Response to comments on "Mercury vapor air-surface exchange measured by collocated micrometeorological and enclosure methods – Part I: Data comparability and method characteristics"*by* W. Zhu et al.

Anonymous Referee #1:

We thank the reviewer forthe thoughtful and constructive comments that improved the readability of our manuscript. We have incorporated the recommendations in the revised manuscript. Our point-to-point response to those comments and questions is given below (in blue). Corresponding revision was added in the manuscript.

Overall comments:

The solar radiation was measured at 3-m height; we all know solar radiation (here Iam referring to UV light, especially for UV-B) is a critical factor for Hg emission fromsoil, and penetration of solar radiation under flux chamber is not 100%. For thick poly-carbonate chamber, the UV penetration could be down to 30%. Do you think the solarradiation measured at 3-m height can represent the UV light intensity in the chamber?Different DFCs were made by different materials, quartz (I guess this should the thickone) and poly-carbonate film. I understand it might be complicated, is it possible for theauthors to include discussions related to this question, and to report the penetration ofUV-B under DFCs cover?

Response:We agree with the reviewer that outside solar irradiation is not represent of the chamber internal irradiation condition. This is a limitation of DFC method because an ideal chamber material that allows both light transmission and manufacturability does not exist.The UV-B transmission for quartz chamber (5 mm) ~90%, and ~30% for polycarbonate. The application of polycarbonate sheet for our NDFC fabrication is due to the NDFC is designed with strict physical shape and dimension, which does not permit the use of thin Teflon film and quartz (Lin et al., 2010).

For the point raised by the review of different material used here, we developed an algorithm based on total solar radiation to correct the flux bias due to light loss, which was presented in our companion paper Part 2. The flux bias has been corrected for the data published in this paper.

Lin, C.-J., Zhu, W., Li, X., Feng, X., Sommar, J., and Shang, L.: Novel dynamic flux chamber for measuring air–surface exchange of Hgo from soils, Environ. Sci. Technol., 46, 8910-8920, 2012.

Zhu, W., Sommar, J., Lin, C.-J., and Feng, X. B.: Air-surface exchange of Hg0 measured by collocated micrometeorological and enclosure methods - Part II: bias and uncertainty analysis, Atmos. Chem. Phys., Submitted for publication.

Specific comments:

Comment #1: The authors used many abbreviations in the manuscript, could theauthors add an overall table to make this clear?

Response:the abbreviations used in this paper was aimed to clarify the flux calculation from different methods. We have clearly explained the symbols below each equation in the revised manuscript, and those symbols were not frequently used in the discussion part.

Comment #2: Page 22275, line 4, "Mercury(Hg). . .. ." a reference is needed

Response: a reference of Lindqvist et al., 1991 has been added.

Comment #3: Page 22276, line 3, suggest to use other word instead of "realized"

Response:the “realized” was changed to “accomplished”.

Comment #4: Page 22277, line 4, correct “per se”

Response: the word “per se” was deleted, see the sentence “Measured fluxes are estimates of unknown quantities of air-surface exchange under field conditions and a reference technique for validating the estimates does not exist.”

Comment #5: Page 22278, line 20, friction velocity, does this mean the atmospheric boundary layeru* or the u* in the NDFC?

Response: the u* represents of “atmospheric boundary layer u*”, it has been clarified in the text.

Comment #6: Page 22279, line 3, “whole-air type” what does this mean?

Response: the “whole-air type” refer to a type of REA systems where a single inlet line is used to draw (whole) air at a high speed to the REA apparatus, where sub-streams are conditionally sampled.

Comment #7: Page 22282, line 3, “DOY” spell out, is this day of year?

Response: it is “day of year”. It has been added into the manuscript.

Comment #8: Line 2-11, can the authors make a clear table to include all the information?

Response:we have revised Table 1 with added information. We reorganized this part to make it more concise. See the text.

Comment #9: Line 21, why is the flow rate 0.75 Lpm? The 2537 cycle here is 5-mins, why not use2.5 mins to obtain higher resolution data than 5 mins?

Response:MM-derived flux require averaging times of 20 min and up depending on the site settings (topography, meteorological conditions etc.). At this site, we have identified 20 min as a suitable time (Sommar et al., 2013). As described in the same paper, coupling our REA system with 2537 only allows the analyser to be operated at 0.75 L/min due to back pressure. To get as robust samples as possible given the premises, 5-min is the sampling duration of choice for the REA and gradient-based system.

Sommar, J., Zhu, W., Shang, L., Feng, X., and Lin, C.-J.: A whole-air relaxed eddy accumulation measurement system for sampling vertical vapour exchange of elemental mercury, Tellus B, 65, 19940, 2013.

Comment #10: Page 22283, line 6-10, could the authors add some details for the operation of synchronized DFCs? If I understand this correctly, one 2537 was used to measure Hgconcentration in following processes: 1. inlet of TDFC for 5 mins (2.5-min cycle) 2.outlet of TDFC for 5 mins. 3. inlet of NDFC for 5 mins. 4. inlet of NDFC for 5 mins.

Response:Yes, it is. We have reorganized this part in the manuscript.

Comment #11: Line 26-27, did the authors measure Hg concentrations at same location to determinesystem blank for MM methods?

Response:We did very careful and thoroughwork on evaluating all the systems blanks. Not mentioned, the MM system blanks were evaluated by sampling zero air before and after the experiments (line 21-23). No significant contamination/carry-over bias was present in either MM-system. More important is the evaluation of MM systematic channel bias (including blanks) that is accounted for in Part I and II.

Comment #12: Page 22284, line 15-17, what parameters were used in this study?
Response:we used flux observed from different methods and environmental parameters.

Comment #13: Line 20, "^{o}C" for temperature?
Response:thank you for correcting, we changed ^{o} to ^{o}C.

Comment #14: Page 22285, line 26, I understand this might need additional work; however, conditional probability function (CPF) can better present the data than Hg concentration wind rose. This is just a suggestion.
Response:We agree with the reviewer that conditional probability function can better address the Hg concentration variability. For this study, we focus on flux method comparison to present the characteristic of each method. We thank the reviewer for the great suggestion, we will do conditional probability function analysis in the further work on atmospheric Hg distribution in the North China Plain.

Comment #15: Page 22286, line10-11 and 14-15, these two sentences are similar please rephrase.
Response: thank you for pointing out this. The sentence of line 10-11 has been removed and incorporated into line 14-15.

Comment #16: Line 16, the temporal variation of what?
Response:the temporal variation of Hg^0 flux, it has been added to the manuscript.

