
- 553 Fu, X., Feng, X., Qiu, G., Shang, L., and Zhang, H.: Speciated atmospheric mercury and its potential source in
- 554 Guiyang, China, Atmos. Environ., 45, 4205–4212, https://doi.org/10.1016/j.atmosenv.2011.05.012, 2011.
- 555 Fu, X., Feng, X., Liang, P., Deliger, Zhang, H., Ji, J., and Liu, P.: Temporal trend and sources of speciated
- atmospheric mercury at Waliguan GAW station, Northwestern China, Atmos. Chem. Phys., 12, 1951–1964,
- 557 https://doi.org/10.5194/acp-12-1951-2012, 2012.

- 558 Gratz, L. E., Ambrose, J. L., Jaffe, D. A., Shah, V., Jaegle, L., Stutz, J., Festa, J., Spolaor, M., Tsai, C., Selin, N. E.,
- 559 Song, S., Zhou, X., Weinheimer, A. J., Knapp, D. J., Montzka, D. D., Flocke, F. M., Campos, T. L., Apel, E.,
- 560 Hornbrook, R., Blake, N. J., Hall, S., Tyndall, G. S., Reeves, M., Stechman, D., and Stell, M.: Oxidation of
- 561 mercury by bromine in the subtropical Pacific free troposphere, Geophys. Res. Lett., 42, 10494–10502,
- 562 https://doi.org/10.1002/2015GL066645, 2015.
- Giorgi, F.: Dry deposition velocities of atmospheric aerosols as inferred by applying a particle dry deposition
 parameterisation to a general circulation model, Tellus, 40B, 23–41,
 https://doi.org/10.1111/j.1600-0889.1988.tb00210.x, 1988.
- 566 Gustin, M. S., Huang, J., Miller, M. B., Peterson, C., Jaffe, D. A., Ambrose, J., Finley, B. D., Lyman, S. N., Call,
- K., Talbot, R., Feddersen, D., Mao, H., and Lindberg, S. E.: Do we understand what the mercury speciation
 instruments are actually measuring? Results of RAMIX, Environ. Sci. Technol., 47, 7295–7306,
 https://doi.org/10.1021/es3039104, 2013.
- 570 Hedgecock, I. and Pirrone, N.: Mercury and photochemistry in the marine boundary layer-modelling studies
 571 suggest the in situ production of reactive gas phase mercury, Atmos. Environ., 35, 3055–3062,
 572 https://doi.org/10.1016/S1352-2310(01)00109-1, 2001.
- Holmes, C. D., Jacob, D. J., and Yang, X.: Global lifetime of elemental mercury against oxidation by atomic
 bromine in the free troposphere, Geophys. Res. Lett., 33, L20808, https://doi.org/10.1029/2006gl027176, 2006.
- 575 Holmes, C. D., Jacob, D. J., Mason, R. P., and Jaffe, D. A.: Sources and deposition of reactive gaseous mercury in
- the marine atmosphere, Atmos. Environ., 43, 2278–2285, https://doi.org/10.1016/j.atmosenv.2009.01.051, 2009.
- 577 Holmes, C. D., Jacob, D. J., Corbitt, E. S., Mao, J., Yang, X., Talbot, R., and Slemr, F.: Global atmospheric model
- 578 for mercury including oxidation by bromine atoms, Atmos. Chem. Phys., 10, 12037–12057,
 579 https://doi.org/10.5194/acp-10-12037-2010, 2010.
- 580 Horowitz, H. M., Jacob, D. J., Zhang, Y., Dibble, T. S., Slemr, F., Amos, H. M., Schmidt, J. A., Corbitt, E. S.,
- 581 Marais, E. A., and Sunderland, E. M.: A new mechanism for atmospheric mercury redox chemistry: implications
 582 for the global mercury budget, Atmos. Chem. Phys., 17, 6353-6371, https://doi.org/10.5194/acp-17-6353-2017,
 583 2017.
- Horvat, M., Kotnik, J., Logar, M., Fajon, V., Zvonarić, T., and Pirrone, N.: Speciation of mercury in surface and
 deep-sea waters in the Mediterranean Sea, Atmos. Environ., 37, S93–S108,
 https://doi.org/10.1016/S1352-2310(03)00249-8, 2003.
- 587 Howard, D., Nelson, P. F., Edwards, G. C., Morrison, A. L., Fisher, J. A., Ward, J., Harnwell, J., van der Schoot, M.,
- 588 Atkinson, B., Chambers, S. D., Griffiths, A. D., Werczynski, S., and Williams, A. G.: Atmospheric mercury in
- 589 the Southern Hemisphere tropics: seasonal and diurnal variations and influence of inter-hemispheric transport,
- 590 Atmos. Chem. Phys., 17, 11623-11636, https://doi.org/10.5194/acp-17-11623-2017, 2017.
- Huang, J., Miller, M. B., Edgerton, E., and Sexauer Gustin, M.: Deciphering potential chemical compounds of
 gaseous oxidized mercury in Florida, USA, Atmos. Chem. Phys., 17, 1689-1698,
- 593 https://doi.org/10.5194/acp-17-1689-2017, 2017.
- 594 Kim, S. H., Han, Y. J., Holsen, T. M., and Yi, S. M.: Characteristics of atmospheric speciated mercury

