Answers to Anonymous Referee #2

General comments

1. This paper […] would be a useful contribution to enhance our current understanding of CH4 dynamics in wetland systems, but the paper needs to be further revised before being considered for publication in this journal. As this paper aims to describe a method in determining ebullition and diffusion from the concentration trace of autochamber measurements, the authors should spend more time in describing the methodological details of the calculation (e.g. determination of diffusive flux) and justifying the use of this approach compared to other existing ones (e.g. Goodrich et al 2011) in estimating ebullition.
   We added a flow chart showing the presented algorithm and a more detailed explanation of it to section *"2.2 Flux calculation and separation algorithm"*. We also added a more detailed discussion about advantages compared to other direct or indirect separation approaches (justification) to section *"3.3 Overall performance"* (please see also 24/28. Comment reviewer #1).

2. The authors spent a substantial proportion of time examining the temporal variability of diffusive, ebullition, and total CH4 fluxes – but these does not prove that the algorithm is working successfully. There is a need to provide further validation of this method in separating diffusion and ebullition through field testing, e.g. the use of bubble traps. Such comparison should be done in this paper to provide a more affirmative testing of the algorithm, rather than in future studies as suggested by the authors in the last paragraph.
   The laboratory experiment showed than the algorithm works to filter ebullition events. In addition we now refer to eddy covariance measurements at the same field site during the same study period, showing comparable total CH_4 emissions (please see also answer to 23. Comment reviewer #1). We think that Bubble traps are not providing a sufficient validation of short term or single ebullition events occurring at the measurement. On the one hand, measurements of total (automatic chamber) and ebullition flux (bubble traps) would be spatially separated. On the other hand, a number of authors showed that ebullition events occur erratically in time and space (e.g. Lindgren et al. 2016; Anthony and Anthony 2013; see pictures below showing bubbles trapped in ice at the study site during January 2016). Thus, the spatial and temporal difference between separated AC measurements and bubble trap measurements will introduce a spatial and temporal error into the comparison of calculated fluxes using the presented approach and measurements of e.g. bubble traps. This bias can be only reduced by a sufficient number of spatial (bubble traps as well as automatic chambers) and temporal measurement repetitions and data aggregation. Both requirements, needed for a low bias will, however, only yield in a comparison of either spatially or temporally (month to years) aggregated comparison, but are not a proper validation of single ebullition events or short term separations of ebullition and diffusion (e.g. hours or days). If the bubble traps, however, would be installed underneath the chambers, to overcome mentioned spatial and temporal heterogeneity, the chamber measurements would be substantially biased, since they would not include ebullition events trapped by the bubble traps. We therefore do not agree on the statement that bubble trap measurements are a suitable method for validating the accuracy of the presented calculation algorithm regarding single measurement or short term (<1 year) flux separation results.

To better address this issue and show the huge advantage of the presented approach we added a discussion regarding direct (e.g. bubble traps) and indirect (data processing based approaches) methods to estimate the CH_4 flux components to section *"3.3 Overall performance"* (please see also answers to 24/28. comment reviewer #1*)*.

3. Also, are there any drawbacks of using IQR of concentration change to detect ebullition events? If ebullition occurs continuously through the measurement period, would the proposed method fail to identify ebullition events due to a consistently, large magnitude of concentration change?
   As stated in the MS: *"... flux separation might be hampered due to a steady flux originating from other processes than diffusion through peat and water layers, such as the steady ebullition of micro bubbles (Prairie and del Giorgio 2013; Goodrich et al. 2011)."*. A consistently large magnitude in concentration changes during the chamber measurements will not hamper the total flux calculation. However, flux separation might be hampered. Out of 14828 valid measurements the algorithm was unable to calculate a valid diffusive flux for 170 measurements. This equals less than 1.15 % of all measurements. Taken into account that the presented measurement site is characterized by rather large CH4 emissions (e.g. Franz et al. 2015) and consistently occurring ebullition events, this problem seems to be negligible for the presented site, but might be of relevance in other wetland ecosystems. We therefore stated the need to apply presented algorithm to other wetland ecosystems within section *"4. Conclusions"* of the MS.