Comment #17: Line 16-17, please re-write, it is difficult to follow.
Response:to consolidate the explanation, Line 16-17 has been deleted and incorporated into pg 92, Line 14-16.

Comment #18: Line 24, what the IQR is? Please spell out.
Response: the IQR means interquartile range, it has been spelled out in the manuscript.

Comment #19: Page 22288 line 1, was, however, 3.5 times higher than that "measured" by TDFC. Here, I have some questions in series. What are the penetration of UV-B through thick quartz chamber and thin polycarbonate film? If the numbers are different, how did the authors compare the data measured by these two different chambers? Is there any way to correct this influence? Can this help to explain that NDFC measured higher flux than the number measured by TDFC?
Response:We agree with the reviewer that UV-B has a significant effect in stimulating Hg^0 emission.
Q1: The thickness of the quartz is ~5 mm, the UV-B transmission for such a chamber is ~90%. However, the UV-B transmission of the PC chamber is ~30% (Lin et al., 2010).
Q2: The choice of chamber material is crucial in DFC measurement. For chamber fabrication quartz, thin Teflon film, and polycarbonate sheet have been used in the previous studies. For the novel chamber in this study, the physical shape is critical in obtaining an optimal performance. Quartz and thin Teflon film were found not suitable for fabrication. The data presented in this study (Part-1) was corrected for flux bias, a calculation algorithm presented in our Part-2 paper, albeit the solar radiation correction was treated as total irradiation (not separated into UV-B and visible light) measured by a pyranometer, we think this is the most direct way to compare the chamber data. The measurement results presented in Fig. 6 demonstrated even if the NDFC UV-B penetration is lower than the TDFC, the measured fluxeswere very comparable.
Q3: We have developed an algorithm (multivariate regression model) to correct the flux forbias due to attenuation of radiation by the chamber material.

Q4: The data presented here has been corrected for the flux bias (Zhu et al., 2014), it clearly demonstrated that NDFC tendency observe a higher flux due to the internal controlled flow condition, "In the present study, TOT of TDFC is 50% lower than that of the NDFC. Moreover, the footprint of the traditional type is in square measure merely two thirds of the NDFC and the mass transfer, an elevation in fluxes derived by this type is expected", however, the direct measured flux is comparable.

Lin, C.-J., Zhu, W., Li, X., Feng, X., Sommar, J., and Shang, L.: Novel dynamic flux chamber for measuring air–surface exchange of Hgo from soils, Environ. Sci. Technol., 46, 8910-8920, 2012.

Zhu, W., Sommar, J., Lin, C.-J., and Feng, X. B.: Air-surface exchange of Hg0 measured by collocated micrometeorological and enclosure methods - part II: bias and uncertainty analysis, Atmos. Chem. Phys., Submitted for publication.

Comment #20: Line 10, how did the authors normalize the data?

Response: we normalized the Hg^0 gradient using Hg^0 concentration difference between the two sampling level divided bythe corresponding height difference. The unit of the presented normalized gradient is ng m^{-4}, i.e. ng m^{-3} m^{-1}.

Comment #21: Line 13, what does "marker's color" mean? I understand the authors present the datain another paper; however, could the authors briefly discuss the uncertainties in thispaper?

Response:Thank you for the suggestion. The "marker's color" means the turbulence data quality. We have described the data quality segregation in the manuscript and detailed those information in the Part 2 paper.

Comment #22: Line 21, is this real "observed flux" or estimated flux?

Response:the "observed" has been corrected with "estimated".

Comment #23: Page 22289, line 5-7, based on the figures, the Hg emission wasn't enhanced whenthe precipitation occurred? This is different from previous Hg emission measurements;most studies have reported Hg emission was enhanced when water was applied, anythoughts?

Response:The key component in all MM-flux system is the 3-D sonic anemometer. When its sending head or the receiving heads become wet, the sensors may not function properly (indicated by diagnostic warnings). It takes a while for the sensor to start to work again after a rainfall even if water droplets are manually wiped by the operator. There is therefore a gap in MM-data for this period. For the TDFC, during the shower, we did not move the chamber, so there is no tendency of increasing fluxes. After the rain stopped, we moved both NDFC and TDFC to the surrounding area. The shower lasts for ~10h, it was around midnight when the anemometer got dry and we relocated the DFCs,we did not observed large increase peak like those reported after immediate rain (Lindberg et al., 1997), this could be because of low temperature when relocate the chamber.

Lindberg, S. E., Zhang, H., Gustin, M., Vette, A., Marsik, F., Owens, J., Casimir, A., Ebinghaus, R., Edwards, G., Fitzgerald, C., Kemp, J., Kock, H. H., London, J., Majewski, M., Poissant, L., Pilote, M., Rasmussen, P., Schaedlich, F., Schneeberger, D., Sommar, J., Turner, R., Wallschlager, D., and Xiao, Z.: Increases in mercury emissions from desert soils in response to rainfall and irrigation, Journal of Geophysical Research-Atmospheres, 104, 21879-21888, 1999.

Comment #24: Line 18, what does the sampling mean here? Sampling method? Sampler?

Response:the "sampling" at here do mean "sampling method", we have corrected this in the revision paper.

Comment #25: Line 20-27, just a comment, sources and sinks of Hg from surfaces are related tosurface

types, surface conditions, Hg soil content, and the environmental conditions.
Response:We do agree with the reviewer. That's why a large area with homogeneous surface was chosen for the experiment.

Comment #26: Page 22290, line 23-26, could the authors please explain this in more detail?
Response:we have modified this part to make it clearer. It was revised following "However, when the sensible heat flux becomes small (small temperature gradient) approximately at $|H| < 20$ w m^{-2}, the correlation coefficient diminishes drastically and the fall-off in slope ($F_{AGM}/F_{MBR} = 0.35 - 0.36$) implying that MBR flux tendency to be significantly overestimated when the temperature gradient becomes very small. These MBR flux data during small scalar gradient time (often during dawn and dusk transition periods) are of questionable quality and should be considered for omission."

Comment #27: Page 22291, line 25, areal?
Response: the "areal" has been corrected with "area".

Comment #28: Page 22294, line 25-27, the reason of a good correlation for integrated flux over time is the way the integrated flux was calculated. The integrated flux at time t was calculated as the flux from t-1 to t adding to the integrated flux at t-1, therefore, both integrated fluxes (MBR and NDFC) are showing increasing trend. This might not be a good way to present the data, the better way to explain the data is to use longer time average (eg. daily, or every three hours)
Response:we than the reviewer for the suggestion of compare DFC and MBR flux.In the revised paper, we using deviation of cumulative flux between DFC and MBR flux, see revised figure 12 and corresponding discussion the revised paper.

