

## Response to comments from Referee #1

### Anonymous Referee #1

Overall, the discussion paper provided some insights into the potential sources and atmospheric circulation patterns affecting speciated atmospheric mercury at a remote high elevation site in western China. I can understand from the discussion paper that this site location is an important background site because the air emissions transport to the site covers a large area (including Asia, Europe, and north Africa), and it will be able to monitor the regional air quality as countries, such as China and India, continue to rapidly develop. The paper provided some discussion of the regional-scale (Indian Summer Monsoon) and localized (mountain valley breeze effects) atmospheric patterns that may influence mercury concentrations at the sampling site, which should always be discussed when conducting air quality analysis studies. However, the discussion paper currently lacks the data to support some of the explanations of the mercury results and more detailed discussions are needed throughout the paper, as described in the specific comments below. In addition to these issues, there were many errors in the discussion paper that need to be corrected before it can be accepted for publication in ACP.

**Response:** We thank the reviewer very much for recognizing the significance of our work and providing suggestions to our paper. Shangri-La, as the reviewer pointed out, is indeed a unique location to study the regional atmospheric transport because this site is nearby important source regions and located in a high-altitude area of southwestern China. The Indian summer monsoon has profound influence to the climate in Shangri-La. Therefore, we thought that it is important to monitor and report the distribution of atmospheric mercury at this unique site. The reviewer's specific comments are addressed as followed:

Pg 11042, line 15: "...Backward trajectory analysis of air masses associated with TGM levels". This statement is vague. What TGM levels are you referring to? Did you mean high TGM levels?

**Response:** We have checked and revised to "high atmospheric mercury" (P1, L26). The atmospheric mercury concentrations more than  $2.5 \text{ ng m}^{-3}$  should be a high TGM level in Shangri-La area.

Pg 11042, line 21-23: You should mention that Hg needs to be in an oxidized form before it enters ecosystems through wet deposition.

**Response:** We thank the reviewer for pointing this out and have revised the wording as suggested (P1, L2).

Pg 11043, line 2-5: The range of mean or median concentrations in remote sites should be reported, instead of the range because the range of GEM, GOM, and PBM that you are reporting for remote sites are considered very high. Are you including sites in the polar region as remote? In this case, GOM and PBM may be elevated during atmospheric mercury depletion events. But you have not referenced any monitoring studies conducted in polar regions.

**Response:** The Polar sites are not included in the original manuscript because the polar region has distinct meteorological conditions compared to the sites at low-latitude. However, we agree with the reviewer that the paper will be more comprehensive by including mercury observations in the Polar regions. In the North Atlantic, the GEM, PBM and GOM median concentrations are  $1.6 \text{ ng m}^{-3}$ ,  $11.3$  and  $3.2 \text{ pg m}^{-3}$ , respectively. In Antarctica, the mean annual GEM concentration of  $0.93 \pm 0.19 \text{ ng m}^{-3}$  is in good agreement with recent southern-hemispheric measurements. The GOM range has been revised with additional references in the revised manuscript (P2, L7-13).

Pg 11044, line 1-5: Can you describe more clearly the importance of the three studies mentioned? Is the first sentence related to the second sentence since it was mentioned that the Tibetan glaciers are melting more rapidly due to black carbon and Hg concentrations in snow packs are elevated? Are you implying that the melting of snow due to black carbon would release Hg more rapidly into the environment?

**Response:** We meant to indicate that the air emissions including Hg and black carbon from South and Southeast Asia can be transported to Tibetan plateau and deposit on glaciers through Indian summer monsoon. In this study, we did not consider to measure the mercury flux of snow melting, but the previous studies already indicated that the high black soot and mercury should be from south Asia and Southeast Asia. Therefore, Shangri-La is an important observational site for studying the long-range transport of Hg emitted from these sources. We have made this clear in the revised manuscript (P3, L1-9).

Figure 1: What is the source of data for the anthropogenic emissions? Please state whether they are annual emissions.

**Response:** The data are the 2010 mercury inventory estimated by AMAP. We have made this clear in the figure caption (P18, L1).

Pg 11045, line 9-10: Please include the source of the Hg emissions data.

