Response to Reviewer #3's Comments

Anonymous Referee #3:

Major comments:

The study by Qin et al. investigates the atmospheric TGM concentrations at two islands and over the coastal oceans in east Asia, and explores the effect and contributions of anthropogenic emissions based on atmospheric tracer ratios and a receptor model. The finding from this study is valuable for understanding the cycling of atmospheric Hg in coastal ocean regions. This study provides many observational data, and does an insightful analysis of the datasets. The manuscript is well written and I broadly agree with the interpretations of the new data. I think the manuscript is currently in good quality. I have provided a number of minor comments that hope to be considered by the authors.

We sincerely thank for your in-depth comments and helpful suggestions on this manuscript. Based on the specific comments, we have responded to all the comments point-by-point and made corresponding changes in the manuscript as highlighted in the track change mode. You have raised a number of issues and we quite agree. We feel the substantial revisions based on your comments have greatly improved the quality of this manuscript. Please check the detailed responses to all the comments as below.

Specific comments:

1. the abstract: the levels and distribution patterns of atmospheric TGM are one of the major contributions of this study, which are better to be briefly summarized in the abstract.

Response: Thanks for your suggestion. The abstract has been revised to emphasize the distribution patterns and levels of TGM. The following sentence was added in Line 32-34.

"The mean concentrations of TGM were 2.32 ± 1.02 ng/m³ (Bohai Sea), 2.55 ± 0.55 ng/m³ (Yellow Sea), and 2.31 ± 0.81 ng/m³ (East China Sea), respectively, with coastal regions exhibiting significantly higher values than open ocean areas due to continental outflows."

2. line 39: these numbers should be linked to the seas investigated in this study.

Response: The text has been clarified to explicitly associate each percentage with its corresponding sea in Line 44-47.

"To assess the potential impact of anthropogenic emissions on the sea-air exchange fluxes of mercury, anthropogenic contributions to TGM were artificially removed, then the fluxes would be increased by 207.1% in the Bohai Sea, 33.4% in the Yellow Sea, and 6.5% in the East China Sea, respectively."

3. line 228: better to specify the equations for the calculation of Schmidt number of elemental mercury. In addition, provide the methods for detection of wind speed and water temperature.

Response: Thanks for the suggestion. The Schmidt number calculation and instrumentation details have been added in the revision (Line 304-308).

"The Schmidt number for Hg ($S_{C_{Hg}}$) was calculated as:

$$S_{C_{Hg}} = v/D_{Hg}$$

where v is seawater kinematic viscosity (Wanninkhof, 2014) and D_{Hg} is the diffusion coefficient of Hg (Kuss et al., 2009)."

The wind speed measurements at Huaniao Island and Juehua Island employed the Vaisala WXT530 surface weather station (Vaisala, Finland) with a resolution of 0.01 m/s. The surface seawater temperature was measured using a YSI EC300 portable conductivity meter (YSI, USA) with a resolution of 0.1°C. During the voyage, wind speed and surface seawater temperature data were acquired by the Finnish Vaisala AWS430 shipborne weather station with resolutions of 0.01 m/s and 0.1°C, respectively. The information has been added in Section 2.4.

4. line 235-239 and 247-248: this study describes well the spatial and temporal distribution patterns. As far as I have concerned, recent long-term continuous observations of TGM in mainland China have already showed significant declines in the past decades (e.g., Feng et al., 2024, NSR). My question is whether long-term changes in TGM in Chinese coastal oceans is similar to the mainland. I therefore suggest the author compare with previous observations and show the trends of TGM in coastal oceans in east Asia.

Response: Thanks for your valuable suggestions. The following table compares the GEM/TGM concentrations observed in this study and previous research in the offshore areas of East Asia. We found that existing observations struggle to clarify the long-term trends of TGM in marine environments, primarily due to two reasons: 1) Insufficient observational frequency—for example, previous studies on Bohai Sea only reported TGM data from spring and autumn of 2014; 2) Difficulty in comparing TGM concentrations across different research cruises, as sampling locations, timing, and cruise durations (typically spanning only a few weeks) varied significantly among different studies.

To infer trends in marine TGM concentrations, long-term monitoring data from coastal cities could serve as a reference. For instance, one study at Chongming Island near the coastal East China Sea revealed a marked decline in GEM concentrations from 2014 to 2016, with a rate of -0.60 ± 0.08 ng m^{-3} yr^{-1}; Research at Dianshan Lake, Shanghai, also indicated a decline rate of

-0.32 ± 0.07 ng m^{-3} yr^{-1} for GEM concentrations from 2015 to 2018. These findings suggest a potential downward trend in TGM concentrations across the offshore regions of East Asia.

