Response to Reviewer#3's comments

Anonymous Referee #3:

This manuscript presents an analysis of speciated atmospheric mercury (Hg), i.e. gaseous elemental mercury (GEM), particulate-bound mercury (PBM), and gaseous oxidized mercury (GOM), during June 2015 to May 2016 at the Dianshan Lake Station (DSL), Shanghai, east China. The topic is relevant to the Atmospheric Chemistry and Physics. However, the scientific contribution is hindered by a lack of methodology description in certain sections, some debatable analysis, some overreaching conclusions, and the quality of presentation. My specific comments and suggestions are listed below.

We sincerely thank for the reviewer's 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 red color. The reviewer has raised a number of issues and we quite agree. We feel the substantial revisions based on the reviewer's comments have greatly improved the quality of this manuscript. Please check the detailed responses to all the comments as below.

1. re-emissions from surfaces.

It is concluded that "GEM at DSL exhibited high concentrations in both warm and cold seasons, which was due to the strong re-emission fluxes from natural surfaces in summer…" (L506) However, there is no quantitative analysis to support this claim. The authors pointed out that "It has to be noted that since no tracers for the natural emissions (e.g. soils, vegetations, and ocean) were available in this study, the identification of natural mercury sources was not possible." (L390). The reviewer believes that temperature could be used to identify reemission with factor analysis approaches, such as Principal Component Analysis (PCA).

Response: Thanks for the suggestion. We applied the PCA to identify the potential sources of GEM by adding temperature. Four factors are resolved, which totally explained 75.32% of the variance as shown in the table below.

Factor 1 accounted for 34.15% of the total variance with high loadings for SO_2, SO_4^{2-}, NH_4^+, K^+, Pb, Se, and As, which is explained as coal combustion mixed with biomass burning. Factor 2 accounted for 14.85% of the total variance with high loadings for temperature, O_3, and NH_3, which was explained as surface emissions. Factor 3 explained 13.43% of the total variance and showed high loadings for Ni and V, which indicated the contribution of ship emissions. Factor 4 explained 12.89% of the total variance and showed high loadings for Fe and Ca, indicating the contribution of cement production.

In the revised manuscript, we have added PCA analysis to perform source apportionment with PMF.

Table R1. PCA analysis for GEM at DSL.

	Factor 1	Factor 2	Factor 3	Factor 4
GEM	0.50	0.25	0.11	0.07
SO_2	**0.69**	-0.20	-0.18	0.35
NO_2	0.38	-0.49	0.35	0.39
SO_4^{2-}	**0.84**	0.13	0.15	0.00
NH_4^+	**0.88**	-0.12	0.18	0.07
K^+	**0.77**	-0.25	0.04	0.39
Pb	**0.80**	-0.17	0.04	0.32
Se	**0.87**	-0.05	0.01	0.29
As	**0.82**	-0.23	0.06	0.33
O_3	0.06	**0.79**	-0.30	0.03
NH_3	0.03	**0.73**	0.36	-0.04
Temperature	-0.23	**0.82**	0.17	-0.03
Ni	0.24	-0.02	**0.85**	0.22
V	-0.03	0.11	**0.90**	-0.05
Fe	0.50	-0.12	0.24	**0.74**
Ca	0.26	0.08	0.00	**0.90**
Explained variance %	34.15	14.85	13.43	12.89

2. formation of GOM and PBM, local vs. regional events.

a. L359-369, CO and SNA concentrations were used to show that "GOM/PBM ratio was a reliable tracer for assessing the relative importance of regional/long-range transport vs. local atmospheric processing." (L368). Because CO and SNA concentrations are available, perhaps there is little need to use the GOM/PBM ratio, or GEM/CO ratio could be used as in previous studies of atmospheric Hg.

Response: Thank for the suggestion. We used CO and SNA to supplement the results based on the application of the ratio of GOM/PBM. The results by using CO and SNA or GOM/PBM are generally consistent, thus our study can be served as a reference that the GOM/PBM ratio could be used a tracer for assessing the relative importance of regional/long-range transport vs. local atmospheric processing even when data such as CO and SNA are not available.

b. Local vs. regional events. The authors stated that "Fig. 12 also demonstrated the increases of GOM along with the ratios of GOM/PBM. The lower ratios of GOM/PBM were associated with lower temperature and O_3 concentrations, indicating the more. probable long range/regional events during the cold seasons with relatively weak photochemistry. On the contrary, the higher ratios of GOM/PBM were associated with higher temperature and O_3 concentrations, indicating the more probable local events during the warm seasons with relatively strong photochemistry." (L460) This reasoning seems to suggest that "long range/regional events" and "local events" dominate during the cold and warm seasons respectively. The authors may want to present evidence that the study site was shielded from long range transport in some seasons, because most receptors are under the influence of long-range transport all the time. In my view, air mass from all directions would have higher temperature and O_3 concentrations in summer thus "relatively strong photochemistry" than those in winter, regardless "local" or "long range/regional events". Therefore, higher temperature and O_3 concentrations in warm season would indicate "relatively strong photochemistry" but not "local events". Similarly, lower temperature and O_3 concentrations in cold season would indicate "relatively weak photochemistry" but not "long range/regional events". The authors may want to amend this section and the related conclusions.