**Response:** The data are the 2010 mercury inventory estimated by AMAP. We have made this clear in the revised manuscript (P4, L3).

Pg 11045, line 23: Include the percent accuracy of the Hg vapor analyzer based on manual injection calibrations.

**Response:** We appreciate that reviewer's point and have reported the accuracy as suggested (P4, L16).

Pg 11046, line 3: Please clarify whether this denuder-based system is separate from the TGM measurements using the Hg vapor analyzer.

**Response:** The denuder-based system is not separated from the TGM measurements using the Hg vapor analyzer. This has been made clear in the revised manuscript (P4, L23).

Pg 11046, line 6-7 and 9-10: Were the PBM and GOM samples analyzed immediately after the sampling cycle? It doesn't seem feasible to replace the impactor and quartz particulate filter after every sampling and analysis cycle when the PBM and GOM samples are being

collected every 2 hours. Were the PBM and GOM samples were collected continuously? How many PBM and GOM samples were collected each day? These procedures are also different from the automated Hg speciation system (Tekran models 1130 and 1135 used in Hg monitoring networks, e.g. AMNet), which does not require replacement of the impactor or the quartz particulate filter after every sampling cycle. Please clarify the procedures.

**Response:** Yes, the PBM and GOM samples were collected continuously and analyzed immediately after the sampling cycle. For continuous measuring PBM and GOM, we prepared additional denuders, filter holders, impactors and filters for collecting PBM so that the sampling cycle can be continued. Each new filter for PBM collection was placed in a filter holder with an impactor placed before the filter in inlet line. A unused KCL-coated denuder was also installed in a separate sampling line with an impactor. Once a two-hour sampling period was completed, the prepared PBM and GOM sampling lines were installed swiftly for the next sampling cycle. The replaced filter and denuder were the immediately heated to 900 and 500 °C using a pyrolyzer for three heating cycles (15 min) to convert PBM and GOM into Hg<sub>0</sub>, which is analyzed by the Tekran 2537A. The typical replacement time of each denuder and filter was very short (almost 10 minutes). Using this sampling protocol, twelve GOM and twelve PBM samples can be collected a day. We have made this clear in the revised manuscript (P4, L22-P5, L6).

Pg 11046, line 20-30: Please clarify that the manual analysis of the denuders and quartz filter were conducted because it is different from the automated Hg speciation system used in other Hg monitoring networks. How long was the analysis cycle and were the denuders and trap analyzed immediately after sampling? Describe how the quartz filter which collects PBM < 2.5\_μm was introduced into the traps and analyzed. If the denuders and quartz filters were not analyzed immediately after sampling, how were the samples stored and how long after sampling were the samples analyzed?

**Response:** The GOM and PBM were sampled simultaneously and each analysis cycle was two hours. During each cycle, we had approximately one hour for PBM and GOM detection after thermal pyrolysis and about one hour for preparing the next sampling installation for swift sample switch. The quartz filter was introduced into a quartz trap via a crochet hook. The procedure is similar to the filter replacement of RPF for automated speciation system. The connection dimension of the quartz trap is the same as the denuder and therefore it was easy to replace the trap after completing a GOM analytical cycle. The manual procedure of PBM and GOM measurement was similar to the automated speciation system. The denuder and filter after the air sampling were analyzed immediately. We have made this clear in the revised manuscript (P5, L8-18).

Pg 11047, line 15-17: What is the range of values for the IMI and what do the values indicate? What value is considered a high IMI or low IMI? Is this value correlated with the rainfall intensity?

**Response:** The IMI is defined as the difference in the 850 hPa zonal winds between a southern region of 5–15 °N, 40–80 °E and a northern region of 20–30 °N, 70–90 °E. The value of IMI represents the intensity of Indian summer monsoon in terms of differential wind speed. When IMI > 0, the study region was considered under the influence of Indian Summer

Monsoon. Zero IMI indicates weak air movement. Negative IMI indicates northerly wind that push the air back the Indian Ocean. The value of IMI is highly correlated with the rainfall intensity. The higher IMI indicates higher chance to bring Indian Ocean water vapor to inlands. We have made this clear in the revised manuscript (P5, L30-P6, L5).

