

Interactive comment on “First Measurement of Atmospheric Mercury Species in Qomolangma Nature Preserve, Tibetan Plateau, and Evidence of Transboundary Pollutant Invasion” by Huiming Lin et al.

Anonymous Referee #1

Received and published: 8 October 2018

This manuscript by Huiming Lin et al. presents the first record of atmospheric mercury species (GEM, GOM, PBM) during the Indian monsoon transition period in the Qomolangma Nature Preserve, located at the southern edge of the Tibetan Plateau along the border with the Indian subcontinent. Higher GEM concentrations during the monsoon period are attributed to air masses originating from east Nepal and Bangladesh, i.e. transboundary transport of Hg. Given the projected increase in Hg emissions in South and South-East Asia, monitoring data from downwind remote sites are essential. I think that this manuscript could make a valuable addition to the literature. However,

C1

and in agreement with reviewer #2, I strongly suggest an update of the references list (imprecise citations throughout the manuscript) along with other edits (see below).

Lines 38-40 (and throughout the manuscript): Could you please add standard deviations every time you refer to a mean concentration? Additionally, did you perform a statistical test to demonstrate that there is indeed a significant difference between ISM and non-ISM concentrations?

Lines 42-44: I don't think that GOM concentrations of ~ 20 pg/m³ are “considerably” higher than values in other clean or polluted regions. Concentrations of 1-20 pg/m³ are often reported at background/remote sites (e.g., Sprovieri et al. 2016) while hundreds of pg/m³ have been reported at urban/polluted sites (e.g., Duan et al. 2017; Han et al. 2018; Guo et al. 2017; Das et al. 2016).

Lines 49-52: To me, GEM concentrations reported in this study are at the lower end of concentrations reported in the Northern Hemisphere (Sprovieri et al. 2016). However, I do agree that international cooperation to limit Hg emissions is of utmost importance.

Line 61: I think reference to a review paper on Hg chemistry and atmospheric cycle is more appropriate here (e.g., Selin 2009).

Line 63: Recent modeling studies suggest a shorter lifetime in the atmosphere: 0.3-1 year (Selin 2009; Horowitz et al. 2017).

Line 64: Again, reference to Fang et al., 2009 is not appropriate here. Cite the original paper or a review paper.

Lines 73-74: Add Sprovieri et al. (2016) here.

Line 110: You could also briefly discuss future projections here (e.g., Pacyna et al. 2016).

Lines 120-122: I agree that this is the first study in the QNNP, but not the first one on the impact of the monsoon on Hg concentrations in Asia (e.g., Sheu et al. 2010; Yin

C2

et al. 2018; Wang et al. 2018; Zhang et al. 2014, 2016). This should be more clearly stated.

Section 2.2: What is the time resolution of GEM measurements (e.g., 5 or 15 minutes)? If 5 minutes, concentrations are most likely biased low and should be adjusted upwards (Slemr et al. 2016; Ambrose 2017).

Line 202: Why did you use an arrival height of 1500 m a.g.l.? According to lines 159-161, the height of the boundary layer is ~ 2000 m during the day and ~ 350 m at night. This means that your back trajectories are well within the convective boundary layer during the day, but above the nocturnal boundary layer. Surface measurements at night are likely decoupled from what is happening in the residual layer and have a fairly restricted footprint. I am worried that these night-time trajectories may not be a good indication of source regions, especially given the complexity of the site. It is of common practice to use a height of 0.5 PBL.

Lines 254-258: I agree with the overall PBM decrease but you should perhaps add a sentence here saying that higher PBM concentrations during ISM2 will be addressed later in the manuscript (Section 3.3.2).

Line 261: Add Sprovieri et al. (2016) here.

Lines 275-277: See previous comment; GOM concentrations are “at the upper end of” (and not “much higher than”) values in clean regions and are not higher than concentrations reported in polluted regions (e.g., Duan et al. 2017; Han et al. 2018; Guo et al. 2017; Das et al. 2016).

Lines 307-311: Please add standard deviations. I would like to see something like the 95 % confidence interval for the mean on Figure 3.

Line 343: Add here what’s written lines 159-161 (“the height of the atmospheric boundary layer changes significantly in one day from ~ 350 m above ground level during the night to ~ 2000 m during the day”).

C3

Lines 344-363: I am not really convinced by the arguments here. Do you expect higher GEM concentrations in the afternoon to be due to local emissions? Have you checked whether you have such an increase every day, i.e., no wind direction influence? Or more or less emissions under more or less radiation? You seem to have all the data needed to perform a more thorough analysis. Could it be due to the boundary layer height? Is the boundary layer lower during the monsoon period? Is there any correlation with radiation or temperature? You could perhaps investigate the correlation between delta-GEM and delta-temperature or something like that.

Lines 378-388: In Figure 4, could you please use something else than shades of green. It is hard to tell the difference between <1.5 and >1.5 ng/m³.

