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Atmospheric Chemistry and Physics Discussions

Interactive comment on "Atmospheric mercury in the southern hemisphere tropics: seasonal and diurnal variations and influence of inter-hemispheric transport" *by* Dean Howard et al.

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We thank the anonymous reviewer for the positive comments and for the very helpful suggestions. These have made many points in the paper much clearer. Please find below the comments and responses, numbered by individual comment. Within each entry we provide a) the reviewer comment, b) our response, and c) changes made to the manuscript. (Note: the attached supplement shows this in a much clearer format).

1. a. The authors declare night time deposition events as a strong evidence for a "multi-hop" mercury transport model. However, they present no evidence for day time reemissions necessary for being able to talk about "hops". b. This is an important

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point and we thank the reviewer for highlighting this. We agree that re- emission of GEM following the nocturnal depletion was not covered in the text and have revised it accordingly. Emission of GEM in the morning precedes the break-up of the nocturnal boundary layer (as evidenced by diurnal-component radon), suggesting emission from the surface. As further evidence of emission, in the early morning GEM concentrations overshoot what could be considered the background GEM concentration, observed in the mid-afternoon when boundary layer mixing is at its greatest. It is reasonable to hypothesise that this early morning emission of GEM is partly made up of the relatively volatile fraction of newly-deposited mercury, based on experience in Arctic AMDEs. Further, observed net GEM fluxes during similar depletion events at another Australian site showed that these depletions were not significant long-term sinks of GEM. We have reworded this section to make this clearer. c. Page 10, Line 9: Reworded most of Section 3.2, through to Page 12, Line 16.

2. a. Isn't there a discrepancy between a "multi-hop" model and an atmospheric GEM lifetime of 6 – 12 months mentioned in the "Introduction"? b. We don't believe there is a discrepancy, as the prompt recycling process would not be resolved in the methods used to estimate GEM lifetime. We have reworded this section to define atmospheric lifetime as the mean time before which GEM is permanently removed from the atmosphere. As, under a "multi-hop" model, deposited GEM is rapidly re-emitted, this does not represent a permanent sink and would not affect calculation of mean atmospheric lifetime as determined from mass-balance approaches. c. Page 2, Line 23: Removed "The low atmospheric . . . inter-hemispheric transport processes." Page 2, Line 20: Inserted "The low atmospheric reactivity and low solubility of the elemental form (GEM) results in low wet/dry deposition rate and scavenging of GEM from the atmosphere. These attributes result in atmospheric transport being the dominant distribution mechanism through the environment, with long-range transport possible across hemispheric scales. Differences in background atmospheric mercury concentrations between the hemispheres are hence dependent on emission rates, deposition rates, inter- hemispheric transport processes, and atmospheric mercury lifetimes. The atmospheric life-

time is defined here as the mean time after emission that GEM is removed from the atmosphere (Lindberg et al., 2007) and is estimated from mass–balance approaches utilising hemispheric background concentration and source/sink data (e.g. Slemr et al., 1985). The atmospheric lifetime of GEM is currently estimated at 5–12 months (Holmes et al., 2006; Selin et al., 2007; Holmes et al., 2010; Horowitz et al., 2017)."

3. a. Or between a "multi-hop" model and chapter 3.2.1 on long-range transport? b. We agree with the reviewer here that the wording used seemed to attempt to replace the model of long-range transport with one of continual deposition and reemission. This was not intended, as differing processes can take place within the planetary boundary layer and free troposphere. As re-emission is difficult to differentiate from emission in GEM flux studies it is possible that evidence for this prompt recycling process has not been clear in the past. Our intention is to highlight, through the evidence of the nocturnal depletion events, that surface deposition/re-emission may be more important to atmospheric mercury cycling than previously believed. We have reworded multiple sections to make this clearer. c. Page 10, Line 23: Removed "Such a phenomenon . . . over long distances." Page 12, Line 12: Inserted "It is important to note that, due to inhibited mixing at the top of the nocturnal boundary layer, the extent of this depletion is limited to within tens to hundreds of metres above the surface. Beyond this, movement of free-tropospheric air continues to enable long-range transport of GEM. Nevertheless, were this phenomenon of rapid, bidirectional exchange with the surface to occur it would have a significant impact on our understanding of atmospheric mercury transport as it can impact the relative importance of intermediate and regional-scale sources, as well as expected time scales for observed decreases in environmental mercury following actions proposed under the Minamata Convention (Lindberg et al., 2007)." Page 1, Line 16: Removed "These cycles provide . . . over long distances." Page 1, Line 16: Inserted "These cycles provide strong further evidence supportive of a "multi-hop" model of GEM cycling, characterised by multiple surface depositions and re- emissions, in addition to long-range transport through the atmosphere." Page 14, Line 11: Removed "Such a phenomenon . . . over long distances." Page 15, Line 14: Inserted "Analy-

ses using diurnal-component radon suggest the rapid increases around sunrise are partly due to volatilisation of newly-deposited mercury, such as seen in other NAMDEs and Arctic AMDEs. The extent of this multi-hop phenomenon may be widespread, which would have a significant impact on our understanding of atmospheric mercury transport, the delivery of atmospheric mercury to the environment, and the legacy of anthropogenic emissions of mercury."

4. a. Section 2.2: Please state the standard conditions (pressure, temperature) at which the Hg concentrations are reported. b. This should have been included and was overlooked. c. Page 5, Line 5: Inserted "Reference volumes are reported at 1 atm and 0 âŮęC."

5. a. Section 3.1: Are the latitudinal differences statistically significant? b. The values themselves are, however additional uncertainty as a result of the instrumentation and differences in sampling periods suggests that this is not the case. c. Page 7, Line 10: Added "These differences are statistically significant (Student's t-test, $p < 0.0001$), though differences in the sampling periods introduces additional uncertainty due to seasonal variation at the sites. Further, an analysis of systematic instrument uncertainty for the Tekran 2537 by Slemr et al. (2015) showed this to be $\sim$10 %."