Pg 11047, line 26: What tool or software did you use to conduct the cluster analysis? There are different types of cluster analysis. Please indicate which type was used.

**Response:** HYSPLIT4 was used for the cluster analysis

(<http://www.arl.noaa.gov/ready/hysplit4.html>). This has been made clear in the revised manuscript (P6, L10-11).

Pg 11048, lines 1-12: More details on the cluster analysis method should be included or if you believe this method has been used many times in previous studies, please provide several references, e.g. were the latitude and longitude endpoints clustered separately? How many clusters were chosen? What do you mean by the endpoints of trajectories in the same cluster were averaged (I assume the latitude and longitude were averaged separately)?

**Response:** We agree with the reviewer's comment and have included the references for the cluster analysis in previous studies (P6, L24-25).

Pg 11049: Please include Wij (weighting factor) in Equation 1, since it is part of the equation.

**Response:** We have included the weighing factor in the revised manuscript as suggested (P6, L29).

Pg 11050, line 8-9: The Koch et al. reference is for two European locations (Mace Head, Ireland, and Zingst, Germany). Please check references carefully and include background TGM measurements from North America.

**Response:** We appreciate the reviewer's careful review. We have checked the references and revised as suggested(P7, L29-30).

Pg 11050, lines 24-30: Do you have data to support the effect of mountain valley breezes on diurnal TGM concentrations? Previous studies have also reported higher TGM concentrations and wind speeds during the daytime and lower TGM at night for non-elevated sites as well. But these results were not indicative of mountain valley breezes. The explanation is a good theory for a high elevation site, but it needs to be better supported with data.

**Response:** The meteorological parameters along with the mountain valley were not measured because of limited conditions. In the discussion, we attempted to give the data a rational explanation based on previous report (P8, L16).

Pg 11051, lines 1-7: These explanations need to be supported with data. How low was the relative humidity in the afternoon and how high were the GOM concentrations? You mentioned the oxidation of Hg<sup>0</sup> but do you have the data to support this, e.g. measurements for oxidants of Hg? Could the higher GOM concentrations be due to the mountain valley breezes discussed in the previous paragraph? Without the data and more detailed discussion, I find the last sentence of this paragraph not convincing and too vague.

**Response:** The relative humidity in the afternoon (14:00~20:00) was 81%, much higher than the 58% at night and in the morning (21:00-13:00). Meanwhile, the mean GOM concentrations in the afternoon was  $9.22 \text{ pg m}^{-3}$  compared to the  $7.34 \text{ pg m}^{-3}$  during other period of the day. Previous study in the Rocky Mountains reported that the buildup of GOM is limited to the occasion when dry air is present. Oxidants of Hg was not measured in this study. However, Shangri-La has stronger solar radiation in the afternoon and therefore the oxidation of GEM is a possible cause for the high observed GOM according to previous reports (Lindberg et al., 2002;Goodsite et al., 2004;Fain et al., 2009). The TGM concentration did not show a distinct diurnal pattern and was most likely caused by the meteorological conditions. We have provided the statistics of RH to support the analysis(P8, L18-26).

Pg 11051, line 22: If you are going to mention correlation, please provide the correlation coefficient.

**Response:** In ISM seasons, the correlation ( $r$  value) between TGM and RH was -0.83 respectively. The data have been supplemented in the revised manuscript (P9, L10).

Pg 11051, lines 23-24: What is RH/AT? Is it the ratio of two parameters or is it either parameter (RH or AT)? Please explain why higher RH causes lower TGM concentrations or provide some references to support this finding. RH likely has no effect on GEM because it is not water soluble. GOM is more water soluble, but GOM concentrations are typically 100 times lower than GEM. Why do you think RH will have a significant effect on TGM?