Table. The GEM/TGM concentrations in this study and other literature.

Location	Site description	Sampling time	TGM/GEM (ng/m^3)	Reference
BS	Nearshore islands	2020 (winter)	2.32 ± 1.02	This study
YS	Sea	2020 (winter)	2.55 ± 0.55	This study
ECS	Sea	2020 (winter)	2.31 ± 0.81	This study
ECS	Sea(Huaniao island)	2020(autumn)	1.85 ± 0.74	This study
BS	Sea	2014 (spring)	2.51 ± 0.77	Wang et al. (2016a)
BS	Sea	2014 (autumn)	3.64 ± 2.54	Wang et al. (2016a)
YS	Sea	2014 (spring)	1.89 ± 0.64	Wang et al. (2016a)
YS	Sea	2014 (autumn)	1.59 ± 0.44	Wang et al. (2016a)
YS	Sea	2012(spring)	1.86±0.40	Ci et al. (2015)
YS	Sea	2012(autumn)	1.84±0.50	Ci et al. (2015)
YS	Sea	2010(summer)	2.61±0.50	Ci et al. (2011)
ECS	Sea (Huaniao island)	October 2013 to January 2014	2.25 ± 1.03	Fu et al. (2018)
ECS	Sea	2014 (spring)	1.61 ± 0.32	Wang et al. (2016a)
ECS	Sea	2013(summer)	1.61 ± 0.32	Wang et al. (2016b)
ECS	Sea	2013(autumn)	2.20 ± 0.58	Wang et al. (2016a)
SCS	Sea	2015(autumn)	1.52±0.32	Wang et al. (2019)
SCS	Sea	2007(summer)	2.62±1.13	Fu et al. (2010)

5. line 261-263: this statement is very speculative and in contrast with the discussions in sections below. The good relationship observed could be affected by many factors. Generally, ocean Hg emissions are largely controlled by wind speed, while the effect of temperature and solar radiations is relatively small.

Response: Thank you for your comments. We agree with the reviewer that wind speed and wind turbulence are the most direct factors influencing oceanic mercury release, which can be reflected by the discussions related to Figure 3. Accordingly, we have revised the statement as "The TGM diurnal pattern displayed strong concordance with temperature and solar flux (Figure 2a)." in Line 348-350.

6. line 280-281: have any of previous studies provided solid evidence that increasing temperature would facilitate strong ocean Hg emissions?

Response: Thanks for your comments. The air-sea exchange of Hg^0 is driven by concentration gradients across the atmospheric and seawater interface (Soerensen et al., 2013). From a physical perspective, Hg^0 inherently possesses high volatility. Elevated temperatures intensify molecular thermal motion, accelerating the volatilization of mercury from the liquid phase to the gas phase, thereby increasing the risk of oceanic mercury release. This is reflected in the calculation of mercury's air-sea exchange flux. According to the two-film theory model (Wanninkhof and Oceans, 1992), the air-sea exchange flux of mercury is calculated as:

$$F = K_W(C_W - C_a/H')$$

where Kw represents the gas exchange velocity. Kw is determined by the formula (Nightingale et al., 2000):

$$K_w = 0.31u_{10}^2(S_{C_{Hg}}/660)^{-0.5}$$

where $S_{C_{Hg}}$ is the Schmidt number of Hg0 in seawater, calculated as:

$$S_{C_{Hg}} = v/D_{Hg}$$

with D being the aqueous-phase diffusion coefficient of Hg^0, calculated as:

$$D = 0.0011e^{-(11.06\ kJmol^{-1})/RT}$$

Where T is the seawater temperature. Notably, under constant conditions, higher seawater temperatures lead to an increase in D, a decrease in the Schmidt number (Sc) of Hg^0 in seawater, and consequently an increase in the mass transfer coefficient Kw, resulting in higher air-sea exchange flux of mercury.

From a chemical perspective, the production of Hg^0 in seawater primarily originates from the photochemical reduction of divalent mercury (Hg^{2+}) (Costa and Liss, 1999; Andersson et al., 2011), which is closely linked to parameters such as light intensity and temperature (Ci et al., 2016; Mason et al., 2001). Both observational and modeling studies demonstrate that rising temperatures enhance oceanic mercury release. For example, observations in the Baltic Sea revealed that seawater Hg^0 concentrations and air-sea exchange fluxes peaked at midday, aligning with temperature fluctuations (Osterwalder et al., 2021). Model simulations indicated that uniformly raising sea surface temperature (SST) by 1 °C increased global Hg^0 evasion, particularly in high-latitude regions (1–8%), where relatively low SST originally limited Hg^0 evasion (Huang and Zhang, 2021).

