Response to reviewers' comments on "Sensitivity analysis of an updated bidirectional air-surface exchange model for mercury vapor" by Zhang et al.

Dear Editor,

We appreciate the helpful comments provided by three reviewers to our paper and we have incorporated their recommendations in the revised manuscript. We appreciate the reviewers for providing the very detailed and constructive remarks. Our point-by-point response to the reviewer' comments is given below. The corresponding changes relating to the reviewers' comments have also been indicated in the response. I hope that the revised manuscript meets the publication standards of *Atmospheric Chemistry & Physics*.

I look forward to hearing from you soon.

Sincerely,

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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 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 ng m^{-3}, 11.3 and 3.2 pg m^{-3}, respectively. In Antarctica, the mean annual GEM concentration of 0.93±0.19 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 ^{0}C using a pyrolyzer for three heating cycles (15 min) to convert PBM and GOM into Hg0, 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_m 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 ^{o}N, 40–80 ^{o}E and a northern region of 20–30 ^{o}N, 70–90 ^{o}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 Hg0 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 pg m^{-3} compared to the 7.34 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_2 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_2 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? Fa ̈n 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^{-3}).

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 R2 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 zero IMI 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 clarifies 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 ± standard deviation concentrations of ...", to clarify which statistic follows the ±.

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 (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 (g km^{-2} 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 92^0E-140^0E, 10^0S-$23^0$26`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^{-3} in Guiyang, Fu et al., 2011; 6.74_0.37 ng m^{-3} 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

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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.
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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.

Anonymous Referee #2

The manuscript entitled “Observation and analysis of speciated atmospheric mercury in Shangri-la, Tibetan Plateau, China” details the atmospheric mercury concentrations at a site in south-central China. This site is part of the Global Mercury Observation System (GMOS) and reporting of the data is potentially important. Unfortunately the paper does little to advance our understanding of atmospheric mercury cycling or atmospheric chemistry and physics in general. The results are not surprising, could have been arrived at by some quick HYSPLIT back-trajectories, and are not at a level that should be reported in a journal with a high impact factor like AC&P. Also several statements are not backed up by data or even a clear explanation. For example, the authors suggest that a diurnal pattern of GOM could possibly be caused by oxidation of Hg0 with stronger solar radiation. However, the authors suggest no mechanism or offer any measurements to substantiate this statement. This oxidation typically occurs in the Arctic, at coastal sites, or areas with high atmospheric halogen concentrations. As far as I can tell this site is none of these. Also in the conclusions the authors state “The TGM concentration was higher during the daytime mainly due to the diurnal surface temperature shift” but there is no data to support this.

Response: We thank the reviewer for recognizing the significance of our observation effort and for providing the recommendations that help us improvement the manuscript. The Shangri-La site is a unique area to study regional transport from South Asia because it is a high altitude site near the

border between China and the important source regions in India and Southeast Asia that The Indian summer monsoon can also influence the climate in Shangri-La. Therefore we thought it is also influenced by the Indian Summer Monsoon. This is the primary motivation of this monitoring effort not to mention that there have been few Hg data in the study region. New findings regarding the anthropogenic impacts of Hg emission from South Asia are reported in the manuscript. This is a site that is fundamentally different from the Arctic sites and therefore we did not include the halogen chemistry typical of the Arctic environment. We have presented additional data analysis in the revised manuscript to address the reviewer's specific comments and technical concerns.

Pg 11045 Line 10: "< 10 g km-2 of Hg annually" needs a citation
Response: we already made the citation in the description of Fig. 1 and gave the source of data (AMAP/UNEP, 2013) (P4, L3).