Comment #29: Page 22295 line 1-2, I am wondering is this possible due to the UV-B influence. Look atHg^0 concentrations in detail, we can find this surface was functioned as a sink ratherthan source from 20:00-8:00. I am wondering is that possible the daytime emissionwas from night deposited? And At 10:00 am, the natural soil surface received enoughsolar radiation, and showed a peak at 11:00 am then Hg emission started to decreaseafter that due to lack of available Hg. However, because the penetration of UV-B under chambers was not high enough till 2:00 pm, it peaked at 2:00 pm. I know it iscomplicated, just some ideas.
Response:The DFCs measurement demonstrated that Hg^0 exchange between soil and atmosphere at nighttime 20:00-8:00 is bidirectional. Even the ambient Hg^0 concentration shown a clear diel pattern with decreasing from afternoon to night, we believe that this is primarily due to the regional source. We agree with the reviewer that nighttime deposition could contribute to daytime emission. We have gave a hypothesis that MM P1was emission from the night deposition via frost, dewfall and dry deposition. The processes could be complicated, we have incorporated the reviewer's suggestion into the discussion in section 3.4.2.

Comment #30: Line 15-18, the authors should read Choi and Holsen, 2009 Environ Pollution, page1673-1678.
Response:we have added the "Choi and Holsen, 2009".

Comment #31: Page 22296, there is a problem from their PCA results, in Table 3, some factors are onlyexplained by one variable. For example, factor 4 IC#2, this factor is only correlated toREA flux, this cannot help explain the data. People usually selected the factor numberonce the eigenvalue reaches to 0.9-1,

and there is no factor explained by a singlevariable. It depends on the situation, to reduce the number of factor might not influencethe meaning of factors; however, in some cases, in-properly using PCA might misleadthe results. I suggest to redo the PCA or move all PCA to SI not emphases in thissection.
Response: we thank the reviewer for the comment. We agree that the data point may restricted the results of PCA, we have remove the PCA analysis in the revised paper.

Comment #32: Table 3, how many data points were used to run PCA?
Response:the data point used for IC #1 and IC #2 is 1218 and 465, respectively. The Table 3 has been moved to supporting information, see Comments #31.

Comment #33: Figure 1, this figure is busy, and the resolution is low, could the authors provide a highresolution one. It is a very good figure, but cannot be read very well.
Response:we thank the reviewer for the suggestion, the figure 1 resolution has been changed into a good reading condition.

Anonymous Referee #2:
We deeply appreciate the reviewer'scarefully review of our manuscript.We have incorporated thethoughtful and constructive recommendations in the revised manuscript. Our point-to-point response to the comments is given below (in blue). Corresponding revision was added in the manuscript.

I. Major comments/suggestions:
1.The scientific contribution is of this manuscript is to some extend buried by thewriting style, including unnecessary materials in Section 2 (Material and methods), redundancy in the Results and Discussion section, some general statements or incorrectstatements, and overlap with Part 2 (Zhu, et al., 2014a). Some clarification and editorial issues are listed in sections II and III. My major concern is the structure of themanuscript. The detailed information of equations and repeated discussion in differentsections would be appreciated for a thesis or a report, while a concise style might bemore appropriate for a journal paper.
Response: We thank the reviewer for highlight the scientific merits of this study. For the manuscript structure and detailed revision, we have revised the manuscript following the reviewer's specific comments as discussed following.

Comment #I-1: All general descriptions as well as equations and related explanation in pages 78-81 could be omitted. Interested readers can find detailed methodology descriptions inpapers referenced within.
Response:Firstly, as the methodological theory is strongly linked to the results and discussion, we choose in a paper of this type to present the basic theory and equations. Moreover, in the Hg literature, there exist slightly different equations to calculate, e.g., AGM flux and in some instances a correction to mitigate for failure of obtaining energy balance closure can be found (see Sommar et al., 2013 for a discussion and references). Secondly, we present a bottom-up assessment of the flux bias and associated uncertainty in our companion paper Part 2.The analysis is fully based upon theseequationsand essential to provide the reader.
Sommar, J., Zhu, W., Lin, C. J., and Feng, X.: Field approaches to measure Hg exchange between natural surfaces and the atmosphere - a review, Critical Reviews in Environmental Science and Technology, 43, 1657-1739, 2013.

Comment #I-2: The manuscript could be shortened by removing sentences simply stating

numberspresented in tables, e.g. pg 86, L7-9; pg 88, L20-24; pg 89, L11-14.
Response:we thank the reviewer for the suggestion. All these sentences have been carefully revised or deleted in the revision manuscript.
The pg 86, L7-9 was revised as "The Hg^0 flux variation range and corresponding average flux measured by DFC techniques at soil surfacewere shown in Table 1. A broader variation range and mean flux was obtained from NDFC."
The pg 88, L20-24 was deleted.
The pg 89, L11-14 is statistic analyzed result of table 1, the sentence has been rewritten to a concise version.

Comment #I-3: The authors may want to consolidate some subsections to remove redundancyregarding to results and discussion, thus to improve the readability. Some examples are listed below:
Item Occurrences
Q1: Correlation between DFC flux and meteorological parameters Pg 86, L11; pg 95, L19
Q2: Correlation between AGM and MBR fluxes Pg 90, L22; pg 93, L26
Q3: Comparison between TDFC and NDFC fluxes: before correction similarity Pg 87, L14; pg 87, L26; pg 93, L24; after correction 3.5 times Pg 87, L18; pg 88, L1
Q4: Comparison of DFC and MM temporal variation Pg 86, L15-17; pg 92, L14-16
Response: we thank the reviewer for the suggestion.
Q1: Pg 86 L11 has been deleted and incorporated into Pg 95.
Q2: Pg 90, L22, we have rewritten this part to consolidate it as: "However, when the sensible heat flux becomes small (small temperature gradient) approximately at $|H| < 20$ w m^{-2}, the correlation coefficient diminishes drastically and the fall-off in slope ($F_{AGM}/F_{MBR} = 0.35-0.36$) implying that MBR flux tendency to be significantly overestimated when the temperature gradient becomes very small. These MBR flux data during small scalar gradient time (often during dawn and dusk transition periods) are of questionable quality and should be considered for omission."P93, L26. The repeated information in L26 has been deleted.
Q3: the pg 87 L12-19 has been removed and integrated with pg87 L26 and pg 88 L1.
Q4: the page 86, L15-17 has been deleted and combined into pg 92, L14-16.