In the revision, we have explained this in Line 557-560:

Higher temperature not only favored the production of DGM in seawater (Costa and Liss, 1999; Andersson et al., 2011; Mason et al., 2001) but also promoted the escape of DGM from the water surface into the atmosphere (Osterwalder et al., 2021; Huang and Zhang, 2021).

7. line 284-285: note that the natural emissions in this study is mainly associated with seawater emissions. while the discussions here is mainly reasonable for soil Hg emissions.

Response: Thanks for the comments. The references for this statement are now replaced with more relevant references that explicitly address mercury marine processes in Line 374-376.

"Previous studies suggested wetting processes may promote the reduction of Hg^{II} to Hg^{0} in surface seawater, while higher wind speed accelerated its evasion (Lin et al., 2010; Soerensen et al., 2013)"

8. line 327-330: I thought the authors should determine the TGM/BC ratios based on the slope of the correlations between TGM and BC. It seems the ratios are calculated by observed levels of TGM and BC. Note that BC is not long-lived atmospheric pollutants and ready to deposit more quickly than TGM, and this is why high TGM/BC ratios was observed at locations far from sources.

Response: Thanks for the comments. We agree with the reviewer that BC is not a long-lived atmospheric pollutant and deposits more rapidly than TGM. The content in lines 327–330 originally aimed to discuss the differences in TGM/BC between terrestrial and marine environments from an observational perspective. However, the wording in the original conclusion was overly definitive. We have therefore revised the statement in Line 431-434.

"On one hand, lower contribution of anthropogenic sources to TGM in the coastal environment compared to the urban environment was expected. On the other hand, BC deposited more quickly than TGM, thus also elevating the TGM/BC ratios was at locations far from sources.".

9. line 370: it is interesting to provide quantitative analysis of the contributions of anthropogenic sources. My question is whether the results from this study agrees or is different from previous studies. Recently, several studies quantify the contributions of anthropogenic emissions to atmospheric TGM in rural areas in China (including HNI) based on Hg isotope approaches. I would therefor suggest the authors compare their results with previous isotope and modelling studies.

Response: Thanks for your valuable suggestions and we do agree that the results in this study should be compared to studies based on Hg isotope approaches. This study found that anthropogenic sources contributed 38%, 26%, and 35% to TGM in the Yellow Sea, East China Sea, and Huaniao Island, respectively. For Dianshan Lake in Shanghai and Juehua Island in Liaoning Province, the anthropogenic contributions rose to 47% and 59%, respectively. In comparison, previous isotope-based source apportionment studies have revealed anthropogenic contributions of 29% and 42% to TGM in remote areas like Changbai Mountain and Ailao Mountain (Wu et al., 2023). In general, the isotope-based results indicated that the relative contributions of anthropogenic emissions to surface GEM in remote China and urban China were around 30% and 49%, respectively (Fu et al., 2021; Feng et al., 2022; Wu et al., 2023). Notably, the anthropogenic contributions to TGM in the Yellow Sea, East China Sea, and Huaniao Island

from this study align closely with isotope-derived values from China's remote regions, while the Dianshan Lake findings correspond with urban isotope results. The elevated contribution observed at Juehua Island (59%) may be attributed to its proximity to the mainland (only 10 km away) and the sampling period occurring during the winter heating season, where continental transport influences were significant (Li et al., 2023). Furthermore, the values obtained in this study fall within comparable ranges to modeling study estimates (typically 33% to 41% on average)(Chen et al., 2014; Wang et al., 2018).
The comparison between this study and previous studies have been added in Line 524-536.

10. line 381-384: see my previous comments on TGM/BC ratios. Are there any strong relationship between TGM and BC over the seas? If yes, better to use relationship slopes to estimate the contributions of anthropogenic emission over the seas, but not using the island results.
Response: Thanks for your valuable comments. We have now checked the relationship between TGM and BC over the seas. As shown in the figure below, the concentrations of TGM and BC observed over the ocean in this study did not exhibit a strong correlation. This was indeed anticipated, as TGM was influenced not only by anthropogenic sources but also significantly by natural sources, particularly over the ocean, while BC primarily originated from anthropogenic emissions. In addition, the cruise observation covered a wide oceanic area. Different oceanic regions had different relationship between natural vs. anthropogenic mercury. Thus, we used other approaches to quantify the contributions of anthropogenic emission over the seas in this study.