- concentrations (TGM, Hg(II) and Hg(p)) in Seoul, Korea, Atmos. Environ., 43, 3267–3274,
 https://doi.org/10.1016/j.atmosenv.2009.02.038, 2009.
- Kim, P. R., Han, Y. J., Holsen, T. M., and Yi, S. M.: Atmospheric particulate mercury: Concentrations and size distributions, Atmos. Environ., 61, 94–102, https://doi.org/10.1016/j.atmosenv.2012.07.014, 2012.
- 599 Kuss, J.: Water-air gas exchange of elemental mercury: An experimentally determined mercury diffusion
- 600 coefficient for Hg⁰ water-air flux calculations, Limnol. Oceanogr., 59, 1461-1467,
- 601 https://doi.org/10.4319/lo.2014.59.5.1461, 2014.
- Kuss, J., and Schneider, B.: Variability of the gaseous elemental mercury sea-air flux of the Baltic Sea, Environ.
 Sci. Technol., 41, 8018–8023, https://doi.org/10.1021/es0716251, 2007.
- 604 Kuss, J., Zülicke, C., Pohl, C., and Schneider, B.: Atlantic mercury emission determined from continuous analysis
- of the elemental mercury sea-air concentration difference within transects between 50°N and 50°S, Global
 Biogeochem. Cycles, 25, GB 3021, https://doi.org/10.1029/2010GB003998, 2011.
- Kuss, J., Krüger, S., Ruickoldt, J., and Wlost, K.-P.: High-resolution measurements of elemental mercury in
 surface water for an improved quantitative understanding of the Baltic Sea as a source of atmospheric mercury,
- 609 Atmos. Chem. Phys., 18, 4361-4376, https://doi.org/10.5194/acp-18-4361-2018, 2018.
- Landis, M. S., Stevens, R. K., Schaedlich, F., and Prestbo, E. M.: Development and characterization of an annular
 denuder methodology for the measurement of divalent inorganic reactive gaseous mercury in ambient air,
 Environ. Sci. Technol., 36, 3000–3009, https://doi.org/10.1021/es015887t, 2002.
- 613 Laurier, F. J. G., Mason, R. P., Whalin, L., and Kato, S.: Reactive gaseous mercury formation in the North Pacific
- 614 Ocean's marine boundary layer: A potential role of halogen chemistry, J. Geophys. Res., 108, 4529,
 615 https://doi.org/10.1029/2003JD003625, 2003.
- Laurier, F., and Mason, R.: Mercury concentration and speciation in the coastal and open ocean boundary layer, J.
- 617 Geophys. Res., 112, D06302, https://doi.org/10.1029/2006JD007320, 2007.
- Lee, G.-S., Kim, P.-R., Han, Y.-J., Holsen, T. M., Seo, Y.-S., and Yi, S.-M.: Atmospheric speciated mercury
- concentrations on an island between China and Korea: sources and transport pathways, Atmos. Chem. Phys., 16,
- 620 4119-4133, https://doi.org/10.5194/acp-16-4119-2016, 2016.
- 621 Lindberg, S. E., Brooks, S., Lin, C. J., Scott, K. J., Landis, M. S., Stevens, R. K., Goodsite, M., and Richter, A.:
- Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise, Environ. Sci. Technol., 36,
 1245–1256, https://doi.org/10.1021/es0111941, 2002.
- Liss, P. W., and Slater, P. G.: Flux of gases across the air-sea interface, Nature, 247, 181–184,
 https://doi.org/10.1038/247181a0, 1974.
- Liu, B., Keeler, G. J., Dvonch, J. T., Barres, J. A., Lynam, M. M., Marsik, F. J., and Morgan, J. T.: Temporal
- variability of mercury speciation in urban air, Atmos. Environ., 41, 1911–1923,
 https://doi.org/10.1016/j.atmosenv.2006.10.063, 2007.
- 629 Liu, B., Keeler, G. J., Dvonch, J. T., Barres, J. A., Lynam, M. M., Marsik, F. J., and Morgan, J. T.: Urban-rural
- 630 differences in atmospheric mercury speciation, Atmos. Environ., 44, 2013–2023,
- 631 https://doi.org/10.1016/j.atmosenv.2010.02.012, 2010.