Pg 11046 Line 3 – 19: Were the measurements conducted with a Tekran speciation unit? If not then there needs to be significantly more information provided. What denuders from what company were used? What filters (part number) from what company were used for PBM? Were there separate sampling streams for GOM and PBM? Were samples collected manually then desorbed later? If so were multiple denuders used? This can introduce bias and data on the variability between the denuders needs to be provided. Was the same 2537A used to analyze the PBM and GOM and GEM? If so there should be gaps in the data, and if not there is some error that needs to be discussed. Has this system been used for other studies? If not then a diagram of the instrument set up is required. Also the dates when GOM and PBM were measured needs explicitly stated.
Response: The sampling of GOM and PBM in this study was performed manually followed by detection using Tekran 2537A, not with the Tekran speciation unit. The sapling method was similar to the automated process via Tekran speciation system. Both 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. An 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 then immediately heated to 900 ^{0}C and 500 ^{0}C using a pyrolyzer for three heating cycles (15 min) to convert PBM and GOM into Hg0, 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, L25-P5, L6).

Pg 11049 Line 12: "level" should be "levels"
Response: The text has been changed as suggested (P7, L12).

Pg 11049 Line 19: "TGM mean concentration at the SAWRS was slightly higher" this is HUGE understatement. Nearly twice as high is not "slightly higher" and should not be called a "background" site in any way. If it must be called a "background" site then it should be referred to as a "Chinese background" site because the concentrations are much too high to be considered a global background

site.

Response: Yes, SAWRS is a regional background site, the TGM level at SAWRS was much higher than the global background value, because it is closed to the southwest China which is huge mercury sources area. We have checked and revised as suggestion (P7, L19).

Pg 11050 Line 1: "possibly the weak local" should be "possibly weak local"

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

Pg 11050 Line 3: "prevailing westerly," should be "prevailing westerly wind,"

Response: The text has been changed as suggested (P7, L24).

Pg 11050 Line 4: "measured at in Korea" should be "measured in Korea"

Response: The text has been changed as suggested (P7, L23).

Pg 11050 Line 6: How is 2.04 ng m^{-3} at Cape Hedo a higher concentration than the 2.55 ng m-3 mean measured at SAWRS?

Response: We meant that the 2.04 ng m^{-3} at Cape Hedo is also higher than the background concentration in the Northern Hemisphere. We have made the statement clear in the revised manuscript. (P7, L22-26)

Pg 11050 Lines 11 – 14: Higher TGM levels with northerly winds and elevated TGM associated with southerly winds? Is there something missing here? Are winds from the north and south both high in TGM? If so what is the range in concentrations for the southerly winds?

Response: The higher TGM concentration was associated with winds coming from the north and south as shown in Fig. 3 (2.5~2.7 ng m^{-3} for northerly winds and 2.5~2.6 ng m^{-3} for southerly winds, compared to 2.3~2.5 ng m^{-3} for winds from the east and west). This has been made clear in the revised manuscript (P8, L3- 9).

Pg 11050 Lines 11 – 12: ": : :northerly winds that carried domestic emissions from West China: : :" Were there any tracers measured to provide proof that these were domestic emissions from this area? I need more convincing here, the evidence is not definitive and needs support.

Response: The explanation of the doubt is in the first paragraph of section 3.4. In this paragraph, we discussed the figure 11, we note that Cluster 4, although relatively infrequent (4%), was associated with the highest TGM concentrations (mean = 3.9 ng m^{-3}) due to the passing of air masses over known source regions in Sichuan province. The area has a high background TGM level (3.98 ng m^{-3}) caused by industrial and domestic coal combustion, smelting industries, cement production, and biomass burning (P11, L17-21).

Pg 11050 Lines 16 – 17: "low TGM concentrations." What is meant by this? What is the range or mean?

Response: Here we meant that the concentration associated with the winds from the east and west was lower (<2.5 ng m^{-3}). This has been made clear in the revised manuscript (P8, L5).

11050 Line 18 – 19: "The air mass in the east-west direction: : :" What mountains did the air mass

need to cross in each direction? What are the elevations compared to the measurement site? Distance from site? Is the site in a valley? This should be described better in the methods section: : :

Response: We agree with the reviewer that the site description can be more specific. The SAWRS is located at a mountaintop in Hengduan Mountain area, the mountain ranges of Hengduan Mountain area run in the north-south direction. Kawa Karpo and Gongga Mountain both are more than 6000 m a.s.l. and almost 150 Km away form site. Kawa Karpo is located in the northwest of SAWRS and Gongga Mountain is northeast of SAWRS. Therefore the air masses in the east-west direction needed to cross high mountains to reach the SAWRS. We already added the description in the section of Materials and methods as suggested (P3, L25-31).