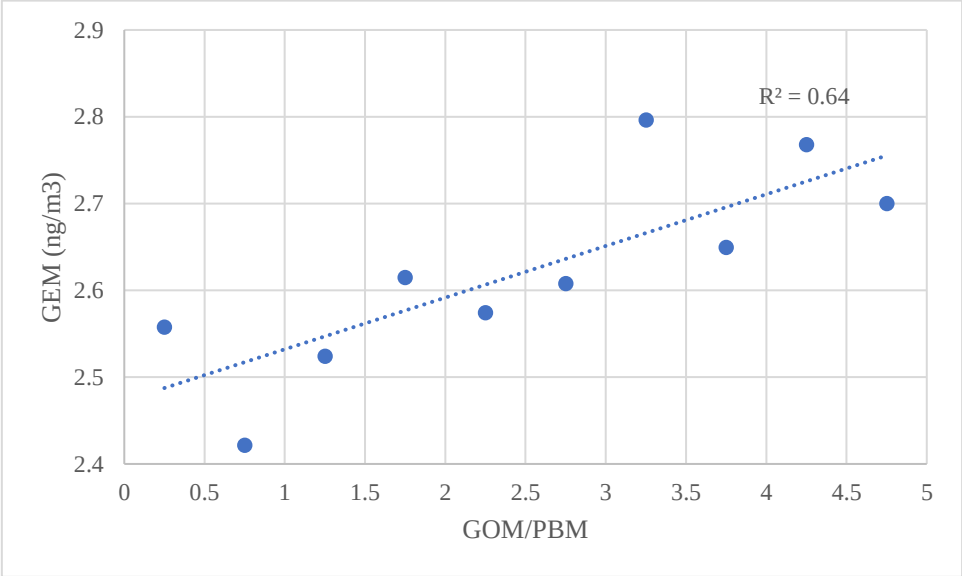
Response: Thanks for the reviewer's in-depth comments and we totally agree. We revised the description as "Fig. 12 also suggested that the lower ratios of GOM/PBM were associated with lower temperature and O_3 concentrations, indicating relatively weak photochemistry during the cold seasons. On the contrary, the higher ratios of GOM/PBM were associated with higher temperature and O_3 concentrations, indicating relatively strong photochemistry during the warm seasons" in the revision.

c. GEM levels. It was concluded that "GEM as a function of the GOM/PBM ratios indicated that when the quasi-local sources dominated, GEM concentrations were relatively higher than those events under the regional/long-range transport conditions" (L522-524). L370-382, "In the GOM/PBM ratio bins of less than 2.5, GEM fluctuated with the mean values less than 2.6 ng/m^3. The mean GEM concentration increased from 2.61 ng/m^3 in the GOM/PBM ratio bin of 2.5-3.0 to 2.8 ng/m^3 in the bin of 3.0-3.5, and then remain relatively stable when the GOM/PBM ratio bins higher than 3.0." Indeed, this is the only noticeable increase in GEM (a 7% increase from the bin of 2.5-3.0 to that of 3.0-3.5) among 10 bins, while the change in GEM concentrations between the first and last bin is less than 5%. Nonetheless, the authors stated that "Generally, GEM showed an increasing trend as the GOM/PBM ratios increased while both SNA and CO decreased. The elevation of GEM concentrations tended to be associated with the impact of quasi-local sources. In contrast, under the high SNA and CO conditions when GOM/PBM ratios were lower, GEM was relatively low, suggesting its formation. was not favored via the regional/long-range transport." The authors may need to report the statistical trend (e.g. regression) if any, and clarity what type of GEM "formation was not favored via the

regional/long-range transport", and explain why GEM would change with the strength of photochemistry. In case GEM is high in summer due to re-emission when GOM is also high due to higher oxidant levels, then the common course is meteorological conditions, namely ambient temperature, an analysis taking into account seasonal trend of all variables involved could be more appropriate, instead of the seemingly speculative approach employed.

Response: Thanks for the insightful comments. The regression analysis between GEM and GOM/PBM was shown in the figure below. Correlation coefficients of the regression reached 0.64, suggesting the positive correlation between GEM concentrations and GOM/PBM ratios. By stating "GEM formation was not favored via the regional/long-range transport" in the original writings, we meant to say that when the impact of long-range transport was important, GEM concentrations were observed to be relatively low. The usage of the word "formation" might lead to misunderstandings here, so we revised the sentence as "In contrast, under the high SNA and CO conditions when GOM/PBM ratios were lower, GEM showed relatively low concentrations. This suggested the regional/long-range transport didn't favor the elevation of GEM concentrations.".