- Liu, N., Qiu, G., Landis, M., Feng, X., Fu, X., and Shang, L.: Atmospheric mercury species measured in Guiyang,
- Guizhou province, southwest China, Atmos. Res., 100, 93–102, https://doi.org/10.1016/j.atmosres.2011.01.002,
 2011.
- Mamane, Y., Perrino, C., Yossef, O., and Catrambone, M.: Source characterization of fine and coarse particles at
 the East mediterranean coast, Atmos. Environ., 42, 6114–6130, doi:10.1016/j.atmosenv.2008.02.045, 2008.
- 637 Mao, H., Talbot, R., Hegarty, J., and Koermer, J.: Speciated mercury at marine, coastal, and inland sites in New
- England Part 2: Relationships with atmospheric physical parameters, Atmos. Chem. Phys., 12, 4181–4206,
- 639 https://doi.org/10.5194/acp-12-4181-2012, 2012.
- 640 Mao, H., Cheng, I., and Zhang, L.: Current understanding of the driving mechanisms for spatiotemporal variations
- 641 of atmospheric speciated mercury: a review, Atmos. Chem. Phys., 16, 12897–12924,
 642 https://doi.org/10.5194/acp-16-12897-2016, 2016.
- Mao, H., Hall, D., Ye, Z., Zhou, Y., Felton, D., and Zhang, L.: Impacts of large-scale circulation on urban ambient
 concentrations of gaseous elemental mercury in New York, USA, Atmos. Chem. Phys., 17, 11655–11671,
 https://doi.org/10.5194/acp-17-11655-2017, 2017.
- 646 Marumoto, K., and Imai, S.: Determination of dissolved gaseous mercury in seawater of Minamata Bay and 647 estimation for mercury exchange across air-sea interface, Mar. Chem., 168, 9-17. 648 https://doi.org/10.1016/j.marchem.2014.09.007, 2015.
- 649 Mason, R. P., Choi, A. L., Fitzgerald, W. F., Hammerschmidt, C. R., Lamborg, C. H., Soerensen, A. L., and
- 50 Sunderland, E. M.: Mercury biogeochemical cycling in the ocean and policy implications, Environ. Res., 119,
- **651** 101–117, doi:10.1016/j.envres.2012.03.013, 2012.
- Mason, R. P., Hammerschmidt, C. R., Lamborg, C. H., Bowman, K. L., Swarr, G. J., and Shelley, R. U.: The air-sea
- exchange of mercury in the low latitude Pacific and Atlantic Oceans, Deep Sea Res. Part I, 122, 17-28,
- 654 https://doi.org/10.1016/j.dsr.2017.01.015, 2017.
- Narukawa, M., Sakata, M., Marumoto, K., and Asakura, K.: Air-sea exchange of mercury in Tokyo Bay, J.
 Oceanogr., 62, 249–257, https://doi.org/10.1007/s10872-006-0049-3, 2006.
- Nho-Kim, E. Y., Michou, M., and Peuch, V. H.: Parameterization of size-dependent particle dry deposition
 velocities for global modeling, Atmos. Environ., 38, 1933–1942, https://doi.org/10.1016/j.atmosenv.2004.01.002,
 2004.
- 660 Poissant, L., Pilote, M., Xu, X., Zhang, H. and Beauvais C.: Atmospheric mercury speciation and deposition in the
- 661 Bay St. François wetlands, J. Geophys. Res., 109, D11301, https://doi.org/10.1029/2003JD004364, 2004.
- Pryor, S. C., and Sorensen, L. L.: Nitric acid-sea salt reactions: Implications for nitrogen deposition to water
 surfaces, J. Appl. Meteorol., 39, 725–731, https://doi.org/10.1175/1520-0450-39.5.725, 2000.
- Radke, L. F., Friedli, H. R., and Heikes, B. G.: Atmospheric mercury over the NE Pacific during spring 2002:
- Gradients, residence time, upper troposphere lower stratosphere loss, and long-range transport, J. Geophys. Res.,
- 666 112, D19305, https://doi.org/10.1029/2005JD005828, 2007.
- 667 Read, K. A., Neves, L. M., Carpenter, L. J., Lewis, A. C., Fleming, Z. L., and Kentisbeer, J.: Four years
- 668 (2011–2015) of total gaseous mercury measurements from the Cape Verde Atmospheric Observatory, Atmos.

- 669 Chem. Phys., 17, 5393-5406, https://doi.org/10.5194/acp-17-5393-2017, 2017.
- 670 Rutter, A. P., and Schauer, J. J.: The effect of temperature on the gas-particle partitioning of reactive mercury in
- 671 atmospheric aerosols, Atmos. Environ., 41, 8647–8657, https://doi.org/10.1016/j.atmosenv.2007.07.024, 2007.
- 572 Sander, R., Keene, W. C., Pszenny, A. A. P., Arimoto, R., Ayers, G. P., Baboukas, E., Cainey, J. M., Crutzen, P. J.,
- Duce, R. A., Hönninger, G., Huebert, B. J., Maenhaut, W., Mihalopoulos, N., Turekian, V. C., and Van Dingenen,
- R., Inorganic bromine in the marine boundary layer: a critical review, Atmos. Chem. Phys., 3, 1301–1336,
- 675 https://doi.org/10.5194/acp-3-1301-2003, 2003.
- 676 Schroeder, W. H., and Munthe, J.: Atmospheric mercury An overview, Atmos. Environ., 32, 809–822,
 677 https://doi.org/10.1016/S1352-2310(97)00293-8, 1998.
- Selin, N. E., Jacob, D. J., Park, R. J., Yantosca, R. M., Strode, S., Jaegl é, L., and Jaffe, D.: Chemical cycling and
 deposition of atmospheric mercury: Global constraints from observations, J. Geophys. Res., 112, D02308,
 https://doi.org/10.1029/2006JD007450, 2007.
- 581 Shah, V., Jaegl & L., Gratz, L. E., Ambrose, J. L., Jaffe, D. A., Selin, N. E., Song, S., Campos, T. L., Flocke, F. M.,
- 682 Reeves, M., Stechman, D., Stell, M., Festa, J., Stutz, J., Weinheimer, A. J., Knapp, D. J., Montzka, D. D.,
- 583 Tyndall, G. S., Apel, E. C., Hornbrook, R. S., Hills, A. J., Riemer, D. D., Blake, N. J., Cantrell, C. A., and
- 684 Mauldin III, R. L.: Origin of oxidized mercury in the summertime free troposphere over the southeastern US,
- 685 Atmos. Chem. Phys., 16, 1511-1530, https://doi.org/10.5194/acp-16-1511-2016, 2016.
- Sheu, G. R., and Mason, R. P.: An examination of the oxidation of elemental mercury in the presence of halide
 surfaces, J. Atmos. Chem., 48, 107–130, https://doi.org/10.1023/B:JOCH.0000036842.37053.e6, 2004.
- 688 Slemr, F., Angot, H., Dommergue, A., Magand, O., Barret, M., Weigelt, A., Ebinghaus, R., Brunke, E.-G.,
- 689 Pfaffhuber, K. A., Edwards, G., Howard, D., Powell, J., Keywood, M., and Wang, F.: Comparison of mercury
- 690 concentrations measured at several sites in the Southern Hemisphere, Atmos. Chem. Phys., 15, 3125-3133,
- 691 https://doi.org/10.5194/acp-15-3125-2015, 2015.
- Soerensen, A. L., Skov, H., Jacob, D. J., Soerensen, B. T., and Johnson, M. S.: Global concentrations of gaseous
 elemental mercury and reactive gaseous mercury in the marine boundary layer, Environ. Sci. Technol., 44,
 7425–7430, https://doi.org/10.1021/es903839n, 2010a.
- Soerensen, A. L., Sunderland, E. M., Holmes, C. D., Jacob, D. J., Yantosca, R. M., Skov, H., Christensen, J. H.,
 Strode, S. A., and Mason, R. P.: An improved global model for air-sea exchange of mercury: high concentrations
- 697 over the North Atlantic, Environ. Sci. Technol., 44, 8574–8580, https://doi.org/10.1021/es102032g, 2010b.
- 698 Soerensen, A. L., Mason, R. P., Balcom, P. H., and Sunderland, E. M.: Drivers of surface ocean mercury
- concentrations and air-sea exchange in the west Atlantic Ocean, Environ. Sci. Technol., 47, 7757-7765,
 https://doi.org/10.1021/es401354q, 2013.
- 701 Soerensen, A. L., Mason, R. P., Balcom, P. H., Jacob, D. J., Zhang, Y., Kuss, J., and Sunderland, E. M.: Elemental
- mercury concentrations and fluxes in the tropical atmosphere and ocean, Environ. Sci. Technol., 48,
- 703 11312–11319, https://doi.org/10.1021/es503109p, 2014.
- 704 Sprovieri, F., Pirrone, N., Gärdfeldt, K., and Sommar, J.: Mercury speciation in the marine boundary layer along a
- 705 6000 km cruise path around the Mediterranean Sea, Atmos. Environ., 37, S63-S71,