Pg 11051 Line 3 – 4: "Another possibility is the oxidation of Hg0 caused by stronger solar radiation." What mechanism is being suggested here? This typically occurs in the Arctic, at coastal sites, or sites with high atmospheric halogen concentrations, but I do not think this site falls into these categories, so is there evidence to back up this statement?

Response: Because we did not measure the oxidants of Hg in this study, but SAWRS is located in a very high elevation area (3580m a.s.l) which has much stronger solar radiation compared to the low elevation area, therefore we deduce that the oxidation of GEM could be a possible reason of high GOM according to previous reports in polar area. Additionally, the low relative humidity could be benefit to elevate GOM from GEM. And previous study at a high elevation research laboratory in the Rocky Mountains reported that the build-up of GOM is limited to the presence of dry air (P8, L18-26).

Pg 11051 Line 11: What does the term "generally" mean? Were they statistically significantly lower? If so what are the statistics?

Response: We agree with the reviewer that the statement can be more specific. At SAWRS, AT, RH and RF were lower during non-monsoon period compared to the values in monsoon period. We have also revised the text to reflect the specific description (P8, L32-33).

Pg 11051 Line 12 – 13: "associated with high WS, from Tibetan Plateau caused the lower observed TGM." How did these air masses "cause" lower TGM? Fewer sources? More forests? What proof is there?

Response: We agree with the reviewer that the statement can be more specific. The statement was based on the results of cluster analysis (section 3.4). The reasons for the lower TGM concentration were fewer anthropogenic emissions and possibly the greater degree of air dilution caused by the stronger wind. We have made this clear in the revised manuscript (P10, L19-27).

Pg 11051 Line 13 – 14: "The TGM level was highest in spring: : :" Is this statistically significant? If so what is the p value? N?

Response: The higher TGM level in spring is statistically significant via independent sample test, the p value=0, N=69532

Pg 11051 Line 17 – 18: "Elevated TGM" What does this mean? Are the concentrations above the mean? Above 2.5 ng m-3?

Response: We thank the reviewer for pointing this out to help improve the clarity. The criterion for

the "elevated" level is above 2.5 ng m^{-3}. We have made the criterion more explicit in the revised manuscript (P9, L3-6).

Pg 11051 Line 22: What is a "general negative correlation"? Is it a statistically significant correlation? If so what are the r and p values?

Response: The negative correlation is statistically significant (r=-0.83, p value=0) via independent sample test and we have deleted the "general" in the revised manuscript (P9, L9-10).

Pg 11051 Line 25 – 26: "enhanced Hg uptake by vegetation: : :" What vegetation surrounds the site? What is the process by which this occurs?

Response: Thank you very much for pointing out this. In our new manuscript, we did not think "enhanced Hg uptake by vegetation". Because, SAWRS is surrounded by alpine forest and the major vegetation kinds in south area of SAWRS are alpine and evergreen broad-leaved trees and what the enhanced Hg uptake by vegetation just happened during ISM period is impossible. In our new manuscript, we think during ISM period, clean Indian Ocean air mass and water vapor can flow into inland and lead to high RH and low TGM. In addition, during rainfall formation, cumulus process also could cause dilution of Hg in atmosphere. We already deleted this part and supplemented the other rational explanation in revised manuscript (P9, L8-15).

Pg 11051 Line 26 – 27: ": : :which has been observed several earlier study in Chang- Bai Mountain and in the Northeast US: : :" should be ": : :which has been observed by two earlier studies in the Chang-Bai Mountains in China and the Rocky Mountains in the US: : :" If the authors are making this statement, significant discussion is needed. The site in the US was a very high elevation site, far above tree line and quite there was quite a bit of discussion. Please put this study into perspective with those studies and tell us in detail why this may be a vegetation effect.