The seasonal relationship between temperature and Hg concentrations have been discussed in details in Section 3.1. Generally, the GEM concentrations increased as the temperature increased, which is interpreted to be likely from the surface emissions. GOM concentration showed a clearly positive correlation with temperature, which might be related to the in situ oxidation of GEM under high temperature.



3 PBM formation

1) L476, "Fig. 13 shows the statistical pattern of the variation of PBM and GOM in the ascending bins of PM2.5. It was obvious that the concentrations of PBM increased with the concentrations of PM2.5, which was due to both primary emissions and the subsequent process of Hg species adsorbed on particulate matters." The authors may want to provide more evidence or citation to support that increasing PBM with increasing PM2.5 concentrations "was due to

both primary emissions and the subsequent process of Hg species adsorbed on particulate matters"

Response: Thanks for the suggestion. Previous studies have shown that PBM can be emitted directly from various anthropogenic sources such as coal-fired power plants and industries (Liu et al., 2018;Wu et al., 2016). In addition, a number of studies concluded that gas-particle partitioning of TGM is an important pathway of PBM formation in urban areas of China (Shon et al., 2005;Fu et al., 2015). The high levels of particles in Eastern China probably facilitated the formation of PBM (Fu et al., 2015).

In the revised manuscript, we have added references in the related paragraphs in Section 3.4.2.

2)L480, "When PM2.5 concentrations were at relatively low levels under 75 μg/m3, GOM concentrations increased with PM2.5. However, when PM2.5 concentrations exceeded 75 μg/m3, GOM exhibited a slightly decreasing trend as PM2.5 increased. It seemed that when the concentration of PM2.5 reached a certain value, the formation of GOM was inhibited to some extent, which was likely due to the adsorption of GOM onto the particles." "Statistical analysis showed that when PM2.5 reached a certain value, GOM was inhibited to some extent due to the gas-particle partitioning process." (L34-35) Those statements/conclusions seem to be debatable. As seen in Fig 13, when PM2.5 concentrations exceeded 75 μg/m3, GOM decreased till PM2.5 reached 105μg/m3 then increased, instead of a slightly decreasing trend. The authors may need to report the statistical trend (e.g. regression) if any and rephrase the statements about inhibitive effects in discussion and conclusion, or omit this passage. Based on an analysis of a three-day episode (December 30, 2015 to January 1, 2016), the authors concluded that "Under high PM2.5 concentrations, high humidity and low temperature conditions, the gas-particle partitioning process was obvious at DSL, which might be an important pathway for the formation of PBM." (L532) The authors may want to provide more in-depth analysis of the reliance of PBM on PM2.5 concentrations when temperature and relative humidity levels favor formation of PBM, and justify the use of a three-day episode in winter to represent the year-long study period. My scan of Fig 6 suggests that only in autumn the PBM levels increase with decreasing ambient temperature, not in other three seasons including winter (the three-day episode).

Response: Thanks for the comments. We do agree with the reviewer that the description about the trends of GOM and PM2.5 seems to be debatable. Thus, the sentence has been changed as "However, when $PM_{2.5}$ concentrations increased to 75-105 μg/m^3, GOM exhibited as obvious decreasing trend as $PM_{2.5}$ increased." and added the sentences "when $PM_{2.5}$ concentrations exceeded 105 μg/m^3, GOM exhibited a slightly increasing trend as $PM_{2.5}$ increased. High $PM_{2.5}$ concentrations in China always related to severe anthropogenic emissions (Auzmendi-Murua et al., 2014), so the moderate increasing trend of GOM in these bins should be attributed to the impact of strong primary emissions." in the revision.

Previous study in an island between China and Korea has demonstrated that the partition coefficient of mercury gas-particle partitioning in atmosphere was negative correlated with atmospheric temperature and positively correlated with relative humidity (Lee et al., 2016). From the seasonal statistical analysis in Fig 6, the autumn PBM levels showed a clear decreasing trend with increasing ambient temperature, while the spring and winter PBM

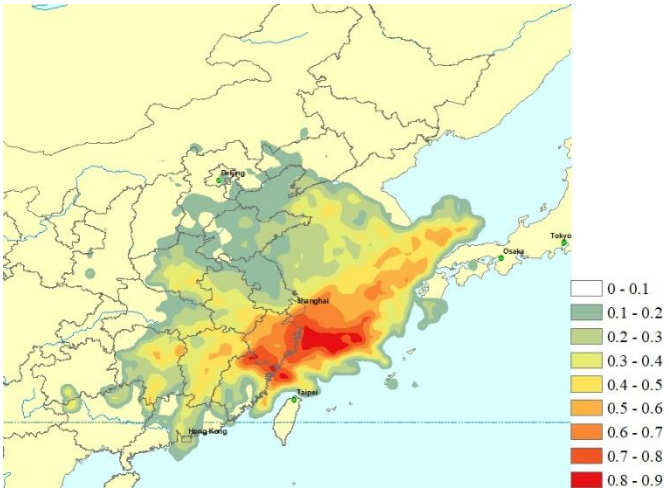
showed a moderate decreasing trend with increasing ambient temperature. We think that this phenomenon can demonstrate the influence of gas-particle partitioning process and temperature on the PBM concentration during the whole study period to some extent.

