Response to Reviewer #1's Comments

Anonymous Referee #1:

Major comments:

The authors conducted about one month (12/2/2020-1/1/2021), 3 weeks (10/14/2020-11/4/2020), and 2 weeks (12/29/2019-1/16/2020) of TGM measurements at two island sites, JHI and HNI, and on a cruise ship. They estimated anthropogenic contributions to ambient concentrations of TGM using PMF and linear regression analysis. They also estimated sea-air exchange flux of Hg° using an air-water exchange flux equation (Wanninkhof, 1992, JGR). Over the past couple of decades since the Tekran series has been deployed globally, numerous long-term datasets of speciated, operationally defined, mercury concentrations have been available and used to study atmospheric Hg cycling, which has generated a large body of research in the literature. While the authors performed a comprehensive analysis with what they got, the short-term nature of their datasets limited their ability to provide substantial insights into atmospheric Hg budgets. The study also presents several methodological concerns.

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. PMF application and interpretation issues. There are multiple concerns regarding the PMF application and interpretation:

1.1 The authors used PMF to identify the factors contributing to ambient TGM at DSL and JHI. DSL was a long-term (2015-2019) monitoring site over land near the coast in Shanghai, while the field campaign at HNI, an island site near DSL, took place a year later. The authors should clearly specify the time periods used in their PMF analysis for the two sites in Section 2.5.

Response: Thanks for your comments regarding the PMF modeling. We do agree that inconsistent observation periods can affect the results. In this regard, we now use observational data which are consistent between DSL and JHI. We have clearly specified the time periods used in the PMF analysis for the two sites in Section 2.5.

The revised manuscript includes the following additions in Line 289-292.
At DSL, we selected observational data from October to December, 2020 (totaling 1,080 valid data points) for PMF modeling to align with the HNI observational campaign. At JHI, observational data from December 2 to 30, 2020 (totaling 675 valid data points) were used for

PMF analysis.

1.2 The study assumes that the empirical relationship between anthropogenic TGM contributions and BC concentrations (derived from 2015–2019 land-based data) is applicable to TGM data at an island site over 100 km away and one year later. This assumption is highly questionable, as empirical relationships may not necessarily hold across different locations and timeframes. At the very least, the authors should acknowledge the potential uncertainty introduced by this approach.

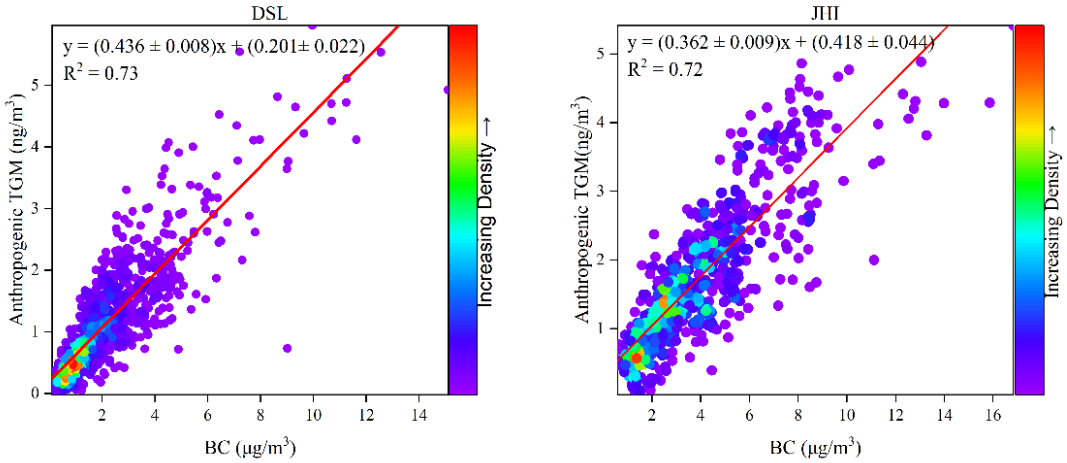
Response: Thanks for your comments. We do agree with you that inconsistent observational periods may result in great uncertainties. As responded to the previous comment, we have now used consistent observational periods at both sites to avoid this issue. As for the potential uncertainty introduced by this approach, we now consider the uncertainty of the derived regression equations, which is described in the next response.

1.3 The size of the dataset for JHI appears too small for PMF. It was a month-long campaign. What was the temporal resolution they had for their datasets? Fig. 5d seems to show about 20 data points. If this represented the number of data points for a single variable used for PMF analysis, then their results would be questionable. The authors should reference Zhang et al. (2009, https://doi.org/10.1016/j.atmosenv.2009.07.009), which discusses the minimum sample size required for PMF applications. That reference is just an example from a large body of literature on the topic. In fact, I am wondering, if the authors used the 2015-2019 data for DSL PMF analysis, how come only a handful of data points were shown in Fig. 5c?