4. Also would the proposed method be able to identify both "major" and "minor" ebullition events? The lab test does not provide a definitive answer to these questions, and further tests with a greater variety of conditions (e.g. pulse vs. continuous injection) are required.
   Due to the variable ebullition filter (IQR-filter), the proposed method identifies is able to identify "minor" and "major" ebullition events. This is also shown by the laboratory experiment which includes simulated ebullition events of different strength. However, ebullition flux components are given as flux integrated over the entire measurement (which might include bigger and smaller ebullition events not separated from each other). Calculated ebullition fluxes (integrated over a single measurement) ranged from approx. zero (0.0002 µmol m^{-2} s^{-1}) to 0.6780 µmol m^{-2} s^{-1}, which supports the capability of the algorithm to identify "minor" and "major" ebullition events.

5. Since the algorithm only estimates ebullition as the difference between total CH4 flux and diffusive flux, accurate quantification of the diffusive components becomes very important. What is the minimum detectable flux of this system?
   The precision of the sensor for CH_4 measurements is 0.6 ppb (10sec records). In principle the minimum detectable flux would be a (near) zero flux. The lowest valid diffusive flux measured during the study period was 0.0012 µmol m^{-2} s^{-1}, which equals a concentration change over 10 minutes of 1.14 ppb (this detection limit might change due to the user defined parameter setups made in the script (e.g. set p-value)).

6. In open-water systems, CH4 flux is expected to be lower than that in vegetated wetlands – if CH4 concentration change within the 15-second interval is not observable, the calculation of diffusive flux might be biased that further causes inaccuracies in the estimation of ebullition. Why not determine the ebullition component directly from the time series of headspace CH4 concentration? These issues should be addressed before the algorithm could be trusted and applied in other autochamber systems for separating the CH4 flux components.

   In the case of low concentration changes the calculated diffusive CH_4-flux using the presented approach would be around zero. The reason therefore is that the calculation algorithm is not based on the r^2 as quality criteria, which was e.g. used by Goodrich et al. (2011), who discarded fluxes for steady flux analysis (diffusion) with an $r^2 < 0.8$. A flux will be identified as long as the regression slope is significant. In the case of low but valid diffusive fluxes, the potential bias to separated fluxes will be small as well and the influence on the ebullition flux component rather negligible. If diffusive flux calculation is not possible due to invalid fluxes (non-significant regression slope), the measurement system (e.g. chamber design or measurement frequency and duration) could be adapted (e.g. smaller chamber volume with bigger basal area), since non-observable fluxes are rather a problem of the used measurement device (chamber system) than the presented flux calculation and separation approach (which can be only as good as the data it is based on). Thus the detection limit might be reduced.

Specific comments

7. L41-42 – Were the quartiles and IQR referring to concentrations within one measurement period?

   Yes. The IQR is referring to the entire single measurement (without death band) and the quartiles are referring to the specific MW subset of the single measurement. If the IQR would refer to the specific MW, the diffusive CH_4 emission present within subsections of the measurement without ebullition events could be not identified due to the highly sensitive IQR-criteria.

   If the quartiles would refer to the entire MW, a bigger ebullition event during the measurement would result in a diffusive flux component, which includes smaller ebullition events, cause the filter criteria would be not sensitive enough.

8. L70 – In open-water systems, I assume vegetation is absent. If this is the case, plant-mediated transport will not be one of the CH4 release mechanisms.

   We agree and changed the sentence to:

   "In wetland ecosystems, CH_4 is released via three main pathways: I) diffusion (including "storage flux", in terms of rapid diffusive release from methane stored in the water column), ii) ebullition and iii) plant-mediated transport (e.g., Goodrich et al. 2011; Bastviken et al. 2004; Van der Nat and Middelburg 2000; Whiting and Chanton 1996)."

9. L108 – How does one define "medium" and "major" ebullition events? Are there any objective criteria for such categorization?

   We changed the sentence by adding the threshold given by Goodrich et al. (2011): *"However, the static threshold to determine ebullition events, as well as low-resolution measurements, limited the approach to estimates ebullition events characterized by a sudden concentration increase ≥ 8 nmol mol^{-1} s^{-1}, which prevents a clear flux separation.".* The threshold given in Goodrich et al. equals an increase due to ebullition of 1.273 ppm per 15s record for the measurement system presented in our MS (bigger chamber volume). Thus a number of smaller ebullition events would be not taken into account. However, it is of course hard to give objective criteria for our categorization of *"major"*, *"medium"* or *"minor"* ebullition events. These categories might be different from measurement site to measurement site as well as the used chamber design (size and volume). This is the main reason why we want to introduce a variable threshold for flux separation.

