**General comments**:

The authors present a first paper using the relaxed eddy accumulation (REA) method to measure flux over an Arctic snow covered surface, providing useful results for those looking to undertake similar Arctic mercury flux studies in the future. However, thorough editing of the text for grammar and clarity should be undertaken; in many areas, sentence fragments are present (see technical corrections for examples), and this should be corrected prior to publication, to increase the comprehensibility of the text. In addition, I would also caution that the authors appear to be extrapolating their conclusions from a relatively limited data set, and many of the conclusions drawn from their results do not appear to be adequately supported by the presented results. In their Fig. 7a, GEM fluxes appear to be predominantly near 0, which means that any derivation of relationships based on this data set must be approached with caution, and broad sweeping statements about the importance of various factors should be avoided (see specific comments below).

**Specific comments**:

1. *Use of the REA method*:

The manuscript presents a study using a technique (REA) which had not previously been used for this type of Arctic GEM flux work; however, the authors fail to explain why they have chosen this technique. In a “first use” study such as this it is important to explicitly outline the benefits of this technique over more traditionally used or other available methods, as well as the potential short-comings. In its present state, it is unclear why this work using the REA method is a benefit to the Arctic Hg flux literature, or why one may choose to do a REA based Hg flux study rather than use a chamber or AGM type method.

2. *Exclusion of data (primarily $CO_2$ flux to determine b section, and one Results reference)*:

It is not immediately clear how the limits were chosen for various parameters to allow for inclusion or exclusion of data (in “$CO_2$ flux to determine b” section). In total, the authors state that they excluded 74% of the collected data, which seems rather extreme, especially lacking adequate justification for the exclusion criteria. The authors state that they have estimated an uncertainty for b (<<10%), and state that the uncertainty is then assumed to be insignificant, but provide no methodology for that estimation, or justification for the assumption of insignificance. In this section, the authors also include a “z/L” criteria as a means by which data were included/excluded from the presented results, but this “z/L” is neither defined, nor described. Various parameters are given, however the choice to discard data should be more thoroughly explained and justified, especially when it means that very little of the originally collected data is included and used to derive the final relationships presented in this work. In the Results and Discussion (pg. 7, line 18 – 19), the authors propose a means by which some data may be falsely interpreted as outliers, but it is unclear whether this was a problem in the presented data set, and, if so, how was this dealt with?

3. *Uncertainty in presented data (pg. 6, lines 12 – 14)*:

The authors provide an estimate of the uncertainty “of the two concentration determinations”, but it is unclear what precisely they mean by “the two concentration determinations”; is this the uncertainty of Hg flux based on the instrumental detection limit of 0.1 $ng/m^3$? If so, this needs to be clarified (and units should be included with the appropriate numbers). In addition, it is stated that the uncertainty of the flux (which is given as 0.14) becomes 28% at the 95% confidence interval (unclear how this was derived), and then in the following sentence the authors give a GEM uncertainty of 10% above 0.5 $ng/m^3$, but this is attributed to another study, so it is unclear how this fits in with any of their data, or uncertainty in their data, and precisely what the uncertainty on any of the provided results would be. In addition, no factors besides flux and b are given any consideration with respect to uncertainty, which makes it impossible to know how robust any of the presented results/relationships are.

4. *Results and Discussion*:

I believe that this section of the manuscript should be thoroughly reviewed by the authors, and revised prior to publication, as many conclusions/relationships appear to have been drawn from GEM flux results that, as presented, typically seem to be approximately zero (Fig. 7a). In instances where the authors are calling on certain events (eg/increases in flux) these events should be explicitly stated, for clarity.