Response: We apologize for the lack of clarity in the original manuscript. Regarding the original Figure 5c and 5d, the data points represent daily averages, that's why only a small number of points is shown in the figure.

For Juehua Island (JHI), the data used for PMF analysis were measured from December 2 to 30, 2020, with a 1-hour temporal resolution, totaling 675 data points. For Dianshan Lake (DSL), the PMF analysis utilized observational data from October to December, 2020, with a 2-hour temporal resolution, totaling 1080 data points. Thus, the dataset sizes for both sites meet the requirements for PMF analysis.

In the revised Figure 5c and 5d as shown below, we have now used the original dataset for plotting. It is found that the regression equations based on the hourly datasets are quite close to those based on the daily datasets. In addition, the revised Figure 5c and 5d also show the regression equations with uncertainties. In the revised manuscript, we have re-calculated the contributions of anthropogenic vs. natural TGM and added the uncertainties based on this approach.



In Line 501 - 504, the concentrations of anthropogenic and natural TGM are revised based on the newly derived formulas.

In Line 497-499, the following writings are added.
Due to the uncertainties of regression slopes and intercepts of the regression formulas, this approach caused around 5% uncertainties on differentiating the anthropogenic and natural fractions of TGM.

1.4 The rationale behind selecting certain tracers is unclear. Why did the authors use V for the shipping emission tracer? The tracers for the cement industry were Ca and Fe, which could very well be indicative of dust. Why was BC not used in the PMF analysis?

Response: Thanks for your comments. V was chosen as an indicator for shipping emissions because V has been recognized as a typical tracer of heavy-oil combustion, which is commonly used in marine vessels (Viana et al., 2009). As such, previous studies have widely adopted V as a key marker for ship-related emissions (Chang et al., 2018; Zhao et al., 2013; Pandolfi et al., 2011).

In the revised manuscript, we have added the following clarification in Line 474-475.
"V has been considered a typical tracer of heavy-oil combustion, which is commonly used in marine vessels (Viana et al., 2009)."

Regarding calcium (Ca), it was a major airborne component from cement production since there are a lot of cement industries in China. However, it is indeed true that Ca and Fe are well indicative of dust. In the revision, we have labeled this factor as "dust and cement production". (Line 475).

As for Black carbon (BC), if it was also included in the PMF analysis, it is expected that BC always correlated with anthropogenic TGM since BC and anthropogenic TGM would be assigned to the same factor. In this study, we intentionally excluded BC from the PMF analysis and

retained it as an independent variable, thus the PMF results and BC measurements were independent with each other. Then, we found out that anthropogenic TGM derived from PMF correlated strongly with BC at two sites. On one hand, this corroborated the reasonability of the PMF modeling results. On the other hand, the results suggested the relationship between anthropogenic TGM and BC was robust when evaluated at two sites.

2. TGM/BC ratio. The authors highlighted the TGM/BC ratio in the abstract as a key finding. However, this ratio appears to be just another variable rather than a novel result that provides additional insights.

Response: Thanks for your suggestion. In the revised manuscript, we have removed the description of the TGM/BC ratio from the abstract.

3. Sea-Air exchange flux calculations. The study recalculates sea-air exchange flux after removing anthropogenic contributions from ambient data. However, the purpose of this recalculation remains unclear. This issue also relates to the statement in the abstract (Lines 39–40), which needs further clarification.

Response: Thanks for your comments. In this study, we recalculated the air-sea exchange flux after removing anthropogenic contributions to quantify the impact of human-driven sources on oceanic mercury release. The purpose of this recalculation is to assess potential changes in the marine mercury flux under the scenario of reduced anthropogenic emissions.

In the revision, this sentence (Line 44-47) is revised as "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.".

4. Unsupported assertions. Assertions throughout the manuscript lack supporting evidence or citations. Below are a few examples:

4.1 Line 256: The term "elucidation" is misleading, as the statement is purely speculative.

Response: We agree with the reviewer that the original phrasing overstated the conclusiveness of the analysis. The sentence (Line 341-342) has been revised as "This divergence may be one of the reasons why the TGM concentration in the YS was higher than that in the ECS."

4.2 Lines 261–263, 265–266 (JHI), 266–268, 287: Assertions require supporting evidence or references.

(1) Lines 261–263

Response: From the calculation of oceanic mercury release fluxes, under otherwise constant conditions, higher temperatures lead to greater mercury release from the ocean (Wanninkhof and Oceans, 1992). This is because 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.

