

Interactive comment on “Air–surface exchange of Hg⁰ measured by collocated micrometeorological and enclosure methods – Part 1: Data comparability and method characteristics” by W. Zhu et al.

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Anonymous Referee #2: We deeply appreciate the reviewer's carefully review of our manuscript. We have incorporated the thoughtful 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 extent buried by the writing style, including unnecessary materials in Section 2

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(Material and methods), redundancy in the Results and Discussion section, some general statements or incorrect statements, 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 the manuscript. The detailed information of equations and repeated discussion in different sections would be appreciated for a thesis or a report, while a concise style might be more 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 #1-1: All general descriptions as well as equations and related explanation in pages 78-81 could be omitted. Interested readers can find detailed methodology descriptions in papers 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 these equations and 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 #1-2: The manuscript could be shortened by removing sentences simply stating numbers presented 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 Hg⁰ flux variation range and corresponding average flux measured by DFC techniques at soil surface were shown in Table 1. A broader variation range and mean

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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 #1-3: The authors may want to consolidate some subsections to remove redundancy regarding to results and discussion, thus to improve the readability. Some examples are listed below: Item Occurrences Q1: Correlation between DFC and meteorological parameters Pg 86, L11; pg 95, L19 Q2: Correlation between AGM and MBR Pg 90, L22; pg 93, L26 Q3: Comparison between TDFC and NDFC: 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 $w \approx 2$, the correlation coefficient diminishes drastically and the fall-off in slope () 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.” Pg 93, L26. The repeated information in L26 has been deleted. Q3: the pg 87 L12-19 has been removed and integrated with pg 87 L26 and pg 88 L1. Q4: the page 86, L15-17 has been deleted and combined into pg 92, L14-16.

Comment #1-4: 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 understand some discussions presented in this manuscript, while the differences within and between DFC and MM methods will be repeated in Part 2 to facilitate the investigation of 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 are

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conclusions in Part 2 presented in this manuscript without relevant methodology and discussion, for instance, reasons of dissimilarity in the DFC and MBR (pg 87, L1-3), reasons of variability in REA and other MM methods (pg 90, L2-5), reasons of disparate AGM and MBR (pg 91, L1-20), reasons of different AGM and MBR in the two campaigns (pg 95, L3-13). Response: we thank the reviewer for the suggestion and have revised the illustration accordingly. The two companion papers are 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 that have 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 the NDFC in this manuscript is a bit confusing partially due to some unfounded statements, e.g. Comment #1-5: Pg 76, L17, “a novel designed DFC (NDFC) based on surface wind shear condition (friction velocity) rather than on artificial wind shear to account for natural shear conditions.” Pg 78, L2, “a novel DFC (NDFC) design capable of controlling the internal shear over measurement surface (Lin et al., 2012). The NDFC internal wind shear condition was precisely controlled to relate to the applied wind shear rate to the atmospheric boundary shear condition (therefore wind condition)”. It

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is not clear how to implement this technique when the flushing rate was indeed fixed in the NDFC operation (pg 78, L7) and the monitoring of atmospheric boundary shear condition is not mentioned. Even with the highly variable friction velocity available, the “precisely controlled” “internal flushing condition” “to relate the applied flushing rate to the atmospheric boundary shear condition” would need a closed-loop system which was not available in this paper and the NDFC paper (Lin et al., 2012). Those statements also contradict equations 1 and 2 which have a fixed flushing 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 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 Eq 2.

Comment #1-6: Pg 86, L16 “DFCs flux was derived from Hg₀ mass balance calculation every 20 min, different from the MM flux that relied on atmospheric turbulence processes.” This sentence contradicts other statements that the modified DFC taking into account turbulence, e.g. pg 76, “based on surface wind shear condition (friction velocity) rather than on artificial fixed flushing to account for natural shear conditions”, pg 87, “the well-developed turbulence (higher friction velocity, Fig. 2) during daytime caused the corrected Hg₀ 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 #1-7: Pg 88, L2-6, “Given that DFC of conventional types cannot reproduce atmospheric turbulence. NDFC is more preferable for the determination of net Hg₀ gas exchange over soils.” This sentence seems to be over-promoting the NDFC when

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in fact no DFCs can “reproduce atmospheric turbulence” regardless of corrections. Response: the “reproduce atmospheric turbulence” has been corrected with “be rescaled with natural surface shear stress”.

3. pg 86, L20, “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, the average flux is slightly positive”. The reasoning here seems questionable; kurtosis and/or skewness themselves are not related to the sign (positive or negative) of a population or sample mean. 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 Hg₀ flux from NDFC flux to be approximately 3.5 times higher than the TDFC flux”. This assessment seems unfounded. Fig 6 had a slope of 2, i.e. one flux is twice as high as the other, or one time higher. Also, the slope of 1.1 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 lower when 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 Hg₀ flux from NDFC flux to be approximately 3.5 times higher than the TDFC flux” has been corrected as “the corrected Hg₀ 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 Pearson correlation and other method of correlation assessment may produce different value. Typically Spearman rank correlation and Kendall rank correlation would pro-

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duce 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 same sets 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 fluxes specifically for MBR vs. NDFC, though the correlation between individual hourly data points is weak, the fluxes integrated over time show strong agreement." Perhaps it should read "Figure 12". Furthermore, the readers might be interested to see if the same could be said with the scatterplots of hourly and cumulative fluxes 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 at time t , the data points are not independent of each other. Consequently, the authors may want to remove the regression equation and r values and to include scatterplots of hourly and cumulative fluxes 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 on correlation among variables which is presented in Table 2, instead of identifying major factors affecting the air-surface exchange processes. In addition, the authors seem to