- 706 https://doi.org/10.1016/S1352-2310(03)00237-1, 2003.
- Sprovieri, F., and Pirrone, N.: Spatial and temporal distribution of atmospheric mercury species over the Adriatic
 Sea, Environ. Fluid Mech., 8, 117–128, https://doi.org/10.1007/s10652-007-9045-4, 2008.
- 709 Sprovieri, F., Hedgecock, I. M., and Pirrone, N.: An investigation of the origins of reactive gaseous mercury in the
- 710 Mediterranean marine boundary layer, Atmos. Chem. Phys., 10, 3985–3997,
 711 https://doi.org/10.5194/acp-10-3985-2010, 2010.
- 712 Steffen, A., Lehnherr, I., Cole, A., Ariya, P., Dastoor, A., Durnford, D., Kirk, J., and Pilote, M.: Atmospheric
- 713 mercury measurements in the Canadian Arctic Part 1: A review of recent field measurements, Sci. Total Environ.,
- 714 509–510, 3–15, https://doi.org/10.1016/j.scitotenv.2014.10.109, 2015.
- 715 Strode, S. A., Jaegl é, L., Selin, N. E., Jacob, D. J., Park, R. J., Yantosca, R. M., Mason, R. P., and Slemr, F.: Air-sea
- 716 exchange in the global mercury cycle, Global Biogeochem. Cycles, 21, GB1017,
 717 https://doi.org/10.1029/2006GB002766, 2007.
- 718 Tseng, C. M., Liu, C. S., and Lamborg, C.: Seasonal changes in gaseous elemental mercury in relation to monsoon 719 cycling over the northern South China Sea. Atmos. Chem. Phys., 12, 7341-7350, 720 https://doi.org/10.5194/acp-12-7341-2012, 2012.
- 721 UNEP: Global Mercury Assessment: Sources, Emissions, Releases and Environmental Transport, UNEP
 722 Chemicals Branch, Geneva, Switzerland, 2013.
- Valente, R. J., Shea, C., Humes, K. L., and Tanner, R. L.: Atmospheric mercury in the Great Smoky Mountains
 compared to regional and global levels, Atmos. Environ., 41, 1861–1873,
- 725 https://doi.org/10.1016/j.atmosenv.2006.10.054, 2007.
- 726 Wang, Y. Q., Zhang, X. Y., and Draxler, R. R.: TrajStat: GIS-based software that uses various trajectory statistical
- analysis methods to identify potential sources from long-term air pollution measurement data, Environ. Model.
- 728 Softw., 28, 938–939, https://doi.org/10.1016/j.envsoft.2009.01.004, 2009.
- Wang, S., Schmidt, J. A., Baidar, S., Coburn, S., Dix, B., Koenig, T. K., Apel, E., Bowdalo, D., Campos, T. L.,
- 730 Eloranta, E., Evans, M. J., DiGangi, J. P., Zondlo, M. A., Gao, R. S., Haggerty, J. A., Hall, S. R., Hornbrook, R.
- 731 S., Jacob, D., Morley, B., Pierce, B., Reeves, M., Romashkin, P., ter Schure, A., and Volkamer, R.: Active and
- videspread halogen chemistry in the tropical and subtropical free troposphere, Proc. Natl. Acad. Sci. U.S.A.,
- 733 112, 9281–9286, https://doi.org/10.1073/pnas.1505142112, 2015.
- Wang, C., Wang, Z., Ci, Z., Zhang, X., and Tang, X.: Spatial-temporal distributions of gaseous element mercury
 and particulate mercury in the Asian marine boundary layer, Atmos. Environ., 126, 107–116,
- 736 https://doi.org/10.1016/j.atmosenv.2015.11.036, 2016a.
- 737 Wang, C., Ci, Z., Wang, Z., Zhang, X., and Guo, J.: Speciated atmospheric mercury in the marine boundary layer 738 Yellow of the Bohai Sea and Sea, Atmos. Environ., 131, 360-370, 739 https://doi.org/10.1016/j.atmosenv.2016.02.021, 2016b.
- **755** https://doi.org/10.1010/j.atmosenv.2010.02.021, 20100.
- 740 Wang, C., Ci, Z., Wang, Z., and Zhang, X.: Air-sea exchange of gaseous mercury in the East China Sea, Environ.
- 741 Pollut., 212, 535–543, https://doi.org/10.1016/j.envpol.2016.03.016, 2016c.
- 742 Wanninkhof, R.: Relationship between wind speed and gas exchange over the ocean, J. Geophys. Res., 97,