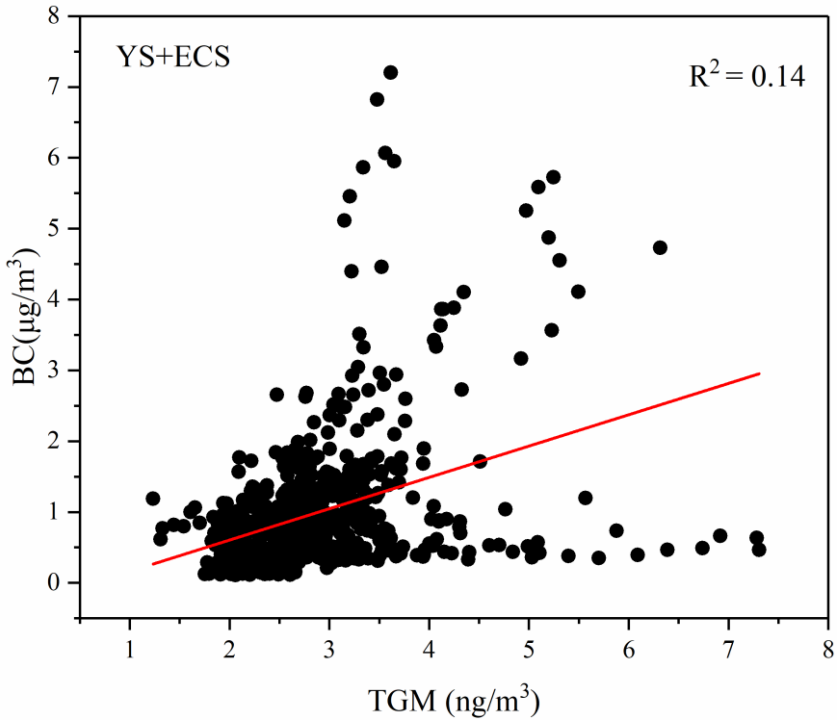

Figure. Scatter plot of TGM and BC concentrations over the ocean and their correlation

11. Line 422-423: better to show the values or ranges of previously observed DGM in the seas.

Response: Thanks for your suggestions. We have now added the concentration ranges of DGM from previous literature in Line 549-551.

"The DGM concentrations measured during this winter cruise campaign (22.9-39.7 pg/L) were significantly lower than those recorded previously during summer and fall in similar regions (52.4-63.9 pg/L) (Ci et al., 2011; Ci et al., 2015; Wang et al.,2016a)."

12. Line 440-441: I would suggest the authors to provide detailed information regarding the DGM and air TGM concentrations, wind speed, water temperature, and calculated exchange flux at each of the sampling sites in the supporting information. This would be valuable for future studies in air-sea Hg exchanges.

Response: Thanks for your suggestions. We have added a new table in the supplementary materials, which includes parameters such as DGM and TGM concentrations, wind speed, water temperature, and exchange flux, as shown in the table below.

Table S1. DGM and TGM concentrations, wind speed, water temperature, and sea-air flux at each sampling site during the cruise campaign.

Date (LT)	Longitude (deg)	Latitude (deg)	DGM (pg L^{-1})	TGM (ng m^{-3})	SST (℃)	WS (m s^{-1})	F (ng m^{-2} h^{-1})
2019/12/29 6:30	122.669	25.481	52.4	2.20	22.4	9.1	10.28
2019/12/29 13:30	121.771	26.002	50.8	2.47	19.8	6.7	4.93
2019/12/29 17:30	121.351	26.303	43.9	2.35	19.5	4.7	2.03
2019/12/29 22:00	120.905	26.609	55.4	2.97	17.8	3.7	1.51
2019/12/30 1:00	120.510	26.843		3.62	15.6	1.8	
2020/1/1 14:00	121.623	27.561	50.4	2.17	14.8	8.1	6.44
2020/1/1 16:00	121.927	27.396	47.5	2.17	15.5	6.6	4.04
2020/1/1 22:00	122.745	27.150	38.2	2.54	15.7	6.7	3.01
2020/1/2 3:30	123.362	26.653	40.1	2.49	16.5	5.1	1.92
2020/1/2 9:00	124.197	26.541		2.49	18	5.7	
2020/1/2 19:00	125.803	26.871	22.8	2.03	19.3	1.9	0.14
2020/1/3 2:30	125.000	27.451	24.3	2.33	17.7	4.7	0.85
2020/1/3 8:30	124.251	27.943	33.0	2.07	17.7	2.4	0.36
2020/1/3 14:00	123.565	28.481	35.2	2.30	19.3	3.3	0.76
2020/1/3 18:30	123.006	28.831	30.2	2.15	17.4	2.5	0.34
2020/1/3 22:00	122.647	29.121	39.1	2.85	14.3	6.7	2.85
2020/1/5 2:30	124.535	29.858	35.5	2.19	14	8.9	4.81
2020/1/5 9:00	125.392	29.270	30.1	2.08	17.8	6.1	2.07
2020/1/5 14:30	126.144	28.704	23.5	2.11	19.1	7	1.98
2020/1/5 21:00	126.998	28.151	20.1	2.39	20.1	7.8	1.84
2020/1/6 2:30	127.601	28.802		3.46	19.7	7.1	

