Interactive comment on “Fluxes of gaseous elemental mercury (GEM) in the High Arctic during atmospheric mercury depletion events (AMDEs)” by Jesper Kamp et al.

Anonymous Referee #2

Received and published: 13 September 2017

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.

>>>>>>
>>>>>>

> We agree that a more detailed argument for choice of measurement method is needed. Therefore following text has been inserted into the manuscript at page 3 line 5:
> *Chamber methods are attractive methods for measuring fluxes because of their low cost and simplicity but suffers from a number of weaknesses. They only capture the flux over a small area, the chamber affects the surface over which the measurement is taken and they can modify physical properties such as light and temperature (Bowling et al., 1998, Fowler et al., 2001). This implies that the measured flux will differ from the natural flux. The AGM is not altering the surface; however, it requires a homogeneous surface several hundred meters upstream the measurement site. Furthermore, it is assumed that the vertical profile is only a product of the vertical turbulent transport; nevertheless fast chemical reactions can affect the profile. The most direct flux measurement technique is the eddy covariance (EC) technique (Buzorius et al., 1998) but close to the surface this technique only works for fast responding monitors (sampling frequency >5 Hz), which is not available for Hg. Therefore, we chose to employ the relaxed eddy accumulation (REA) method (Businger and Oncley, 1990) which is based on EC and the method does not affect the surface. Oncley et al. (1993) reported results with agreement within 20% for EC and*

REA and a study by Hensen et al. (1996) shows agreement between EC and REA within 10%, a difference that is reported not to be significant because the main error for REA is the determination of the concentration difference.

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?
>>>>>>>
>>>>>>>

We agree it is unclear how the various parameters to exclude data is chosen. A more detailed explanation for the choice of the limits of b and the uncertainty is needed, as well as an explanation of z/L. Furthermore we have made further studies of the uncertainty of b and have adjusted our estimates and the discussion of uncertainty. Therefor the section 2.5 has been changed to the following text:

We determine the proportionality factor b used to calculate fluxes of GEM from CO_2 fluxes assuming fluxes of all gases are transported by the turbulence in a similar way. CO_2 flux can oppose to GEM be measured using the more direct EC method, thus b can be estimated from the measured CO_2 flux and CO_2 concentrations using Eq. 1.
Close to the REA flux system, an enclosed CO_2 gas analyzer (LI-7200, LI-COR Inc.) was mounted on the boom with the inlet directly below the ultrasonic anemometer 6.08 m above ground and above the GEM sample inlets. The gas analyzer measures CO_2 and H_2O concentration at 10 Hz to derive the EC flux of CO_2 and H_2O. The CompactRIO compiles all data from the gas analyzer, valve positions and meteorological data from the REA system. The flux of CO_2 was measured in order to determine b from the CO_2 flux and back-calculations of CO_2 concentration in updrafts and downdrafts in each measuring interval (Gao, 1995;Ruppert et al., 2006). For each interval, b is used to determine the REA flux of GEM.
Meteorological conditions or parameters, such as temperature, wind direction and speed, heat fluxes, relative humidity, pressure, and water vapor were measured for further analysis of the GEM fluxes. The Monin-Obukhov length (L) was calculated in order to estimate stability, as atmospheric stratification is expected to affect the surface exchange. In order to ensure data from a well-developed turbulent flow field and a reasonably constant wind direction, wind speeds below two m s^{-1} were discarded.
For an ideal Gaussian joint probability distribution of the vertical wind speed and the scalar concentration, b has a well-defined value of 0.627 (Wyngaard and Moeng, 1992). However, experimentally determined b's for fluxes of heat, moisture and CO_2 typically

range from 0.5 to 0.7 (e.g. (Katul et al., 1996), Ammann and Meixner, 2002, Sakabe et al., 2014).
As mentioned a fixed "dead band" of 0.076 m/s is introduced. Adding a "dead band" will affect the magnitude of b. In many applications, a dynamic dead band scaled with standard deviation of the vertical velocity w (σ_w) is used, which gives a smaller but relatively constant b (Hansen et al.2013) according to Eq. 3:

$$b = b_0 exp \frac{-0.75 \cdot \omega_0}{\sigma_w} \qquad (3)$$

Where b_0 is b without the dead band and ω_0 is the dynamic dead band. However, for practical reasons (limitation on processing time for data control and data collection) we used a fixed dead band causing a b, which varies with σ_w. The standard deviation of w measured in present study varied between 0.03 and 0.4 m/s. According to eq. 3, this will cause a variation of b (~ 0.2-0.8) depending on the size of b_0. Several researchers have studied the dependence of b_0 on the atmospheric dimensionless stability parameter z/L (L is the Monin-Obukhov length and z is the measurement height, z/L < 0 indicates unstable, z/L >0 stable and z/L= 0 neutral conditions). The majority of the studies (Andreas et al, 1998; Ammann and Meixner 2002 and Salkabe et al., 2014) showed an increase in b_0 with increasing z/L, however for the most part they refer to a limited stability range (-1.5< z/L <1.5). In the high Arctic, we often find very stable as well as neutral and slightly unstable stratification. In order to keep the estimated b values within a well-investigated stability range, data are discarded if they fall out site the stability range (-1.5< z/L <1.5). If b in a given experiment differs too much from the expected value, the probability distribution is likely to differ from the Gaussian distribution, thus in the present experiment, data was discarded in periods where b derived from T or CO_2 was below 0.2 and above 0.8.
After data filtration, only 26% of the total 1653 measurements were approved during the campaign. We are aware that this is a very strict filtration; however, this ensures that the data used for the analysis are solid.
Several studies have been dedicated to investigate the implications on the flux related to b (e.g. Andreas et al., 1998; Ruppert et al., 2006; Sakabe et al., 2014) and the standard deviation of b is often estimated to be around 10% (e.g. Amman and Meixner, 2002; Sommar et al., 2013; Zhu et al., 2015). However, b is calculated based on measurements of CO_2 fluxes, thus the uncertainty of b must be related to the uncertainty of the measured flux. It is not trivial to estimate the uncertainty of EC fluxes. Finkelstein & Sims (2001) suggested to use direct calculation of the variance of the covariance for calculating the random sampling error in EC measurements. They tested measurements at several type of surfaces and found the relative error to be approximately 25-30% for trace gas fluxes. However, one could argue that this method is only revealing how constant the flux measurement is and not how accurate the measured flux is. A more correct way to estimate the error is to measure the flux in parallel towers (Post et al., 2015). This is very expensive and very rarely carried out. Hence, here we use the general relative standard deviation of CO_2 fluxes on 25-30% estimated by Finkelstein &Sims (2001). Using error propagation theory on eq.1 the uncertainty of b (u_b) can be estimated as the combined relative uncertainty of the measured flux (25%) and the relative uncertainty of the measured concentration of CO_2 (1%, (Li-Cor)) from following equation:

$$u_b(y) = \sqrt{\sum_{i=1}^{n} u(x_i)^2}$$

Where $u(x_i)$ is the standard uncertainty. The uncertainty of b is ≈ 25%. To estimate the total uncertainty of the GEM flux we also have to consider the uncertainty of the measurements of the GEM concentration. This was found to be 10% by Skov et al. 2004, which used same type of instrument for GEM measurements. The uncertainty of the GEM flux can now be determined from the combined uncertainty of the concentration measurements and uncertainty of the estimated b: $\sqrt{0.1^2 + 0.1^2 + 0.25^2} \approx 0.30$ *and the uncertainty of the flux becomes ≈ 60% at 95% confidence level.*

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.
>>>>>>
>>>>>>

The section of uncertainty has been revised (see text under section 2 above)

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.
>>>>>
>>>>>

We have changed figure 7, so it becomes more clear. We have added numbers to Fig. 7 to refer to the events and refers to the numbers in the discussion. Furthermore, we have added an extra figure showing the relation between the fluxes and temperature.
The whole section has been thoroughly revised and rewritten.

