

Atmos. Chem. Phys. Discuss., referee comment RC1
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Comment on acp-2021-357

Anonymous Referee #2

Referee comment on "First Observation of Mercury Species on an Important Water Vapor Channel in the Southeast Tibetan Plateau" by Huiming Lin et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-357-RC1>, 2021

The manuscript entitled 'First Observation of Mercury Species on an Important Water Vapor Channel in the Southeast Tibetan Plateau' by Lin et al. presents ~5 months of speciated mercury concentrations (using online and offline sampling) at Nyingchi during the period preceding and during the Indian Summer Monsoon (ISM). This site is located in an important water vapor channel and thus is ideal for investigating the transport of pollution to the Tibetan Plateau. The authors divide the ISM into three periods, then use back trajectory clustering analysis and principal component analysis to investigate the sources and source regions affecting mercury concentrations. The authors found the PISM periods to be affected by westerly circulation with higher levels of GEM, a distinct diurnal pattern, with long-range transport and local emissions being important factors. While the ISM period was affected by transport from the Bay of Bengal and the Indian Ocean, with lower levels of all mercury species, a different diurnal pattern compared to PISM, and local emissions, meteorology, and snowmelt. They concluded wet deposition and uptake by vegetation to be responsible for the low concentrations observed during the ISM. This manuscript presents the first results from this location and coupled with their previous study from Qomolangma Natural Nature Preserve present an important analysis of pollution entering the Tibetan Plateau. However, there are points where the manuscript could be improved. Their interpretation is sound although requires more discussion. While the manuscript is readable, there are improvements to the language that would aid in the readability. Overall, I recommend the publication of this manuscript after addressing the major revisions outlined below.

General Comments

It is important to make a distinction between which species of mercury the authors are referring to in a specific context. Often 'Hg concentrations' are stated when it isn't completely clear which species (GEM, GOM, or PBM) or which measurement technique (Tekran vs passive samplers) is being referred to in that context.

Throughout the text, the authors write 'under the control of' or 'control period' when referring to transport/circulation patterns. While this is understandable after several

readings and sometime thinking about the meaning, this phrasing can be reworded to be more concise and readable. This would go a long way to improving the ease of readability of this manuscript.

The authors make a great effort to characterize the sources and transport patterns of GEM using clustering of back trajectories and PSCF. However, I was quite perplexed to find that no effort had been made to couple back trajectories to GOM or PBM concentrations.

The GEM passive samplers data are presented although discussed only briefly. This is an underutilized dataset in this manuscript, the large variations in the data warrant further analysis.

The results of the PCA analysis, at least to me, indicate that long-range transport is the dominant source of GEM while local emissions are more important for GOM and PBM. This is a key result from this study which is listed and mentioned briefly. The author proposes yak dung to be an important local source yet only speculate and do not provide any references that show this could be a source of GOM or PBM. A similar comment for the snowmelt factor, during ISM1, snowmelt is a source of GEM and GOM. From Fig. 2, it appears this factor could be occurring only during a short period (the large spike in GEM and GOM at the end of ISM1), which could be investigated in more detail (e.g., was there snow on the ground during this time, what was the wind direction, temperature, RH, solar radiation during this time?). Expanding on the PCA analysis could give more insight into the local sources of Hg species at Nyingchi.

One practical note, please follow ACPs guidelines on the placement of figures and figure captions 'Figures and tables as well as their captions must be inserted in the main text near the location of the first mention (not appended to the end of the manuscript)'. It wasn't practical to change between text and figures, especially when the captions were also in a different location. Also, please put a line between references in the bibliography, it was quite difficult to find a certain reference when they are all bunched together. The references need to be properly formatted as well.

Specific Comments

Line 29: I feel there is a better word than 'infected' which can be used here. Possibly 'influenced'.

Lines 33-36: The authors separate the ISM into three periods but list an average for the entire ISM. Maybe it could be beneficial to list averages for all three periods or list the periods in descending order? There is also a significant overlap between the standard deviations for parameters between periods. Have the authors performed any statistical tests like a t-test or Wilcoxon Rank Sum test to test for significant differences?