Response: Thank you very much for your attention! We already deleted this part and supplemented the other rational explanation in revised manuscript (P9, L8-15).

Pg 11051 Line 28: "winter months were" should be "winter months was"

Response: The text has been changed as suggested (P9, L15).

Pg 11052 Line 2: What does "not substantial" mean? What was the mean, range, and standard deviation?

Response: We meant that the variability of WS was not significant and have made it clear in the revised manuscript (P9, L17).

Pg 11052 Line 4 – 6: What does "substantial seasonal variability" mean? Are there significant differences? If so what are the p values?

Response: It is mean the variation was distinct. P value=0 via two- tail test.

Pg 11052 Line6: "5 times of the" should be "five times the"

Response: The text has been changed as suggested (P9, L23).

Pg 11052 Line 6: ". Similar pattern: : :" should be "A similar pattern: : :"

Response: The text has been changed as suggested (P9, L23).

Pg 11052 Line 10: "were high in autumn and winter: : :" what does "high" mean? How high? Compared to what?

Response: Compare to the PBM level (29.14 pg m^{-3}) in spring and summer, the PBM level (37.78 pg m^{-3}) was higher in autumn and winter.

Pg 11052 Line 14: "Trajectories analysis" should be "Back-trajectory analysis"

Response: The wording has been revised as suggested (P10, L3).

Pg 11052 Line 15 – 16: "air mass from Burma, Vietnam, Laos, Kampuchea and Thailand, likely of biomass burning origins." Is there any data to back up this statement? Other atmospheric tracers? Information on forest fires or biomass burning of any kind?

Response: We appreciate the reviewer's insightful comment. The satellite images of MODIS for biomass fires provide a good representation of the biomass burning activities (https://earthdata.nasa.gov/data/near-real-time-data/firms/active-fire-data#tab-content-7) and we agree with the reviewer that the primary burning season is in spring season. Therefore, high PBM level in autumn could be caused by the local anthropogenic sources, but previous measurement of atmospheric Hg in Southeast Asia reported quite high TGM concentrations (Sheu et al., 2013) . This suggested that the rapid development of industry in the Indochina Peninsula might also have contributed to the elevated atmospheric Hg concentrations to some extent. We already revised it as suggested (P10, L3-12).

Pg 11053 Line 1: "three high Hg events". Why are two highlighted in red and one in blue?

Response: We thank the reviewer for pointing this out and would like to clarify it. The event marked in red represent the causes of these events are not only impacted by local airflow but also long-term transport; while blue just represent the local airflow impact.

Pg 11053 line 11: "Figure 1" There are no back-trajectories on Figure 1.

Response: We thank the reviewer for catching this. It should be Figure 10 and it has been corrected in the revised manuscript (P11, L2).

Pg 11053 Line 13 – 14: "The elevated concentration in this event was likely to be contributed by the domestic emissions." How do you know this?

Response: The peak cannot be explained by the transport of air masses as indicated by trajectory analysis and therefore it was suspected to be caused by domestic emissions that are closer to the monitoring site.

Pg 11055 Line 5: Northern Hemispheric background is 1.5 – 1.7 ng m^{-3} not 1.5 – 2.0 ng m^{-3}.

Response: The values have been revised as suggested (P12, L22).

Pg 11055 Line 10: "level" should be "levels"

Response: The word has been changed as suggested (P12, L26).

Pg 11055 Line 11 – 12: "The TGM concentration was higher during the daytime mainly due to the diurnal surface temperature shift." This was not proven in the manuscript. Thorough discussion needed. Is this referring to the cold air drainage off of the mountains? If so are there any measurements of boundary layer height? Balloon launches? Modeling?

Response: Thank you very much for your attention! Yes, the relationship between surface temperature and TGM had not been discussed in the revised manuscript. We already deleted the wording and revised as suggested (P12, L27-28).

Pg 11055 Line 13 – 14: "and the in situ photochemical productions of might have contributed to the occasional high GOM concentrations." First, this should be "and in situ photochemical production might contributed to occasionally high GOM concentration." And second, this is not supported by any data and therefore cannot be a main finding of the paper.