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.
>>>>
>>>>

We are aware of the reduction of GOM and possible subsequent emission of GEM and this was also our initial hypothesis. We investigated the relation between solar radiation and GEM emission but found no clear relation. However we found a possible correlation between temperature and GEM emission, which we think could be an important information to the scientific society measuring Hg fluxes in the Arctic, and we did not think it would be right to ignore this. We have inserted a figure showing the relation between GEM flux and radiation and the GEM flux and temperature, stating we are aware that this is the temperature in the atmosphere, since we unfortunately did not measure the snow temperature. We have changed the text in the discussion so it becomes more clear that the relation between GEM flux and temperature as well as radiation was investigated. Also we now refer more clear to other studies which also found a (reduction/emission???) temperature relation. Furthermore, we have changed the text on page 2 line 30-31 to:

This is most likely due to photoreduction of GOM and subsequent emission of GEM; however, it is also possible that a correlation between solar radiation-induced parameters such as heat flux or temperature change and GEM fluxes exists, making it relevant to look into temperature and heat flux as well as radiation in relation to GEM flux.

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).
>>>>
>>>>

We have added a figure (Fig. 9) showing the relation between GEM flux and atmospheric temperature. We found no clear correlation between GEM flux and radiation, but we are aware of the relation. It is true that the relation between GEM and temperature and latent and sensible heat flux is not straightforward. This is because other parameters are also affecting the size of the flux. This has been written more clearly in the text now.

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.
>>>>>
>>>>>

The event on April 30 is an extreme event caused by a strong change in the meteorological conditions (possible a front passing) and as we have pointed out in the text this should not be a part of the general analyses. We have tried to make it more clearly in the text.
It is true that many other parameters are influencing the flux and concentration of GEM. We have tried to make this more clear in the result and discussion.

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.
>>>>
>>>>

It is true many parameters are affecting the flux and we have tried to show that especially for GEM the temperature is special since we don't see the same relation between CO_2 and temperature, however the relation between CO_2 and wind speed is the same as for GEM and wind speed.

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).
>>>>>>
>>>>>>

We have changed part of the conclusion and made it more subtle:
The results of this study supports to some extent the general understanding of the AMDE mechanisms where GEM oxidation is followed by deposition of GOM, which is partly reduced to GEM and reemitted into the atmosphere. Furthermore, the data indicates that heating of the snow surface influences formation of GEM and reemission of GEM.

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.

>>>>
>>>>

The relation between CO2 and GEM was suggested by the editor. We have now added figure 10, which shows the co2 and GEM flux in relation to wind speed and it by visual observation we see an anti-correlation between the two fluxes. It is explained more careful in the text

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).
>>>>>
>>>>>

We agree that this could be more clear and the discussion has in general been cleaned so it is follows the recommendation of the reviewer.

The phrase opposite concerning Oswalds observations is changed to: "*On the other hand, Osterwalder et al. (2016) observed emission during unstable conditions, a small deposition during stable conditions and deposition during neutral conditions.*"

Regarding the strong stratification and assumptions for different flux measurement techniques, we expect the reader to be familiar with the basics of the different techniques, but see answer to comment 1 where justification for the method is added. To the introduction the following has been added: "*Furthermore, strong stratification violates the assumption of gradient measurements, thus REA is in our opinion the best possible option to measure GEM flux.*"

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.
>>>>>
>>>>>

We agree that this should be more clear. This has been revised accordingly (see previous comments and text).

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.
>>>>>
>>>>>

There is an error in table 1. The range should be -8.1 to 179.2 ng m-2 min-1. This is now corrected both in the main text and the table.