6. a. Page 2, line 17: "existence"? b. The wording here has been altered. c. Page 2, Line 17: Removed "resulting from its existence . . . environmental conditions."

7. a. Page 4, line 7: Reference "Köppen Aw" is missing in the list of references. b. "Köppen Aw" was intended as the category type reported by Peel et al., though this was not made clear. c. Page 4, line 7: Changed reference to "Köppen category Aw, as reported by Peel et al., 2007".

8. a. Andreae and Merlet (2001) is a review article citing largely work by others. They did not determine the mercury emission factor from biomass burning. Please reword or cite the original work. b. The use of "determined" was incorrect and unintended. c. Page 8, Line 5: Changed "determined by Andreae and Merlet (2001)" to "reported by

Andreae and Merlet (2001, and references within)”

9. a. Page 8, line 23: a net sink b. In response to this comment and a similar one from Reviewer 2, we have altered the text to better describe air–sea GEM exchange. c. Page 8, Line 23: Removed “Air-sea exchange of GEM . . . from soil and vegetation.” Page 9, Line 11: Inserted “Air-sea exchange of mercury is complex, with the ocean generally considered a net sink for atmospheric mercury (Mason and Sheu, 2002; Song et al., 2015). Reduction of mercury within the photolytic zone can give rise to increased concentrations of elemental mercury and hence evasion of GEM to the atmosphere (Soerensen et al., 2014). Terrestrial surfaces are also commonly sources of GEM; Nelson et al. (2012) modelled terrestrial mercury emission fluxes over Australia that were generally between 8 and 44 μg m-2 a-1 from soil and vegetation. Figure 3 does not show a strong difference in concentration distributions between the two source regions.”

10. a. Page 9, line 23: Year of the reference “Howard et al.”? b. This was an error; the correct reference should have been Howard and Edwards (2017). c. See 1c.

11. a. Page 11, caption of Fig.4: Median values are usually points – please reword. What is IQR? b. The shading shows nocturnal periods, hence the wording should have noted that the shading edges are sunset/sunrise times. IQR refers to inter-quartile range and was not defined. c. Changed caption to “Edges of shading denote median sunset/sunrise times for each season. Data have been split into stability categories based on diurnal-component radon quartiles at sunrise (marked in top panels). Lines are median values and error bars indicate inter-quartile ranges.”

11. a. Page 11, line 5: Mean values should be given with their standard deviations or errors and the number of measurements. b. These have been included. c. Page 12, Line 30: Included “Mean values for each were 1.08 $\pm$ 0.12 ng m-3 (n = 3048), 0.97 $\pm$ 0.13 ng m-3 (n = 81073) and 0.90 $\pm$ 0.10 ng m-3 (n = 46191), respectively.”

12. a. Figure 5: Vertical line at bottom for NH wet season merges with the blue line.

Please make it more distinct. b. This has been fixed. c. Altered Figure 5.

Please also note the supplement to this comment:
https://www.atmos-chem-phys-discuss.net/acp-2017-307/acp-2017-307-AC1-supplement.pdf

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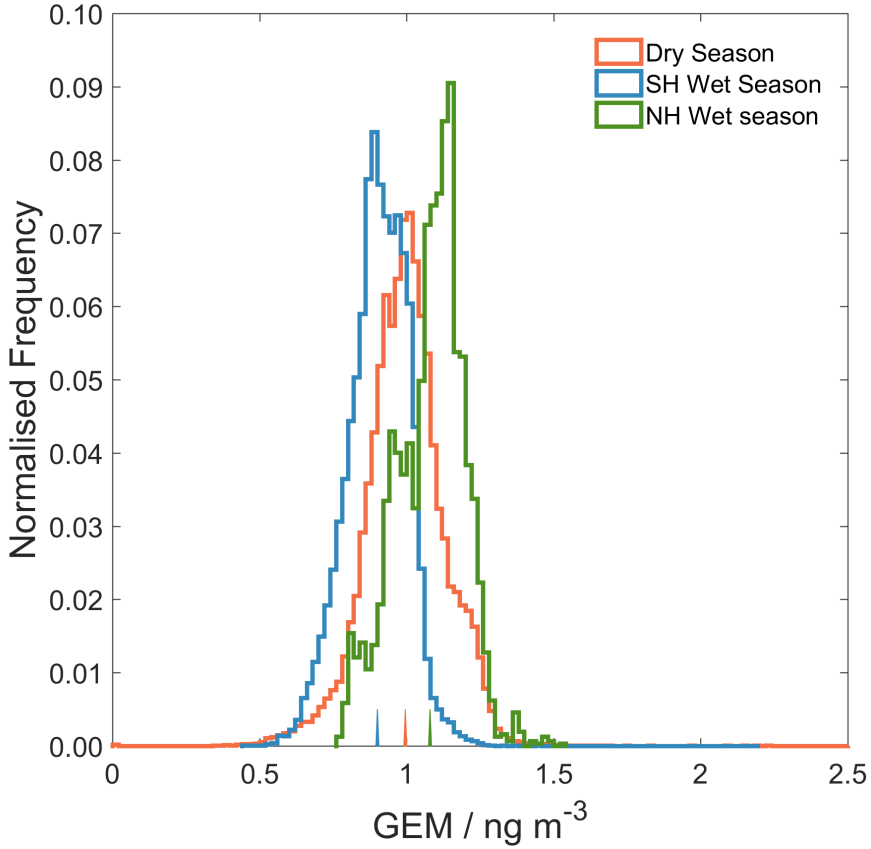

Fig. 1.

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