Response: We have made the editorial revision as suggested (P12, L29-30). Regarding the photochemical production, it is a logical reasoning from the strong solar irradiance at the site and was not included as the primary finding in this work.

Table 1. Some indication of the number of samples should be given.

Response: We have added the indication of the number of samples (P17).

Fig. 1. The countries should be delineated and labeled.

Response: We agree and have labeled the countries (P18).

Fig. 2. Five-min mean implies that many samples were taken and a mean was taken. Is this correct? If not then "Five-min mean" should be "Five-min samples".

Response: Yes, because we set Tekan 2537A to get a data every five minutes.

Fig 3. "roses" should be "rose"

Response: We thank the review for catching the typo and have corrected it in the revised manuscript (P20).

Fig 4. Second and third sentences are results and should not be in the figure legend.

Response: We agree with the reviewer's suggestion and have shortened the caption (P21).

Fig 5. Not sure what the second sentence in the legend is saying and third sentence is a result and should not be there.

Response: We agree with the reviewer's suggestion and have shortened the caption (P22).

Fig 6. Sentence two and three are results.

Response: We agree with the reviewer's suggestion and have shortened the caption (P23).

Fig 7. What do the boxes indicate and why are they different colors?

Response: We appreciate the reviewer for pointing out this. Highlighted in red represent the causes of these events are not only impacted by local airflow but also long-term transportation; while blue just represent the local airflow impact.

Fig 8 and 9 legends have results that should not be there.
Response: We agree with the reviewer's suggestion and have made new figures (P25, P26).

Fig 10 is not referenced in the text.
Response: This has been addressed in an earlier comment and we thank the reviewer for pointing this out (P10, L33).

Fig. 11 should be more clearly described in the text and only the information needed to interpret the figure given in the legend. As it is it makes no sense.
Response: We agree with the reviewer's suggestion and have shortened the caption (P28).

Fig. 10 to 14 could be deleted.
Response: These Figures are very important to analyze the long range transport of Hg and potential resources for SAWRS.

References

AMAP/UNEP: Technical Background Report for the Global Mercury Assessment 2013, in, Arctic Monitoring and Assessment Programme, Oslo, Norway/UNEP Chemicals Branch Geneva, Switzerland, 2013.

Anonymous Referee #3

This manuscript provides speciated atmospheric Hg data collected at a high-elevation site in China. As this site is located in a region that could see air flow from both East Asia and South Asia, I consider these Hg data are valuable. Nevertheless, these data are not well analyzed and presented in this manuscript so I saw little advance in improving our understanding of regional Hg transport and cycling. Some statements or conclusions made by the authors are not back up by data. For example, the author suggested that the high TGM value observed in June-July was caused by the biomass burning activity in the Indochina Peninsula because air masses were coming from this region to the sampling site. However, the major biomass burning season in the Indochina Peninsula is spring. In June-July, biomass burning activity is very low or not existing. In fact, the authors did not provide any data to demonstrate the influence of biomass burning activity on air quality at the sampling site. Therefore, I think this manuscript needs a major revision before being considered for publication in Atmospheric Chemistry and Physics. Below is a list of comments and suggestions:
Response: We thank the reviewer for recognizing the importance of the data presented in this work and providing us the valuable suggestions for us to improve the paper. The reviewer's review comments and recommendations are well received. We have carefully addressed the technical points and believed that the paper has been substantially improved.

1. Page 11041, line 15: "Backward trajectory analysis of air masses associated with TGM levels : : :" This sentence is not clear. What TGM levels are talking about? High levels or low levels?
Response: We have checked and revised to "high atmospheric mercury" (P1, L26). We mean the high TGM level. The atmospheric mercury concentrations more than 2.5 ng m^{-3} should be a high TGM level in Shangri-La area. We made the back trajectories of the upper quartile and lower quartile of

TGM observations at the SAWRS and analyzed the long range transport of Hg in section 3.4.