Lines 410-412: How can you explain that GEM concentrations in air masses originating from the Tibetan Plateau were the highest?

Lines 415-417: “The clusters were similar to most of the clusters during the PISM period; however, the GEM concentrations in these clusters were higher than those during the PISM period”. Could you explain why?

Lines 452-454: What about Bangladesh? Additionally, you don’t really explain why GEM concentrations increase during the ISM period.

Line 464: Could you please add the dates for ISM2 here and/or add ISM2 in Figure S3?

Lines 464-466: Large amounts of PBM “may have been released”. In this section and throughout the manuscript, please use the conditional tense to express conjectures/hypotheses.

Line 471: The discussion is about PBM here, not GOM. Remove reference to GOM.

Line 478: Can you explain this high value? Where did the air masses come from?

Lines 484: As mentioned above, 1.3 ng/m³ is at the low end of GEM concentrations

C4

reported in the Northern Hemisphere. I agree that there is indeed an influence from South Asia, but concentrations on the QNNP are still fairly low during the PISM. I feel like you should slightly nuance your position.

Lines 487-495: Could you possibly add a comparison between PISM and ISM periods in Figure 7? This comparison is the core of your manuscript.

Line 503: “significant” rather than “considerable”.

Line 507: Not true everywhere (e.g., Martin et al. 2017).

Line 516: Do you know if India, Nepal and Bangladesh have ratified the Minamata Convention on Hg? Check here: <http://mercuryconvention.org/Countries/Parties/tabid/3428/language/en-US/Default.aspx>. Hg emissions are projected to increase in India (Pacyna et al. 2016), what about Nepal and Bangladesh? You can perhaps strengthen the discussion here.

Lines 526-528: Is there a significant difference?

Lines 544-546: Again, concentrations reported here during PISM are at the low end of concentrations reported in the Northern Hemisphere. Additionally, concentrations are similar to those recently reported at Nam Co station on the Tibetan Plateau (Yin et al. 2018).

Figure 1: I assume that the red star within the QNNP is the location of the monitoring station. What about the two other red stars (Lhasa and Xigaze)? Do they represent cities and potential emissions? You should perhaps use a different type of star (monitoring site vs. cities) and make it clear in the caption.

Figure 2: Could you please add on this Figure the different periods (ISM1-5) you're referring to in Table 1?

Figure 3: I can't read the yellow axis, it is too bright. Please use another color. Ad-

C5

ditionally, what do you mean by GOM or PBM? Is this GOM, PBM, or the sum of the two? It is hard to see the dots and the diurnal cycle for GOM/PBM.

Figure 4: Which one is GOM, which one is PBM? Add a), b), c) on the Figure and caption.

Figure 6: Could you please explain in the caption what these values are? Probability of air passes passing through these regions?

Table 2: I think you can focus on Asian sites or refer to Figure 1 in Yin et al. (2018). The concentration reported for Nam Co station is incorrect (Yin et al. 2018).

Figure S4: Could you please add PISM, ISM1-5? Additionally, instead of April-August, is it possible to plot fires during PISM, ISM1-5? It would make it easier to identify whether fires are indeed more frequent in the area of interest during ISM2.

References:

Ambrose, J. L. 2017. “Improved Methods for Signal Processing in Measurements of Mercury by Tekran[®] 2537A and 2537B Instruments.” *Atmos. Meas. Tech.* 10 (12): 5063–73. <https://doi.org/10.5194/amt-10-5063-2017>.

Das, Reshmi, Xianfeng Wang, Bahareh Khezri, Richard D. Webster, Pradip Kumar Sikdar, and Subhajit Datta. 2016. “Mercury Isotopes of Atmospheric Particle Bound Mercury for Source Apportionment Study in Urban Kolkata, India.” *Elem Sci Anth* 4 (0): 000098. <https://doi.org/10.12952/journal.elementa.000098>.

Duan, Lian, Xiaohao Wang, Dongfang Wang, Yusen Duan, Na Cheng, and Guangli Xiu. 2017. “Atmospheric Mercury Speciation in Shanghai, China.” *Science of The Total Environment* 578 (February): 460–68. <https://doi.org/10.1016/j.scitotenv.2016.10.209>.

Guo, Junming, Shichang Kang, Jie Huang, Qianggong Zhang, Maheswar Rupakheti, Shiwei Sun, Lekhendra Tripathi, et al. 2017. “Characterizations of Atmospheric Particulate-Bound Mercury in the Kathmandu Valley of Nepal,

C6

South Asia.” *Science of The Total Environment* 579 (February): 1240–48. <https://doi.org/10.1016/j.scitotenv.2016.11.110>.

