Response to reviewers' comments on "Observation and analysis of speciated atmospheric mercury in Shangri-la, Tibetan Plateau, China" by Zhang et al.

Dear Editor,

We appreciate the helpful comments provided by the reviewers to our paper and have incorporated their recommendations in the revised manuscript. The constructive remarks indeed improve the quality of the paper. Our point-by-point response to the reviewers' comments is given below. The corresponding changes 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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Reviewer #1:

Abstract L25-26: The statistics in Table S2 and S3 do not support the enhanced wet scavenging of GOM and PBM during the ISM period. The ISM period is between May and Sep (mainly occurring in the summer). No statistical differences in the mean GOM between summer and spring (not ISM) and between summer and fall (not ISM). No statistical differences in the mean PBM between summer and spring (not ISM) and between summer and winter (not ISM). It is more accurate to state that the mean values were lower but statistical differences were not found.

Response: We thank the reviewer for pointing this out and would like to clarify it. We meant to state that somewhat lower PBM and GOM levels during the ISM period were also observed and attributed to the enhanced wet scavenging. The high GOM and PBM could be caused by local photochemical transformation under low RH and the domestic biofuel burning during cold seasons. We have revised the discussion to reflect this (P1, L25-28).

P9, L31 explains that the lower TGM in the winter was due to stagnant winds that limited regional transport. But when you explain why GOM was elevated in the winter, you state that it was due to air masses traveling with relatively high wind speeds (P10, L6). The two statements contradict each other. Was it stagnant winds or high wind speeds in the winter? Wind transport does not seem to explain why TGM concentrations were low and GOM concentrations were high at the same time.

Response: Yes, the wind speed was highest (2.34 m s^{-1}) in winter due to the strengthening Westerlies. During the period, dry air masses associated with high wind speed, from Tibetan plateau caused the lower observed TGM. However, the air masses carried by the Westerlies occasionally passed through

several strong source regions in South and Southeast Asia as well as local anthropogenic sources from domestic biofuel burning for house heating, and potentially transported mercury to the SAWRS. We have revised the discussion and wording (P9, L32-34; P10, L10-12).

P13, L1 in the conclusion also contradicts with the results. In the results, it was stated that high TGM in the spring and late April was due to regional transport from Northwest China (P9, L16-22 and Table S1). But ISM is associated with air masses from South Asia and occurs from May to Sep. Which air mass direction coincided with higher TGM?

Response: We meant to highlight separate transport events during the spring when Northwest China was occasionally an important source region for the high TGM levels observed in Shangri-La. Most of air masses with high Hg were form South Asia and Southeast Asia as discussed in section 3.3 and shown in Fig. 8. From June 19 to July 2 as shown in Fig. 9, the air mass was mainly from the Bengal and Burma because of the ISM, suggesting trans-boundary transport possibly caused by anthropogenic sources in the region and the industrial cities (Fig. 1) such as Yangon (Burma's capital), Bangkok (Thailand's capital) (P11, L7-13). Therefore there is no contradiction.

Conclusion: Avoid using the word "caused" because the study has not establish any causal relationships.

Response: We thank the reviewer for pointing this out and have revised the wording as suggested (P13, L5; P13, L6).

Table S4 caption: It should be the mean TGM concentrations associated with clusters 1 and 2 and clusters 2 and 3 that are not significantly different, and not clusters 1 and 4, 2 and 4, and 3 and 4.

Response: We thank the reviewer for pointing this out and have revised the wording as suggested (Table S4).

Reviewer #2:

Major comments

This is the third round of reviews for the manuscript "Observation and analysis of speciated atmospheric mercury in Shangri-la, Tibetan Plateau, China." Although, some sections are much improved, unfortunately, there are still significant language issues. I have tried to point out some of them, but it is still is not in a final publishable form even after three reviews. The authors have done a good job showing how the denuders were analyzed (for GOM) but still have not reported exactly how they analyzed the filters for PBM (e.g., were the filters removed from the filter holders, placed in a non-annular denuder and heated in a Lindberg Furnace?). Although they state that their method is identical to a Tekran speciation unit, this is not true. Having to remove a filter each time a sample is collected is not an identical analysis method, it is similar but not identical.

The manuscript could also use a reorganization. Section 3.3 and the first paragraph of Section 3.4 are well written and really work to substantiate the findings. Instead of talking about distribution and influences of potential regional sources and the backtrajectory analysis in separate sections, the results of these two parts should be integrated and the backtrajectories used to more clearly make the point. As the paper is organized right now, statements are made in Section 3.1 and seem unsubstantiated until Section 3.3 – 3.4. The paper would be much clearer if the statements were immediately justified and these discussions were combined.

Further major revisions to the language, method description, and discussion are needed before this manuscript can be published.

