

Review of MS No.: acp-2016-517: “Five-year records of Total Mercury Deposition flux at GMOS sites in the Northern and Southern Hemispheres”, by F. Sprovieri et al.

(Review by Mark Cohen, Sept 27, 2016)

General Comments

The authors present a detailed description of 2011-2015 mercury deposition measurements made at selected sites in Europe, Russia, China, Mexico, and the Southern Hemisphere. Tropical and Southern Hemisphere data have been heretofore particularly scarce, and so it is welcome to see these data presented, alongside data from the Northern Hemisphere. It is a difficult task to explain spatiotemporal variations in mercury wet deposition observations, and the authors are to be commended for their thoughtful discussion of the various factors that may have contributed to site-to-site variability. While there are some areas for potential improvement that can be considered, as discussed below, this is an excellent paper and should certainly be published.

Specific Comments

- The title is misleading, suggesting that both dry and wet deposition is being reported. Perhaps the title could be reworded to be something like the following: “Five-year records of mercury wet deposition flux at GMOS sites in the Northern and Southern Hemispheres”
- Page 2, Lines 8-11. Could mention that dry deposition is often estimated via models using measurements of ambient concentrations of mercury and meteorological parameters.
- Page 2, Lines 29-30. What is the gradient in northern Europe?
- Page 3, Lines 7-8; and page 5, Lines 6-8. Why were these particular 17 sites chosen out of the 43 monitoring stations worldwide? Why were some sites excluded?
- Table 1. Several questions and suggestions to consider:
 - (a) “Elev.” – could give units (m-asl) in the table. The units are given later in the text, but for clarity, could be included in the table.
 - (b) The Sampling frequency could be included, e.g., 2-weeks for some sites, etc.
 - (c) What is the meaning of sites listed as “M/S” and “S/M”?
 - (d) If the site is a member of a national/regional network, this network could be listed.
 - (e) The years of data collection could be noted for each site (e.g., 2013-2014, etc.)
 - (f) In my opinion, would be very helpful to show the sites on a global map, perhaps with insets with close-ups as needed for clarity (e.g., for Europe)

- Table 2, and associated text. It is not clear to me what “ndays” data mean, and why it would be less than ~365 days per year. At some points in the manuscript, it seems that it might be being implied that if there was no precipitation during a given period (e.g., page 6, lines 20-21), then that period would not be reported as being a day of sampling for that year? But I don’t think that this is what you mean. My understanding of wet deposition samples is that the collector is in the field for a certain period (e.g., 2 weeks) and any precipitation that falls during this period is collected. So, in the usual case, if the site is operational, then the sampling generally occurs for the entire year, i.e., ~365 days. There may be some sampling periods where no precipitation is collected, but this is still a “sample” to be counted in the number of sampling days for that year. So, it would be helpful to clarify what is meant for each site, for each year, when the number of sampling days is less than 365. Was the site “closed” for the non-sampling days, i.e., the collector was not being operated? And if this is the case, and since there are seasonal patterns to precipitation and mercury wet deposition, it is not clear to me that normalizing the measured deposition by the number of sampling days is a reasonable approach. In other words, the periods when the sampler was “on” would not necessarily be representative of the “average”. I’m not sure if it’s really useful to present data for fragments of years, given the seasonality, and given that the dates of collection are not given. In my opinion, it might be best just to give the data for a site when an entire year of samples was collected (or at least *most* of the year).

- Page 5, Lines 14-15: Additional description could be given in the text regarding the “bulk-modified” sites, e.g., at least a few sentences describing the sampling protocol at these sites. E.g., what does “bulk-modified” mean? Also, should be noted that bulk-collection sites collect some dry deposition.

- Page 5, Lines 28-30. As noted above in comments on Table 1, it would be helpful to give the sampling frequency of each site.

- Figure 1. Several comments/suggestions:

- (a) Figure is too small to read easily. One suggestion would be to switch the x/y axes, i.e., put the sites along the x-axis on the bottom, and the flux on the y-axis. And then, use the whole width of the page, so that the data can be more easily distinguished. Another suggestion might be to use symbols rather than bars.

- (b) In the text, you refer to European sites extensively, and it would be helpful if these were grouped in the Figure. I know that you ordered the sites by latitude. But, in my opinion, you refer so many times to the “European sites” and refer to trends, etc., that it is really inconvenient to have to filter out the Chinese sites, etc.

- (c) Could consider showing a scatter plot of deposition flux vs. latitude instead, or in addition.

- Page 7, Lines 4-5. Seems that there are 11 sites in the Northern Hemisphere, rather than 10? And you discuss the European sites and Chinese sites extensively, but not the Russian site LIS.

Why is this? Also, here you say 7 European sites, but Figures 4-7 for European sites show only 6 sites. Could mention at some point why is the CMA site in Italy is not included in Figures 4-7.