Comment #I-4: The overlap between this manuscript and Part 2 seems to be beyond a few lead-ins.Those overlaps hinder the ability of each manuscript (this and Part 2) to be a stand-alone paper. My impression is that the readers need to read Part 2 to under-stand some discussions presented in this manuscript, while the differences within andbetween DFC and MM methods will be repeated in Part 2 to facilitate the investigationof the causes of discrepancy. For example, the methodology of uncertainty analysis was not presented in this manuscript, but the results were (Fig 4). Similarly, there areconclusions in Part 2 presented in this manuscript without relevant methodology anddiscussion, for instance, reasons of dissimilarity in the DFC fluxes (pg 87, L1-3), reasons of variability in REA and other MM methods (pg 90, L2-5), reasons of disparate AGM and MRB fluxes (pg 91, 1-20), reasons of flipped AGM and fluxes in the twocampaigns (pg 95, L3-13).
Response:we thank the reviewer for the suggestion and have revised the illustration accordingly. The two companion papersare aimed to illustrate flux characteristics (Part I) and flux uncertainty (Part II). In this Part-I paper, we have presented flux data characteristics and discuss the causes for various discrepancies if such could be identified. In Part II, we performed a rigorous analysis of results to provide precision requirements of the Hg analysis as well as uncertainty/bias of concentration differences and chamber/micrometeorological exchange parameters. This facilitates thorough Hg flux error estimates thathave not been reported in earlier literature. The authors deeply regret that the Part I & II manuscripts were not submitted at the same time

because of project scheduling. Part II has now been submitted.

The quantified uncertainty within MM and DFC that could result the discrepancy was presented in the part 2.

(1)We gave an overall flux uncertainty in the Fig.4 to show the flux quality, which is important to understand the observed discrepancy among the MM methods.

(2) pg 22287 L1-3 has been deleted.

(3) pg 22290 L2-5; (4) pg 22291 L1-20; and (5) pg 22295 L3-13 cited our quantitative flux uncertainty analysis in Part-II paper that supports the discrepancy between each method. The citations are not overlapped discussion in the two papers.

2. The so-called NDFC has advantages over the TDFC. However, presentation of theNDFC in this manuscript is a bit confusing partially due to some unfounded statements,e.g.

Comment #I-5: Pg 76, L17, "a novel designed DFC (NDFC) based on surface wind shear condition(friction velocity) rather than on artificial fixed flow to account for natural shear conditions." Pg 78, L2, "a novel DFC (NDFC) design capable of controlling the internal shearflow over measurement surface (Lin et al., 2012). The NDFC internal flow conditionwas precisely controlled to relate to the applied flushing flow rate to the atmosphericboundary shear condition (therefore wind condition)". It is not clear how to implement this technique when the flow rate was indeed fixed in the NDFC operation (pg 78, L7) and the monitoring of atmospheric boundary shear condition is not mentioned. Evenwith the highly variable friction velocity available, the "precisely controlled" "internal flowcondition" "to relate the applied flushing flow rate to the atmospheric boundary shearcondition" would need a closed-loop system which was not available in this paper andthe NFDC paper (Lin et at., 2012). Those statements also contradict equations 1 and2 which have a fixed flow rate.

Response: the operation of the TDFC and NDFC for flux measurement was similar executed at a fixed flushing flow rate at 15 L min^{-1}. The atmospheric boundary shear condition (parameter friction velocity) was measured using the collocated eddy correlation system (Fig. 1). As discussed in our previous paper (Lin et al., 2012), the NDFC internal flow was precisely controlled at a constant operation flow rate. The flux under atmospheric boundary shear condition can be estimated based on the overall mass transfer coefficient. The statement of "to relate the applied flushing flow rate to the atmospheric boundary shear condition" does not mean that the operation is under a varied flow rate. It is actually running with a fixed flow rate that stated in Eq 1 and Eg 2.

Comment #I-6: Pg 86, L16 "DFCs flux was derived from Hg^0 mass balance calculation every 20min, different from the MM flux that relied on atmospheric turbulence processes." Thissentence contradicts other statements that the modified DFC taking into account turbulence, e.g. pg 76, "based on surface wind shear condition (friction velocity) rather thanon artificial fixed flow to account for natural shear conditions", pg 87, "the well-developed turbulence (higher friction velocity, Fig. 2) during daytime caused the correctedHg^0 flux from NDFC flux to be approximately 3.5 times higher than the TDFC flux".

Response: the Pg 86 L16 part has been removed.

Comment #I-7: Pg 88, L2-6, "Given that DFC of conventional types cannot reproduce atmosphericturbulence.NDFC is more preferable for the determination of net Hg0 gas exchangeover soils." This sentence seems to be over-promoting the NDFC when in fact no DFCscan "reproduce atmospheric turbulence" regardless of corrections.

Response:the "reproduce atmospheric turbulence" has been corrected with "be re-scaled with natural surface shearstress".

3.pg 86, L20, “Probability plots of both DFC datasets showed positive kurtosis (3.0and 4.1) and skewness (1.6 and 2.1) (Fig. 5). As a consequence, the average flux isslightly positive”. The reasoning here seems questionable; kurtosis and/or skewnessthemselves are not related to the sign (positive or negative) of a population or samplemean. The authors may want to clarify the meaning of positive kurtosis and skewness,and rephrase the sentence.

Response:we thank the editor for the suggestion. The sentence has been rephrased as “Probability plots of both DFC datasets showed positive kurtosis (3.0 and 4.1) and skewness (1.6 and 2.1) (Fig. 5) as a consequence of stronger emission and friction velocity at daytime”.

4.Pg 87, L18, “the corrected Hg^0 flux from NDFC flux to be approximately 3.5 timeshigher than the TDFC flux”. This assessment seems unfounded. Fig 6 had a slopeof 2, i.e. one flux is twice as high as the other, or one time higher. Also, the slope of1.1 indicates the two DFCs had similar fluxes, thus the corrected NDFC fluxes be one time “higher than the TDFC flux” and the NDFC fluxes when flux >0, but lowerwhen flux<0. Furthermore, Figure 6 caption seems incorrect regarding to the markers.If the discussion refers to Table 1 (2.2 vs. 7.6, 2.5 times higher), please clarify.

Response:“the corrected Hg^0 flux from NDFC flux to be approximately 3.5 times higher than the TDFC flux” has been corrected as “the corrected Hg^0 flux from NDFC flux to be approximately 2.5 times higher than the TDFC flux”.