10. L134-136 – I think the hypothesis can be further refined. The flux separation algorithm might help tease out the contribution of diffusion and ebullition to overall flux, but itself could not be used to reveal the spatial and temporal dynamics – this is rather achieved by the AC system.
    We partly agree. Both, the chamber system and the used algorithm complement each other (please see also answer to 26. Comment reviewer #1 and changes in 3.3). Spatial dynamics are of course a result of the spatially distributed chambers and not of the flux separation algorithm. However, temporal dynamics for the diffusive and ebullition flux components are obtained through the presented algorithm, since the chamber measurements itself only deliver temporal dynamics of total CH_4 fluxes. Therefore we modified our hypothesis to:

    "We hypothesize that the presented flux calculation and separation algorithm can reveal together with the presented AC system, concealed spatial and temporal dynamics in ebullition- and diffusion-associated CH_4 fluxes. This will facilitate the identification of relevant environmental drivers.".

11. L161-165 – Only 4 chambers along the transect? No replications? Also, what is the shape of the chamber?
    Yes, the study site consist of 4 Chambers along a transect. However, the spatial difference between the chambers along the transect is not within the scope of this MS, since the study site only acts as test data set, wherefore 4 chambers are assumed to be sufficient. The same accounts for replications. The chamber shape is shown in Fig. 1 and now also stated in the MS (section 2.1):

    "The AC system consists of four rectangular transparent chambers, installed along a transect from the shoreline into the lake."

12. L171-173 – What is the rate of gas flow within the AC system? Do the chambers equipped with a vent tube for pressure equalization?
    We added the gas flow rate of 5 l per minute to the MS. The chambers are not equipped with a vent for pressure equalization. Possible effect due to this measurement method specific limitation were tried to be reduced by a rather slow and soft chamber closure as well as by the applied death band at the beginning of each measurement. This might be a limitation of the presented AC system (chamber system), but it is not a limitation for the presented flux separation algorithm. Quiet the converse, the good overall agreement of the in parallel performed eddy covariance measurements (please see also answer to 23. comment reviewer #1) with the shown chamber measurements of the total CH4 emissions indicate that either the chamber measurements are not affected, or that the flux calculation approach was able to discard pressure related artifacts.

13. L173-175 – What did "overcompensation" exactly mean? Did you refer to the drop in CH4 concentration in the chamber headspace – this looked strange to me.
    Yes exactly. We added a short explanation for this to the MS (please see also answer to 1. comment reviewer #1).
    Would this mean that the fan is not effective enough in homogenizing the headspace air?
    Yes, this means that the fan is homogenizing the comparable huge chamber volume (1.5 m^3) within 15 to 30 sec. A stronger fan might trigger ebullition by stir up the water underneath the chamber. However, as stated in the MS: *"To avoid measurement artefacts (e.g., overcompensation), being taken into account as start or end concentration, measurement points representing an inherent concentration change smaller or larger than the upper and lower*

quartile ± 0.25 times IQR were discarded prior to calculation of the total CH_4 flux.", the presented algorithm is accounting for this artifact. Since the chamber headspace homogenization is a purely chamber design related limitation, it is not a limitation of the presented flux separation algorithm. A faster homogenization would simply result in a non-discarding of concentration records for the total flux calculation, and thus not affecting the flux separation result.

14. L192-196 – The equation deriving CH4 flux does not look right – the unit of CH4 flux shown is umol C m-2 s-1, but in the calculation molar mass of CH4 is used?
    We corrected *"µmol"* into *"µg"* within the equation.

15. L200-204 – Would appreciate a more in-depth description of the protocol here, as this is the crucial part of the paper. How did the variable moving window work? Were fluxes calculated for various durations of MW within the 10-min deployment period, as long as the rigid outlier test was passed? If this was the case, which one would be chosen to represent diffusive fluxes?
    Yes, various diffusive fluxes might be calculated for one flux measurement using different durations of this measurement. We added a flow chart to the MS, to better explain the used algorithm (please see also answer to 1. comment reviewer #1). Furthermore, we rewrote section 2.2 and added more details.

    "The resulting numerous $CH_{4_{diffusion}}$ fluxes calculated per measurement (based on the moving window data subsets) were further evaluated according to different exclusion criteria: (i) range of within-chamber air temperature not larger than ± 1.5 K; (ii) significant regression slope ($p \leq 0.1$, t-test); and (iii) non-significant tests ($p > 0.1$) for normality (Lillifor´s adaption of the Kolmogorov-Smirnov test), homoscedasticity (Breusch-Pagan test) and linearity. In addition (iv) abrupt concentration changes within each MW data subset were identified by a rigid outlier test, which discarded fluxes with an inherent concentration change outside of the range between the upper and lower quartile ± 0.25 times (user defined) the interquartile range (IQR). Calculated $CH_{4_{diffusion}}$ fluxes which did not meet all exclusion criteria were discarded. In case of more than one flux per measurement meting all exclusion criteria, the $CH_{4_{diffusion}}$ flux with a starting CH_4 concentration being closest to the atmospheric CH_4 concentration was chosen."