**Response:** RH or AT is mean RH or AT, but we also realize AT should not be a significant parameter to the monthly variation of Hg, we already deleted it. We agree with the reviewer that RH itself may not have a significant effect on GEM. However, we would like to point out that high RH can decrease the Hg emission from surface and enhance wet deposition of Hg, which could contribute a relative lower TGM level (Seo et al., 2012;Poissant and Casimir, 1998). Therefore, given the significant anti-correlation between RH and TGM observed in this study, it is likely that the high RH was possibly a contributing cause to the low TGM level in Shangri-La. This has been clarified in the revised manuscript (P9, L8-15).

Pg 11051, line 24-27: Do you have data to support that the decrease in TGM is due to enhanced Hg uptake by vegetation? E.g., dry deposition rates, CO<sub>2</sub> data was used in Obrist et al. (2008), etc. Can you find some studies that have estimated how much Hg is removed from the atmosphere by vegetation uptake and include these results?

**Response:** Thank you very much for your attention! As you thought, we did not measure the deposition rates and CO<sub>2</sub> data, so our deduction should be reluctant. We already deleted this part and supplemented the other rational explanation in revised manuscript (P9, L8-15).

Pg 11052, lines 8-10: Your data shows that GOM and RH are strongly negatively correlated. What does the strong negative correlation suggest in terms of potential atmospheric processes and sources? Faïn et al. (2009) discussed about the potential transport of free tropospheric (dry) air masses that are elevated in GOM to the Rocky Mountains site. Does this apply to the SAWRS, why or why not? If it does, please discuss this mechanism and why the GOM concentrations at SAWRS are much lower than those observed at other sites affected by the

subsidence of free troposphere (125- 145 pg m<sup>-3</sup>).

**Response:** The air at the SAWRS is dry (55%) with strong solar irradiance in winter. This favors GOM production from GEM according to previous reports. However, importantly, the SAWRS was built in the heavily forested valleys, and the air flow in Shangri-La was dominated by westerlies which perform the highest wind speed of 2.34 m/s in the study, especially the south tributary of westerlies, the air flow from south and southeast Asia could transport to SAWRS along with the valleys, therefore the GOM level at SAWRS could be effected primarily by local photochemical transformation or meteorological conditions, compared to the subsidence of free troposphere. This has been clarified in the revised manuscript(P9, L20-32).

Pg 11052, lines 18-20: The weak correlation with rainfall doesn't support the scavenging of PBM and GOM by precipitation; therefore, you should not state that the correlations indicate scavenging by precipitation. At  $r = -0.18$ , the  $R^2$  is only 0.036, indicating only 3.6% of the variance in GOM is explained by precipitation. It is even lower for PBM.

**Response:** Yes, the negative correlation between GOM and PBM and rainfall was not distinct, given the weak correlation in the study, just indicate that the rainfall could has a certain role to the low GOM and PBM in ISM period, even the weak scavenging might be a possible reason. In fact, the rainfall events were appeared in sampling period of spring (May 8-18) and summer (July 10-25), this could be the reason that we measured the low GOM and PBM due to the scavenging of precipitation. We already revised the wording in this paragraph (P10, L3-12).

Pg 11052, line 23: What do you mean by “possible ISM months”?

**Response:** Thanks a lot. The word “possible” was deleted already (P10, L16).

Pg 11052, lines 23-26 and Fig. 7: You should also explain what a negative and zeroIMI value indicate.

**Response:** Zero IMI indicates weak air movement. Negative IMI indicates northerly wind that push the air back the Indian Ocean(P6, L3).

\*Section 3.3: Can you conclude there was a strong effect of the ISM on TGM concentrations? It appears only one of the 3 elevated TGM events were related to the ISM because of the southerly airflows. You mentioned that the ISM is associated with higher rainfall. Can you add more discussion on how the rainfall events affected TGM, GOM, and PBM?

**Response:** Yes, the precipitation events occurred predominantly during the ISM period ( $IMI > 1$ ). however the high Hg peaks were not appeared frequently, this could be related to the location and topography of Shangri-La, which is in the Tibetan Plateau, when the air flow from south and southeast Asia climb up to Shangri-La, the air flow will move speed slackened and formed cloud, perhaps the cumulus process could cause dilution of the air masses, and we can see that the wind speed was low of 1.63 m/s in summer. Therefore the wet deposition of Hg mainly happened before the air flow arrive in Shangri-La. Additionally, In ISM period, the wet air masses were mainly form Indian ocean and could dilute the mercury concentration in air, this two reasons might be why there were not many high Hg peaks appeared in ISM

period. The relationship between RF and PBM and between RF and GOM is discussed in Section 3.2 and we already added the discussion on how the rainfall events affected TGM (P10, L19-28).