5. Pg 93, L23, correlation. Because of the substantial departure from normal distributions (Figures 5 &7; pg 98, L10), the use of Pearson correlation (in tables, figures and main body) should be justified. Alternatively, Spearman rank correlation and Kendall rank correlation could be employed.

Response:We recognized that the algorithms of Person correlation and other method of correlation assessment may produce different value. Typically Spearman rank correlation and Kendall rank correlation would produce a more representative correlation coefficient when the data distribution is highly skewed. In our datasets, even though the data is deviated from normal distribution, the data range covers a sufficiently broad range with highest probability density near the central value such that Pearson correlation is representative (Hauke and Kossowski, 2011). Given that Pearson correlation is the most applied correlation method well received by most technical reader, we chose to use Pearson method.

Hauke J., Kossowski T., Comparison of values of Pearson’s and Spearman’s correlation coefficient on the samesets of data. Quaestiones Geographicae 30(2), Bogucki Wydawnictwo Naukowe, Poznań 2011. DOI 10.2478/v10117-011-0021-1, ISBN 978-83-62662-62-3, ISSN 0137-477X.

6.Pg 94, L23, “Figure 11a and b shows scatterplots of hourly and cumulative fluxspecifically for MBR vs. NDFC, though the correlation between individual hourly datapoints is weak, the fluxes integrated over time show strong agreement.” Perhaps itshould read “Figure 12”. Furthermore, the readers might be interested to see if thesame could be said with the scatterplots of hourly and cumulative flux for MBR vs.TDFC. More importantly, the correlation of two cumulative fluxes may violate the independency requirement. Because the cumulative fluxes at time t+1 depend on fluxes attime t, the data points are not independent of each other. Consequently, the authorsmay want to remove the regression equation and r values and to include scatterplotsof hourly and cumulative flux for MBR vs. TDFC.

Response:The “Figure 11a and b” has been corrected with “Figure 12a and b”. For the figure, we aimed to illustrate that NDFC flux showed advantages in bridging the gap with MM fluxes. In the revised paper, we compared the difference between cumulated MBR and cumulated NDFC/TDFC flux, it clearly demonstrated the advantage of NDFC method.

7.Pg 96, L1-15. As presented, the use of PCA seems unnecessary and the interpretation of the PCA results seems questionable. The discussion was focused oncorrelation among variables which is presented in Table 2, instead of identifying majorfactors affecting the air-surface exchange processes. In addition, the authors seemto have reached contradicting conclusions, "The environmental variables also significantly modified the gradient-MM fluxes (factor loading > 0.3)", and "Two separate PCAwas resolved for gradient fluxes variance (factor 2). The two factors are not contributed from the environmental variables (factor loading < 0.1), suggesting that theMM fluxes and their temporal characteristics are likely influenced by turbulent transportprocesses . . ." Furthermore, in both IC1 and IC2, only the first two factors had morethan one loading > 0.4. In other words, factors 3-5 in IC1 and factors 3-4 in IC2 failed tobe valid factors when there is one loading > 0.4 hence that factor only represents onevariable. In cases like this, all 4 or 5 factors may become uncertain. This is likely dueto the limitation of the dataset. Consequently, I would suggest remove this paragraph.

Response:We thank the review for the insight and agree that the dataset for PCA analysis is limited. In the revised paper, this section has been removed for the succinctness of the paper.

8.Fig 3. Wind rose. The height of 3 m above ground for meteorological measurementscould be too low to represent regional movement of air mass. An alternative could bedatasets from a nearby airport or air flow directions from trajectory models with smallgrids.

Response:The meteorological data were aimed to elucidate the influence of environmentalfactors on the observed fluxes and therefore it is more representative to use in-situ observational data. The experiment site is located in a large flat area and theselected 3-m sampling height is representative.

9. Fig 3. The pollutant rose as presented offers little information about the distribution of directional concentrations. The authors may want to consider the use of percentiles (e.g. 25%, 50%, 75% and 95%, see Figure 4b in http://www.mdpi.com/2073-4433/4/4/472).

Response: We thank the reviewer for the comment and would like to clarify this. Fig. 3 was utilized to illustrate the general wind pattern and the distribution of ambient Hg concentration at the measurement site instead of showing directional distribution that illustrates the source direction.

II. Clarification issues

Comment #II-1: Pg 74, L13 & Pg 95, L14, the reviewer did not find any results or discussion about "sensitivity". Perhaps "correlation" is more appropriate.

Response:We have changed the "sensitivity" to "correlation" in both places.

Comment #II-2: Pg 75, L9-13, "Hg0 is subject to bi-directional exchange between atmosphere andnatural surfaces through complex and yet not well understood processes, re-emittingpreviously deposited Hg back to the atmosphere (Bash, 2010; Gustin and Jaffe, 2010).Recent estimation indicates that annual natural emission accounts for two-thirds ofglobal release of atmospheric Hg (Pirrone et al., 2010)." It is not clear whether "re-emitting previously deposited Hg" is part of the "natural emission". The authors maywant to tidy up those loosely defined terms.

Response:We thank the reviewer for the suggestion, the "re-emitting previously deposited Hg" was treated as bulk natural emission as we cannot discriminate it concurrently. We have rephrased the discussion.

Comment #II-3: Pg 75, L19, "representing the smallest scale (< $0.1 m^2$)". 1) in case you were notsure that areas covered any DFCs ever existed were < $0.1 m^2$, perhaps "in the orderof $0.1 m^2$" could be more conservative, 2) perhaps "representing the smallest scale asthe areas covered by the devices are typically in the order of $0.1 m^2$" could be moreappropriate.

Response:We thank the reviewer for the consideration and have revised the sentence as suggested.

Comment #II-4: Pg 77, last paragraph before section 2. "Real fluxes are per se unknown underfield conditions and it is impossible to validate flux measurements by any (reference)technique". In that case, the reviewer is curious on how to "quantify the bias of theexamined flux measurement methods using statistical analyses".

Response:Eventhough there is no standard or preferred field Hg^0 flux measurement technique, deeper understanding of the pros and cons associated with the currently available techniques is however required.We serious regret the delay of submitting Part II. It has however been submitted in the mid last month and should hopefully be available for discussion soon.

Comment #II-5: Pg 78, L5, please explain the meaning of "wind condition"

Response:The "wind condition" has been corrected as "wind shear condition", it means the near surface wind resulted sear stress which promote Hg emission from soil.