Response: We appreciate the reviewer's detailed and helpful comments. Yes, the filters were removed from the filter holders, placed in a non-annular denuder and heated in a Lindberg Furnace to 900 ^{0}C and 500 ^{0}C using a pyrolyzer for three heating cycles (15 min) to convert PBM into GEM, which is then analyzed by the Tekran2537A. The configuration of GOM and PBM analysis were similar to Tekran speciation unit. A new filter was used for each PBM sample in our manual method, compared to a filter being used for multiple times in Tekran speciation unit. The discussion of potential regional sources was based on and continued from the preceding analysis of the back trajectories and therefore we are confident regarding the identified source locations of Burma, Bengal bay, north India, west Sichuan Province and west Yunnan Province. We have also gone through another round of editorial revision in addition to those pointed out by the reviewer.

Specific comments

1) P2 L11: need citations after "respectively"

Response: We thank the reviewer for pointing this out and have added the citations as suggested.

2) P2 L13: need citations after "respectively"

Response: We thank the reviewer for pointing this out and have added the citations as suggested.

3) P2 L19: "help understand" should be "help with understanding"

Response: We have revised the wording to "help in understanding."

4) P2 L29: should be "corner of the Tibetan"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

5) P2 L30: should be "mainland" not "Mainland"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

6) P2 L33: "Recently, there" should be "There"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

7) P3 L5: "pollutants emissions" should be "pollutant emissions"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

8) P3 L17: "aimed to establish" should be "aimed at establishing"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

9) P3 L19 - 20: "The data within GMOS network provide global monitoring data…" should be "The global monitoring data within the GMOS network can be used…"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

10) P3 L22: The citation Gay et al., 2013 is about AMNet. Not GMOS, I suggest a citation about GMOS.

Response: We thank the reviewer for pointing this out and have changed the citation into the website of GMOS for a more complete introduction of GMOS.

11) P3 L31: "away from" should be "of"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

12) P3 L31: "The largest point sources…" In a previous comment I asked what these were and this has not been addressed. Are they coal fired power plants? Cement manufacturing? Gold mining? Just how large are these sources?
Response: We thank the reviewer for pointing this out. Kunming city is a major industrial city with large coal fired power plants. The source intensity has been shown in Fig. 1.

13) P3 L33: "are to east" should be "are east" and what is the distance from Shangri-La.
Response: We thank the reviewer for pointing this out and have revised the wording as suggested. Some other large cities (Chengdu, Guiyang and Chongqing) are east of Shangri-La (~1000 km).

14) P4 L26: "A denuder-based sampling unit was for sampling PBM and GOM and it was separated from the TGM measurements using the Hg vapor analyzer" should be "A denuder-based sampling unit (separate from the TGM sample train) was used for sampling PBM and GOM."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

15) P5 L14 – 15: "other is undergoing" should be "other was undergoing"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

16) P5 L16 – 17: "samples can be collected a day." Should be "samples were collected per day."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

17) P5 L17: "filter after air sampling were analyzed immediately" should be "filter were analyzed immediately after air sampling."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

18) P5 L19 – 28: Still not fully addressing the reviewers concerns! Specifically detail what was done to the quartz filter. On page 5 of the SI it does not show or state how the quartz filter was handled during the analysis by saying "A separate PBM quartz trap of identical connector" does not address this concern. Was the filter placed in this "trap"? How? Were tongs used? If so were the tongs Teflon coated? I see a hollow denuder in Fig S6, was this used? Was the filter analyzed in this? If it was how was the filter placed in the denuder? These issues are the reasons the reviewers are having so many issues.
Response: We would like to further clarify the point here. The size of hollow quartz denuder (trap) for PBM analysis is identical to the denuder for GOM analysis. And yes, the filter was placed in the trap using a removal/installation tool (Nichrome wire with a hook at one end, Tekran P/N: 30-13540-00, Filter Change Tool). The procedure of placing filter is similar to the procedure of replacing filter for RPF in Tekran 1135 unit. Gently bend a quartz filter membrane so that the normally flat disk surface is curved. Slide the curled filter down the tube and locate it in the middle of the trap using the installation

tool. We added the description in revised paper as suggested (P5, L23).

19) P5 Line: Please reference discussion in Supporting Information
Response: We thank the reviewer for pointing this out and have added the reference as suggested.

20) P5 L24: "using a pyrolyzer for three heating cycles" – in Fig. S5 this is a Lindberg Blue clamshell furnace and not a "pyrolyzer". What was a heating cycle? The cycles on the furnace would be heating to the 500 or 900 then completely cooling and heating again. This is not possible with the current set up, in order to be ready for the next sample collection.
Response: We thank the reviewer for pointing this out. A heating cycle is the period when the 2537A analyzes a sample (5 minutes). When we performed the GOM and PBM analysis, the heating temperature of furnace was set up to 500 ^{0}C for the GOM analysis first. Upon completing the GOM analysis, the heating temperature of furnace was set up to 900 ^{0}C for PBM analysis. In fact, the temperature increased from 500 ^{0}C to 900 ^{0}C very quickly (<10 minutes). We had two hours to analyze GOM and PBM, the time was more than enough to prepare the next sample collection. We have revised the wording for the "pyrolyzer" (P5, L25).