Pg 11053, lines 16-17: "...were grouped into four clusters (Fig. 11) to understand the regional transport pathways." This statement is very vague. What is the purpose of analyzing the back trajectories using cluster analysis?

**Response:** Cluster analysis of backward trajectories is a useful tool to show the synoptic air transport pattern and has been applied extensively. The cluster analysis in this study showed the prevailing air flow arriving at Shangri-La and facilitated the identification of potential Hg source regions. We have made this clear in the revised manuscript (P11, L8-10).

Pg 11053, lines 17-19: The average trajectory for cluster 1 in Fig. 11 does not appear to pass over Siberia. Please state the results carefully.

**Response:** We thank the reviewer for pointing this out and have revised the text accordingly (P11, L14).

Pg 11054, lines 13-15 and Fig. 13: The back trajectories associated with the lower quartile of TGM don't appear very different from those associated with the upper quartile of TGM (Fig. 12). They also do not appear to be from random directions (same airflow directions in Fig. 13 as Fig. 12). Please explain in more detail how they are different. If Fig. 12 and Fig. 13 are similar, it suggests the air masses don't really contribute to elevated Hg.

**Response:** We appreciate the reviewer's insight and would like to clarify this point. Actually, Fig. 12 and Fig. 13 represent different information. We re-made the seasonal plots (Fig. 13 and Fig. 14) from the upper and lower quartile of TGM. In the revised manuscript, the back trajectories associated with the lower quartile of TGM is longer and higher. These indicated that the air masses were traveling well above the planetary boundary layer where ground based emission may not be incorporated in the air masses during low TGM period. This has been clarified in the revised manuscript (P11, L30-P12, L 6).

Pg 11054, lines 19-21 and Fig. 14: Did you conduct a seasonal PSCF analysis? The study that you mentioned is related to the seasonal changes of air movement caused by monsoons. If you want to say the PSCF analysis supports the previous study, you should present the seasonal PSCF results.

**Response:** We did not conduct a seasonal PSCF analysis because a significant portion of the trajectories reaches the ground level and must be rejected from the trajectory analysis. PSCF values are based on conditional possibility and therefore a larger number of trajectory endpoints will yield a greater power of the statistics. In this PSCF analysis after rejecting the grounded trajectories, 484 trajectories and 58080 endpoints were included and will give representative source regions despite the trajectories in all seasons were included in calculating the PSCF values. In fact, in new manuscript, seasonal changes of air movement is displayed in Fig. 13-Fig. 14.

Pg 11055, line 7: Be more specific in the conclusion about which meteorological factors



affected the seasonal variation of TGM.

**Response:** We agree with the reviewer on this comment and have list the meteorological factor in the conclusion section (P12, L24).

Pg 11055, lines 9-10: You have not described in the discussion how the moist air from ISM affected GOM and PBM concentrations. Please include more discussion about this before making this conclusion.

**Response:** The precipitation events occurred predominantly during the ISM period (IMI>1). However the high Hg peaks were not appeared frequently, this could be related to the location and topography of Shangri-La, which is in the Tibetan Plateau, when the air flow from south and southeast Asia climb up to Shangri-La, the air flow will move speed slackened and formed cloud, perhaps the cumulus process could cause dilution of the air masses, and we can see that the wind speed was low of 1.63 m/s in summer. Therefore the wet deposition of Hg mainly happened before the air flow arrive in Shangri-La. Additionally, In ISM period, the wet air masses were mainly form Indian ocean and could dilute the mercury concentration in air, this two reasons might be why there were not many high Hg peaks appeared in ISM period. The discussion regarding how the moist air from ISM affected GOM and PBM has been added in section 3.2 (P10, L19-28).