Comment #II-6: Pg 78, L7, and other places, the term "footprint" in environmental studies often refersto an area much larger than what is covered by a DFC because the inlet lines sampleair outside the chambers.

Response:The "footprint" in DFC measurement refers to "the surface area covered by the chamber".

Comment #II-7: Pg 81, L19, "350 km from Beijing". It would be more informative to state the provinceand distance to any nearby Hg sources, instead of distance to the capital

Response:It has been revised as "which is a semi-rural agricultural station approximately 50km fromJinan, Shangong Province".

Comment #II-8: Pg 83, L13, the reviewer could not find any description of "EC flux corrections" inthis or any other sections

Response:The pg 84 L7-10 is the EC flux correction, the sentence has been rephrased as "A series of standard data corrections were implemented following (Sommar et al., 2013b) including the Webb-Pearman-Leuning (WPL) correction. Moreover, tests were applied on 20-min fast time (10 Hz) series raw data to qualitatively assess turbulence for the assumptions required of applying MM methods (steady-state conditions and the fulfillment of similarity conditions)."

Comment #II-9: Pg 83, L26, "low blank were observed for both DFCs", please state whether the DFCfluxes were blank corrected.

Response:It has been changed to "Chamber blanks performed at the field site were consistently low for both DFCs…and not subtracted upon calculation of fluxes...".

Comment #II-10: Pg 84, L20 and Fig 2, precipitation, please clarify mm (cumulative) or mm/time(precipitation rate).

Response:We have clarified precipitation amount as "event-based rainfall".

Comment #II-11: Pg 84, L24, "every 20 min", if once "every 20 min", please provide sampling duration (e.g. 1 min); if continuous monitoring, please provide sampling frequency (e.g.1 hz) and averaging intervals (e.g. 1 min). Also in this paragraph, soil temperature is missing. Furthermore, please 1) identify measurements that were not carried out at 3 m above ground if any, 2) provide the distance between the weather station and the DFCs, 3) consider move this section to 2.1, in case the friction velocity is needed but not estimated by the DFCs.

Response:The sampling is at 1Hz, it has been added in the revised paper. The soil temperature was added in the revised paper. The weather station is close to flux chamber sampling site same as the previous studies, the soil temperature could represent the natural soil temperature. The friction velocity is measured by eddy covariance system providing supplementary data for NDFC flux estimation.

Comment #II-12: Pg 85, L24, "The medians were elevated compared to the hemispheric background,but nevertheless appeared representative of a semi-rural area of North China plain(Zhang et al., 2013)." Please provide range of hemispheric background values andsemi-rural area of North China plain Hg levels.

Response:The corresponding concentration has been added. "The medians were elevated compared to the hemispheric background (1.5 -1.7 ng m-3), but nevertheless appeared representative ofa semi-rural area of North China plain(~3.2 ng m-3, Zhang et al., 2013)".

Comment #II-13: Pg 86, L2-4, "The angular dependence of the ambient Hg^0 level indicates the relative impact of regional anthropogenic Hg sources in mainland China (Zhang et al., 2013)." This sentence is rather ambiguous. Mainland China is enormous in terms of geographic coverage. Please comment on the locations of major Hg sources nearby or in the region, and whether the directional distribution of Hg^0 reflects the transportation by air flows.

Response:we agree with the reviewer and have deleted it in the revised paper.

Comment #II-14: Pg 86, L10-15, "fluxes positively correlated with solar irradiation and soil temperature", "flux was gradual and similar to irradiation and soil temperature", I would suggestto 1) consolidate those two sentences, 2) reference a table/figure or provide r and pvalues, because solar radiation and soil temperature are not plotted in Fig 4.

Response:Pg 86 L11 "fluxes positively correlated with solar irradiation and soil temperature" has been deleted. The specific correlation between flux and environmental factors was presented in table 2.

Comment #II-15: Pg 87, L11, "the surface soil Hg content within the methodological footprint range",please specify such a range, or did you mean the "the surface soil Hg content underthe two DFCs placed 2 m apart is largely homogeneous".

Response:The methodological footprint was assessed in Section 3.4.1 "footprint of flux measurement". The soil samples were collected and analyzed within this area.

Comment #II-16: Pg 87, L13, "In addition, NDFC measured flux calculated from Eq. (1) was presented in gray squares. The data were significantly positive correlated (R = 0.93, R =0.95 for NDFC fluxes calculated with Eq. (2) and Eq. (1) $p < 0.01$)". The first sentencecould be removed. Please rephrase the second sentence to clarify the correlationsamong the three datasets, TDFC, NDFC and DNDFC after correction.

Response:As the reviewer suggested, this part has been combined with the coming paragraph as the following "The data were significantly positive correlated (R=0.93, R=0.95 between TDFC and NDFC fluxes calculated with Eq.2 , Eq.1, respectively; $p<0.01$)."

Comment #II-17: Pg 87, L22, please clarify the meaning of "positive influence".
Response:the "positive influence" indicate that flux increased with applied flushing flow, the sentence has been revised following "The DFC flushing flow rate was identified to have substantial positive influence."

Comment #II-18: Pg 88, L17, "an higher scale of gradient variability", please provide statistical support, e.g. coefficient of variation.
Response:we have provided the statistical support. "Hg0 concentration gradients were observed in the similar rangesof -0.49 to 0.33 and -0.48 to 0.25 ng m-4 in both campaigns (Table 1 and Fig. 4), though the more occasionally shifting conditions of weak and developed turbulence in IC #1 tend towards promotinga higher scale of diurnal gradient variability (IC #1 vs IC #2 standard deviation: 0.09 vs 0.06)."

Comment #II-19: Pg 89, L2, please explain "low quality turbulence". If you have assessed the qualityof turbulence, please provide the methodology. If you have assessed the quality of theturbulence measurements, please rephrase.
Response:We assessed following the method of "The basic flag system of Mauder and Foken (2004) was utilized to indicate weak anddeveloped turbulence, where, quality indices of 0, 1, and 2 denote high, moderate and low quality", section 2.4.
Mauder, M., Cuntz, M., Drüe, C., Graf, A., Rebmann, C., Schmid, H. P., Schmidt, M., and Steinbrecher, R.: A strategy for quality and uncertainty assessment of long-term eddy-covariance measurements, Agricultural and Forest Meteorology, 169, 122-135, 2013.

Comment #II-20: Pg 89, L6-7, suggest provide the net fluxes in Table 1.
Response:On Pg 89, sentence L6-7 was has been deleted, it was repeated information with section 3.4.3 which discussing the cumulative flux observed from various methods.

