Response to the Referees' comments

Reviewer 1

Accepted as is.

Reviewer 2

Specific comments:

1. Line 186: Using 12 trajectories to represent one GOM or PBM measurement is a large source of uncertainty. Simply mentioning that other studies had done the same thing doesn't address the uncertainties in this current study. The trajectory directions can change significantly over a 12 hr period, but this variation is not reflected in the GOM and PBM measurements. More discussion is needed on how these uncertainties would affect the PSCF results.

> Although many studies have used much finer meteorological data for 12hr- or even 24hr-measurements we agree that there may be uncertainties existing when different time scales for measurements of pollutants and meteorological data are used. While we cannot quantify these uncertainties we checked to see how large the trajectory directions changed during 12hr period from 1st to 5th sampling period. The results are shown in Fig. 1R to Fig. 5R below. It turns out that the divergence of trajectory directions during 12hr is considerably smaller than the divergence of trajectories with different starting heights (200 m and 500 m in this study). Therefore, we do not think that the possible source areas identified by PSCF are seriously misidentified in this study.
>
> We have added additional discuss about possible uncertainties in section 3.2.2 PSCF results, as follows.
>
> *"It should be noted that different temporal resolutions for trajectories (hourly) and concentrations (every 12 hr) were used for GOM and PBM. Since trajectory directions can significantly change over the course of 12 hrs there is a possibility that some source areas could be misidentified, especially for more distant regional sources. However, upon investigation it was determined that over the 12 hr sampling periods the trajectories did not diverge significantly at this sampling site for most sampling periods."*

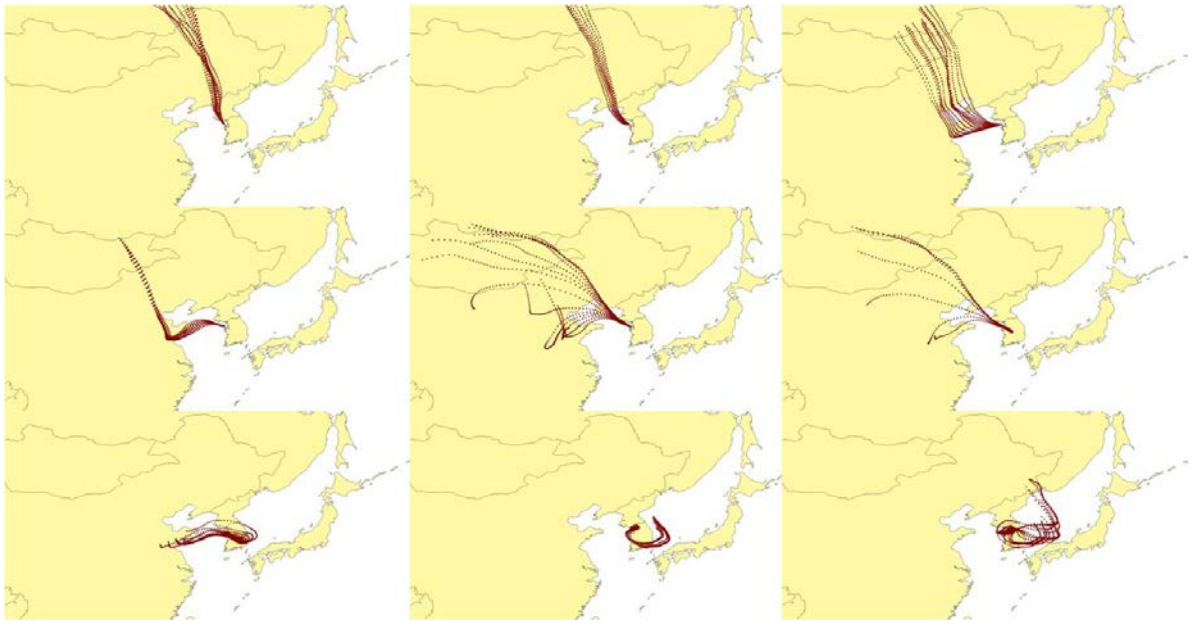

Figure 1R. The divergence of trajectories during each 12-hr for the 1st sampling period.