Lines 36-37: While the passive sampling was for one year, stating the annual average here can be misleading since this information isn't in the abstract. It could also be beneficial to indicate the seasonal averages or variations instead of just an annual average.

Lines 37-38: The authors should indicate the sampling area was clean compared to other high-altitude sites.

Lines 38-40: These sentences describe only half of the diurnal pattern in the respective periods. It could be beneficial to state other diurnal features present during the different periods. For instance, simply add that during the PISM afternoon concentrations were lower (which is still due to boundary layer dynamics) and that low concentrations of GEM were observed during the morning in the ISM due to vegetation effects.

Line 42: Maybe 'circulation patterns' would fit better here than 'airflow fields'?

Lines 42-43: The authors should indicate that westerly circulation occurs during the PISM.

Lines 45-47: It would be helpful to know during which periods the different factors were dominant.

Line 47: I feel the abstract is missing one sentence stating how this research will be valuable, similar to the wording on lines 121-122.

Line 50: This sentence requires a reference.

Line 55: Are GOM and PBM undergoing chemical reactions that lead to their wet and dry deposition? To my knowledge, this is due to their water solubility (GOM and PBM) and low vapor pressure (GOM). Maybe the authors could be more specific in their description here.

Line 57: 'physicochemical' instead of 'physiochemical'. I also make this mistake which is why I caught it.

Line 60: 'effects'

Line 63-67: I am surprised the Arctic Monitoring Assessment Programme is not listed here (Arctic Monitoring and Assessment Programme | AMAP) as this is an important Hg monitoring network covering North American and European Arctic. Also, it might be beneficial to the reader if references for individual networks are listed with the acronym, similar to the AMNet.

Line 66: The semicolon may be removed and replaced with 'and the'. In my opinion, this will improve the readability of the sentence.

Lines 80-81: As currently constructed, this sentence isn't representative of the text in Chen et al. (2016). From Chen et al. (2016) 'The total fuel-related atmospheric mercury emissions amount to 859.12 t, to which coal, oil products and biomass contribute 85.77%, 9.06% and 5.17%, respectively.' So, it appears coal contributes 86 % of fuel combustion emissions. This sentence should be reworded to reflect this.

Line 112: The Tekran speciation units are quite uncertain in terms of collection efficiency (Maruszczak et al., 2017; Huang et al., 2017; Gustin et al., 2015), therefore I would recommend removal of the phrase 'high-precision' from this sentence.

Maruszczak, N., Sonke, J. E., Fu, X., and Jiskra, M.: Tropospheric GOM at the Pic du Midi Observatory – Correcting Bias in Denuder Based Observations, *Environ. Sci. Technol.*, 51, 863–869, <https://doi.org/10.1021/acs.est.6b04999>, 2017.

Huang, J., Miller, M. B., Edgerton, E., and Sexauer Gustin, M.: Deciphering potential chemical compounds of gaseous oxidized mercury in Florida, USA, *Atmos. Chem. Phys.*, 17, 1689–1698, <https://doi.org/10.5194/acp-17-1689-2017>, 2017.

Gustin, M. S., Dunham-Cheatham, S. M., Huang, J., Lindberg, S., and Lyman, S. N.: Development of an Understanding of Reactive Mercury in Ambient Air: A Review, *Atmosphere*, 12, 73, <https://doi.org/10.3390/atmos12010073>, 2021.

Line 117: When referring to 'cluster analysis', do the authors mean PCA or clustering of back trajectories?

Line 119: 'sources'

Line 131: It could be helpful to the reader if the authors state the temperature for the PISM and the ISM since the manuscript revolves around these periods.

Line 134: Other than the YZB Grand Canyon, what are the other water vapor channels?

Lines 134-135: Similar comment as above but for precipitation.

Line 141: Can the authors give some examples of this unique high-altitude distribution pattern of biomes and vegetation in the area? This would aid the reader and help explain the interpretation that vegetation effects have a significant effect on GEM concentrations.