3) contribution of the shipping sector

It is concluded that "shipping emissions" contributed to 20% of GEM based on PMF (L30, L526). However, there is a lack of data support. The authors may want to a) identify the port on Fig 1, b) find out the Hg emission value at and near this port (I believe that researchers from Tsinghua University have some related publications), and compare the shipping sector emission with emissions from other sources, c) for each sample, plot the time series of contribution to GEM by this factor and the wind or air mass direction to verify that this factor indeed represents the shipping sector.

Response: Thanks for the suggestion. The ports in the YRD region has been added in Fig 1. However, Hg emissions at the port level are not available as the public emission inventories are always grided.

As the reviewer suggested, we have extracted the time-series of GEM concentrations from the shipping factor based on the PMF modeling. We use this time-series into the PSCF modeling. The figure below showed the potential sources regions were mainly located over the East China Sea, which indicated Factor 2 from PMF should be representative of the shipping sector.



2. The title reads, "Characteristics of atmospheric mercury in East China: implication on sources and formation of mercury species over a regional transport intersection zone". The authors may want to justify the use of data from a single site to represent 1) atmospheric Hg in East China, and 2) regional transport intersection zone, or rephrase the title. Similarly, some conclusions are a bit over-reaching, e.g. "shipping emission was found to be an important source (19.6%) of atmospheric mercury in East China" (L30)

Response: Thanks for the suggestion. The title is changed as "Characteristics of atmospheric mercury in the suburbs of East China: implication on sources and formation of mercury species over a regional transport intersection zone".

In the original abstract, the sentence before the writing "Besides the common anthropogenic emission sectors, shipping emission was found to be an important source (19.6%) of atmospheric mercury in East China" is "Six sources and their contributions to anthropogenic GEM were identified.". The PMF modeling only resolve source apportionment to anthropogenic mercury. However, based on the original data analysis and additional PCA analysis, we found the observed mercury has sources from natural emissions. Thus, the number 19.6% only pointed to anthropogenic emissions, but not all the mercury emissions. In the revised abstract, we have changed "atmospheric mercury" to "anthropogenic mercury" for clarification.

3. Seasonal trend of Hg species

The differences in seasonal mean TGM concentrations could be too small (max=2.88 ng/m3, min=2.63 ng/m3, the difference is 9%; summer 2.87 ng/m3, spring 2.73 ng/m3, difference 5%) and statistically not significant to support the conclusion of "GEM concentrations were elevated in both cold and warm seasons" (L21) and "GEM at DSL exhibited high concentrations in both warm and cold seasons, which was due to the strong re-emission fluxes from natural surfaces in summer and enhanced coal combustion for residential heating over northern China in winter." (L506). The authors may want to conduct comparison of multiple means (e.g. under ANOVA) for each of the three Hg species, and rephrase the discussion and conclusion in case the seasonal means are not statistically significant. Furthermore, please comment on the reasons of small difference among the four seasonal means if coal combustion for residential heating is enhanced in winter and re-emission fluxes from natural surfaces is strong in summer

Response: The table below showed the p-value by t-test between seasons for GEM concentrations, which suggested that the GEM concentration in autumn is statistically different from that of spring, summer, and winter ($p<0.05$), while there are no significant differences among spring, summer, and winter ($p>0.05$). In this regard, we revised the description as "Statistical test showed no significant differences of the seasonal variations of GEM concentrations among spring, summer, and winter were observed (Table S1). This was different from many urban and remote sites in China, such as Guiyang, Xiamen, and Mt. Changbai, where GEM showed significantly high concentrations in cold seasons than those in warm seasons (Feng et al., 2004;Xu et al., 2015;Fu et al., 2012). The relatively high GEM concentrations during the cold season in China should be attributed to the increases of energy consumption (Fu et al., 2015). In this study, GEM concentration in summer was comparable to that in winter, which was likely attributed to the strong natural mercury emissions (e.g. soils, vegetations, and water) due to elevated temperature in summer (Liu et al., 2016)" in the revision.

Table R1. P-value between seasons for GEM concentration

	spring	summer	autumn	winter
spring				

summer	p>0.05			
autumn	p<0.05	p<0.05		
winter	p>0.05	p>0.05	p<0.05	

4. Methodology is missing at numerous places. e.g. 1) please explain how to plot "Mean concentrations of (b) GEM, (c) PBM, and (d) GOM as a function of wind speed and wind directions" (L700) in the Method section and why Figures 5(b), (c) and (d) do not represent the frequency of wind directions shown in Figure 5 (a), in the Results section. 2) details of the backward trajectory simulation should be provided, including the model being use, run time, and start height, each with a justification, 3) where and how to use the "weighting function (wij)" (L162), 4) PMF, the treatment of missing data if any, and the total number of samples, 5) what is "the uncertainty of the jth pollutant on the ith measurement" (L187) in your study, 6) how to evaluate whether PMF is able to reproduce the observed Hg concentrations (e.g. time series and/or seasonal means), 7) how was hourly or bi-hourly PBL (assume it means Planetary Boundary Layer) height measured or estimated, 8) how was "secondary inorganic aerosols (SNA)" (L359) or "SNA (sulfate, nitrate, and ammonium) in PM2.5" (L715) monitored, 9) how to conduct analysis with bi-hourly Hg data and hourly data of meteorological parameters and other air pollutants.