Pg 11055, lines 11-12: Your discussion of the diurnal TGM trend in pg 11050 did not include temperature. Why are you mentioning in the conclusion that it is mainly due to a diurnal temperature shift?

**Response:** Thank you very much for your attention! Yes, the relationship between surface temperature and TGM had not been discussed, we have checked and revised as suggestion (P12, L27).

We deeply appreciate the reviewer's effort in providing the editorial comments below and have revised the text according to the reviewer's suggestions.

Technical corrections:

Pg 11042, line 2: "This study reports the speciated concentrations..."

**Response:** The text has been revised as suggested (P1, L14).

Pg 11042, line 6: "Gaseous Oxidized Mercury, GOM)", to be consistent with Gaseous Elemental Mercury in previous line

**Response:** The text has been revised as suggested (P1, L17).

Pg 11042, line 8: "...potential influence of the Indian summer monsoon (ISM) and westerlies on the atmospheric transport of mercury."

**Response:** The text has been revised as suggested (P1, L19).

Pg 11042, line 9: "The mean  $\pm$  standard deviation concentrations of ...", to clarify which statistic follows the  $\pm$ .

**Response:** Thanks a lot. We have checked and revised as suggestion (P1, L20).



Pg 11042, line 14: “low PBM and GOM levels were attributed to...”. Wet scavenging is a form of deposition. Did you mean dry and wet deposition? If it is only wet scavenging, you can delete deposition because it is redundant.

**Response:** The sentence has been revised to “Low PBM and GOM levels during the ISM period were attributed to the enhanced wet scavenging” (P1, L24).

Figure 1 caption: “The location of SAWRS, anthropogenic Hg emissions ( $\text{g km}^{-2}$ ) and major cities in Asia with a large population and industrial production.” Please check for grammar.

**Response:** The figure caption has been revised to “Map showing the location of SAWRS, anthropogenic Hg emissions ( $\text{g km}^{-2} \text{y}^{-1}$ ) and major cities in Asia” (P18, L1).

Pg 11044, line 24: “The SAWRS is a remote highland site located in Hengduan Mountains area southeast of the Tibetan Plateau.”

**Response:** The text has been revised as suggested (P3, L23-24).

Pg 11045, line 4: “Other large cities (Chengdu, Guiyang and Chongqing) are east of Shangri-La.”, a close parentheses is needed.

**Response:** The parenthesis has been added as suggested (P3, L29).

Pg 11045, line 6: “Southeast Asia is due southeast.” Southeast Asia seems too broad of an area. Why do you need to mention this? Is Shangri-La not part of southeast Asia?

**Response:** India and Bengal are located to the west of SAWRS and Southeast Asia (In this study, it defined as the region  $92^{\circ}\text{E}$ - $140^{\circ}\text{E}$ ,  $10^{\circ}\text{S}$ - $23^{\circ}26'\text{N}$ ).

Pg 11045, line 8: “There are no large-scale industrial activities and fossil fuel consumption in the area.”

**Response:** The text has been revised as suggested (P3, L34).

Pg 11045, line 13: “The inlet of the heated Teflon...”

**Response:** The text has been revised as suggested (P4, L7).

Pg 11045, line 24-25: The sentence was already stated at the beginning of this paragraph.

**Response:** The text has been revised as suggested (P4, L18).

Pg 11046, line 3: “The annular denuder tubes...”

**Response:** The text has been revised as suggested (P4, L23).

Pg 11046, line 21: You repeated the reference to Landis et al.

**Response:** The repeated reference has been eliminated (P5, L8).

Pg 11047, line 17-18: Please check the HYSPLIT website for the correct reference citation.

**Response:** The citation of HYSPLIT has been updated (P6, L4).

Pg 11048, lines 13-15: The description of PSCF needs to be moved down, before the PSCF

equation.

**Response:** We have moved the PSCF text to before PSCF equation (P6, L29).

Pg 11048, lines 16-18: This sentence needs to be moved up to the cluster analysis section.

**Response:** The text has been revised as suggested (P11, L10-12).

Pg 11048, line 25: "Overall, more than ..."