Han, Deming, Jiaqi Zhang, Zihao Hu, Yingge Ma, Yusen Duan, Yan Han, Xiaojia Chen, Yong Zhou, Jinping Cheng, and Wenhua Wang. 2018. “Particulate Mercury in Ambient Air in Shanghai, China: Size-Specific Distribution, Gas–Particle Partitioning, and Association with Carbonaceous Composition.” *Environmental Pollution* 238 (July): 543–53. <https://doi.org/10.1016/j.envpol.2018.03.088>.

Horowitz, H. M., D. J. Jacob, Y. Zhang, T. S. Dibble, F. Slemr, H. M. Amos, J. A. Schmidt, E. S. Corbitt, E. A. Marais, and E. M. Sunderland. 2017. “A New Mechanism for Atmospheric Mercury Redox Chemistry: Implications for the Global Mercury Budget.” *Atmos. Chem. Phys.* 17 (10): 6353–71. <https://doi.org/10.5194/acp-17-6353-2017>.

Martin, L. G., C. Labuschagne, E.-G. Brunke, A. Weigelt, R. Ebinghaus, and F. Slemr. 2017. “Trend of Atmospheric Mercury Concentrations at Cape Point for 1995–2004 and since 2007.” *Atmos. Chem. Phys.* 17 (3): 2393–99. <https://doi.org/10.5194/acp-17-2393-2017>.

Pacyna, J. M., O. Travnikov, F. De Simone, I. M. Hedgecock, K. Sundseth, E. G. Pacyna, F. Steenhuisen, N. Pirrone, J. Munthe, and K. Kindbom. 2016. “Current and Future Levels of Mercury Atmospheric Pollution on a Global Scale.” *Atmos. Chem. Phys.* 16 (19): 12495–511. <https://doi.org/10.5194/acp-16-12495-2016>.

Selin, Noelle E. 2009. “Global Biogeochemical Cycling of Mercury: A Review.” *Annual Review of Environment and Resources* 34 (1): 43–63. <https://doi.org/10.1146/annurev.enviro.051308.084314>.

Sheu, G.-R., N.-H. Lin, J.-L. Wang, C.-T. Lee, C.-F. O. Yang, and S.-H. Wang. 2010. “Temporal Distribution and Potential Sources of Atmospheric Mercury Measured at a High-Elevation Background Station in Taiwan.” *Atmospheric Environment* 44: 2393–2400.

C7

Slemr, F., A. Weigelt, R. Ebinghaus, H. H. Kock, J. Bödewadt, C. A. M. Brenninkmeijer, A. Rauthe-Schöch, et al. 2016. “Atmospheric Mercury Measurements Onboard the CARIBIC Passenger Aircraft.” *Atmos. Meas. Tech.* 9 (5): 2291–2302. <https://doi.org/10.5194/amt-9-2291-2016>.

Sprovieri, F., N. Pirrone, M. Bencardino, F. D’Amore, F. Carbone, S. Cinnirella, V. Mannarino, et al. 2016. “Atmospheric Mercury Concentrations Observed at Ground-Based Monitoring Sites Globally Distributed in the Framework of the GMOS Network.” *Atmos. Chem. Phys.* 16 (18): 11915–35. <https://doi.org/10.5194/acp-16-11915-2016>.

Wang, Xun, Che-Jen Lin, Xinbin Feng, Wei Yuan, Xuewu Fu, Hui Zhang, Qingru Wu, and Shuxiao Wang. 2018. “Assessment of Regional Mercury Deposition and Emission Outflow in Mainland China.” *Journal of Geophysical Research: Atmospheres* 0 (ja). <https://doi.org/10.1029/2018JD028350>.

Yin, Xiufeng, Shichang Kang, Benjamin de Foy, Yaoming Ma, Yindong Tong, Wei Zhang, Xuejun Wang, Guoshuai Zhang, and Qiangong Zhang. 2018. “Multi-Year Monitoring of Atmospheric Total Gaseous Mercury at a Remote High-Altitude Site (Nam Co, 4730 M a.s.l.) in the Inland Tibetan Plateau Region.” *Atmospheric Chemistry and Physics* 18 (14): 10557–74. <https://doi.org/10.5194/acp-18-10557-2018>.

Zhang, H., X. Fu, C.-J. Lin, L. Shang, Y. Zhang, X. Feng, and C. Lin. 2016. “Monsoon-Facilitated Characteristics and Transport of Atmospheric Mercury at a High-Altitude Background Site in Southwestern China.” *Atmos. Chem. Phys.* 16 (20): 13131–48. <https://doi.org/10.5194/acp-16-13131-2016>.

Zhang, H., X. W. Fu, C.-J. Lin, X. Wang, and X. B. Feng. 2014. “Observation and Analysis of Speciated Atmospheric Mercury in Shangri-La, Tibetan Plateau, China.” *Atmospheric Chemistry and Physics*.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, <https://doi.org/10.5194/acp-2018-806>,

2018.

C9