Response:

1) Figure 5 (b), (c), and (d) were plot by the software R-studio. The radii of the circle in Figure 5 (b), (c), and (d) represent the value of wind speed. Figure 5 (b), (c), and (d) showed the relationship of Hg concentrations with wind speed and wind directions. As for Figure 5(a), the radii of the circle represent the frequency of wind directions but not the value of wind speed. In the revised manuscript, we have stated more clearly in the caption of Figure 5.

2) The details of the backward trajectory simulation have now been added into the Method section.

The HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) model is applied for calculating air mass backward trajectories (Draxler and Rolph, 2012). The model was run online at the NOAA ARL READY Website using the meteorological data archives of Air Resource Laboratory (ARL). The meteorological input data used in the model was obtained from NCEP (National Centers for Environmental Prediction)'s global data assimilation system (GDAS) with a horizontal resolution of 0.5° × 0.5°. In this study, 72-hours back trajectories were calculated at 500m AGL (above ground level) and the cell size was set as 0.5°×0.5°.

3) w_{ij} is an empirical value to reduce the uncertainties of PSCF values in certain grid cells that are associated with a small number of endpoints. The values of wij have been shown in Eq. (2) in the manuscript and w_{ij} is multiplied by $PSCF_{ij}$ to derive the weighted PSCF values.

In this revision, we have stately clearly about the calculation.

4) we added the details of PMF model runs in the Method section. In this study, the number of factors from 3 to 8 was examined with the optimal solutions determined by the slope of the Q value versus the number of factors. For each run, the stability and reliability of the output were assessed by referring to the Q value, residual analysis and correlation coefficients between observed and predicted concentrations. Finally, a 6-factor solution, which showed the most stable results and gave the most reasonable interpretation, was chosen. Before running the model, a data set including unique uncertainty values of each data point was created and inserted into the model, the error fraction was assumed to be 15% of concentrations for GEM and 10% of concentrations for other compounds (Xu et al., 2017), the missing data were excluded and the total number of samples is 3526.

5) The error fraction was set as 15% of concentrations for GEM and 10% of concentrations for other compounds (Xu et al., 2017).

6) In this study, the number of factors from 3 to 8 was examined with the optimal solutions determined by the slope of the Q value versus the number of factors. For each run, the stability and reliability of the output were assessed by referring to the Q value, residual analysis and correlation coefficients between observed and predicted concentrations. Finally, a 6-factor solution, which showed the most stable results and gave the most reasonable interpretation, was chosen. These procedures guaranteed the PMF was able to reproduce the observed Hg concentrations.

7) The data of the height of planetary boundary layer (PBL) were retrieved from the U.S. National Oceanic and Atmospheric Administration (https://ready.arl.noaa.gov/READYamet.php).

8) Water-soluble inorganic anions (SO_4^{2-}, NO_3^-, Cl^-) and cations (K^+, Mg^{2+}, Ca^{2+}, NH_4^+) in $PM_{2.5}$ were simultaneously monitored by the Monitor for Aerosols and Gases in ambient Air (MARGA) at the temporal resolution of one hour. The details of the instrument have been described in the first paragraph of Section 2.3.

9) To conduct data analysis with bi-hourly Hg data and hourly data of meteorological parameter, we first converted hourly meteorological data to bi-hourly meteorological data. Briefly, the hourly meteorological data was converted into the X and Y vectors. Then wind speed at the X and Y vectors were averaged and final we can get the average wind speed and wind direction based on the trigonometric function.

5. GOM and PBM could be included in PMF source apportionment. The results (e.g. sample by sample GEM, GOM, PBM, CO, SNA etc contributions in each factor) might help the identification of age of the air mass, as well as sources and processes related with Hg.