2. Page 11043, line 2-5: Wide range of background concentrations of GEM, GOM and PBM are cited for remote sites. However, I won't consider a remote site background if it often sees GEM concentrations greater than 2 ng/mˆ3, considering the Northern Hemisphere background GEM level is 1.5-1.7 ng/mˆ3.

Response: We agree with the reviewer's point that 2 ng m^{-3} is not typical of background site and have re-word the phrase in the manuscript. Consider the high remote location of the SAWRS, the TGM level was anticipated to be close to the global background. This suggests that the site is under the influence of regional emissions and/or long-range transport. We have made this clear in the revised manuscript (P2, L7-13).

3. Page 11045, line 3-4: In this sentence, it said that Kunming city is _650 km northwest of SAWRS. However, it is clear from Fig. 1 that Kunming city is located to the southeast of SAWRS.

Response: We thank the reviewer for catching the typo and have corrected it in the revised manuscript (P3, L29).

4. Page 11045, line 6: I will say that Southeast Asia (e.g. Indochina Peninsula) is located to the south, instead of southeast, of SAWRS.

Response: We agree with the reviewer on this comment and revised it as suggested (P3, L30).

5. Fig. 1: Font size of the city names is too small to read.

Response: We agree with the reviewer on this comment and have changed the font size in Figure 1 (P18).

6. Page 11045, line 9-10: What is the source of the Hg emission data?

Response: The source of Hg emission data is from AMAP emission inventory for 2010, we have cited the source as suggested (P4, L3).

7. Page 11045, line 13-14: Is the Tekran 2537A set up on the roof of a building?

Response: The Tekran 2537A was set up in the room with the inlet of air flow from 5 m above the roof of the building. This has been made clear in the revised manuscript (P4, L8).

8. From Fig. 2 I can tell that TGM was monitored between December 2009 and November 2010. However, it is not mentioned in the section of "Sampling methods and analysis".

Response: We thank the reviewer for pointing this out and have provided the measurement period in the revised manuscript (P4, L7).

9. Page 11046, line 3: Is this the denuder-based system a manual system separate from the Tekran 2537A or is it connected to the 2537A (but it is not Tekran 1130/1135)? The whole description about this system is very confusing. Please provide a detail and clear description.

Response: The sampling of GOM and PBM in this study was performed manually followed by detection using Tekran 2537A, not with the Tekran speciation unit. The sapling method was similar to the automated process via Tekran speciation system. Both 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. An 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 ^{0}C and 500 ^{0}C using a pyrolyzer for three heating cycles (15 min) to convert PBM and GOM into Hg0, 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, L18).

10. Page 11046, line 13: The inlet of the denuder-based system was 1.5 m above ground, but the inlet of the TGM system was 10 m above ground. So the TGM system and the denuder system were not measuring speciated Hg at the same elevation above ground?

Response: Yes, our observation room is a little cabin witch has a vaulted roof, it is not feasible to put the denuder system on the roof. So we just can measure the GOM and PBM on the ground, but the denuder system was quite closed to inlet of sample line, it should not affected the measurement of Hg species. And the mixing of air is good enough to make the GOM and PBM measurement representative.

11. Page 11047, line 15-16: What is the range of IMI values? Could it be a negative value? How to decide a value is high or low? Higher values indicate stronger westerlies?

Response: Thanks for pointing out this. The IMI is defined as the difference in the 850 hPa zonal winds between a southern region of 5–15 ^{o}N, 40–80 ^{o}E and a northern region of 20–30 ^{o}N, 70–90 ^{o}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).

12. Page 11048, line 13-18: It seems to me that line 16-18 should be put in front of line 13-15. Please check.

Response: We agree and have made the change in the revised manuscript (P6, L27).

13. Page 11049, line 2: What is nij? Is it the same as Nij?

Response: We thank the reviewer for catching the inconsistency. It is the same and has been made consistent in the revised manuscript (P6, L29-33).