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have reached contradicting conclusions, "The environmental variables also significantly modified the gradient-MM fluxes (factor loading > 0.3)", and "Two separate PCA was resolved for gradient fluxes variance (factor 2). The two factors are not contributed from the environmental variables (factor loading < 0.1), suggesting that the MM fluxes and their temporal characteristics are likely influenced by turbulent transport processes . . ." Furthermore, in both IC1 and IC2, only the first two factors had more than one loading > 0.4 . In other words, factors 3-5 in IC1 and factors 3-4 in IC2 failed to be valid factors when there is one loading > 0.4 hence that factor only represents one variable. In cases like this, all 4 or 5 factors may become uncertain. This is likely due to 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 measurements could be too low to represent regional movement of air mass. An alternative could be datasets from a nearby airport or air flow directions from trajectory models with small grids. Response: The meteorological data were aimed to elucidate the influence of environmental factors 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 the selected 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

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And 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 and natural surfaces through complex and yet not well understood processes, re-emitting previously deposited Hg back to the atmosphere (Bash, 2010; Gustin and Jaffe, 2010). Recent estimation indicates that annual natural emission accounts for two-thirds of global 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 may want 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 (< 0.1m²)”. 1) in case you were not sure that areas covered any DFCs ever existed were < 0.1 m², perhaps “in the order of 0.1 m²” could be more conservative, 2) perhaps “representing the smallest scale as the areas covered by the devices are typically in the order of 0.1 m²” could be more appropriate. 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 under field 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 the examined flux measurement methods using statistical analyses”. Response: Even though there is no standard or preferred field Hg₀ 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.

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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 shear stress which promote Hg emission from soil.

Comment #II-6: Pg 78, L7, and other places, the term “footprint” in environmental studies often refers to an area much larger than what is covered by a DFC because the inlet lines sample air 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 province and 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 50 km from Jinan, Shangong Province”.

Comment #II-8: Pg 83, L13, the reviewer could not find any description of “EC flux corrections” in this 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 DFC fluxes 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”.

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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 moving this section to 2.1, in case the friction velocity is needed but not estimated by the DFCs. Response: The sampling is at 1 Hz, 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 and semi-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 of a 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 Hg₀ 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 Hg₀ 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 temper-

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ature”, I would suggest to 1) consolidate those two sentences, 2) reference a table/figure or provide r and p values, 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 under the 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 sentence could be removed. Please rephrase the second sentence to clarify the correlations among 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 ranges of -0.49 to 0.33 and -0.48 to 0.25 ng m⁻⁴ in both campaigns (Table 1 and Fig. 4),

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though the more occasionally shifting conditions of weak and developed turbulence in IC #1 tend towards promoting a 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 quality of turbulence, please provide the methodology. If you have assessed the quality of the turbulence measurements, please rephrase. Response: We assessed following the method of “The basic flag system of Mauder and Foken (2004) was utilized to indicate weak and developed 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 \dot{Q}_{net} 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 conññed distribution” while other methods had less conññed distribution, by range or by coefññcient 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 air or soil temperatures to support the claim of “warmer IC2”, 2) clarify whether Baya and Van Heyst, 2010; Gustin, 2011, assumed “that the soil Hg_0 eññux was

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higher during the warmer IC#2”. Response: The sentence has been revised following the suggestion. “Even though not measured, it is credible to assume that the soil Hg_0 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 affect the 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 $\dot{Q}_{\text{sensible}}$ ” were associated with “periods at dawn, dusk and during nighttime” in your IC1 and/or IC2 Response: Sensible heat flux generally displays a clear diurnal cycle 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 change transitions.

Comment #II-25: Pg 90, L26-27, “The MBR method becomes uncertain and may significantly overestimate \dot{Q}_{net} ”, please explain why “AGM \dot{Q}_{net} were on an average 26.1% lower than MBR \dot{Q}_{net} 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 Hg_0 (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 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. $m s^{-1}$) (Fritsche et al., 2008b;Foken, 2008).”

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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", please reference 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, instead of 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 "a large 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) 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 discus-

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sion. 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 the diurnal 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 direct linkage 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 $W m^{-2}$ (filled circles)". 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/higher than the 10th and 90th percentiles, respectively. Also, whiskers are missing for fluxes over wheat canopy in all three MM subplots. Response: The caption has been revised as "Box and whisker plots of diurnal Hg₀ flux patterns measured with various techniques. The two box horizontal border lines represent 25th, and 75th

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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 style has much room for improvement. The reviewer found many examples of awkward sentence structure, run-on sentences, ambiguous references (not citations, but use of words 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 Hg0 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 in sections 1 and 2, which hinders the readability of the paper. The authors may want to list a few examples each time, perhaps citing the original methodology papers and the most recent applications. When there is more than one paper, you may want to order them 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

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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): Atmospheric Mercury 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 in corresponding plots represent absolute flux uncertainties" make the plots even harder to read. You may want to remove the black bars and enlarge the

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charts. Response: The plot and fonts were adjusted to provide better readability.

Comment #III-16: Figs 5&7, the reviewer could not see 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 sentences have been revised for better readability.

Interactive comment on Atmos. Chem. Phys. Discuss., 14, 22273, 2014.