16. L226 – Were these the volume and area of the chamber or the tub? How much water was added into the tub?
    We added 12 l water to the tub. The volume of 0.114 m^3 represents the total headspace volume (air-filled) of the construction. Tub and chamber volume were each 0.063 m^3. To better address this, we changed the sentence to:

    "In order to artificially simulate ebullition events, distinct amounts (5, 10, 20, 30 and 50 ml) of a gaseous mixture (25 000 ppm CH_4 in artificial air; Linde, Germany) were inserted by a syringe through a pipe into a water filled tub (12 l) covered with a closed chamber (headspace V=0.114 m^3; A= 0.145 m^2)."

17. L231-233 – This assumed that all the added gases would be released as gas bubbles without any CH4 being dissolved in water. How would the authors ensure the absence of dissolved CH4 in water?
    Right, some of the injected CH4 will be dissolved in the water. We therefore added the following sentence: *"To ensure CH4-saturation after the first simulations of ebullition events, the water within the tub was not replaced during the laboratory experiment."*. The water temperature during the lab experiment was approx. 20°C, which results in a solubility for CH4 of ~25mg/l or 300 mg for the 12 l of added water. We injected 4.5 to 51.5 ml of a gaseous mixture with 25 000 ppm. This equals an injection of 115 to 1290 ppm/l per simulated ebullition event (n=20). Since we did

not change the water during the entire measurement, it was assumed that the water was saturated with CH4 after the first simulations (starting with a 50 ml injection). However, of course the issue of dissolved CH4 might be a part of the (small) deviation of the measured ebullition fluxes from the theoretical 1:1-agreement shown in Fig. 3 (now 4).

18. L237-239 – How long was the chamber closed, and how was CH4 ebullition converted to amount (mg m-2) as shown in Fig. 4? Would be useful to show the time series of CH4 concentration as well in the lab test.

    Since the laboratory experiment was carried out manually, the closure time for the performed chamber measurements varied between 5 to 10 minutes. An exemplary time series of measured CH_4 concentrations during one pulse experiment is shown in Fig. 2a (now 3a). The expected concentration changes within the chamber headspace as the result of methane injections were calculated as the mixing ratio between the amount of inserted gaseous mixture (25 000 ppm) and the air filled chamber volume (2 ppm) and related to the chamber/tub basal area.

19. L242-246 – However, increasing the frequency of concentration measurement might make it harder to detect significant concentration changes for quantifying diffusive fluxes, which could be low in open-water systems.

    This important issue is clearly a question of measurement accuracy and detection limits. Thus, it is not directly a problem of the presented calculation and flux separation algorithm. The presented chamber system has a rather large chamber volume, which makes it hard to observe really small concentration changes within a shorter period. However, applying the algorithm to a chamber system, featuring a smaller chamber volume might solve this problem. Hence, low fluxes are a limitation of the measurement system and its specific design (which shows the importance of adapting the measurement system to the measurement site conditions), but does not constitute a limitation of the presented algorithm (which can be only as good as the data it is based on).

20. L251-257 – Would need some elaborations on why this method is better than other existing methods of quantifying ebullition (e.g. Goodrich et al. 2011). Gas traps should still be able to work in shallow water systems?

    We added advantages compared to direct measurements to section 3.3. Gas traps will work as long as the water level is not dropping below a certain level (given by their specific design). The water level at the presented study site, however, dropped several times during the last 10 years below the sediment surface, or stayed only a few cm (<5 cm) above the sediment surface. Hence, during this period the use of bubble traps and especially bubble shields will be limited depending on their specific design. A water level just slightly above the sediment/peat, however, will not necessarily exclude ebullition events (e.g. GAZOVIC et al. 2010). As a result, measurement gaps will occur when using bubble traps instead of the presented algorithm (please see as well section 3.3 given above).

21. L257-260 – It is a bit far-fetching to suggest that this method is "applicable to a broader range of different manual and automatic closed chamber systems, instrumental setups, study designs, and ecosystems" without other solid evidence.

