**Response to comments from referee #3**

**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 arearun in the north-south direction. And Kawakarpo 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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