

Review of Fain et al.

General Comments

Fain et al. present a study of gaseous elemental mercury (GEM) concentrations in snow interstitial air through the snowpack at a high elevation site in the Rocky Mountains. They observed both the diurnal photochemical production of GEM from Hg^{2+} in surface layers and the oxidation and loss of GEM in lower snowpack layers. This manuscript is well written and these data represent an important contribution to the understanding of Hg cycling within a mid-latitude snowpack. To my knowledge, this contribution represents the first in-situ examination of GEM in a mid-latitude snow pack. However, as discussed below, there are some aspects of the data that need to be explored in greater depth and some sections of the manuscript that need to be re-organized for clarity.

Specific Comments

- The authors need to more fully explore the implications of their study with respect to Hg loading in alpine snowpacks and the possibility that Hg in the snowpack represents an important source of Hg to ecosystems after snowmelt. Can the authors estimate the relative flux of GEM from the snowpack versus the amount of Hg wet deposited in snowfall? If this cannot be done quantitatively, the authors should at least qualitatively place the results of their study within this larger framework and discuss the implications of their data more fully in the Conclusions section.
- Although the authors state that they collected data both in 2009 and 2011/2012, only the data from the 2009 study are presented in the figures and discussed in the text. The authors should discuss and present their results from the 2011/2012 sampling campaign as well. By describing the sample collection but not the results, it leaves the reader wondering whether the results from 2011/2012 are contradictory to those from 2009. If there is not room within the manuscript to do this fully, these data could be included in a supplementary section. Alternatively, if it is more appropriate to do so, the authors could focus this manuscript only on the 2009 data and present the 2011/2012 data in a subsequent publication. In addition, the authors chose specific dates as examples (e.g., in Figures 1 and 6) of the phenomena they describe but it is not always clear why these dates were chosen. The authors should describe their methods for choosing to present these data (e.g., they are exemplary of the entire study or represent the most complete data sets, etc.).
- The authors should discuss whether changes in the height of the atmospheric sampling inlet above the snowpack impacted the measured GEM concentrations. It appears that the concentrations measured at this inlet were fairly stable throughout the study period. However, it seems possible that concentrations could have been elevated if the inlet was very close to the surface of the snowpack due to collection of GEM that was photochemically produced and emitted from the snowpack. Did the authors observe such a relationship?
- I would suggest that the authors reorganize section 3.1 so that it is clearer to the reader. It seems that it would make most sense to first describe the general trends in the data (as starts on pg. 15433, line 21) and the GEM concentrations throughout the year (Figures 2 and 3), and then describe the detailed diurnal cycling using the example in Figure 1.

- The authors discuss various oxidants that may cause dark destruction of GEM within the snowpack. This discussion would be aided by the measurements of O₃ and NO that were made throughout the snowpack. If these data do not fit in the manuscript, they could be presented in a supplemental section. In addition, the authors should clearly discuss the balance between diffusive transport of GEM down through the snowpack (resulting in the lag in max GEM concentrations), advective transport of GEM up through the snowpack due to high surface winds, and oxidation of GEM within the snowpack. What factors (oxidants? Meteorology? Snow physics?) control the relative importance of these processes in the different snow layers?
- The authors should further explore the correlations that they observe between GEM in the upper snow layers, solar radiation, and recent snowfall. Are these correlations statistically significant (Figure 5)? In addition, there are peaks in GEM on days 95 and 96 that are associated with very low amounts of solar radiation. The authors hypothesize that photochemical reduction produces the GEM in the upper snow layers, but these data may suggest that there are other significant reductants of GEM in the upper snow layers.
- The authors should more fully discuss the results of the snowpit THg sampling. Although the authors state that they sampled THg in the snowpits throughout the season (p. 15426, line 22), they only discuss THg concentrations in two pits sampled on March 9 and April 27 (section 3.4). It also does not seem correct that March 9, 2009 was the start of the snow season. Were other pits sampled during the season? If not, why do the authors believe that these snowpits were representative of the snowpack throughout the season? In addition, because Figure 7 was not included in the PDF version of this manuscript, it is difficult to assess the trends in THg concentration described by the authors.

Technical Corrections

Pg. 15425, line 17: Suggest that “Hg storage reservoir” should be “Hg storage reservoirs”.

Pg. 15426, line 10: Suggest that “Hg emission” should be “Hg emissions”.

Pg. 15427, line 16: Suggest that “interstitial snow air” should be “snow interstitial air”.

Pg. 15428, lines 10-11: I would suggest that the authors indicate that the sampling proceeded from X height inlet to X height inlet. This will make the sampling sequence clearer.

Pg. 15430, line 1: Suggest that “for total Hg” should not be stated twice in this sentence.

Pg. 15430, lines 7-10: The authors should discuss the results of measurement of bottle blanks for the snow THg samples. This is especially important because the authors did not clean the bottles using BrCl and because concentrations of THg in snow can be extremely low.

Pg. 15433, line 15: Suggest that “12:00am” should be “12:00pm”.

Pg. 15433, line 22: Suggest that “Sects. 2 and 3” should be “Sects 3.2 and 3.3”.

Pg. 15434: Figure 3 does not clearly show the “strong diurnal concentration fluctuations” in GEM as the authors state (line 13). Because each day is represented by such a small space on the figure, it is very difficult to observe the diurnal cycles on this annual plot.

Pg. 15436, line 10: Suggest that “Fig. 2b” should be “Fig. 2a”.

Pg. 15436, lines 14-17: This sentence is confusing. I would suggest that the authors rewrite it so that their meaning is clearer.

Pg. 15437, line 1: Suggest that “20-days” should be “20-day”.

Pg. 15438, line 12: Suggest that this sentence should read “... in the 0-30 cm depth layer in response to fresh snowfall.”

Pg. 15438, lines 13-17: This sentence is long and confusing. The authors should restate so that their meaning is clearer.

Pg. 15439, lines 21-26: I would suggest that the authors switch the order of these last two sentences to improve the logical flow of their arguments.

Pg. 15440, lines 22-23: This section may be confusing to readers. I would suggest that the authors add a clause to this statement indicating that the ratio should be constant if changes in GEM concentration are driven by snow physics and wind-induced advection.

Pg. 15440, line 25: Suggest this sentence is changed to read: "... thereby allow assessment of GEM chemical sinks ..."

Pg. 15441, line 6: It appears on Figure 6 that the GEM/CO₂ ratios in the lower snow layers are lower than the normalized value in the upper layer. However, the ratios (as plotted) are not negative.

Pg. 15441, lines 18-20: Does Figure 3 include data from NWT or from another site located further south? This statement is confusing because it suggests that the data presented in Figure 3 are from another site.

Pg. 15442, line 1: Suggest that "ozone is" should be "ozone are".

Pg. 15442, lines 18-19: The authors should add references to this statement.

Pg. 15443, line 1: Suggest that "4-days" should be "4-day".

Pg. 15443, line 23: Suggest that "understandings" should be "understanding".

Pg. 15445, line 2: Suggest that "dynamics" should be "dynamic".

Pg. 15445, line 10: Suggest that "... may indicate a potential GEM production.." should be "...may indicate potential GEM production..."

Figures:

Figure 4: It is difficult to tell the different colored lines apart on this figure and in the figure legend. I would suggest that the authors make the lines in the legend thicker.

Figure 5: I would suggest that the authors use the same temporal reference frame for both the y-axis labels and the figure legend (i.e., 1 day after versus 1 day before).

Figure 7: This figure was not included in the online PDF version of this manuscript.