    We changed the sentence to:

    "As a result of this, the presented, data processing based approach will be applicable as long as the underlying closed chamber measurements deliver continuous data sets for CH_4 concentration and air temperature.
    Accounting for the few prerequisites (high resolution closed chamber measurements) as well as mentioned advantages, an application of the presented approach to open-water areas of a broad range of wetland ecosystems and closed chamber systems is stated. ".

22. L280-281 – Not sure about the claim that diffusive flux shifted to a daytime maximum was valid. Higher CH4 flux was still observed during the night period between midnight and 6 am.
    We changed *"daytime"* for *"early morning hours"* within the paragraph.

23. L281-286 – This might be tested by measuring CH4 concentration at different water depths. While thermal mixing might be weaker during daytime, this might be compensated by stronger wind and mechanical mixing. Low wind speed at night might contribute to lower diffusive fluxes owing to poor mixing of air above water surface. Also, temperature in July and September did not differ that much – why was the diurnal pattern different between these two months? Further discussion is needed.
    Actually the diurnal cycle changed twice (as shown in Fig. 6 (now 7)): once from July to August, and back again from August to September. We therefore rewrote the misleading sentences*:*

    "However, compared to the diurnal variability of the total CH_4 fluxes, a pronounced shift of maximum CH_4 emissions from early morning to nighttime hours was revealed for the diffusive flux component during August 2013 (Fig. 5 and 6 (now 6 and 7)).While maximum diffusive fluxes during July were recorded during early morning hours (approx. 3:00 to 6:00), a shift to the nighttime was observed for August (max. from 21:00 to 0:00). During September maximum fluxes shifted back to the early morning, with maximum fluxes between 0:00 and 9:00 (Fig. 6 (now 7))."

24. Table 1 – No details about these statistical tests were given in the methodology section. How were differences among chambers tested – which post hoc test was used?
    We added details about the statistical test (balanced case; Tukey HSD test), used to identify significant difference between the chamber positions to Tab. 1. The other used statistical tests/measures are indicated within the table caption. The decision on whether or not a dependency of the flux components from the different environmental variables is considered to be significant was made according to the average coefficient of determination from regression analysis of all daily data subsets. This was needed due to present seasonality in the flux data. That means that even though a dependency is indicated as being significant, it might be not true for some daily data subsets, and vice versa. The same accounts for given average NSE values, which might be higher but also lower for the different daily data subsets.
    This is, however, of minor importance regarding the accuracy of the presented flux calculation and separation algorithm, for which the relative difference between found dependencies (for daily data subsets) in flux components compared to the total flux is more important.

25. Please justify the choice of $p < 0.1$ in detecting statistical significance – the norm is to use $p < 0.01$.
    We decided to use $p<0.1$, because field study measurements are usually related to a higher uncertainty (as shown, CH4 emissions at the study site are characterized by a high spatiotemporal heterogeneity), compared to laboratory experiments, wherefore it might be advantageous to use a more sensitive testing to detect potential environmental drivers (Type I error vs. Type II error). In addition, we do not agree on the statement of a standard or norm of $p<0.01$. Different p-value (e.g. 0.05) are given in the statistical literature, without the aim to state a dogmatic standard p-value to be used (e.g. Field et al. (2000); Fisher (1956); Coehen (1992)).
    Moreover, we were rather interested in showing the relative difference and not making a statement about the (absolute) significance of our results. That means that we intended to show two things: 1.) that environmental variables seems to influence the separated flux components differently (no clear driver for erratically ebullition events but dependencies for diffusive flux components) and 2.) that trends or tendencies observed for the total CH4 flux become more clear for the diffusive CH4 flux, when separating data noise originating from the ebullition flux component.

Technical corrections

26. L80-81 – “if aiming to identify relevant environmental drivers of CH4 emissions” is grammatically incorrect. Modify as “if relevant environmental drivers of CH4 emissions are to be identified”
    Done.

27. L139 – “Exemplary” field data? Not sure about the purpose of this heading.
    We changed this heading into *“2.4 Exemplary study site”*. This now includes the site description of the study site used to gain the test data set as well as a description of the performed environmental measurements.

28. L161 – Change “installed as transect” to “installed along a transect”
    Done.

29. L175 – Fig. 3 appears below Fig. 2?
    We checked, refreshed and corrected all figure and table references made in the MS.