Line 149: These dates are different from the ones listed in the abstract.

Line 155: 'drawn in' instead of 'sucked' and 'into' instead of 'in'.

Lines 157-160: Having worked with the Tekran instruments, I understand what is meant when the authors describe the sample collection procedure, however, a reader unfamiliar with this procedure could misinterpret the text. The time required to collect and analyze one sample is two hours, one hour for collection and one hour for analysis. This isn't stated clearly here, I suggest rephrasing these sentences to make this clearer to the reader.

Lines 165-167: Can the authors elaborate on the method from Slemr et al. (2016)?

Line 170: Again, these dates are different from the abstract. These dates need to be reconciled. Also, why is a day not stated here when it is other places.

Lines 173-174: The authors need to state a more precise sampling interval for the passive samplers.

Line 175: What is a DMA-80? Can the authors give more information on this instrument?

Line 199: Would 'air parcels' be a better term than 'matter' in this context?

Lines 202-203: What is the typical boundary layer height at Nyingchi? Are there times when the boundary layer is below 1000 m? Have the authors varied the arrival height to see its effect on air mass origin? Have the authors calculated trajectories longer than 72 hours? For GOM and PBM, this length is reasonable, however, for GEM the lifetime is much longer and could be affected by sources further away than 72 hours. While the input meteorological data is at a time resolution of 6 h, the HYSPLIT model can interpolate these data and produce hourly trajectories. This would increase the uncertainty but would allow for measurements of GOM and PBM to be integrated with these trajectories. Have the authors explored such an analysis? Do the authors mean 'simulated' instead of 'stimulated'?

Lines 204-205: The last sentence in this paragraph needs to be reworded.

Lines 206-212: The description of PSCF needs to be expanded. What was the threshold percentile? What was the arbitrary weighting function used? These parameters need to be stated for this research to be reproducible.

Lines 218-222: Can the authors elaborate on the tests and procedures used for determining the optimal solution for the PCA analysis? For example, what are the Kaiser-Meyer-Olkin measure of sampling adequacy and Bartlett's test of sphericity used for? What was the outcome? Please define MSA. Were there multiple elbows in the scree plots?

Line 228: The text states 'daily' here and in other places, but the rightmost y-axis label in Fig. 2 gives units of 'nm² hour'. Can the authors please clarify this discrepancy?

Lines 231-232: What are the criteria for dividing the ISM into three periods in terms of precipitation? Please elaborate on these criteria and the reasoning behind the selection of the timing of the different periods.

Lines 232-235: Please see my comments about listing the concentrations for different ISM periods from the abstract.

Line 235: I think the words 'locally monitored' can be omitted.

Line 237: Same but for 'decisive'.

Line 243: I feel there is a better reference for the chemical properties of GEM than Horowitz et al. (2017), which deals with modeled redox chemistry of Hg. Possibly a review paper, or references from a review paper, might be more appropriate here.

Lines 244-246: Is this total precipitation or an average during these periods? It is interesting that GOM decreased by roughly half while PBM only decreased by ~25 %.

Lines 249-252: This is an important result of a previous study. During the PISM, GEM is mainly from long-range transport, while during the ISM local emissions is an important source of GOM and PBM (from the PCA analysis). These local emissions could be important for total Hg in rainwater.

Line 255-258: It was stated in the site description that westerly circulation patterns are dominant from September to April and that ISM circulation patterns are dominant from May to August. Was this information obtained through trajectory analysis or previous knowledge from the site? This information is again presented here and used to explain the higher passive sampler GEM concentrations in the later part of the sampling period. I am curious if any trajectories were calculated for the passive sampler period? This could be used to directly support the abovementioned statements. The large variations in the passive sampler period, in my opinion, warrant further investigation. What were the meteorological conditions or transport patterns under high and low concentrations?

Lines 258-260: I agree this is most likely the case, given the Hg emission inventory and trajectory clusters plotted in Fig. 5. Calculating trajectories for the entire passive sampler period would directly show this.

Lines 260-262: This is nice since it gives the reader context, however, maybe it would benefit the reader to move it to the beginning of this paragraph.