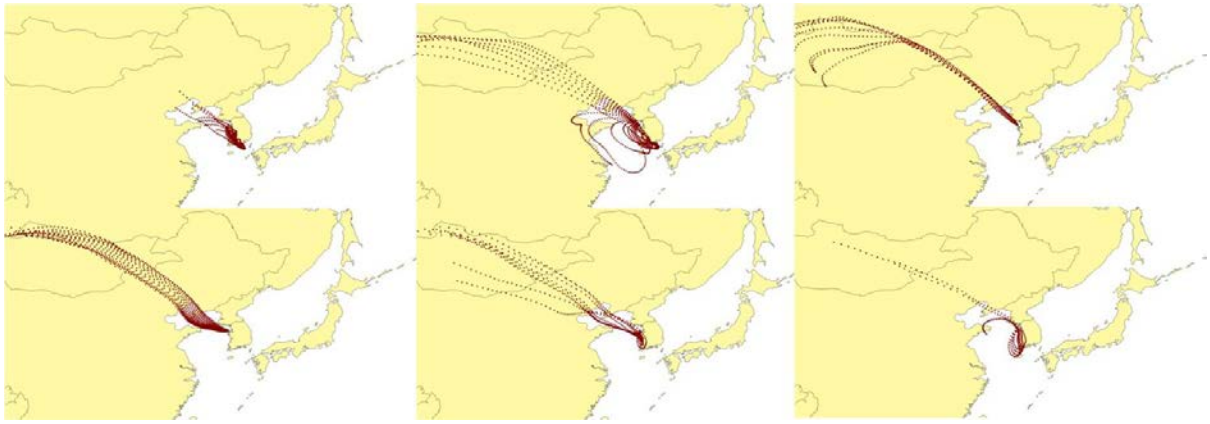

Figure 2R. The divergence of trajectories during each 12-hr for the 2nd sampling period.