30. L209-210 – The phrase “smaller or larger than the upper and lower quartile 0.25 times IQR” is confusing – do you mean something like outside of the range between the upper and lower quartile 0.25 times IQR?
    Yes. To avoid confusion on this important issue, we changed the sentence to:

    “Abrupt concentration changes within the MW were identified by means of a rigid outlier test, discarding fluxes with an inherent concentration change outside of the range between the upper and lower quartile ± 0.25 times the interquartile range (IQR)”.

31. Figure 3 – This was not exactly a scatterplot of concentrations – perhaps a time series plot would be more appropriate.
    We agree and changed the figure caption to:

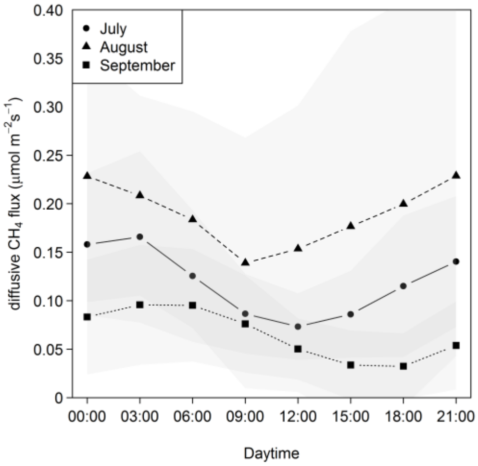
    “Time series plot of recorded concentrations (ppm) within the chamber headspace for (a) a simulated ebullition event and (b) an exemplary CH_4 measurement.”

32. Figure 5 – Why would the bars (I assume is CH4 flux) in the top graph have different colors? What do gray and black colors represent (the bars, not the pie chart)?
    Fig. 5a shows the measured total CH_4-flux as a subdivided bar diagram. This means that the grey part of the bar shows the respective ebullition flux component and the black part the diffusion flux component. Thus, it is possible to show the flux components proportions throughout the entire study period. To make this clearer we rewrote the figure caption:

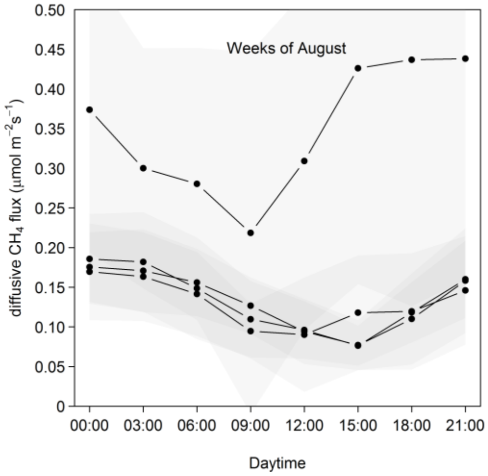
    *“**Fig. 6:** Time series of (a) total CH_4 emissions with proprotions of ebullition (grey bar) and diffusion flux components (black bar) during the study period from July until September 2013. Figure 5b and 5c show the seperated flux components ((b) ebullition and (c) diffusion) togetehr with the development of important environmental parameters, which are assumed to explain their specific dynamics ((a) water level, (b) RH and wind speed and (c) sediment (solid line) and water temperature (dashed line)). Pie charts represent the biweekly pooled diurnal cycle of measured CH_4 fluxes. Slices are applied clockwise, creating a 24-hour clock, with black and light grey slices indicating hours with CH_4 flux above and below the daily mean, respectively.”.*

33. Figure 6 – Please show the error bars for the data points. Also, change “Juli” to “July”, and “montly” to “monthly”

We refreshed the figure (see below) and changed *“Juli”* to *“July”* and *“montly”* to *“monthly”* within the figure and figure caption. We now included the standard deviation (SD) as a gray shaded area (indicating the average value ± 1SD) around the shown development of monthly averaged diffusive CH4 emissions (please see figure below).



We didn’t include this before, because we thought it might be misleading. As shown for the four weeks of August (please see figures below) and also within Fig. 5 (now 6) of the MS, the magnitude of calculated diffusive CH_4 emissions differs between the months but also the weeks and days of each month. As a result, and despite of the clear diurnal development during each single day, the SD for monthly averages of hourly diffusive CH_4 emission is rather high. This is however, a result of the temporal development in diffusive CH_4 emission during the study period and can’t be used to qualify significant differences between day and night. A high SD is shown in particular for August 2013 and mainly a result of the first week of August. This week shows substantially higher diffusive CH_4 emissions (while still evidencing a diurnal development within these high emissions!) compared to the second, third and fourth week of August.



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