14. Fig. 2: The highest GEM value looked suspicious. What's the data QA procedure?

Response: Tekran 2537A automatically calibrates for Hg^0 every 25 h using an internal permeation source, which provides approximately 1 pg s^{-1} of Hg^0 at 50 $^\circ$C into a zero air stream. External calibration using Tekran 2505 with manual injections of known concentrations of Hg^0 was performed

every 4 months. The percent accuracy of the Hg vapor analyzer based on manual injection calibrations was 96.84%. The precision is <2% with a detection limit <0.1 ng m^{-3}. Whenever we found high Hg values, the Tekran 2537A was examined and we have ruled out the possible faults of instrument such as leaking and contamination. Therefore the reported data are representative of what was present in the air sample. We have also include the QA procedure in the revised manuscript (P4, L7-20).

15. Page 11049, line: The authors compared their mean GEM value to those of several other mountain sites also in China. I think this comparison should be extended to cover mountain sites in other region of the world, such as the Mt. Bachelor Observatory and Storm Peak Laboratory in the USA (Jaffe et al., 2005; Obrist et al., 2008) and Lulin Atmospheric Background Station in Taiwan (Sheu et al., 2010).

Response: Yes, we already covered several remote high elevation sites of high TGM level, including Storm Peak Laboratory in the USA, the GEM level of Lulin Atmospheric Background Station in Taiwan (1.73 ng m^{-3}) was relatively lower compare to Shangri-La (P7, L22-31).

16. Page 11050, line 6: The mean GEM at Cape Hedo is lower, not higher, than that of the SAWRS.

Response: This is a wording confusion. We meant that the 2.04 ng m^{-3} at Cape Hedo is also higher than the background concentration in the Northern Hemisphere. We have made the statement clear in the revised manuscript. (P7, L22-26).

17. Page 11050, line 9-10: Any analysis to support your argument?

Response: The analysis was discussed in the latter sections 3.4 (P11, L4).

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18. Page 11050, line 18-19: I don't understand the authors' logic. Why will the air masses have lower TGM concentrations simply because they need to cross high mountains?

Response: The SAWRS is located at a mountaintop in Hengduan Mountain area, the mountain ranges of Hengduan Mountain area run in the north-south direction. And Kawa karpo and Gongga Mountain (more than 6000m a.s.l, almost 150 Km away form site) are located in the northwest and northeast of SAWRS respectively. Therefore the air masses need to cross high mountains to reach the SAWRS. When the air flow 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 he wind flows from the east and west gave low TGM concentrations (2.3-2.5 ng m^{-3}). We already checked and revised it (P8, L1-16).

19. Page 11050, line 19: Figure "5" should be Figure "4".

Response: We thank the reviewer for catching this and have correct the figure number (P8, L9).

20. Page 11050, line 24-28: Any data to support the existence of a diurnal valley breeze system influencing the sampling site? The diurnal pattern of RH or water vapor mixing ratio should be

included in Fig. 4 because this may be helpful for the identification of the valley breeze system.

Response: We did not measure the meteorological parameters along with the mountain valley because of limited conditions, we just give a reasoning result according to previous reports. We made the statistics of RH, The relative humidity in the afternoon (14:00~20:00) was 81.47% which was much higher than the night and morning (21:00-13:00) of 58.24%, the diurnal pattern of RH is included in Fig. 4. This indicate that the dry air masses form high elevation should be moved down to the low elevation area (P8, L18-26).

21. Page 11050, line 25-26: In fact, air near the valley, not mountaintop, heats up faster in daytime.

Response: We thank the reviewer for pointing this out and have revised the text accordingly (P8, L13).

22. Page 11051, line 1-7: This argument needs to be supported by data. Otherwise, it's not convincing.

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 pg m^{-3} compared to the 7.34 pgm-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).

23. Page 11051: I think the authors need to perform statistical tests to see if the seasonal differences in TGM, GOM and PBM concentrations are statistically significant?

Response: Yes, we already made statistics test and see At 95% confidence, except spring and winter (Sig.=0.19, by two- tail test) among other season, TGM concentrations are statistically different(Sig. =0.00 for all). Except spring and summer (Sig.=0.53, by two- tail test), among other season, GOM concentrations differences are statistically significant (Sig. =0.00 for all) (P8, L30; P9, L20) .