Comment #II-21: Pg 89, L17, please explain how the "MBR method giving the most confined distribution" while other methods had less confined distribution, by range or by coefficient of variation.
Response:While flux by MBR-method is calculated using the kinematic heat flux, AGM rely on the transfer velocity. Transfer velocity is a function of friction velocity. It is well known that EC measurements of friction velocity generally include more scatter than for e.g. heat fluxes and the reasons for this is described in Foken (2008). The transfer velocity term also include corrections for stability that is not required in MBR.
Foken, T.: Micrometeorology, Springer-Verlag, Berlin, Heidelberg, 306 pp., 2008.

Comment #II-22: Pg 89, L25-27, the reasoning is confusing, you may want to 1) cite average airor soil temperatures to support the claim of "warmer IC2", 2) clarify whether Baya andVan Heyst, 2010; Gustin, 2011, assumed "that the soil Hg^0 efflux was higher duringthe warmer IC#2".
Response:The sentence has been revised following the suggestion. "Even though not measured, it is credible to assume that the soil Hg0 efflux was higher during the warmer IC#2 due to higher temperature (Table 1)(Baya and Van Heyst, 2010;Gustin, 2011)."

Comment #II-23: Pg 90, L17, please explain why "changes in concentration with time" would affectthe MBR but not AGM.
Response:The changes in concentration with time will influence concentration gradient, which will do influence both MBR and AGM flux.

Comment #II-24: Pg 90, L20-24, please explain whether “small sensible heat fluxes” were associatedwith “periods at dawn, dusk and during nighttime” in your IC1 and/or IC2

Response:Sensible heat flux generallydisplays a clear diurnalcycle with maxima at mid-day. Small fluxes prevailing from dawn and dusk. In addition, specifically at dawn and dusk time there are a higher tendency in heat flux sign changetransitions.

Comment #II-25: Pg 90, L26-27, “The MBR method becomes uncertain and may significantly overestimate flux”, please explain why “AGM fluxes were on an average 26.1% lower thanMBR fluxes during IC #1, but 13.8% higher during IC #2.”

Response:The reason was explained in the coming passage from L27 to Pg 91 L1-L10. “The disparate results may largely stem from micro-methodological issues(Fritsche et al., 2008b). In previous studies using the AGM method to gauge various trace gas fluxes including Hg0 (Edwards et al., 2001;Edwards et al., 2005;Simpson et al., 1997), normalization of Eq. 5 was introduced to mitigate for systematical failure of obtaining energy budget closures (Twine et al., 2000) by a factor of 1.3 - 1.35. The AGM method involves momentum flux, and an atmospheric stability parameterization in the flux calculation.For conditions of weak developed turbulence to a greater extent prevailing under nocturnal stable stratification, where u_* is very low, the AGM and MBR methods are prone to large uncertainties and corresponding fluxes are suggested to be flagged by applying wind or friction velocity thresholds (viz. $u_* < 0.07 - 0.1$ m s-1)(Fritsche et al., 2008b;Foken, 2008).”

Comment #II-26: Pg 91-92, first paragraph of section 3.4.1. This passage is a bit hard to follow.Suggest remove general statements (e.g. L25) and rephrase long sentences (e.g. pg92, L1-5) to make clear the estimated footprint of each method.

Response: This section has been rephrased to improve the readability.

Comment #II-27: Pg 92, L27, “The pattern resembles to extent that of latent heat flux”, pleasereference a table or figure where latent heat flux is presented.

Response:The reference “Liu and Foken, 2001” has been added.

Liu, H., and Foken, T.: A modified Bowen ratio method to determine sensible and latent heat fluxes, Meteorologische Zeitschrift, 10, 71-80, 2001.

Comment #II-28: Pg 93, L3-11. The point of this passage is not very clear. The challenge in qualifying air-surface exchange of Hg is well understood. Therefore, the authors may want to support the discussion with new findings in this study or remove this passage.

Response:The point of this passage is to explain the observed flux pattern. Even though we know that quantifying air-surface Hg flux is challenging, the possible reasons resulted in the flux patterns are discussed here.

Comment #II-29: Pg 93, last line, when p>0.05, the correlation becomes statistically not significant,i.e. the hypothesis of “no correlation between X and Y” could not be rejected, insteadof a “weak correlation”.

Response:The sentence has been rephrased to “REA fluxes were not significantly correlated with fluxes derived by other techniques (R<0.2, p> 0.05)”.

Comment #II-30: Pg 94, L17, “This was likely due to the presence of high eddy diffusivity of heat.”It is unclear what “this” refers to and why high eddy diffusivity of heat would cause “alarge increase” of one flux or a stable flux of another.

Response:The sentence has been revised to “A period of divergence in the magnitude between the derived turbulent exchange parameters (eddy diffusivity of heat and υ_{tr}) resulted in intersected courses of MBR and AGM cumulative flux (17th Nov.).”

Comment #II-31: Pg 95, L10-13, please reference a figure to support your discussion.

Response:Fig. 11b has been added.

Comment #II-32: Pg 96, L27, “the diurnal variation of MM fluxes were biased under the low turbulence condition”, there is a lack of support in Section 3 about factors that bias thediurnal variation, suggest remove.

Response:It has been removed.

Comment #II-33: Pg 97, L4-6, please explain the association between the “poor to moderate” “comparability between individual DFC and MM fluxes” and “the risk of utilizing sporadic (non-diurnally resolved) flux measurements as representative of an ecosystem.”

Response:See Pg 94. line 25 – 29.

Comment #II-34: Pg 97, last paragraph. The discussion seems to be general and lacking a directlinkage to the data and analysis presented in this paper, suggest remove.

Response:This paragraph is of important for future application of each flux quantification method.

Comment #II-35: Table 1. Please clarify “NDFC” or “NDFC after correction”. It might help your discussion to include net fluxes, median absolute deviation or coefficient of variation, dry deposition velocities, kurtosis, skewness, and the results of normality tests, instead of those numbers popping in the main body.

Response:we thank the reviewer for suggestion and have reorganized the main text. It has been clarified in the revised paper, see section 3.3.2.

Comment #II-36: Fig 8 caption, please explain “those plots under sensible heat flux Wm^{-2} (filledcircles)”. The unit of H should be provided too.

Response:The filled circle denotes the data at < 20 W m^{-2} irradiance.

Comment #II-37: Fig 9 caption seems incorrect, 5th and 95th percentiles should be lower/higherthan the 10th and 90th percentiles, respectively. Also, whiskers are missing for fluxesover wheat canopy in all three MM subplots.