- Page 7, Lines 7-9. The trend is not that clear in Figure 1, partly because the Russian and Chinese sites are interspersed in the Figure with the European sites. As noted above, I understand that you've listed the sites according to latitude, but ultimately, I think might be clearer if you group by region first, given that the discussion is predominantly carried out by region. Also, the LON site does not seem to fit the European trend noted.
- Page 7, Line 10. ... no north-south spatial trend has been observed.
- Page 8, Lines 9-12. Wet deposition of atmospheric Hg at any given location...
- Page 8, Lines 9-12. Wet deposition also depends on the type of precipitation (e.g., snow vs. rain), and the height and thickness of the precipitating cloud layer in the atmosphere, and the degree of convection involved. These are included at several points later in the document, but when I read this at this point, it seemed like important factors were being left out.
- Figure 3. As noted at several points in the manuscript, the relative proportion of snow vs. rain (or frozen vs. liquid) precipitation can be an important factor in interpreting the wet deposition data. Are there any site-specific data on this could be shown in Figure 3, or in a different figure?
- Page 9, Lines 1-9. The idea that more wet Hg deposition occurs with more precipitation is mentioned here and at several other points in the document. I think it might be really useful to show Figure(s) that show the deposition flux as a function of precipitation amount. This might be easier to interpret/explain than constantly going back and forth between the separate flux and the precipitation plots.
- Page 11, Lines 26-34. Here, you present arguments that suggest that the relatively high Hg wet deposition at CPT is due at least in part to contributions from local and regional sources. But then on page 26, lines 15-17, you cite a study that purportedly concluded that no significant local anthropogenic influences were found in Hg concentrations at CPT. How can these conflicting situations be reconciled?
- Figures 4-5-6-7. Are the box-whisker plots showing statistics for the sample-by-sample distributions for each season? If so, then it would definitely be important to know the sampling frequency. Might be useful to state what the boxes mean (25%, 50%, 75%?), and what the whiskers mean (5%, 95%?)
- Figure 5. Seems like could reduce y-axis to 0-60 ng/lit to show data more clearly.
- Figures 4-5-6-7: Again, maybe could add a figure that shows flux as a function of precip... This might be very illuminating.
- Figures 4-5-6-7: Why is CMA not included as a European station? I guess because not enough data?

- Figures 4-5-6-7: Would be useful somehow to show degree of solid vs. liquid precip, e.g., in Figure 4, if these data were available.
- Page 13, Line 5: Why were only 91 days sampled at the site? Was this because there was no precipitation, or was this because the site was simply not operated during that time?
- Page 15, Lines 3-11. Here, and in some other places in the document, it seems that you are just restating the information that can be clearly seen in the Figures. The manuscript is pretty long, and perhaps some efficiency could be obtained by omitting at least some of this type of reiteration?
- Page 15, Lines 16-17. As mentioned earlier in other contexts, it might be really helpful here to show a graph of flux vs. precipitation amount.
- Page 17, Line 6. What emissions are larger in the warmer months?
- Page 19, Lines 2-3. Perhaps too much to ask, but would it be possible to show maps of emissions in relation to the sites?
- Page 21, Lines 1-2. Sorry to be repetitive, but again, could maybe show a graph of flux vs. precipitation amount.
- Page 22, Lines 10-15. Not sure what you mean by “washout”. Are you referring to below-cloud scavenging of PBM by falling precipitation? Perhaps you could explain a bit more about the phenomena that you are describing here.
- Page 26, Lines 1-3. Here, and at a few other points, you note patterns in relation to GOM or other measurements. Would a graphic be useful here to show the relationship, e.g., GOM vs. volume-weighted-mean concentration in precip?
- Page 27, Lines 30-31. You state that early models tended to overestimate the influence of local emissions sources. This may or may not have been true, for one or more models, but I feel you’d need to cite a lot of different papers really make this statement. To me, seems like an overly provocative statement, and one that is not really needed for the paper? The general idea that observations are critical for model evaluation is certainly valid, but I don’t think you can (or need to) make this sweeping statement about “early models”. Indeed, Sunderland et al (2016) have recently pointed out that “early models” may have significantly underestimated the influence of local emissions sources!

Sunderland, E. M., C. T. Driscoll, J. K. Hammitt, P. Grandjean, J. S. Evans, J. D. Blum, C. Y. Chen, D. C. Evers, D. A. Jaffe, R. P. Mason, S. Goho and W. Jacobs (2016). Benefits of Regulating Hazardous Air Pollutants from Coal and Oil Fired Utilities in the United States. *Environmental Science & Technology* 50(5): 2117-2120.

- Page 27, Line 31. Is this really all available GMOS wet dep data, or just the data from selected sites for selected years? Also, are the GMOS wet dep data (and other data?) available? Perhaps this could be mentioned?