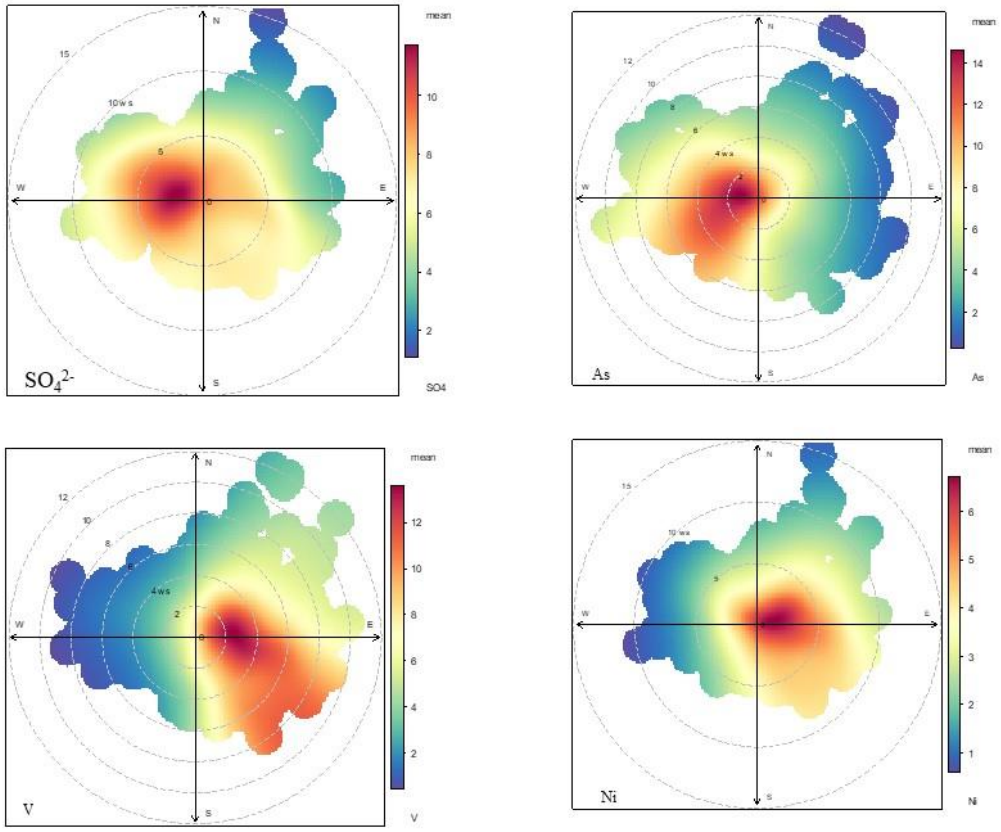
Response: Thanks for this suggestion. Actually, we've tried hard to apportion the major sources of PBM and GOM by using PMF. However, we found the results difficult to explain. We think that the possible reasons are that the concentrations of PBM and GOM fluctuated much stronger than other atmospheric species such as soluble ions, organic/elemental carbon, and elements. Due to the relatively short residence time in the atmosphere, it seems not suitable by digesting

PBM and GOM into the PMF analysis. Thus, we didn't include source apportionment of PBM and GOM in this study.

6. Interpretation of PMF results. This section could be improved by a more in-depth analysis and/or citation of existing literature. Overall, the analysis seems to be subjective at times, leading to invalid or overreaching statements/conclusions based on measurements at one site, a small difference in seasonal or binned concentrations, speculative approaches, or a short episode of a few days. It is recommended to clarify the meaning of local and regional events, whether "local" means local man-made emissions, local re-emission, local photochemical reactions, all of the above; or different item in different sections. Time series (Fig 2) or box plots (Fig 3) of temperature, relative humidity, CO, PM2.5 mass, SNA (sulfate, nitrate, and ammonium, or secondary inorganic aerosols), O3, and GOM/PBM ratios could be employed to depict seasonal trend of those variables, which may help to differentiate the association from causation.

Response: Thanks for the comments. In this study, local sources represented all emission sources in local area. GOM/PBM ratios together with CO, and SNA were used to reveal the relative importance of local sources vs. long-range transport.

In order to validate the PMF results, wind roses of SO_4^{2-}, As, Ni, and V are plotted in the figure below. SO_4^{2-} and As shared similar patterns with high concentrations mainly from the southwest. SO_4^{2-} and As are tracers for coal combustion. PMF identified combustion is the biggest source of GEM. Thus, this confirmed that the major sources of GEM are located in the southwest region. Ni and V showed similar patterns with high concentrations mainly from the northeast, east, and southeast. PMF identified a shipping source from the ocean. The wind rose plots of Ni and V also confirmed the identification of the shipping factor.



Clarification issues

1. The following items could be included in Fig 1, 1) location of the sampling site, 2) location of the shipping port, 3) the name of all provinces within e.g. 2000 or 3000 km of the sampling site, 4) a scale in the lower-right box, 5) the meaning of the upper-right box, 6) the meaning of the short black line.

Response: Thanks for the suggestion. Due to the conversion problems from text to pdf, some items in the figure were not properly shown in the original submission. Now we have corrected the errors and added marks as the reviewer suggested.

1) The location of the sampling site has been marked with a pentagram in Fig. 1.

2） The locations of the shipping ports has been marked by the purple triangle marks in Fig. 1.

3) The name of all provinces within e.g. 2000 or 3000 km of the sampling site have been added in Fig 1.

4) the plotting scale of the lower-right box has been added.

5) The upper-right box showed the compass of the map but failed to display, there must be an error in the file conversion.

6) That line was due to an error in file conversion and it is corrected now.

2. The provincial level Hg emissions in Wu et al. (2016) could be provided as supplemental information.

Response: The provincial level Hg emissions in Wu et al. (2016) have been provided as supplemental information in the revision.