From the perspective of Hg^0 production mechanisms in seawater, 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).

Therefore, in the revised manuscript, we have cited relevant references as supporting evidence, as demonstrated in Line 348-350.

“The TGM diurnal pattern displayed strong concordance with temperature and solar flux (Figure 2a), indicative of significant impacts from natural sources (Osterwalder et al., 2021; Huang and Zhang, 2021; Mason et al., 2001).”

(2) Lines 265–266 (JHI)

Response: The answer to this question aligns with that of the previous response.

(3) Lines 266–268

Response: We have cited field measurements from the HNI (Fu et al., 2018) to support anthropogenic influences on coastal TGM. Also, field measurements from JHI (Li et al., 2023) are cited to support the are primarily influenced by coal combustion for heating in winter.

(4) Line 287

We recognized that the conclusion in the original text—"Thus, relative humidity and wind speed were verified as critical factors influencing the levels of TGM as similar as temperature"—was overly assertive. Therefore, in the revised version (Line 378-379), we have rephrased this statement as "This may explain the observed positive correlations between humidity, wind speed, and TGM concentrations."

4.3 Lines 281–300: This paragraph is speculative and lacks supporting evidence.

Response: Regarding the conclusion in lines 281–282, "the evident correlation between TGM and temperature exemplified the significant effects of natural surface emissions," we have already addressed this in the response of question **4.2(1)** and supplemented it with supporting literature.

The statement in lines 287–288, "Thus, relative humidity and wind speed were verified as critical factors influencing the levels of TGM as similar as temperature," has been revised in the answer of **question 4.2(4)** to be "This may explain the observed positive correlations between humidity, wind speed, and TGM concentrations."

For the unclear explanation in lines 291–294 regarding the phenomenon where TGM concentrations initially rise and then decline with increasing PBL (Planetary Boundary Layer) height, we have refined the reasoning in the revised version (Line 384-390):

"This observed diurnal pattern of TGM may primarily stem from the interplay between temperature-driven natural surface emissions and atmospheric dilution effects. When the PBL height was below 1 km, its increase coincided with rising temperature. Under these conditions, the enhancement of natural surface emissions due to temperature outweighed the dilution effect caused by the developed PBL, leading to increased TGM concentrations. Afterwards, as the PBL height continued to rise, the dilution effect gradually surpassed the temperature-driven emission enhancement, resulting in a decline of TGM concentrations."

Finally, to support the conclusion in lines 295–297—"This was likely ascribed to that the natural release around JHI was weaker than that around HNI, thus the dilution effect of elevated PBL overwhelmed the effect of natural surface emissions"—we have incorporated observational data

from the BS (Wang et al., 2020) and ECS (Wang et al., 2016a). These data corroborate that marine mercury emissions in the BS are significantly lower than those in the ECS.

Line 395-396: Due to the significantly lower marine mercury emissions in the BS (Wang et al., 2020) than in the ECS (Wang et al., 2016a), this phenomenon was likely ascribed to that the natural release around JHI was weaker than that around HNI, thus the dilution effect of elevated PBL overwhelmed the effect of natural surface emissions.

4.4 Line 319: The claim appears overstated.

Response: We appreciate the reviewer's critical perspective. The original phrasing has been revised as "Their concentration time series exhibited moderate agreement, suggesting potential land-sea interactions." in Line 420-421.

4.5 Line 322: Requires a reference.

Response: We have incorporated supporting reference "which was also the major anthropogenic source of TGM (Pacyna et al., 2006; Streets et al., 2011; Liu et al., 2019)." in Line 424-425.

4.6 Lines 426–428: Wouldn't higher temperatures enhance the partitioning of Hg° from water to air? The authors should clarify this mechanism.

Response: Thank you for the comment. We agree with you that higher temperatures enhance the partitioning of Hg^0 from water to air, as Hg^0 inherently possesses high volatility. Elevated temperature intensifies molecular thermal motion, thereby accelerating the volatilization of mercury from the liquid phase to the gas phase.

In the revision (Line 557-560), we have rephrased this sentence as "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)."

4.7 Lines 427–428: Supporting evidence or references are needed for the statement.

Response: 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 peak at midday, aligning with temperature fluctuations (Osterwalder et al., 2021). Model simulations indicate that uniformly raising sea surface temperature (SST) by 1 °C increases global Hg^0 evasion, particularly in high-latitude regions (1–8%), where relatively low SST originally limits Hg^0 evasion (Huang and Zhang, 2021).