Line 272: Is there a better way to say 'monsoon control zones'? See general comments above.

Line 276: I feel there is a better phrase than 'violent' to describe depositional processes. Possibly 'extreme'?

Lines 283-284: 'generally believed' isn't the most appropriate language for a scientific article. Please rephrase.

Line 290: Fu et al. (2016) provide an excellent explanation of the decrease of GEM over the whole ISM and the diurnal profile at night. However, this study was conducted in a different geographical region and at a lower altitude. Can the authors offer any reasoning for why this effect is valid at both locations? For instance, is there similar vegetation at

both sites?

Line 291: This is also a very logical explanation for the decrease in GEM during the ISM, however, this statement requires a reference. Have other locations in India observed enhancements of halogens during the ISM?

Lines 291-293: From Fig. 2, it appears that during the beginning of ISM1 GOM concentrations are lower than ISM2 and on a similar level to ISM3. However, there is a large spike in GOM at the end of ISM1 that could be skewing the average for this period. Has this spike in GOM been investigated in more detail?

Line 297: 'deposit' instead of 'settle' since you are referring to wet deposition.

Figure 4: It is impossible to extract information from these figures. Seven axes on one figure are way too many. The lettering for each panel is also very large compared to the figures themselves. The combination of lines with errors represented by dashed lines and dots of small sizes and similar colors is dizzying and makes interpretation unnecessarily difficult. I do not understand why so many parameters are presented when only the Hg species are discussed briefly in the text.

I would suggest either group the Hg species and meteorological parameters separately or group parameters with a similar diurnal profile together. I would then opt for the former and put the diurnal profile of meteorological parameters in the supplement.

Line 314: Any statement that mentions 'previous research' requires references and citations, both of which are missing from this sentence.

Lines 323-325: Can the authors expound upon this speculation? They have offered yak dung as a possible source of local emissions elsewhere in the text, is there any other possible local sources of Hg that could explain this observation?

Lines 330-331: I am not sure what is meant by 'chemical dissipation', and there was nothing in the references given. Do the authors mean chemical reactions? Also, the references don't support the statements in the sentence.

Line 346: Holmes et al. (2010) isn't an appropriate reference for the reduction of GOM in local snowy mountains. Is there not more specific studies (possible lab or field campaigns) that show this mechanism in more detail?

Lines 346-347: What do the authors mean by 'field GEM source'?

Lines 349-350: Please provide references for the Indian Ocean being a source of halogens.

Figure 5: Making the size of the cluster trajectory is a very nice way of intuitively showing the relative proportion of each cluster occurrence, however, it is difficult to grasp the absolute percentage from the legend (this is just an observation not necessarily a suggestion to change it).

Starting the cluster index at zero is a matter of taste, but it is intuitively easier to understand when the index starts at one.

A color scale or color bar is required for the emissions inventories.

Having all the color scales for GEM the same might make it easier to notice the differences between different periods

Line 360: This sentence needs to be reworded. See general comments above.

Line 365: 'relatively'.

Line 367-369: This information about the cluster turning in the Bay of Bengal is not represented in the cluster average. It might be beneficial to show the individual trajectories for each cluster in the supplement. Also, as currently constructed, the citation to the UNEP reports appears to reference the turn in trajectories. I suggest moving the citations to the end of the sentence, this would alleviate any confusion.

Lines 370-372: This is true for GOM and PBM, however, not for GEM, which as stated above in the text, isn't very water-soluble. This is an example, where specifying which Hg species the authors are referring to would lessen any confusion from the reader's perspective.

Lines 374-375: Showing the individual trajectories for each cluster during this period would directly show what the text is stating, as right now, the statement is not evident from Fig. 5b.

Lines 377-378: HYSPLIT can output precipitation and H₂O mixing ratio at each trajectory step, this information would show what the authors are suggesting — water vapor is increased when air masses arrive from the Indian Ocean.

Lines 383-386: A color bar for the Hg emission inventories would be helpful here.