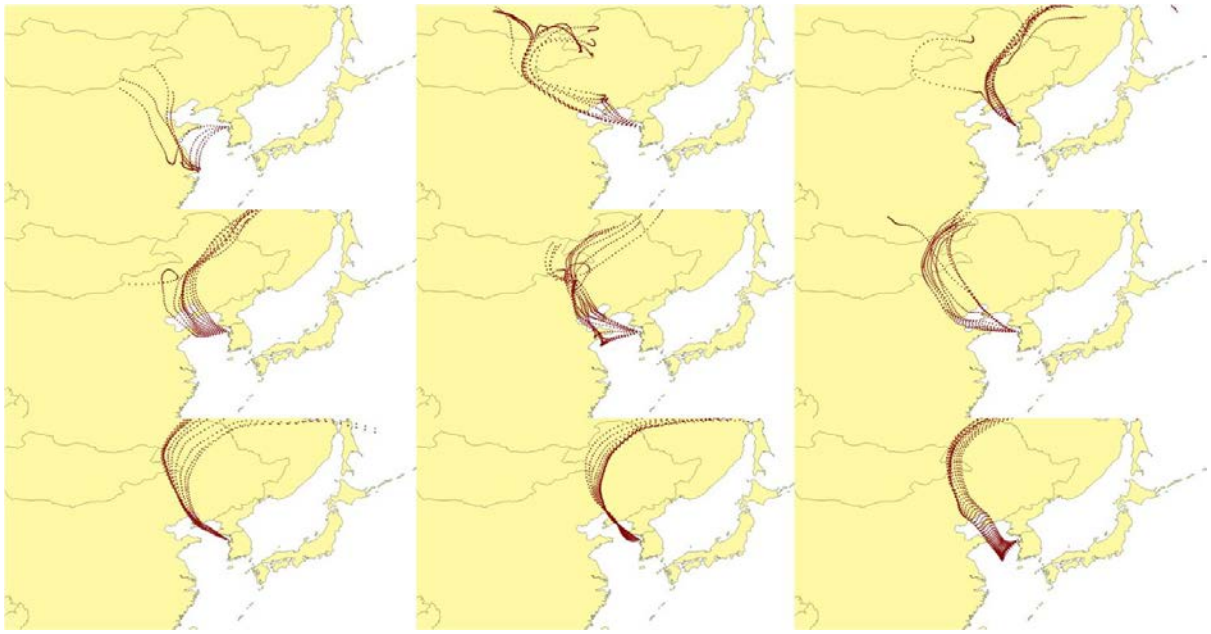

Figure 3R. The divergence of trajectories during each 12-hr for the 3rd sampling period.

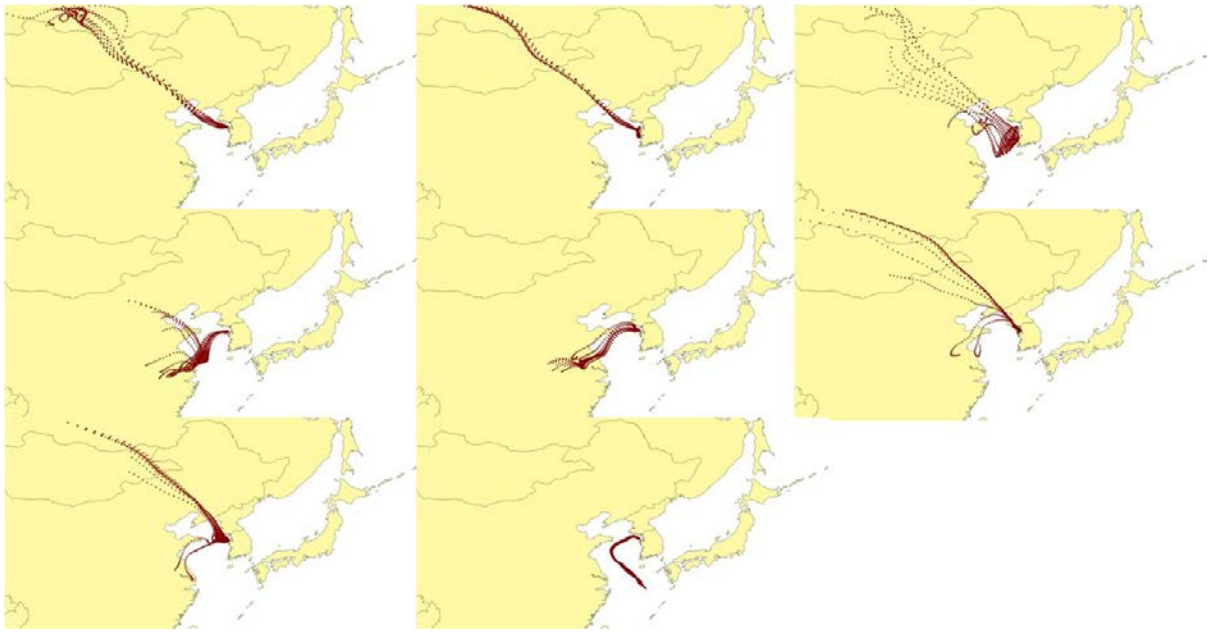

Figure 4R. The divergence of trajectories during each 12-hr for the 4th sampling period.