3. Please provide the distance between the sampling site and the nearest coastal line, and comment on whether the sampling site is capable of capturing the land-sea circulation.

Response: The distance between the sampling site and coastal lines is about 50 km. One recent study reported that shipping emissions influenced the air quality in not only coastal areas but also the inland areas hundreds of kilometers away from the sea in China, as shown in the figure below (Lv et al., 2018). In this regard, our sampling site is well capable of capturing the land-sea circulation.

In the revised manuscript, we have added more descriptions about the sampling site.

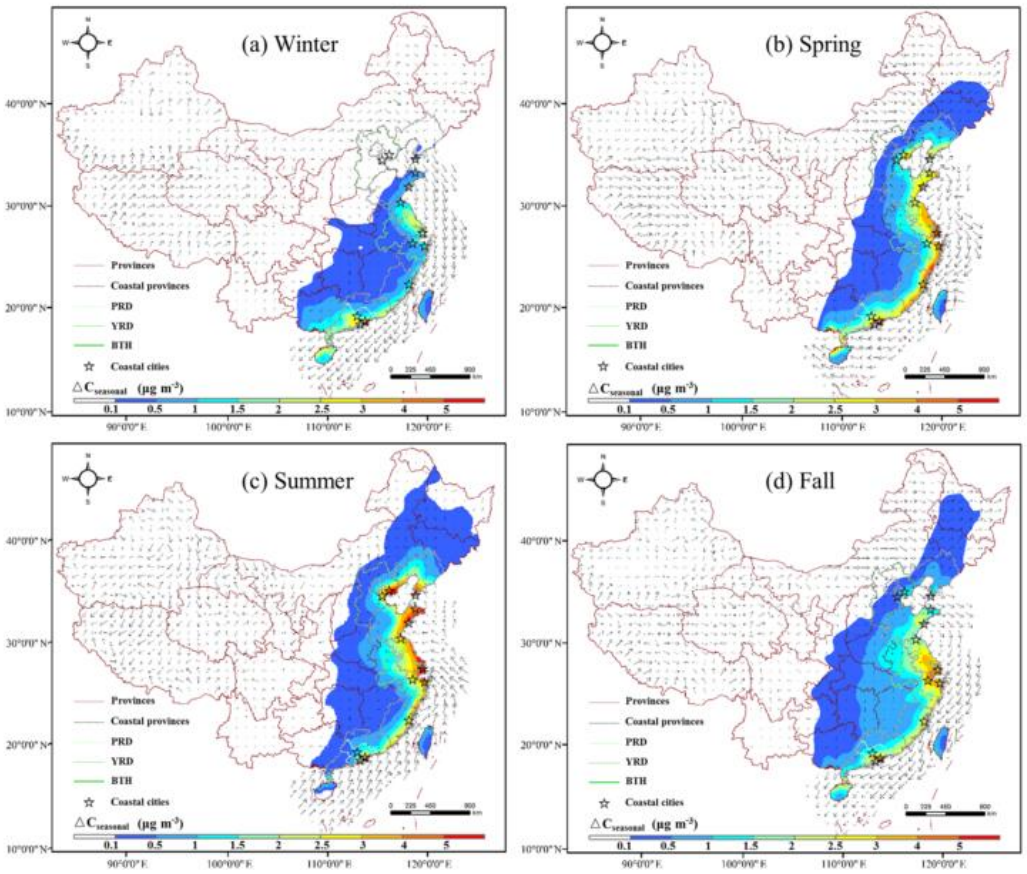


Figure 5. Contributions of shipping emissions to the seasonal mean $PM_{2.5}$ concentrations (base case- no ship case): **(a)** winter, **(b)** spring, **(c)** summer and **(d)** fall. Arrows represent the WRF modeled seasonal mean surface wind field.

4. L147, "meteorological data", please provide the height of the instrument about ground level, and comment on whether the instrument is capable of capturing the meteorological condition in the study area.

Response: The instrument was set up on the top roof of a four-story building, which is a supersite carefully maintained by Shanghai Environmental Monitoring Center (SEMC). The height of the instrument is about 14 m above the ground. The sampling site is located in the rural areas and beside the shore of Dianshan Lake. There are no tall buildings or mountains nearby, making sure that this site is capable of capturing the meteorological conditions in the study area.

5. L149, other pollutants, please provide the reporting or averaging intervals of CO, ozone and PM2.5 mass measurements.

Response: Thanks for the suggestion. The average intervals of CO, ozone, and $PM_{2.5}$ mass measurements were one hour. We revised the description as "Atmospheric ozone (O_3) concentration was continuously measured using Thermo Fisher 49i, which operates on the principle that ozone molecules absorb UV light at a wavelength of 254 nm. The ambient carbon monoxide (CO) and $PM_{2.5}$ concentrations were measured by Thermo Fisher 48Itle and Thermo Fisher 1405F, respectively. All data were averaged at the intervals of one hour." in the revision

6. L205 and L362, please explain what is "secondary formation".

Response: In Line 205, it means the oxidation process of GEM to GOM. In Line 362, it means secondary particle formation. We have expressed more clearly about this term in the revision.