Response:The caption has been revised as“Box and whisker plots of diurnal Hg^0 flux patterns measured with various techniques. The two box horizontal border lines represent 25th, and 75th percentiles from bottom to top, and whiskers indicate 10th and 90th percentiles of Hg0 flux. Bold line and fine line in the box indicate mean and median flux”. Due to the short measurement period the whisker is unavailable for the wheat campaign.

Comment #II-38: Please report p values in any figures where correlation coefficient (r) is presented.

Response:The *p* value has been added.

III. Editorial suggestions

The use of English language is largely satisfactory. However, the overall writing stylehas much room for improvement. The reviewer found many examples of awkwardC7407sentence structure, run-on sentences, ambiguous references (not citations, but use ofwords like “this”, “both”), and unusual word choices. Some examples are listed below.Furthermore, a proof reading by a native speaker could help.

Comment #III-1: The term NDFC was defined at least twice in the main body.

Response: the second NDFC definition was removed.

Comment #III-2: Suggest avoiding the use of first person, i.e. “we”.

Response: we have carefully edited the paper to reduce use of first person.

Comment #III-3: Significant numbers, e.g. wind speed and Hg^0 concentrations, perhaps one decimal is sufficient; for percent differences, integers could be adequate.

Response:for the number presented was based on the precision of the data.

Comment #III-4: Citation in the main body, the number of papers seems a bit excessive especially insections 1 and 2, which hinders the readability of the paper. The authors may want tolist a few examples each time, perhaps citing the original methodology papers and themost recent applications. When there is more than one paper, you may want to orderthem by year of publication.

Response: we have re-ordered the citation of papers in the paper followed the suggestion.

Comment #III-5: In quite a few incidents, a review of others’ work (e.g. pg 87, L4; pg 87, L20; pg95, L15) was placed before your results. You may want to present your results first, followed by a discussion.

Response:We agree with the reviewer, those sentence has been reordered following the suggestion.

Comment #III-6: Pg 74, L13, please define DFC.

Response: DFC has been defined.

Comment #III-7: Pg 76, L17, could read “Lin et al. (2012)”.

Response:It has been changed.

Comment #III-8: Pg 76, L21, “4-day” or “4 days”.

Response:It has been changed.

Comment #III-9: Pg 83, L15, a reference is needed for the SOP by NADP.

Response:The reference has been added.

National Atmospheric Deposition Program (NADP): AtmosphericMercury Network Operations Manual (2011–05) Version 1.0., http://nadp.isws.illinois.edu/amn/docs/AMNet_Operations_Manual.pdf, NADP Program Office, 2204 Griffith Dr., Champaign, IL 61820, 2011.

Comment #III-10: Pg 88, L15 and other places in some tables and the main body, the range expressed as e.g. “-2 –4 m/s” is hard to follow, suggest using e.g. “-2 to 4 m/s”.

Response:It has been replaced.

Comment #III-11: Pg 94, L5-7, those #s could be reported in Table 1.

Response: the dry deposition velocity is easier to be compared when presented in this order.

Comment #III-12: Pg 96, L20-27, the switches from temporal trends to median values of the three MM method then back to temporal trends make the passage hard to follow, please rephrase.
Response:The sentence has been re-ordered as suggested.

Comment #III-13: There is little need to repeat in the main body the content of figure captions regarding to the meanings of some markers.
Response:It has been deleted as suggested.

Comment #III-14: Fig 3, please provide units.
Response:The units were added.

Comment #III-15: Fig 4, the plots and fonts are a bit too small to read; also the "black bars given incorresponding plots represent absolute flux uncertainties" make the plots even harderto read. You may want to remove the black bars and enlarge the charts.
Response:The plot and fonts were adjusted to provide better readability.

Comment #III-16: Figs 5&7, the reviewer could not find the "filled diamond".
Response:The "filled diamond" for TDFC and NDFC were within and out of the box horizontal boarder.

Comment #III-17: Examples of unusual word choices:
Pg Line Words Comments/Suggestions
74 16 driving rephrase:
77 6 benefits "advantages"
77 19 sophisticated remove
82 1 spatial homogeneously rephrase
83 20 limited rephrase
85 18 integral rephrase or remove
86 22 As a consequence consequently
87 3 foundation rephrase
89 16 It is obvious remove
90 24 approximately remove
92 14 many up to x
92 19 there is an obvious lag there is a 2-hr lag
93 5 So they not thus they do not
95 18 statistical correlation Pearson correlation
97 10 next to REA in scale remove
97 13 behavior rephrase
97 13 in turn remove
Fig 5 Caption unbroken solid
Fig 8 Caption empty open
Response:We thank the reviewer for kindly suggestion of word used. The suggested word has been careful considered and replace in the paper.

Comment #III-18: Examples of awkward sentences:
Pg Line
74 24
86 27
87 23-25
97 19
Response: Those sentenceshave been revised for better readability.

Anonymous Referee #3:

Overall comments:
In this manuscript, Zhu et al. performed a comprehensive inter-comparison of fivecontemporary Hg(0) flux quantification techniques. This study is of broad interest tothe audience of this journal and to the scientific community studying environmental fateof Hg. This paper should be acceptable for publication following some minor revisions.In addition, this manuscript still requires grammatical edits throughout.
Response:We thank the reviewer for the positive comments on the scientific importance of this research. All the specific comments have been incorporated the in the revised manuscript. Our point-to-point response to the comments is given below (in blue). Corresponding revision was added in the manuscript.

Specific comments:
Comment #1: Page 22286, line 8: delete the third 'ng m-2 h-1'
Response: It has been deleted as suggested.

Comment #2: Page 22295, line 24: change flux to fluxes
Response: It has been changed as suggested.

Comment #3: Page 22288, line 17: change an to a
Response: It has been changed as suggested.

Comment #4: Page 22289, line 20: change canopies to canopy; change contribute to contributes
Response:thetext has been reworded accordingly.

Comment #5: Page 22291, line 11: change methods to method
Response: It has been revised as suggested

Comment #6: Page 22291, line 15 to 20: rewrite the sentence 'Other gases (e.g. NH3,CH4) that. . .. . .AGM fluxes '
Response:The sentence has been revised as "Other gases (e.g. NH_3, CH_4) that have been studied with this triad of MM-techniques, higher variability in REA flux is generically observed (Nemitz et al., 2001;Fowler et al., 1995;Moncrieff et al., 1998). In addition, systematically fluxes differences between a suite of NH_3-REA systems as well as collocated AGM system inter-compared have been reported (Hensen et al., 2009)".