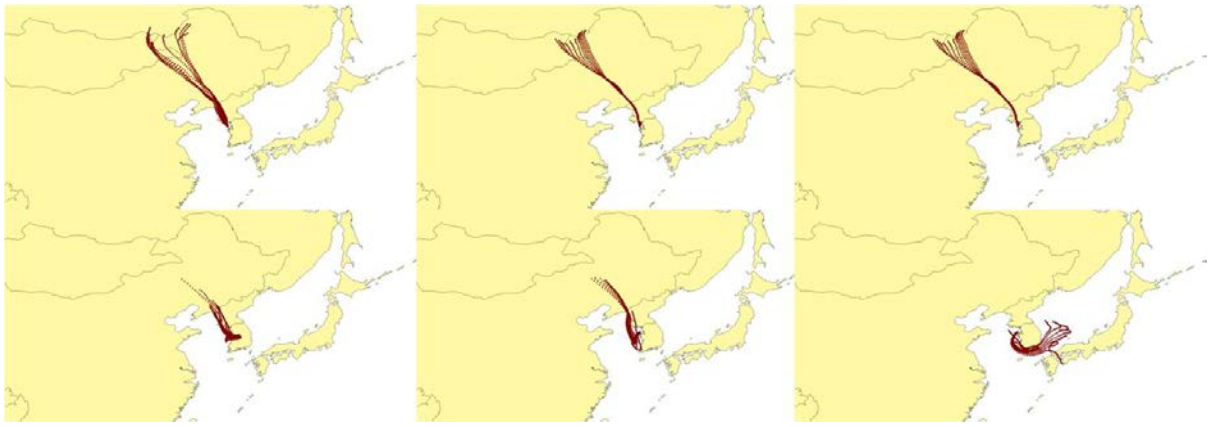

Figure 5R. The divergence of trajectories during each 12-hr for the 5th sampling period.

2. Lines 189-190: I suggest not using “successful” because the modeled sources haven’t been evaluated.

We have deleted “successful”. We have also slightly changed the sentence, as follows.

“Although there have been many studies using different time scales for measurements of pollutants and for meteorological data (Amato and Hopke, 2012; Galindo et al., 2011; Kim et al., 2007) this mismatch might increase the uncertainty for the trajectory-based approaches.”

3. Line 211: Similar to the above comment, using 12 wind directions to represent one GOM and PBM concentration is a large source of uncertainty. Wind directions can change significantly over a 12 hr period but you wouldn’t know the impact to GOM or PBM because there is only one measurement. More discussion is needed on how these uncertainties would affect the CPF results.

Basically, our GOM and PBM concentration measurements represent 12-hr averaged values for 1-hr WD data. As the reviewer indicates the impact of this mismatch on the results is unknown however as we indicated above over the 12 hr sampling periods the trajectories did not diverge significantly at this sampling site for most sampling periods. In response to this comment, we have added the following sentences.

“The 1hr-averaged wind direction (WD) data were used for 12hr-averaged GOM and PBM concentrations for most of periods, so that in total 12 WS and WD were used for one averaged GOM and PBM concentrations to create CPF. It should be noted that there are some unquantifiable uncertainties derived from using different time resolutions between measurements of GOM and PBM and WD data although overall trajectories did not diverge

significantly at this sampling site for most sampling periods"

4. Lines 295-297: The partial correlations from multiple linear regression should be reported. This gives the unique contribution of each independent variable to Kp. It's not the same as performing two separate linear regressions for temperature and RH.

The partial correlation coefficients are -0.389 (p-value<0.001) for temperature and 0.375 (p-value=0.001) for RH. We have added the partial correlation coefficients in the revised manuscript, as follows.

"The multiple linear equation fit the data well (R^2=0.29, R=0.54, p-value<0.001), and both variables of temperature RH were statistically significant (p-value<0.001). The partial correlation coefficients were -0.389 for temperature (p-value<0.001) and 0.375 for RH (p-value=0.001)."

5. Line 303: What was the goodness of fit (R2) for this equation? Gas-particle partitioning models developed in other studies should be mentioned in the results as well. The studies below were able to obtain a higher R2 and validated the model with Kp data at other sites.