5. *The importance of heat/temperature changes*:

On pg. 2 (lines 30 – 31) the authors propose that a correlation between solar radiation induced heat flux/temp change and GEM production/flux exists, due to the observed relationship between solar radiation and GEM concentration increases in surface snow, and then again in the results/discussion (pg. 8, lines 9 – 12) they propose a temperature dependence in their observed results, and in the conclusions (pg. 9, lines 29 – 31) state that their data support the hypothesis that heating of the surface is influencing GEM formation/emission; however, this ignores the generally accepted idea that GEM production in snow (and many other media) is the result of photochemical reduction of $Hg^{2+}$ to produce $Hg^0$. It is possible that this process ($Hg^{2+}$ reduction or subsequent $Hg^0$ emission) from snow may be influenced by heat, but the primary reason for an increase in $Hg^0$ production with increased solar radiation intensity will most likely be due to increases in the extent of $Hg^{2+}$ photoreduction, not as a result of heat induced processes in the snowpack.

In addition, in the results/discussion section, the authors state “After deposition, we speculate that GOM is reduced photolytically to GEM.” (pg. 6, line 23 – 24), but then later state “GOM is reduced at the snow surface when temperature increases (Ferrari et al., 2008).” (pg. 6, line 26).

GOM in the snow will most likely be reduced by some manner of photochemical reduction reaction (whether this be photolytic, or as a result of some other photochemically driven process), and while temperature may influence the extent of this reaction, or movement of GEM from the snowpack, radiation (sunlight) is required for the reaction to proceed. As it is used in the aforementioned statement, the Ferrari et al. (2008) reference is somewhat misleading, and this should be revised. In addition, the authors are using air temperature rather than the snowpack surface temperature to derive relationships with flux, and these will differ. Since the surface snow temperature may be significantly different than the ambient air temperature, it is difficult to make compelling conclusions about temperature dependence on snowpack Hg flux without these snowpack temperatures.

In addition, the authors state that "The highest temperatures were found during events with the largest emissions…" (pg. 7, line 32 – 33), but these events have not been explicitly pointed out to the reader using the dates provided in the figures. From the data, I can see three easily observable GEM flux increase events (April 27, 28, 30); however, in one of these three easily observable emission events (April 28) increasing GEM fluxes occur before temperatures begin to increase, and, at least at the beginning of the GEM flux increase event, temperature is actually decreasing, and this is counter to what the authors have stated in their text. As a result, the text should be revised to explain this divergence from what they are stating to be typical behaviour (if more than the three observable events are being invoked).

The authors use an instance of an upward latent heat flux (presumably indicated as a positive number in Fig. 7e?) on April 27 which coincided with a GEM emission event as evidence to support their GEM flux temperature/water dependence hypothesis; however, this increase in LH flux appears to be quite small, and while it does coincide with an increase in GEM flux, other increases in LH flux which appear to be comparable in magnitude (eg/April 25 – 26) did not result in increased GEM fluxes. Further, the second observed increase in GEM flux (April 28) appears to have occurred with no significant change in LH flux, and the largest observed increase in GEM flux (April 30) occurred with a LH flux decrease (negative value) that was much more significant than any of the LH flux increases observed in the data set. At best, it would appear that the data presented by this work appears to neither support nor dispute the hypothesis of temperature dependence of Hg flux based on LH flux information, but it supplies no compelling evidence to support it. If the latent heat flux argument is to be included, other incidents which were counter to the authors' hypothesis must also be discussed in the text, and adequate reasoning provided as to why these do not give evidence to disprove the proposed hypothesis.

6. *The effects of wind speed on GEM flux*:

The authors state that all large emission events occurred when wind speeds increased (pg. 7 line 10), but it is unclear how wind speed effects and temperature effects are distinguished, or whether the proposed temperature effect on flux (see above) is simply the result of greater wind speeds appearing to coincide with increases in temperature (Fig. 6). With so many variables changing (potentially independently) at the same time, it is not possible to tease apart the relative importance of these on GEM flux by visual observation alone, which is how the proposed relationships appear to have been derived.