- Page 28, Line 1. Having data a “remote” sites with few local or regional sources is important, for sure, but having data a sites that are influenced by local and regional sources are also important for better understanding of Hg atmospheric fate and transport (and model evaluation), etc.

Technical Corrections

- Page 3, Line 1: ... long-term measurements of ambient Hg concentrations and measurements of Hg wet deposition fluxes were lacking...
- Table 2. There is a vertical line in the top of the table (see clip below, with red circle), that I think should be removed.

| | | 2011 | | | | | 2012 | | | | |
|---------------------|-----|--|------------------|--------------|---------------------------------|---|--|------------------|--------------|---------------------------------|---|
| | | Annual Wet Dep. Flux [$\mu g m^{-2} yr^{-1}$] | Rainfall [mm] | ndays [d] | Weighted HgT [$ng L^{-1}$] | Aver. Wet Dep. Flux [$ng m^{-2} d^{-1}$] | Annual Wet Dep. Flux [$\mu g m^{-2} yr^{-1}$] | Rainfall [mm] | ndays [d] | Weighted HgT [$ng L^{-1}$] | Aver. Wet Dep. Flux [$ng m^{-2} d^{-1}$] |
| Northern Hemisphere | NYA | - | - | - | - | - | 0.9 | 238.6 | 350 | 3.8 | 2.6 |
| | PAL | 2.9 | 407.4 | 363 | 7.1 | 8.0 | 1.9 | 278.6 | 332 | 6.8 | 5.7 |
| | RAO | 5.8 | 646.6 | 364 | 8.9 | 15.8 | 6.5 | 621.8 | 366 | 10.4 | 17.8 |
| | MHE | - | - | - | - | - | 0.9 | 393.7 | 113 | 2.2 | 7.6 |
| | LIS | - | - | - | - | - | 0.2 | 17.4 | 18 | 9.7 | 9.4 |
| | CMA | - | - | - | - | - | - | - | - | - | - |
| | ISK | 5.1 | 680.2 | 224 | 7.5 | 22.7 | 8.4 | 1349.7 | 363 | 6.2 | 23.2 |
| | MCH | 2.8 | 264.6 | 119 | 10.6 | 23.6 | 4.8 | 569.4 | 228 | 8.4 | 21.1 |
| | LON | - | - | - | - | - | 0.3 | 88.2 | 19 | 3.9 | 18.1 |
| | MWA | - | - | - | - | - | 0.3 | 79.5 | 127 | 4.3 | 2.7 |
| | MAL | 4.3 | 1543.2 | 222 | 2.8 | 19.5 | 3.2 | 971.5 | 202 | 3.3 | 16.1 |
| Tropics | SIS | - | - | - | - | - | - | - | - | - | - |
| | CST | - | - | - | - | - | 2.4 | 297.1 | 155 | 8.1 | 15.5 |
| Southern Hemisphere | AMS | - | - | - | - | - | - | - | - | - | - |
| | CPT | 0.3 | 133.5 | 119 | 2.1 | 2.4 | 3.8 | 260.3 | 147 | 14.6 | 25.8 |
| | CGR | - | - | - | - | - | - | - | - | - | - |
| | BAR | - | - | - | - | - | - | - | - | - | - |

- Table 3. I think “uom” refers to “units of measurement”, but maybe clearer just to put the units, or spell out “units of measurement. Better yet to include the units directly in the table.
- Page 6, Lines 31-32: ...the number of the sampling days as well as the annual wet deposition flux and average THg wet deposition flux calculated for each year in the period 2011-2015.
- Page 6, Line 32: As noted above, it is really unclear how valid any of the partial-year data are, given that it is unclear if the missing data are from rainy or dry seasons, etc.
- Page 8, Line 17. ... during the 2011-2015 period are is reported in Figure 3
- Page 11, Lines 19-24. Seem like sometimes you refer to sites using the 3-letter abbreviation, and sometimes you refer the sites using the full name of the site. Since the graphics all use the 3-letter abbreviation, maybe better to just use these in the text throughout. Could give the full name the first time it was mentioned, with the abbrev in parentheses, and then just use the abbreviation from then on?
- Page 13, Line 13. “meteorological” is misspelled.
- Page 20, Line 5. “rain” not “rainy”

● Page 22, Line 5. ... The positive or negative correlation between THg concentrations and the precipitation amount has not been obviously observed at MAL where the rainy samples shows a fairly seasonal variability, ~~during all seasons~~ with lowest average rainfall in winter and the highest in fall...

● Page 23, Line 18. ... exhibit a seasonality in annual rainfall, ...

● Page 24, Line 10. “fuels” not “flues”

● Page 24, Line 12. “United States”

● Page 24, Line 12. “waste incinerators”