Rutter, A. P., & Schauer, J. J. (2007). The effect of temperature on the gas–particle partitioning of reactive mercury in atmospheric aerosols. Atmospheric Environment, 41(38), 8647-8657.
Cheng, I., Zhang, L., & Blanchard, P. (2014). Regression modeling of gas-particle partitioning of atmospheric oxidized mercury from temperature data. Journal of Geophysical Research: Atmospheres, 119(20), 11864-11876.

R was already included in the manuscript for this equation, and R^2 has been added in the revised manuscript. We have reviewed the two references as suggested. Cheng et al. (2014) calculated Kp from 10 AMNet sites located in North America, and linear regression models of log(1/Kp) and 1/T resulted in R^2 between 0.04 and 0.53. Rutter and Schauer (2007) obtained an R^2 of 0.77 from Milwaukee and Riverside. The slopes and y-intercepts from both studies were similar to the results of Amos et al. (2012).

In response to this comment, we added the results from the two references the reviewer suggested, as follows.

"In Amos et al. (2012), the coefficients, ß and y_0, ranged from -1600 to -3300 and 6 to 13,

respectively, and R^2 ranged from 0.16 to 0.57 at different monitoring sites. Rutter and Schauer (2007) also determined the relationship between Log(1/K_p) for urban aerosols with inverse temperature and found the slope and intercept were -4250 ±480 and 10 ±2, respectively, with R^2 of 0.77. Another study using datasets from 10 AMNet sites located in North America found R^2 ranged between 0.04 and 0.53. In summary the values derived in this study fall between those reported by other studies."

6. Line 404: Uncertainties related to different aerosol composition and temperature with different wind directions needs to be mentioned here. Both of these factors can affect gas-particle partitioning, which in turn affects K_p or the GOM/PBM ratio.

In response to this comment we have added the following sentences.

"It should be noted that there are uncertainties related to different aerosol composition, particle surface area variations, and/or temperature change which can affect both Kp and the GOM/PBM ratio."

7. Lines 486-488: These results could be very different using the median concentration for each cluster. If the mean and median concentration for each cluster varies significantly, the source contribution results would likely be very different. In this case, which measure is most representative of the concentration for each cluster and should be used to determine the source contributions?

For both TGM and PBM the results are consistent using means and medians. Only for GOM are results using the median different from the results shown in the manuscript. However we obtained consistent results for all three Hg species when maximum, 75th percentiles values, geometric means, and the arithmetic means are used. Based on the totality of these results, we feel the results reported are valid. However, we agree that it is not clear which measure is most representative of the concentration for each cluster.

In response to this comment, we have added the following sentence regarding the issues related to the choice of the representative statistics for each cluster and the future work required to sort this out in the Conclusion and Implication section

"On average, contributions from out-of-Korean sources were similar to Korean sources for

TGM whereas Korean sources contributed slightly more to the concentration variations of GOM and PBM compared to the out-of-Korean sources. However, in general, conclusions using this approach are more uncertain when the concentration ranges are similar between clusters. Additional work is needed with this approach to determine if a different statistic (other than mean) would provide better results when there are not distinct concentration differences between clusters. In addition, uncertainties exist in the source attribution approach based on cluster analysis because the trajectories inevitably overlap between different clusters since the cluster analysis accounts for both variations in transport speed and direction simultaneously. Nevertheless, this new approach can augment existing methods including CPF and PSCF to help identify source contributions to the concentration variations at the sampling site."

8. Lines 466-476: The results here are very inconsistent. Line 466 states, "Compared to the other clusters, the source contributions of clusters 1 and 4, which represent regional transport, were relatively low for all Hg species (Table 4). Cluster 5 contributed more significantly, especially for GOM and PBM, indicating the importance of Korean sources." Later on line 475 states, "Clusters 1 and 5 were used to represent the effect of sources outside of Korea and the cluster 4 was used to indicate the effect of sources in Korea."

Thank you for pointing out this typo. We have changed "clusters 1 and 4" to "clusters 1 and 5" on line 467. Also, "cluster 5" has been changed to "cluster 4" on line 467.