2020/1/6 8:00	127.251	29.650		2.85	20.6	6.1	
2020/1/6 13:00	126.849	30.505		2.61	21.1	7.7	
2020/1/6 17:30	126.498	31.303		3.45	19.1	9.3	
2020/1/6 21:00	126.327	31.898		3.50	19.1	7.1	
2020/1/7 8:00	124.501	31.336	36.1	2.64	17.4	11.9	9.12
2020/1/7 13:30	123.504	31.331	46.6	2.67	19.5	11.7	13.12
2020/1/9 13:30	122.764	31.084	57.4	4.71	10.7	5.5	2.30
2020/1/9 15:00	122.803	31.332	43.7	7.11	10	7.7	1.20
2020/1/9 17:30	122.487	31.361	57.7	5.04	8	7.1	3.27
2020/1/9 19:30	122.485	31.597	51.4	4.09	8.3	7.4	3.42
2020/1/10 10:00	123.989	32.997	31.4	3.90	8.9	6.3	0.97
2020/1/10 15:00	123.056	32.966	36.4	4.20	8.7	5.6	0.98
2020/1/10 21:00	122.003	32.994	27.6	4.32	7	4.4	0.19
2020/1/11 5:00	121.251	34.001	37.2	3.46	7.1	6.4	1.57
2020/1/11 10:00	122.144	33.990	35.9	4.01	8.7	5.4	0.94
2020/1/11 14:30	123.051	34.003	34.5	4.53	8.4	3.6	0.31
2020/1/11 19:30	124.001	33.992	29.8	2.67	8.4	7.3	1.77
2020/1/12 2:00	123.991	34.953	31.8	2.77	5.3	10.2	3.25
2020/1/12 9:00	123.058	34.997	35.9	2.36	4.1	11.3	5.29
2020/1/12 13:30	122.205	35.001	38.3	2.91	3.8	8.5	2.87
2020/1/12 18:30	121.345	34.995	19.3	2.32	4.1	5.2	0.30
2020/1/12 23:00	120.521	34.999	31.4	2.30	3.7	3.7	0.46
2020/1/13 5:30	121.250	35.982	31.6	3.18	1.2	4.5	0.40
2020/1/13 10:00	122.152	35.979	28.7	3.36	3.5	2	0.07
2020/1/13 14:30	123.051	35.985	30.3	3.55	5.5	5.4	0.60
2020/1/13 20:00	123.964	35.979	36.5	2.42	3.9	2	0.17
2020/1/14 2:30	123.967	36.984	42.9	2.65	3.6	9.5	4.59
2020/1/14 6:00	123.381	37.000	26.1	3.01	1.2	8.7	0.92
2020/1/14 10:30	122.752	36.977	21.9	3.28	2.1	8.3	0.22
2020/1/14 19:30	123.962	38.006	33.3	2.62	-0.4	6.1	1.00
2020/1/15 0:00	123.006	37.999	23.8	2.05	0	5.4	0.51
2020/1/15 4:30	122.052	37.998	31.1	2.31	-0.7	8.9	2.07
2020/1/15 8:30	121.201	38.003		1.96			
2020/1/15 13:30	121.523	38.473	17.8	2.47	2.3	3.1	0.05
2020/1/15 16:30	122.077	38.748	20.7	2.82	-0.1	0.8	0.00
2020/1/15 21:30	123.001	38.755	32.0	2.83	0.9	2.4	0.14
2020/1/16 2:00	123.801	38.748	27.4	2.62	2	6.3	0.79
2020/1/16 7:30	123.969	39.499	25.2	3.34	-3.4	5.2	0.03
2020/1/16 11:00	123.234	39.295	26.4	2.81	-1.4	5.6	0.37

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