**Response:** The text has been revised as suggested (P7, L1).

Pg 11049, line 13-14: "(9.7\_10.2 ng m<sup>-3</sup> in Guiyang, Fu et al., 2011; 6.74\_0.37 ng m<sup>-3</sup> in Chongqing, Yang et al., 2009)", there is an extra )

**Response:** The extra ")" has been removed (P7, L13).

Pg 11050, line 1: "...possibly weak local sources." "Shangri-La is located between East Asia and South Asia, which are regions with large Hg emissions (Fig. 1)."

**Response:** The text has been revised as suggested (P7, L21).

Pg 11050, lines 9-10: "The elevated background level of TGM at the SAWRS is likely caused by strong regional sources in Asia."

**Response:** The text has been revised as suggested (P7, L30).

Pg 11050, lines 15-16: "...the weak emissions in the Shangri-La County south of the monitoring site."

**Response:** The text has been revised as suggested (P8, L4).

Pg 11050, line 19: "Fig. 4 displays the average diurnal trend...", are you showing an average diurnal trend?

**Response:** Yes, this figure displays the average diurnal trend. We already revised it as suggestion, thanks a lot! (P8, L9).

Pg 11053, line 11: Figure 1 should be Fig. 10

**Response:** We thank the reviewer for catching this. It has been corrected (P11, L2).

Fig. 11: Please label the average trajectory with the cluster number so that readers can easily identify the trajectory cluster in the figure while reading the discussion.

**Response:** The cluster numbers have been labeled in the upper right corner with different colors.

Pg 11053, line 29: "...Cluster 2, also infrequent, could be..."

**Response:** The text has been revised as suggested (P11, L24).

Fig. 12: "The air masses with high Hg from South and Southeast Asia occurred in autumn and summer", not the winter. Please make sure the caption and the discussions in the text are consistent. "The rest of the air masses were from west Asia..."

**Response:** We re-made the seasonal plots (Fig. 13) from the upper quartile of TGM. We have checked and made sure that the caption and the text are consistent (P31, L1-4).

Fig. 14: SAWRS is repeated in the first sentence of the caption.

**Response:** The text has been revised as suggested (P33, L1-3).

## References

- Fain, X., Obrist, D., Hallar, A., Mccubbin, I., and Rahn, T.: High levels of reactive gaseous mercury observed at a high elevation research laboratory in the Rocky Mountains, *Atmospheric Chemistry and Physics*, 9, 8049-8060, 2009.
- Goodsite, M. E., Plane, J., and Skov, H.: A theoretical study of the oxidation of Hg<sup>0</sup> to HgBr<sub>2</sub> in the troposphere, *Environmental science & technology*, 38, 1772-1776, 2004.
- Graydon, J. A., St. Louis, V. L., Lindberg, S. E., Hintelmann, H., and Krabbenhoft, D. P.: Investigation of mercury exchange between forest canopy vegetation and the atmosphere using a new dynamic chamber, *Environmental science & technology*, 40, 4680-4688, 2006.
- Lindberg, S.: Forests and the global biogeochemical cycle of mercury: the importance of understanding air/vegetation exchange processes, in: *Global and Regional Mercury Cycles: Sources, Fluxes and Mass Balances*, Springer, 359-380, 1996.
- Lindberg, S. E., Brooks, S., Lin, C.-J., Scott, K. J., Landis, M. S., Stevens, R. K., Goodsite, M., and Richter, A.: Dynamic oxidation of gaseous mercury in the Arctic troposphere at polar sunrise, *Environmental science & technology*, 36, 1245-1256, 2002.
- Poissant, L., and Casimir, A.: Water-air and soil-air exchange rate of total gaseous mercury measured at background sites, *Atmospheric Environment*, 32, 883-893, 1998.
- Schwesig, D., and Krebs, O.: The role of ground vegetation in the uptake of mercury and methylmercury in a forest ecosystem, *Plant and Soil*, 253, 445-455, 2003.
- Seo, Y.-S., Han, Y.-J., Choi, H.-D., Holsen, T. M., and Yi, S.-M.: Characteristics of total mercury (TM) wet deposition: scavenging of atmospheric mercury species, *Atmospheric Environment*, 49, 69-76, 2012.