- 743 7373–7382, https://doi.org/10.1029/92JC00188, 1992.
- Witt, M. L. I., Mather, T. A., Baker, A. R., De Hoog, J. C. M., and Pyle, D. M.: Atmospheric trace metals over the
- south-west Indian Ocean: Total gaseous mercury, aerosol trace metal concentrations and lead isotope ratios, Mar.
 Chem., 121, 2–16, https://doi.org/10.1016/j.marchem.2010.02.005, 2010.
- 747 Xu, L., Chen, J., Yang, L., Niu, Z., Tong, L., Yin, L., and Chen, Y.: Characteristics and sources of atmospheric
- 748 mercury speciation in a coastal city, Xiamen, China, Chemosphere, 119, 530–539,
- 749 https://doi.org/10.1016/j.chemosphere.2014.07.024, 2015.
- 750 Ye, Z., Mao, H., Lin, C.-J., and Kim, S. Y.: Investigation of processes controlling summertime gaseous elemental
- 751 mercury oxidation at midlatitudinal marine, coastal, and inland sites, Atmos. Chem. Phys., 16, 8461–8478,
 752 https://doi.org/10.5194/acp-16-8461-2016, 2016.
- 753 Zhu, J., Wang, T., Talbot, R., Mao, H., Yang, X., Fu, C., Sun, J., Zhuang, B., Li, S., Han, Y., and Xie, M.:
- 754 Characteristics of atmospheric mercury deposition and size-fractionated particulate mercury in urban Nanjing,
- 755 China, Atmos. Chem. Phys., 14, 2233–2244, https://doi.org/10.5194/acp-14-2233-2014, 2014.
- 756

757 Figures and Tables



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Figure 1. Map of the South China Sea (a) (1: China, 2: Philippines, 3: Brunei, 4: Malaysia, 5:
Indonesia, 6: Myanmar, 7: Thailand, 8: Laos, 9: Cambodia, 10: Vietnam, 11: Singapore). The
locations of the Pearl River Estuary (PRE), DGM sampling stations and R/V tracks (b). It should
be noted that the black solid points represent the sampling stations, and the number near the black
solid point represents the name of the station.



Figure 2. Time (local time) series of GEM, Hg^P_{2.5}, RGM and some meteorological parameters, including relative humidity (RH), air temperature and wind speed ("A" represents the data measured in the PRE, "B" represents the data measured in the northern SCS, "C" represents the data obtained in the port of Sanya, "D" represents the data measured in the western SCS). It was rainy day on the days of 8 and 26 September 2015.



Figure 3. The concentrations and spatial distributions of GEM (a) and RGM (b) in the MBL of theSCS.







Figure 5. Size distributed concentrations of Hg^{P} (PM₁₀) in the MBL of the SCS, (a) represents all the data excepting the measurements in the PRE; (b) represents all the data. The data shown are the mean and standard error.



Figure 6. Diurnal variation of GEM concentration (mean \pm SD) over the SCS.



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Figure 7. Daily variation of RGM concentration over the SCS.





Figure 8. Daily variation of $Hg_{2.5}^{P}$ in the MBL of the SCS. The light gray area represents the data in the PRE, while the light magenta area represents the data in the nearshore area of the Hainan Island.



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Figure 9. DGM concentrations (a) and sea-air exchange flux of Hg^0 (b) in the SCS.