Technical corrections:
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.
>>>>> **This sentence is now removed since it confuses the reader instead of enlighten.**

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

>>>>> **This sentence is removed as part of the sentence above.**

Pg. 2 line 4 – 5: Sentence fragments; consider adding fragment “These atmospheric mercury depletion events…” to the previous sentence.
>>>>> **Changed as suggested, and made into one sentence.**

Pg. 2 line 8 – 9: “Thus, this is a human health…” sentence fragment.
>>>>> **Changed to one sentence.**

Pg. 2 line 13: Consider revising “…concentration decreases during…” to “…concentration decreases due to…”
>>>>> **Changed as suggested.**

Pg. 2 line 18 – 19: “Vertical gradient…” is repeat of the information presented in previous sentence, consider revising.
>>>>> **We agree, and have removed the sentence**

Pg. 2 line 22 – 24: “This is likely due to…” sentence fragment, consider adding to the previous sentence.
>>>>> **Changed as suggested.**

Pg. 2 line 25: affect, not affects.
>>>>> **Changed as suggested.**

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!
>>>>> **This statement is changed to:** ***“This is most likely due to photoreduction of GOM and subsequent emission of GEM; however, it is also possible that a correlation between solar radiation-induced parameters such as heat flux or temperature change and GEM fluxes exists, making it relevant to look into temperature and heat flux as well as radiation in relation to GEM flux.”***

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.
>>>>> **It is specified, and a section on errors has been added, see above.**

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).
>>>>> **Changed as suggested.**

Pg. 5 line 26 – 28: “The flux of CO_2…” Sentence is confusing, please revise.
>>>>> **The sentence has been rewritten.**

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

Pg. 6 line 7: What is z/L? This is not defined.
>>>>> **A sentence is added.**

Pg. 6 line 26 – 27: “This leads to increased…” Sentence fragment, please revise.
>>>>> **Rephrased as part of revision of section 3.**

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?
>>>>> **A reference to figure 9a is added, which shows no correlation between the flux and solar radiation. We think it is redundant to show a graph with the diurnal pattern, as there is no correlation.**

Pg. 7 line 15 – 18: “This could have occurred…” Sentence fragment, please revise.
>>>>> **Rephrased as part of revision of section 3.**

Pg. 7 line 23 – 24: “These differences can be explained…” Sentence fragment, please revise.
>>>>> **It is specified to differences in emission during different stabilities.**

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.
>>>>> **It is true that the GEM flux was near zero in many cases, so the sentence is rephrased as there are only fluxes close to zero < -20 C with a reference to fig 9b.**

Pg. 8 line 24 – 25: “This could be due to higher wind speeds…” Sentence fragment, please revise.
>>>>> **The two sentences are combined.**

Pg. 8 line 31 – 34: The two sentences about the Manca et al. (2013) study should be combined, as the second is mostly redundant.
>>>>> **Rephrased as part of revision of section 3.**

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.
>>>>> The sentence is coupled to the next and rephrased to specify why the differences are important.

Pg. 9 line 15: “Important parameters are…” Sentence fragment, please revise.
>>>>> **Changed with the previous comment.**

Pg. 9 line 29 – 31: I don’t believe you have provided a compelling case to support the given hypothesis, as the manuscript stands.
>>>>> **We hope the changes made in the manuscript are sufficient to support this sentence.**

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.
>>>>> **We think that the position is already pointed out in the caption and by visual contrast, so nothing has been changed here.**

Pg. 15 Fig. 2 caption: Should be “indicates”, not “indicate”.
>>>>> **Changed as suggested.**

Pg. 16 Fig. 3 caption: Should be "shows" not "show".
>>>>> **Changed as suggested.**

Pg. 19/20 figure labels: authors are not consistent with the way they are writing units in the text vs. the figures (eg/ ng/m3 in text vs. ng m-3 in figures).
>>>>> **All cases have been changed, expect z/L that is a normal term in meteorology.**

Pg. 20 Fig. caption: "mol" not "mole".
>>>>> **Changed as suggested.**

Pg. 21 Table title: Should be "Summary table of…" rather than "Summary table over…"
>>>>> **Changed as suggested.**