Line 391: De Simone et al. (2015) is about modeled Hg emissions from biomass burning and not with anthropogenic emissions. The UNEP reports seem like a better reference for this statement.

Line 393: It would be more appropriate to list the references given in Lin et al. (2019) for yak dung burning instead of just Lin et al. (2019). I wonder why these references were not given in other locations where yak dung is mentioned. The words 'yak dung' does not appear in Huang et al. (2016). Also, the reference for Lin et al. (2019), Lines 730- 733, appears to be incorrectly formatted.

Line 402: Which species of Hg?

Line 407: Can the authors show that many wildfires existed during this period?

Line 410: This is an example of how the phrasing 'controlling the region' needs to be rewritten to describe the transport patterns and air mass circulation.

Line 412: The cluster average does not show this and traj0 is hardly visible. Interestingly, traj1 appears to have the highest concentrations of GEM and arrives from areas with high Hg emissions but is not mentioned in the text. This cluster occurs rather infrequently though. I agree the weakening of the ISM is likely the reason for the increasing pattern in GEM during the ISM3, but this should at least be mentioned.

Line 419: Again, I wouldn't refer to measurements made with the Tekran systems as 'detailed'. The exact chemical identify of GOM and PBM is unknown. Therefore, I would remove this word.

Lines 418-427: In the previous paragraphs in this section, the authors examine the source regions of GEM and transport patterns during different periods. This PSCF muddles this analysis and do not provide any additional or useful information. The PSCF was applied to GEM, please indicate which species of Hg is being referred to here. The smoothing applied

to these figures could be obscuring the analysis. The authors discuss depositional processes during transport affecting Hg concentrations, although this would apply to GOM and PBM and not so much GEM. In my opinion, I would omit the PSCF analysis, as it does not provide gainful information, is not described adequately in the methods section, and contradicts the previous analysis of GEM with trajectory cluster analysis. This is, however, only my opinion.

Lines 429-430: I am confused by the number of factors for each period. For example, from Table 2 there are only two factors that occur during the PISM (long-distance transport and local emissions). There is only one factor that is unique to a period (melt during ISM1) and only local emissions occur during all periods. Please clarify this in the text.

Table 2: The caption for Table 2 needs to be expanded. I can see that numbers in bold indicate a loading over 0.5, this needs to be stated in the caption. Why are certain species omitted from the PCA analysis for certain periods? This was not clear from the methods section. Why is there two ISM1 for local emissions? Please define VE. Would it be possible to remove the underscores from the column headers?

Line 452: A reference is required for this statement.

Lines 453-462: While meteorology is no doubt affecting the behavior of atmospheric mercury, I am confused about how this factor affects mercury at Nyingchi. A different Hg species are excluded from the PCA for ISM1-3 and the only significant variable is GEM during ISM2. It is not clear from the text how meteorology is affecting GEM during this period.

Lines 464-467: Please indicate which period the authors are referring to here as well as the panel in Fig. 3. These two sentences largely say the same thing and cite the same studies, one could reasonably combine them for brevity.

Lines 469-470: Which 'previous simulations'? Please provide a reference. Are the authors referring to Song et al. (2018)? If so, please cite them or combine this sentence with the previous one. Also, the wording 'previous simulations....during the ISM1 period' implies that simulations were performed for GOM during this campaign. Please rectify this.

Line 477: Please see my previous comment about the phrasing 'generally believed'.

Line 480: 'masses' instead of 'mass'.

Line 497: Can the authors provide direction or recommendations for further studies?

Line 502: Similar comment as the previous one.

Lines 503-511: In combination with the previous study from Qomolangma, this study provides important insights into the transport, dynamics, and processes affecting Hg species during the PISM and ISM. I feel that since these two studies are the first in this geographical area, there should be more of a discussion between the differences and similarities between these two sites. The authors mention differences but only briefly.

Conclusions: The Conclusions section is very similar to the Abstract. Please see my Specific Comments from the Abstract section for suggestions and General Comments for topics that should be highlighted or discussed in greater detail, which should be represented in a revised Conclusions sections.