791	Table 1. The GEM, Hg	P _{2.5} and RGM concentrations	in this study and other literature.
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Location		Classification	Sampling time	$\begin{array}{c} \text{GEM} \\ \text{(ng m}^{-3}) \end{array}$	$Hg_{2.5}^{P}$ (pg m ⁻³)	$\begin{array}{c} \text{RGM} \\ \text{(pg m}^{-3}) \end{array}$	Reference
China	SCS	Sea	2015	$1.52\!\pm\!0.32$	3.2 ± 1.8	$6.1\ \pm 5.8$	This study
	BS and YS	Sea	2014 (Spring)	$2.03\ \pm 0.72$	$11.3\ \pm 18.5$	$2.5\ \pm 1.7$	Wang et al., 2016a, b
	BS and YS	Sea	2014 (Fall)	$2.09\ \pm 1.58$	9.0 ± 9.0	$4.3\ \pm 2.5$	Wang et al., 2016a, b
	YS	Sea	2010 (Summer)	$2.61\ \pm 0.50$	NA ^a	NA	Ci et al., 2011
	YS	Sea	2012 (Spring)	1.86 ± 0.40	NA	NA	Ci et al., 2015
	YS	Sea	2012 (Fall)	$1.84\ \pm 0.50$	NA	NA	Ci et al., 2015
	ECS	Sea	2013 (Summer)	$1.61\ \pm 0.32$	NA	NA	Wang et al., 2016c
	ECS	Sea	2013 (Fall)	$2.20\ \pm 0.58$	NA	NA	Wang et al., 2016c
	Northern SCS	Sea	2007	$2.62\ \pm 1.13$	NA	NA	Fu et al., 2010
	Northern SCS	Sea	2003-2005	2.8-5.7	NA	NA	Tseng et al., 2012
	Nam Co	lake	2014-2015	$0.95\ \pm 0.37$	$0.85\ \pm 2.91$	$49.0~{\pm}60.3$	de Foy et al., 2016
	Xiamen	Coastal urban	2012-2013	3.50	61.05	174.41	Xu et al., 2015
Japan	Okinawa Island	Ocean	2004	$2.04\ \pm 0.38$	3.0 ± 2.5	$4.5\ \pm 5.4$	Chand et al., 2008
Korea	Seoul	Urban	2005-2006	$3.22~{\pm}2.10$	$23.9\ \pm 19.6$	$27.2~{\pm}19.3$	Kim et al., 2009
USA	Weeks Bay	Coast	2005-2006	1.6 ± 0.3	2.7 ± 3.4	$4.0\ \pm 7.5$	Engle et al., 2008
Canada	Ontario Lake	Remote area	2005-2006	$1.57\ \pm 0.22$	$4.42~\pm3.67$	$0.99\ \pm 1.89$	Cheng et al., 2012
	Nova Scotia	Coast	2010-2011	$1.67\ \pm 1.01$	$2.32\ \pm 3.09$	$2.07\ \pm 3.35$	Cheng et al., 2013
	Nova Scotia	Coast-rural	2010-2011	$1.38\ \pm 0.20$	$3.5~{\pm}4.5$	$0.4\ \pm 1.0$	Cheng et al., 2014
Australia	ATARS ^b	Coast	2014-2015	$0.95\ \pm 0.12$	NA	NA	Howard et al., 2017
South-we	st India Ocean	Ocean	2007	1.24 ± 0.06	NA	NA	Witt et al., 2010
North Atl	antic Ocean	Ocean	2003	$1.63\ \pm 0.08$	NA	$5.9\ \pm 4.9$	Laurier et al., 2007
West Atla	antic Ocean	Ocean	2008-2010	1.4-1.5	NA	NA	Soerensen et al., 2013
North Pac	cific Ocean	Ocean	2002	2.5	NA	9.5	Laurier et al., 2003
Pacific O	cean	Ocean	2011	1.15-1.32	NA	NA	Soerensen et al., 2014
Mediterra	nean Sea	Sea	2000	1.9 ± 1.0	NA	7.9	Sprovieri et al., 2003
Global O	cean	Ocean	2006-2007	$1.53\ \pm 0.58$	NA	$3.1~{\pm}11.0$	Soerensen et al., 2010a
Adriatic S	Sea	Ocean	2004	1.6 ± 0.4	$4.5\ \pm 8.0$	6.7 ± 11.7	Sprovieri and Pirrone, 2008
Amsterda	m Island	Ocean	2012-2013	1.03 ± 0.08	0.67	0.34	Angot et al., 2014

792 ^a NA: No data available.

793 ^bATARS: Australian Tropical Atmospheric Research Station.

794 Table 2. Correlation coefficients for speciated atmospheric Hg and meteorological parameters (one asterisk

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Speciation	G	EM	R	GM	H	g ^P _{2.5}	Wine	d speed	Air tem	perature	F	RH	P	AR
Speciation	р	r	р	r	р	r	р	r	р	r	р	r	р	r
RGM	0.069	0.294			< 0.01	0.453**	0.123	-0.251	0.053	0.313	0.065	-0.299	< 0.01	0.638**
Hg ^P _{2.5}	< 0.01	0.539**	< 0.01	0.453**			0.037	-0.335*	0.621	0.082	0.434	-0.129	0.432	0.130

denotes significant correlation in p < 0.05, double asterisks denotes significant correlation in p < 0.01).

1 Supplement of

2	Speciated	atmospheric	mercury an	nd sea-air	exchange	of
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3 gaseous mercury in the South China Sea

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Table S1. The concentration (pg m^{-3}) of each size-fractioned particulate mercury (Hg^P) in the South China Sea

41 (SCS) during our cruise.