7. *Conclusions based on flux data trends*:

The authors attempt to determine relationships and draw conclusions regarding the effects of various factors on GEM flux; however, in looking at the data in Fig. 7a, it appears that in most instances, GEM flux is at or very near 0 $ng/m^3$; while this may be a function of the scale being used, there simply does not appear to be adequate data to support strong conclusions, with the data presented, especially with no inclusion of the uncertainty on these data. Further, if the authors plan to make conclusions regarding the dependence of flux on the various factors that they measured, it would be very helpful to have some manner of statistical test to back up these claims, as without them, the invoked trends are neither clear nor compelling. For example, the authors state that depletion events on April 23 – 25 and May 2 – 5 are followed by GEM emissions; however, while this may be supported by the April 27 increase in GEM flux that is visible in the results, there does not appear to be a significant increase in GEM flux following May 5, until the small (almost apparently negligible) increase in flux on May 7, when GEM concentrations are higher again. With the data as it has been presented, this does not appear to support GEM emission post-AMDE as the authors have proposed (pg. 6 line 21).

8. *Correlation between $CO_2$ and GEM*:

The authors state that there is a correlation between $CO_2$ and GEM, but offer no methods used to determine this. Was this decided based on mathematical/statistical analysis? Simple observation? Is this GEM concentration, or GEM flux? Based on a quick visual inspection of the results in Fig. 7a/b and 7c, there does not appear to be a compelling case for simple visual observation of such a trend. If this conclusion is to be included in the paper, there should be a more thorough investigation of the claim, or the methods used to draw this conclusion should be explicitly stated, at the very least.

9. *Comparison of study results with literature results (pg. 7 lines 19 – 24, pg. 8 line 15 – pg. 9 line 21)*:

Overall, the authors' discussion of their results as compared to other literature results is somewhat difficult to follow, and should be revised for clarity. This portion of the manuscript would benefit from the inclusion of more concrete results and explicit discussion (eg/ pg. 7 line 23 – 24: how can your results being in opposition to those from the Osterwalder study be explained by the difference in location? How is the GEM dynamic different in these studies? eg/pg. 8 line 21: what was your net emission, exactly? What were the values found by the other works that are referenced?), and from complete discussion of one topic before moving on to another to improve flow and comprehensibility (eg/ stability conditions are discussed in more than one place). Overall, as a discussion paper, comparison with other studies should be much clearer, allowing the reader to easily place the present study with those already existing in the literature (does it agree with other studies using similar method or not, and why?), and at present this is not the case. It is further unclear, in some instances, how certain discussions relate to the present study. For example, when discussing stable conditions/GEM build-up (pg. 9, lines 3 – 10), the authors state that strong stratification with a build-up of GEM near the surface will result in violation of a basic assumption for the flux gradient method; was this phenomenon expected in your study, and if so, how did you deal with it? If this violation of a basic assumption for the flux gradient method was not observed in your study, why have you included it here? The discussion section (and comparison to other studies) might also be easier to follow if a better introduction to the chosen technique was included (see specific comment #1).

10. *Results figures/tables*:

For the results figures (Fig. 6 and 7), the markers/scales chosen make it almost impossible to see differences in the data with time, except where those differences are very large. Since the authors are attempting to use such differences in various measurements over time to derive information regarding factors influencing GEM flux, it is imperative that the reader be able to see the differences the authors appear to be speaking of, and at present, this is not true in most cases. In addition, where uncertainty is known (eg/flux uncertainty = 28%, as stated in the manuscript) error bars should be provided for the data in these figures, as it is unclear whether changes (eg/in flux over time) might be statistically significant, or not. Also, certain events (eg/ AMDEs as in pg. 9 line 26 – 27: "…during which several AMDEs were observed.") should be explicitly marked on your data, or the dates you are proposing they have occurred should be present in the text and/or figure captions.

In the summary table (Table 1, pg 21) the authors give a flux range of 8 – 190 ng $m^{-2}$ $min^{-1}$ for their data set; however, in looking at the results in Fig. 7a, it is apparent that there were some incidents of negative (depositional) flux (April 27), and there are many instances where the flux appears to be zero. As a result, it appears that the flux range given in the table is either incorrect, or some values were excluded, and if they were excluded, a reason should be given for this, as the table is not particularly informative without it.