21) P5 L26 – 27: Change "the caps were not heated and air cooling was also used to lower the temperature of caps" to "a fan was used to blow cool air on both ends of the denuder."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

22) P5 L34: Change "The blank of denuder and quartz filter" to "The denuder and quartz filter blanks"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

23) P6 L2: "procedure were followed to ensure the data" should be "procedure was followed to ensure data"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

24) P6 L3: "of Tekran 2537A must to be" should be "of the Tekran 2537A must be"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

25) P6 L8: should be "via an automatic"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

26) P6 L15: should be "by air transport of the ISM, and wet air flow from the Indian Ocean will cause high rainfall in southern and southeastern Asia."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

27) P6 L17-18: Remove "A positive IMI also indicates the wet air flow form Indian Ocean will cause high rainfall in southern and southeastern Asia."
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

28) P7 L24 – 25: There is a range of months referred to, but only one spike for each period. Is this a single spike or where spikes typically this high? If they are single spikes the date the spike occurred

should be given.
Response: We would like to point out that there were multiple peaking events in the month (Fig. 7).

29) P8 L17-22: were the differences in concentration by wind direction statistically significant?
Response: We did not perform the statistical analysis for the TGM concentrations and wind direction, but we found that the high TGM concentrations were from north and south of SAWRS.

30) P8 L24: should be "dilution of an air mass"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

31) P8 L26: "and therefore gave" should be "resulting in"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

32) P8 L29: "located at a" should be "located on a"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

33) P9 L9: should be "and photochemical"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

34) P9L13-14: If statistically lower we need a p value, if not then this should be mentioned or discussion removed.
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

35) P9L27: should be "during the ISM" and "air masses"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

36) P9L28: "flow into inland" should be "flow inland" and "In addition, the cumulus"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

37) P9L29: should be "in the atmosphere"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

38) P9L30: should be "from the surface" and "relatively lower"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

39) P9L32: should be "August) likely due"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

40) P9L33: should be "limited regional"
Response: We thank the reviewer for pointing this out. But we have deleted the wording and revised the discussion as suggested by the other reviewer.

41) P9L33-34: So if the wind speeds weren't different then this explanation is not supported by your data and must be deleted.

Response: We thank the reviewer for pointing this out and have revised the discussion and wording (P9, L32-34). The wind speed was highest (2.34 m s^{-1}) in winter due to the strengthening Westerlies. During the period, dry air masses, typically associated with high wind speed, from Tibetan plateau caused the lower observed TGM.

42) P10L27: This section needs incorporated earlier to make the discussion more understandable.
Response: We thank the reviewer for pointing this out and have revised the wording and added the description as suggested (P11, L2-7).

43) P10L29: "values IMI" should be "IMI values"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

44) P10L33: What does "High TGM peaks were not observed consistently" mean?
Response: It means that the high TGM event did not persist throughout the entire ISM period. We have revised the wording to make it clear.

45) P11 L13: This discussion also needs incorporated earlier
Response: We feel that a separate discussion for the trajectory analysis provides a better clearity.

46) P11 L21: should be "moved fast, passing"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

47) P12 L3: should be "from Central"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

48) P12 L6: "were displayed" should be "are displayed"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

49) P12 L8-10: "The winter…" This sentence says that the trajectories were cause by domestic sources, I think this should refer to mercury concentrations instead.
Response: We agree with the reviewer on this comment and have made the change (P12, L13-15).

50) P12 L11: What does "mild air movement" mean?
Response: It means that the air masses moved slowly.

51) P12 L31: should be "on the Tibetan plateau"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

52) P13 L1: should be "levels by occasionally"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

53) P13 L2: should be "while moist air from the ISM likely decreased"
Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

54) P13 L7: Maybe there should be some mention of the very nice cluster analysis results here to support these statements.

Response: We thank the reviewer for pointing this out and have added cluster analysis discussion as suggested (P13, L12-13).

Supporting Info

I have only given this a cursory look, but the text here needs a complete re-write as there are many typographical errors.

Pg 5 last paragraph: "paralyzer" should be "pyrolyzer"

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.

Pg 5: This technique is not identical to the speciation unit, so please remove any reference to "identical."

Response: We thank the reviewer for pointing this out and have revised the "identical" into "similar" as suggested before.

Fig S2: Grass should be Glass

Response: We thank the reviewer for pointing this out and have revised the wording as suggested.