Sampling time				S	ize range (μm)				Hg ^P
(Year/Month/Day)	< 0.4	0.4–0.7	0.7-1.1	1.1–2.1	2.1-3.3	3.3–4.7	4.7–5.8	5.8–9.0	9.0–10	in PM ₁₀
2015/9/4-6	3.03	1.69	2.90	2.47	2.04	1.72	1.55	1.46	1.39	24.35
2015/9/6-8	0.57	0.39	0.36	0.30	0.24	0.26	1.20	0.51	0.60	5.93
2015/9/8-10	0.67	0.65	0.48	0.88	0.52	1.04	0.89	0.49	0.50	8.15
2015/9/10-12	1.00	1.18	1.59	0.73	0.59	0.82	0.94	0.69	0.74	11.05
2015/9/15-17	0.33	0.21	0.45	0.18	0.41	0.50	0.50	0.74	0.60	5.23
2015/9/17-19	0.96	0.39	1.30	0.38	1.14	0.36	0.84	1.46	0.82	10.2
2015/9/19-21	0.46	0.32	0.13	0.49	0.29	0.34	0.29	1.13	0.41	5.15
2015/9/21-23	0.43	0.28	0.30	0.13	0.14	0.14	0.26	0.47	0.30	3.26
2015/9/23-25	0.97	0.68	1.02	1.15	0.99	0.88	0.89	0.92	0.82	11.09
2015/9/25-27	0.49	0.44	0.64	0.53	0.52	0.47	0.62	0.76	0.59	6.75
Average value	0.89	0.62	0.92	0.72	0.69	0.65	0.80	0.86	0.68	9.12

Table S2. The dry deposition flux (ng $m^{-2} d^{-1}$) of each size-fractioned Hg^P in the SCS during our cruise.

Sampling time				S	ize range (μm)				Total dry
(Year/Month/Day)	< 0.4	0.4–0.7	0.7-1.1	1.1–2.1	2.1-3.3	3.3–4.7	4.7–5.8	5.8–9.0	9.0–10	deposition
2015/9/4-6	0.0785	0.0146	0.0251	0.1280	0.2644	0.2229	0.2009	0.6938	0.6605	2.2887
2015/9/6-8	0.0148	0.0034	0.0031	0.0156	0.0311	0.0337	0.1555	0.2424	0.2851	0.7846
2015/9/8-10	0.0174	0.0056	0.0041	0.0456	0.0674	0.1348	0.1153	0.2328	0.2376	0.8607
2015/9/10-12	0.0259	0.0102	0.0137	0.0378	0.0765	0.1063	0.1218	0.3279	0.3516	1.0718
2015/9/15-17	0.0086	0.0018	0.0039	0.0093	0.0531	0.0648	0.0648	0.3516	0.2851	0.8431
2015/9/17-19	0.0249	0.0034	0.0112	0.0197	0.1477	0.0467	0.1089	0.6938	0.3897	1.4459
2015/9/19-21	0.0119	0.0028	0.0011	0.0254	0.0376	0.0441	0.0376	0.5370	0.1948	0.8923
2015/9/21-23	0.0111	0.0024	0.0026	0.0067	0.0181	0.0181	0.0337	0.2233	0.1426	0.4588
2015/9/23-25	0.0251	0.0059	0.0088	0.0596	0.1283	0.1140	0.1153	0.4372	0.3897	1.2840
2015/9/25-27	0.0127	0.0038	0.0055	0.0275	0.0674	0.0609	0.0804	0.3612	0.2804	0.8997
Average value	0.0231	0.0054	0.0079	0.0375	0.0892	0.0846	0.1034	0.4101	0.3217	1.0830