**<u>Technical corrections</u>**:

Pg. 1 line 27 – pg. 2 line 2: These two sentences appear to be contradictory, and it is unclear what the authors are attempting to inform the reader of re: atmospheric lifetime of GEM with the given text.

Pg. 1 line 29: What do you mean by "…the relaxation time of mercury in the atmosphere…"? Please clarify/revise.

Pg. 2 line 4 – 5: Sentence fragments; consider adding fragment "These atmospheric mercury depletion events…" to the previous sentence.

Pg. 2 line 8 – 9: "Thus, this is a human health…" sentence fragment.

Pg. 2 line 13: Consider revising "…concentration decreases during…" to "…concentration decreases due to…"

Pg. 2 line 18 – 19: "Vertical gradient…" is repeat of the information presented in previous sentence, consider revising.

Pg. 2 line 22 – 24: "This is likely due to…" sentence fragment, consider adding to the previous sentence.

Pg. 2 line 25: affect, not affects.

Pg. 2 line 30 – 32: "Thus it is likely that a correlation…" this statement is somewhat misleading, as increases in solar radiation lead to increases in GEM production/emission as a result of an increase in $Hg^{2+}$ reduction, which may have nothing to do with heat!

Pg. 5 line 19: The detection limit you have given is the literature value for the instrumental detection limit, which may be significantly different than your method detection limit. Consider revising to be the method detection limit.

Pg. 5 line 25: "EC" is used without definition in the text (provided in the abstract, but should also be included on first use in the main body of the manuscript).

Pg. 5 line 26 – 28: "The flux of $CO_2$…" Sentence is confusing, please revise.

Pg. 6 line 2: Was b derived based on T (presumably temperature?)? If so, this method has not been described.

Pg. 6 line 7: What is z/L? This is not defined.

Pg. 6 line 26 – 27: "This leads to increased…" Sentence fragment, please revise.

Pg. 6 line 31 – pg.7 line 2: "We observed a clear diurnal pattern…" Did you measure incident solar radiation intensity? If so, this data should be included, and if not, how have you arrived at this conclusion/the timing of max and min sunlight?

Pg. 7 line 15 – 18: "This could have occurred…" Sentence fragment, please revise.

Pg. 7 line 23 – 24: "These differences can be explained…" Sentence fragment, please revise.

Pg. 8 line 3: “At low temperature (< -20 C) the flux of GEM was near zero.” GEM flux was also near zero in many cases when the temperature was > -20 C; this should be discussed.

Pg. 8 line 24 – 25: “This could be due to higher wind speeds…” Sentence fragment, please revise.

Pg. 8 line 31 – 34: The two sentences about the Manca et al. (2013) study should be combined, as the second is mostly redundant.

Pg. 9 line 14 – 15: “Differences in locations between research sites…” Please state why this is important and/or how this is expected to influence your study with relation to others, as there is no context for this statement at present.

Pg. 9 line 15: “Important parameters are…” Sentence fragment, please revise.

Pg. 9 line 29 – 31: I don’t believe you have provided a compelling case to support the given hypothesis, as the manuscript stands.

Pg. 14 Fig. 1: It’s a little bit difficult to find the yellow dot in your figure, consider giving this dot a dark coloured outline to increase visibility where it overlies the white page.

Pg. 15 Fig. 2 caption: Should be “indicates”, not “indicate”.

Pg. 16 Fig. 3 caption: Should be “shows” not “show”.

Pg. 19/20 figure labels: authors are not consistent with the way they are writing units in the text vs. the figures (eg/ ng/m$^3$ in text vs. ng m$^{-3}$ in figures).

Pg. 20 Fig. caption: “mol” not “mole”.

Pg. 21 Table title: Should be “Summary table of…” rather than “Summary table over…”