We have revised the explanation in the updated manuscript and cited relevant literature. Specifically, the original statement "Higher temperature was generally more favorable for the formation of DGM" has been rephrased as "Higher temperatures not only favor the production of DGM in seawater (Costa and Liss, 1999; Andersson et al., 2011; Mason et al., 2001) but also

promote the escape of DGM from the water surface into the atmosphere (Osterwalder et al., 2021; Huang and Zhang, 2021).” in Line 557-560.

4.8 Lines 430–431: Requires citations.

Response: We have now added relevant references to this statement as below (Line 564).

"This suggested that continental inputs, such as river discharge, had a significant influence on DGM levels in nearshore waters (Chen et al., 2020; Kuss et al., 2018; Liu et al., 2016)."

5. Insufficient methodological details. For the ancillary data of ion concentrations, trace gases, and meteorological variables, the authors provided little information on the instruments used, and no information on data quality control and assurance as well as temporal resolution. Also, where were the PBL data from? They were introduced abruptly at one point in the results section.

Response: Thanks for the suggestion. We have added detailed methodological descriptions in Section 2 (Line 219-262).

At JHI, water-soluble ions in $PM_{2.5}$, including sulphate (SO_4^{2-}), nitrate (NO_3^-), ammonium (NH_4^+), chloride (Cl^-), sodium (Na^+), potassium (K^+), magnesium (Mg^{2+}), calcium (Ca^{2+}), alongside the soluble gases such as ammonia (NH_3) and sulfur dioxide (SO_2) were continuously monitored using an In-situ Gas and Aerosol Composition monitoring system (IGAC) (Wang et al., 2022). IGAC operated at a 1-hour temporal resolution and consisted of a wet annular denuder (WAD) and ion chromatography (IC) equipped with columns CS17 and CG17 for cations and AG11-HC and AS11-HC for anions. Ambient air was drawn into a $PM_{2.5}$ cyclone inlet by a built-in pump at a flow rate of 16.7 L/min. The sampled air was separated by passing through the vertically placed WAD to capture water-soluble gases, and airborne particles were collected by a steam scrubber and impact aerosol collector placed downstream. Air samples were dissolved by 30 ml ultra-pure water (18.25 MΩ cm^{-1}) and then divided into two steams. Both aqueous samples (including particles and gases) were injected into the IC system by two separated syringe pumps for analyzing the cations and anions. For quality assurance/quality control (QA/QC) of IGAC, a standardized lithium bromide (LiBr) solution was continuously introduced into aerosol liquid samples during the campaign to validate sampling and analytical stability. Weekly calibrations were performed for the ion chromatography (IC) module using certified standard solutions, with linearity ($R^2 > 0.99$) and detection limits (LODs) validated. Black carbon (BC) in $PM_{2.5}$ was measured continuously using a multi-wavelength Aethalometer (AE-33, Magee Scientific, USA). Meteorological parameters were measured using a Vaisala WXT530 surface weather station (Vaisala, Finland). Surface seawater temperature was recorded by a YSI EC300 portable conductivity meter (YSI, USA) with a resolution of 0.1°C.

At HNI, methods for analyzing meteorological parameters, BC, and surface seawater temperature mirrored those employed at JHI.

During the cruise campaign, the meteorological metrics (e.g., air temperature, wind speed/direction) and surface seawater temperature were collected from the Finnish Vaisala AWS430 shipborne weather station onboard the R/V. AE-33 was also used for BC measurements during the cruise.

At DSL, water-soluble ions in $PM_{2.5}$ and soluble gases were also measured by the IGAC instrument. Trace metals in $PM_{2.5}$ (Al, Ti, V, Cr, Mn, Fe, Co, Ni, Cu, Zn, Ga, As, Sr, Cd, Sn, Sb, Ba, Tl, Pb, and Bi) were continuously measured using an Xact multi-metals monitor (Model Xact™ 625, Cooper Environmental Services LLT, OR, USA). It operated at a flow rate of 16.7 L min^{-1} with hourly resolution. Particles in the airflow passed through a PM2.5 cyclone inlet and were deposited onto a Teflon filter tape, then the samples were transported into a spectrometer for analysis via nondestructive energy-dispersive X-ray fluorescence.

Planetary boundary layer (PBL) height data were obtained from the Global Data Assimilation System (GDAS) archive maintained by the U.S. National Oceanic and Atmospheric Administration (NOAA), available through the READY (Real-time Environmental Applications and Display sYstem) portal (https://www.ready.noaa.gov/archives.php; last accessed: 11 May 2025). The dataset, featuring 1-hour temporal resolution, was processed and extracted using MATLAB R2021b (MathWorks, Natick, MA).

