

Interactive comment on “Reactive mercury flux measurements using cation exchange membranes” by Matthieu B. Miller et al.

Matthieu B. Miller et al.

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Response to Anonymous Referee #1 Received and published: 11 January 2019 This work addresses a significant research gap and provides a new, and seemingly improved methodology for measuring gaseous oxidized Hg fluxes from source material to air. This work has wide-ranging applicability in estimating Hg fluxes from contaminated materials, surface waters, Arctic permafrost, etc. Because it has such a wide applicability, it would be beneficial to researchers if the authors included in the discussion how their findings of both the method's abilities and limitations apply to other environmental settings. For instance, providing information on estimated sample time based on Hg substrate concentration, and detection limits in context of background Hg concentrations would be extremely useful.

Response: We thank this reviewer for the acknowledgement of the fact this is a new method that holds significant promise. We have added the information the reviewer requested at the end of the discussion section. We also thank this reviewer for their careful review and suggestions for improvement.

Overall the manuscript is well written, however it would greatly benefit from a streamlining of the discussion around methodology. Most of the information provided in Sections 2.4 and 2.5 is interesting but distracts from the scientific findings and would be better suited as supplementary material or moved to other parts of the discussion.

Response: We prefer to leave these two sections in the paper for they are important information regarding the method development.

Finally, it can be hard to keep track of all the abbreviations/acronyms used in this study, a legend of sorts would be helpful.

Response: We have eliminated some of the abbreviations that were not necessary. We also wrote out the names of the materials used. We included definition of the abbreviations in between the abstract and the introduction.

Specific comments: L54: add a couple sentences regarding the reactivity of GOM and how this relates to its residence time in the air compared to GEM, and how this relates to the need for accurate active sampling of GOM in a variety of environments

Response: Thank you very much for this comment. This information has been added and the paragraph rewritten so it flows better.

L84: it would be beneficial here to briefly discuss limitation to passive CEM sampling and what gap in understanding measuring fluxes could fill – make it clear to the reader how this method could be applied

Response: We prefer to not to discuss passive sampling here since this paper is focused on active sampling and I think this would be confusing to the readers.

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L99: include Hg content of source material

Response: This information is in Table 1. We made this clearer. LL114-L116: discussion of chamber footprint is confusing here, suggest moving this sentence to where you discuss need to place the flux chamber directly on the surface conditions (_L240)

Response: We prefer to leave this sentence where it is. It seems to fit better where it is.

L163: How did you determine how often filters would be changed? This is later mentioned in the discussion, however it is unclear whether sampling time was determined based on previous measurements or purely based on substrate Hg concentrations.

Response: Flow rates were described in the paper for each type of test and varied from 72 to 24 hours. For materials where fluxes were not above the detection limit given the sampling time it is clear a longer sampling time was needed.

L201: define 'high' ambient GOM concentrations

Response: We have removed this sentence. It did not make sense.

LL208-214: "All fluxes were measured in : : :." This section should be with the experimental set-up.

Response: We have moved this as the reviewer requested.

L260: what's accounting for the 21.4% variation in mean GOM flux measured from the TCL material? Could changing conditions in greenhouse explain any of this variability?

Response: The 21.4% variability between tubs of material is due to the fact that are slight variations in concentrations of the overall material being measured. The good replication for flux measurements on one material demonstrates that the individual tub measurements were consistent. We have added this to the manuscript.

L265: what's considered 'good agreement' - how does this compare to other methods

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(i.e. KCl denuders or other membranes)

Response: We removed good agreement for this component of the sentence was not necessary. r-friendly version Discussion paper LL281-285: A summer flux figure (similar to Figure 7b,c) would be helpful for comparing summer to winter values

Response: These are compared but not in as much detail in Figure 8. We prefer not to add an additional figure. We already have many. If the Editor wants us to do this, then we will.

L325: this would be a good place to discuss how CEM GOM measurements are affected by temperature and humidity, and what accompanying physiochemical and meteorological measurements should be taken with GOM and GEM measurements

Response: We thank the reviewer for this comment and we have added the following to the paper. Limited work in laboratory experiments with HgBr₂ demonstrated that RH did not affect measurement of GOM by the CEM (Miller et al., 2019). No tests have been done to directly address if there is a temperature effect. When making these measurements scientists should measure RH and temperature inside and outside the chamber. Other information that would be useful include soil moisture, precipitation, and solar radiation.

L331: define small difference

Response we have added ~10 pg and referred the reader to Figure 7a.

LL342-362: this seems out of place here, and should be moved to methodology section where defining GOM and discussing sample inlet and filter size (L163)

Response: We have changed the wording of the first sentence and prefer to keep this information here to reinforce our conclusion.

Technical corrections: L214: (see below) - Is this referring to a figure or text?

Response: To address this we changed this to read see Development of Method de-

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scription below.

L290: include reference to Table 1

Response: Thanks for finding this we added reference to Table 1.

L300: should this reference Figure 8?

Response: No the data are in Figure 7, but the regression is not shown. We refer to Figure 7 as the location of the data used to derive the regression equation.

L319: word choice, suggest using 'observation' instead of 'point'

Response: We made this change.

Figure 2: it is not clear where the filter packs are located in the schematic. Also remove double period in figure caption '..'

Response: We think it is clear where the filter packs are they have a very distinct shape. Got rid of the extra period.

Table 1: is substrate concentration total Hg conc.? If so, use the same units as reported in the text ($\mu\text{g g}^{-1}$) and label as Hg. If not Hg, then please include Hg conc.

Response: Yes, substrate concentration is total Hg concentration. We have changed the units in the text to reflect those in the table ng g^{-1} and inserted in the figure caption that Substrate conc. is total Hg concentration.

Interactive comment on Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-360, 2018.

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