a . <i>i</i>	Posit	tion	Date and time	Depth	DGM	PAR	GEM	$T_{\rm w}$	Wind speed	Saturation	Flux
Station	Longitude (°E)	Latitude (°N)	(Year/Month/Day/Local	m	pg 1 ⁻¹	$W m^{-2}$	ng	c	$m s^{-1}$	%	ng m ⁻²
1	114 73	22.22	2015/09/05/10:15	44	57.8	152.10	1.86	27.3	73	1053	7 36
3	115.09	21.59	2015/09/06/04:15	94	41.4	0	1.00	29.2	7.5	1268	6.18
5	115.09	21.55	2015/09/06/08:10	123	37.0	8676	1.10	29.2	7.3	993	4 73
8	115.63	20.64	2015/09/06/14:50	389	34.9	111.08	1.33	30.0	7.1	1021	4 26
10	115.81	20.33	2015/09/06/19:50	657	32.0	0	1 19	29.9	62	976	2.96
11	115.01	19.96	2015/09/07/14:50	960	30.4	88.62	1.19	30.2	5.4	881	2.50
13	114 90	20.28	2015/09/08/07:00	151	37.8	39.76	1.20	29.7	4.6	949	1 91
16	114.63	20.26	2015/09/08/15:30	85	33.0	21.35	2.37	29.7	7.5	501	3 97
18	114.36	21.23	2015/09/08/17:55	91	36.8	10.09	2.37	29.0	11.5	576	10.72
20	114.00	21.25	2015/09/08/23:25	33	66.8	0	2.39	26.4	5.0	927	3.91
20	113.28	21.00	2015/09/09/04:10	41	44.5	0	1.72	25.5	94	835	9.04
21	113.20	20.86	2015/09/09/04:10	91	37.5	76.89	2.09	28.8	10.6	630	9.42
25	113.04	20.80	2015/09/09/14:10	178	30.0	183 30	1.82	20.0	0.7	598	6.20
31	111.95	18.85	2015/09/10/06:40	3/3	33.8	7.02	1.86	30.0	62	661	2.96
34	111.05	19.06	2015/09/10/12:48	178	37.9	189.97	1.50	30.1	7.0	910	4 43
36	111.49	10.10	2015/09/10/12:48	108	40.4	110.82	1.52	30.1	7.0	1015	5 21
30	111.20	19.19	2015/09/10/10:10	25	55.0	0	1.40	30.2	7.5 8 7	1015	0.34
20 20	111.02	19.55	2015/09/10/19.50	23	55.9 65.0	0	1.49	30.1	0.2 3 7	1372	9.54
39 41	110.70	18.90	2015/09/10/22.50	20	41.0	0	1.04	20.1	5.7	047	2.20
41	111.90	18.70	2015/09/11/05.10	20	22.0	22.05	1.30	20.2	0.0	947 607	4.54
44	111.20	18.40	2015/09/11/08.10	97	32.9 25.0	52.95 107.57	1.75	20.2	7.0	702	7 00
40	111.40	10.20	2015/09/11/15.00	1012	22.0	197.37	1.02	20.1	9.0	192 757	7.00 5.06
47	110.60	17.50	2015/09/11/20.00	1015	25.0	0	1.05	20.1	0./ 5 7	737	3.90
49 50	110.65	17.75	2015/09/12/02:20	064	35.0	0	1.60	20.0	5.7	725	2.67
52	110.43	18.13	2015/09/12/10:35	95	41.2	109.70	1.69	30.0	7.8	891	5.97
54	110.28	18.38	2015/09/12/14:10	15	58.9	183.59	1.57	30.3	8.9	13/3	11.61
62	110.50	15.45	2015/09/17/00:10	015	46.6	0	1.82	27.9	0.0	882	4.//
60 57	111.50	15.45	2015/09/17/18:40	1281	29.7	41.79	1.12	28.9	0.3	940 597	2.80
57	112.00	15.45	2015/09/18/08:10	2847	24.1	41.78	1.49	29.9	4.0	587	0.80
55 72	113.95	15.25	2015/09/18/15:55	1073	27.9	139.61	1.38	30.3	4.4	/41	1.25
73	114.00	14.00	2015/09/18/22:30	4356	34.9	0	1.37	29.9	8.5	926	6.02
/1	113.00	14.00	2015/09/19/06:00	2513	30.3	3.8/	1.22	29.9	0.1 5 4	904	2.68
69	112.00	14.00	2015/09/19/13:00	1803	30.7	195.05	1.12	29.7	5.4	1182	2.62
68	111.50	14.00	2015/09/19/19:10	2773	32.1	0	1.08	29.9	6.3	1082	3.10
65	110.50	14.00	2015/09/20/06:00	1951	23.0	0	1.52	29.2	3.4	537	0.58
81	110.50	13.00	2015/09/20/13:45	2546	30.5	200.23	1.40	29.8	5.5	/91	2.16
79	111.50	13.00	2015/09/21/01:30	2333	38.0	0	1.98	29.9	4.2	696	1.54
76	113.00	13.00	2015/09/21/13:50	4033	34.9	165.63	1.37	29.4	3.5	916	1.02
74	114.00	13.00	2015/09/21/21:30	4243	32.1	0	1.31	29.9	2.3	892	0.40
90	114.00	12.00	2015/09/22/08:50	4349	34.7	77.65	1.60	29.9	4.4	789	1.57
107	114.00	11.00	2015/09/22/15:55	3387	36.1	115.95	1.58	29.4	5.1	819	2.20
109	113.50	10.00	2015/09/23/00:30	3050	38.6	0	1.28	29.5	6.9	1085	4.46
111	112.50	10.00	2015/09/23/18:30	1814	29.6	5.12	1.36	29.7	7.2	785	3.58
112	112.00	10.00	2015/09/24/16:40	2357	37.0	33.50	1.48	29.7	10.2	899	9.15
113	111.50	10.00	2015/09/24/20:20	3347	29.2	0	1.44	29.6	9.9	733	6.61
114	111.00	10.00	2015/09/25/00:30	3372	34.8	0	1.46	29.4	10.6	852	9.22
115	110.50	10.00	2015/09/25/05:30	2533	31.5	0	1.46	29.3	11.2	771	9.19
100	110.50	10.50	2015/09/25/08:00	1791	37.1	77.25	1.37	29.3	11.5	966	11.75
99	110.50	11.00	2015/09/25/14:15	1721	38.1	139.76	1.20	29.7	11.7	1140	12.71
102	111.50	11.00	2015/09/25/21:15	3853	35.7	0	1.24	29.7	11.7	1043	11.83
104	112.50	11.00	2015/09/26/05:40	4182	27.0	0	1.22	29.5	11.3	794	8.06
105	113.00	11.00	2015/09/26/08:20	4227	30.4	51.68	1.23	29.6	4.7	888	1.59
92	113.00	12.00	2015/09/26/14:20	4277	38.5	159.27	1.24	30.0	7.3	1134	5.01
94	112.00	12.00	2015/09/27/00:25	4281	31.3	0	1.26	30.1	7.5	908	4.20
96	111.00	12.00	2015/09/27/07:00	2256	34.6	11.87	1.25	29.9	5.8	1002	2.81
97	110.50	12.00	2015/09/27/11:00	2404	40.8	181.18	1.27	30.0	2.2	1171	0.48

56 Table S3. Atmospheric GEM, surface seawater DGM and temperature (T_w) , wind speed (u_{10}) , saturation and air-sea flux of Hg⁰ during our cruise.



Figure S1. The insulated box of the RGM and $Hg_{2.5}^{P}$ sampling unit.



Figure S2. GEM concentrations over the northern SCS and 72-h back-trajectories arriving at some stations.



Figure S3. GEM concentrations over the western SCS and 72-h back-trajectories arriving at some stations.



65 Figure S4. RGM concentrations in the forenoon, afternoon and night.