24. Can this seasonal difference in TGM be due to seasonal difference in natural emissions? Any evidence to say that natural emissions is not a factor here?

Response: We thank the reviewer for pointing this out to help improve the clarity. The natural emissions could be different in different season, but we did not perform the flux measurement in this study.

25. Page 11051, line 22-23: ": : :, a general negative correlation between : : :.". Please perform a correlation analysis and show the correlation coefficient.

Response: In ISM seasons, the correlation coefficient (*r* value) between TGM and the RH was -0.83. We revised the text as suggestion (P9, L10).

26. Page 11051, line 23-24: Why the higher RH in the summer months might have caused the lower

TGM concentrations? Any scientific evidence or reference?

Response: The 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 clarifies in the revised manuscript (P9, L8-15).

27. Page 11052, line 4-16: ": : :: : :.., likely of biomass origins". Well, the major biomass burning season in the Indochina Peninsula is SPRING, especially in March. Therefore, even though these authors observed high PBM with air masses from the Indochina Peninsula in FALL, it is very unlikely to be due to biomass burning. Any data to indicate the air quality at the sampling site was influenced by biomass burning?

Response: We appreciate the reviewer's insightful comment. The satellite images of MODIS for biomass fires provide a good representation of the biomass burning activities (https://earthdata.nasa.gov/data/near-real-time-data/firms/active-fire-data#tab-content-7) and we agree with the reviewer that the primary burning season is in spring season**.** Therefore, high PBM level in autumn could be caused by the local anthropogenic sources, but previous measurement of atmospheric Hg in Southeast Asia reported quite high TGM concentrations(Sheu et al., 2013) . This suggested that the rapid development of industry in the Indochina Peninsula might also have contributed to the elevated atmospheric Hg concentrations to some extent. We already revised it as suggested (P10, L14-28).

28. Page 11052, line 23-24: I can't understand this sentence "The values IMI represent seasonal rainfall anomalies, : : :"

Response: The description was meant to indicate that the higher precipitation associated with the Monsoonal winds, which occurs when the IMI is positive. We have revised the wording to make it clear in the revised manuscript (P5, L29; P6, L4).

29. Fig. 8-13: What are the altitudes of these trajectories? If an air mass passes an anthropogenic Hg emission source region at a high altitude, its Hg concentration may not be influenced by this anthropogenic emission.

Response: We thank the reviewer for pointing out this. We have made the new trajectory plots with endpoint heights information in Fig.8-Fig.14 and re-analysis these figures in new manuscript.

30. Page 11053, line 7-11: Again, biomass burning is unlikely to contribute to the high TGM in June-July because it is not the right season

Response: We thank the reviewer for pointing this out to help improve the clarity. Like the response in question 21, the biomass burning activities from Southeast Asia are not heavy in the wet and warm season. We agree the reviewer's viewpoint and corrected as "increasing consumptions of resource" (P10, L15-28).

31. Page 11053, line 11: Figure "1" should be "10".

Response: We thank the reviewer for catching this. The typo has been corrected in the revised

manuscript (P11, L2).

32. Page 11053, line 18: ": from North Africa and Siberia, : : :". I don't see this line has passed Siberia

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

33. Page 11053, section 3.4: Any statistical test performed to see if these cluster mean TGM values are statistically different?

Response: We have performed what statistical tests and found the TGM mean concentrations associated with different cluster are significantly different (Table S1), particularly in the difference between Clusters 1 and 3, between Clusters 1 and 4, and between Clusters 3 and 4. These results indicate that the TGM concentrations from high altitude (Clusters 1 and 2) were different from the TGM concentrations form low altitude (Clusters 3 and 4), and that the air masses of Clusters 3 and 4 passed Hg emission areas and transported atmospheric Hg to the SAWRS.

34. Fig. 12 and 13: Directions of airflow in Fig. 12 and 13 do not look very different. Can the authors explain in detail how they are different?

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.

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