6. Random citations. Some references seemed to be cited arbitrarily. While citing every study on a given topic is impractical, it is important to acknowledge milestone research appropriately. Here are a few examples. There have been hundreds and thousands of journal articles on PMF applications. Did Qin et al. (2020) develop the PMF approach? Was Gibson et al. (2015) the first to recognize PMF "for its efficacy in elucidating sources profiles and quantifying source contributions"? In lines 280-281, were those studies the first to establish the role of temperature in GEM evasion? In lines 321-322, were those studies the first to identify fossil fuel combustion as a major mercury source?

6.1 There have been hundreds and thousands of journal articles on PMF applications. Did Qin et al. (2020) develop the PMF approach? Was Gibson et al. (2015) the first to recognize PMF "for its efficacy in elucidating sources profiles and quantifying source contributions"?

Response: Thank you for the comments. The original statement "Qin et al. (2020) develop the PMF approach" has been revised, and "Gibson et al. (2015)" is no longer cited as the first to recognize PMF "for its efficacy in elucidating source profiles and quantifying source contributions." The theoretical foundation of the PMF model was originally documented in the study by Paatero and Tapper (Paatero and Tapper, 1994). In the revised manuscript, the citations in lines 267-288 have been modified as follows.

- "The PMF model is recognized for its efficacy in elucidating source profiles and quantifying source contributions (Paatero and Tapper, 1994)."
- "Detailed descriptions can be found in previous study (Paatero and Tapper, 1994)."

6.2 In lines 280-281, were those studies the first to establish the role of temperature in GEM evasion?

Response: The role of temperature in mercury evasion was indeed established earlier. Temperature-driven Hg^0 evasion has been mechanistically characterized since the 1990s (Lindberg et al., 1998; Poissant et al., 2000). We have updated the references to prioritize pioneering studies.

6.3 In lines 321-322, were those studies the first to identify fossil fuel combustion as a major mercury source?

Response: The original citation overlooked earlier work. The text now cites the early global mercury emission inventory identifying fossil fuels as a dominant anthropogenic source (Pacyna et al., 2006; Streets et al., 2011).

7. Uncertainty in sea-air exchange flux calculations using TGM as a proxy for Hg° in sea-air exchange flux calculations could introduce significant uncertainty. While this may be reasonable in a landlocked atmosphere, it can be problematic in the marine boundary layer, where halogen compounds are abundant and subsequently GOM concentrations are probably not negligible at times. For example, Castagna et al. (2018, atmos. Env.) reported GOM reaching well over 100 pg/m3, ~10% of TGM, at times. Note that in the reference cited, GOM was measured using the Tekran series, which has been in the literature suggested to be largely under-biased, primarily by Gustin et al.'s team. The actual GOM concentrations may be even higher.

Response: Thanks for your insightful comments. As noted, Gustin et al. (2015) highlighted that GOM concentrations measured by KCl-coated denuders could be underestimated by a factor of 2 to 3. To investigate the impact of this GOM measurement underestimation on the calculation of air-sea exchange fluxes in our study, we compiled observed GEM and GOM concentrations in China's marginal seas (listed in the table below). We found that even after accounting for the 2- to 3-fold GOM underestimation reported by Gustin et al. (2015), the proportion of GOM relative to TGM (GOM + GEM) in China's marginal seas didn't exceed 1.2%. Therefore, we conclude that the error introduced by substituting TGM for GEM in calculating air-sea exchange fluxes should be negligible in the China's marginal seas.

Table. GEM and GOM concentrations in China's marginal seas

Location	Site description	Sampling time	GEM (ng/m^3)	GOM (pg/m^3)	Reference
China	South China Sea	2015	1.52±0.32	6.1±5.8	(Wang et al., 2019)
	Bohai Sea and Yellow Sea	2014 (spring)	2.03±0.72	2.5±1.7	(Wang et al., 2016b)
	Bohai Sea and Yellow Sea	2014 (fall)	2.09±1.58	4.3±2.5	(Wang et al., 2016b)
	East China Sea (HNI)	2013-2014	2.25 ± 1.03	8 ± 10	(Fu et al., 2018)

Andersson, M. E., Sommar, J., Gårdfeldt, K., and Jutterström, S.: Air–sea exchange of volatile mercury in the North Atlantic Ocean, Mar. Chem., 125, 1-7, https://doi.org/10.1016/j.marchem.2011.01.005, 2011.
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Lindberg, S. E., Hanson, P. J., Meyers, T. P., and Kim, K. H.: Air/surface exchange of mercury vapor over forests - The need for a reassessment of continental biogenic emissions, Atmospheric Environment, 32, 895-908, 10.1016/s1352-2310(97)00173-8, 1998.
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