7. L302-304, "The potential source regions of all year round PBM were mainly from northeastern China, including Jiangsu, Anhui, Shandong, and Hebei provinces." Please justify the classification that Jiangsu and Anhui belong to northeastern China.

Response: Thanks for the comments. The classification of the provinces in the original manuscript is not well defined. We revised the description as "The potential source regions of all year round PBM were mainly from the north of Jiangsu and Anhui provinces, and from northeastern China, including Shandong and Hebei provinces."

8. section 3.2.2 (L333), all eight wind directions should be included in the discussion. The authors many want to point out that there is little difference in the wind direction distribution between the GOM/PBM bins of 1-2 and 2-3.

Response: Thanks for the suggestion. We revised the sentence as "As shown in Fig. 8, the frequency of south, southwest, west, and northeast winds showed no clear trend as the GOM/PBM ratios increased." in the revision.

9. Figure 2, please explain the meaning of the red-dish line.

Response: The red-dashed line represented the mean concentrations of each season. We added the sentence "The red-dashed lines represent the mean concentrations of Hg species in each season." in the caption of Figure 2.

10. Figures 5 (b), (c), (d), what is the meaning of "15 ws"?

Response: "15 ws" in Figures 5(b), (c), (d) meaning the wind speed is 15 m/s. The radii of the circle in Figures 5 (b), (c), (d) represent the value of wind speed.

11. Figures 9 and 12, please explain the meaning of the bars.

Response: The bars in Figures 9 and 12 represent one standard error of the parameters in each bin. It has been clarified in the captions of the figures now.

12. Figure 12 caption, “Temperature as a function of the GOM/PBM ratios” Please explain why temperature would change with changing GOM/PBM ratios in the main body.

Response: This expression is not appropriate. We have changed it as “Temperature variations in each bin of the GOM/PBM ratios”.

13. Figure 13, please explain the meaning of the shaded areas.

Response: The shaded areas in Figure 13 represented one standard error of GOM and PBM concentrations.

Editorial suggestions:

1. The significant number seems to be excessive at times, e.g. one decimal will suffice when presenting Hg concentrations.

Response: Thanks for the comment. We do agree with the reviewer that the significant number is excessive at times. After referring to a number of related papers, we revised the significant number as one decimal for GOM and PBM concentrations and two decimals for GEM concentrations in the revised manuscript.

2. Avoid the use of first person, i.e. “we”.

Response: Thanks for the suggestion. All statements involving the use of first person have been revised in the revision.

3. Please define all abbreviations, e.g. PSCF, PBL, RH, in the main body instead of the headings.

Response: Thanks for the suggestion. All abbreviations have been defined in the main body instead of the headings in the revised manuscript.

4. Inconsistent expressions, e.g. “Equation 1” (L160), “as below” (L163), “shown as Eq. (3)” (L173); “secondary inorganic aerosols (SNA)” (L359), “SNA (sulfate, nitrate, and ammonium) in PM2.5” (L715); RGM (Figure 6).

Response: Thanks for the suggestions. All the inconsistent expressions have been revised in the revision.

5. There are quite a few awkward sentences, e.g. 34-35, L171-172, L268-270, L393 (“Se, As, and Pb, which were typical tracers of coal combustion”), L403-404, L443.

Response: Thanks for pointing it out. The awkward sentences have been adjusted.

6. There are quite a few awkward phrases, e.g. “obvious” (L35 and other places), “comprehensive” (L86, suggest to remove), “Xij is” (L175) should be “where Xij is”, “about” (L305 and other places, suggest to replace with “approximately”), “As similar as Fig. 9” (L452), “CN” in Table 1 should be replaced with “China”.

Response: Thanks for pointing it out. The awkward phrases have been replaced or removed according to the suggestions of the reviewer.

7. The manuscript could be shortened by 1) removing some unnecessary material (e.g. L170-186, those model descriptions could be removed by citing the Users Guide by USEPA or other publications), 2) condense the discussion and conduction sections.

Response: Thanks for the suggestions. We have revised the manuscript as suggested.

8. Figure 2, the seasons seem to be incorrect.

Response: Thanks for pointing this out. The seasons in Figure 2 have been corrected in the revised manuscript.

9. Figure 4 caption, suggest to remove “The red line and black line represented the corresponding diurnal variation of RH and wind speed, respectively.” because there are legends to represent each and all variables.

Response: Thanks for the suggestion. The sentence has been removed in the caption of the figure.

10. Figure 6, the ranges should be, <13, 13-15, 15-17, etc.

Response: Thanks for the suggestion. The presenting of the ranges has been changed in the revision.

11. Figure 8, the ranges should be 0-1, 1-2, 2-3, >3.

Response: The presenting of the ranges has been changed according to the suggestion of the reviewer in the revision.

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