

Answers to Anonymous Referee #1

General Comments

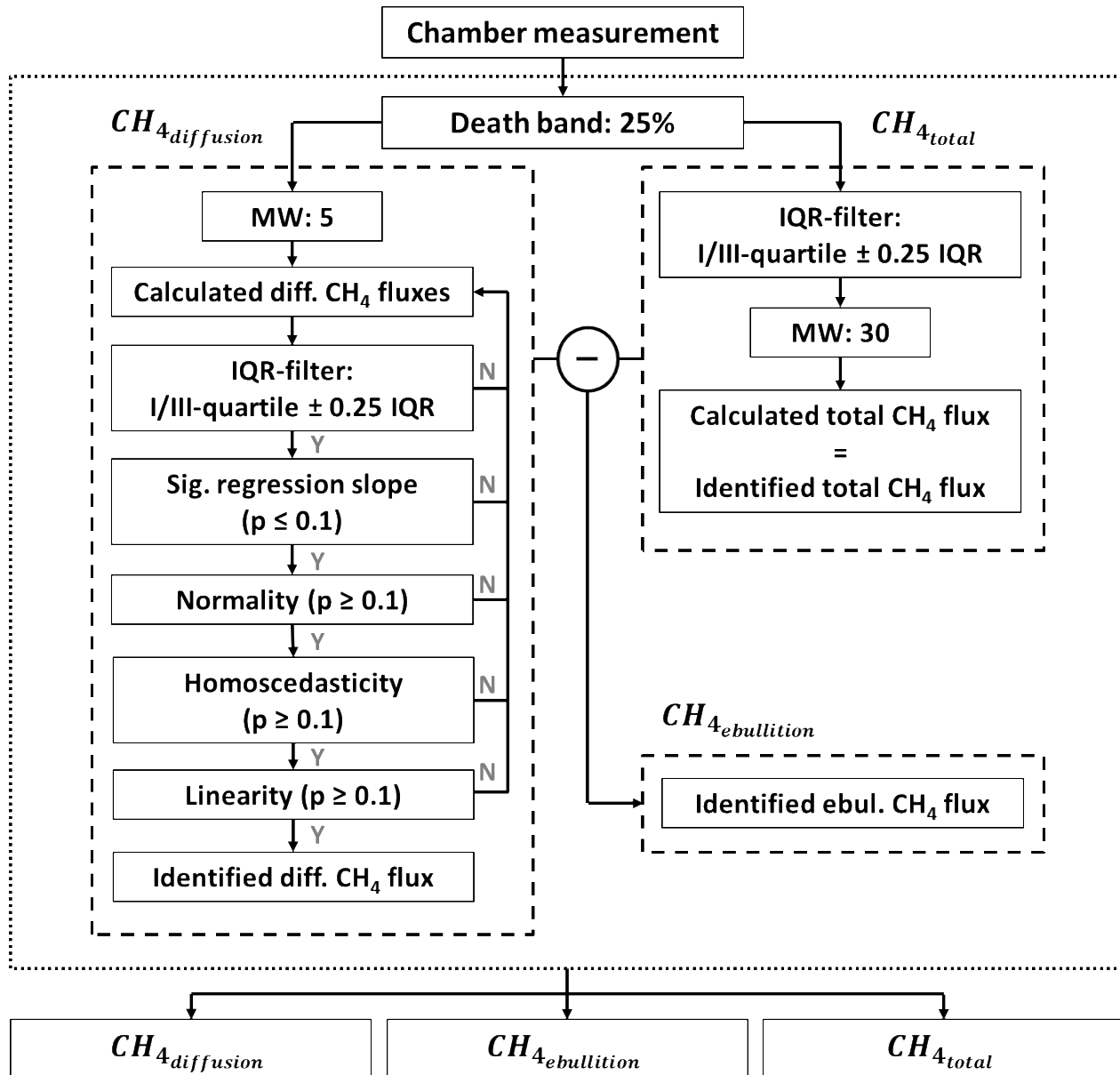
1. I find the study very interesting and useful, but I have a range of suggestions how the paper could be significantly improved. According to the aims and scope of AMT, the journal's aim is to foster scientific discussion about advances in measurement techniques and data processing methods. Therefore, in my mind, the focus of an AMT paper should be to describe a method in a way that others can easily understand it and use it for their own research. With this in mind, I would like to suggest that the authors put more emphasis on explaining the method they use. The paper is a bit short in details about the data processing and the validation of the method and puts more focus on the results from a field campaign. Those are important, but it should be clear that the method is the main point and that the field study served as a test of the method's applicability and potential. We agree and restructured and rewrote the entire section 2.2 which explains the flux calculation and separation approach in more detail now. To further support the more detailed explanation a flow chart was added to the MS as well (please see changed section 2.2 and flow chart below).

"CH₄ flux calculation and separation was performed based on an adaptation of a standardized R-script presented in detail by Hoffmann et al. (2015). Fig. 2 shows a flow chart of the flux calculation algorithm implemented in R and the principle of the performed CH₄ flux separation. To estimate the relative contribution of diffusion and ebullition to total CH₄ emissions, flux calculation was performed twice, once for the total CH₄ flux (CH_{4total}) and once for the diffusive component of the flux rate (CH_{4diffusion}), by adjusting selected user-defined parameter setups of the used R-script (Hoffmann et al. 2015). Prior to each flux calculation a death band of 25 % (user defined) was applied to the beginning of each flux measurement, thus excluding measurement artefacts triggered by the process of closing the chamber. On the remaining flux measurement data a variable moving window (MW) with a minimum size of 5 (CH_{4diffusion}; user defined) and 30 consecutive data points (CH_{4total}; user defined) was applied, generating several data subsets per flux measurement for CH_{4diffusion} and one data subset for CH_{4total}. Subsequently, CH₄ fluxes were calculated for all data subsets per flux measurement using Eq. (1), where M is the molar mass of CH₄, δv is the linear concentration change over time (t), A and V denote the basal area and chamber volume, respectively, and T and P represent the inside air temperature and air pressure. R is a constant ($8.3143 \text{ m}^3 \text{ Pa K}^{-1} \text{ mol}^{-1}$). In case of CH_{4total} the difference between the start and end CH₄ concentration of the enlarged MW (7.5 minutes) was used (δv) instead of the slope of the linear regression fit (CH_{4diffusion}). 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 (user defined) were discarded prior to calculation of CH_{4total}. In case of diffusion the resulting numerous CH₄ 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 $\pm 1.5 \text{ K}$; (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_{4diffusion} fluxes which did not meet all exclusion criteria were discarded. In case of more than one flux per measurement meeting all exclusion criteria, the CH_{4diffusion} flux with a starting CH₄ concentration being closest to the atmospheric CH₄ concentration was chosen. Finally, the

proportion of the total CH_4 emission released via ebullition was estimated by subtracting identified $CH_{4\text{diffusion}}$ from the calculated $CH_{4\text{total}}$ following Eq. (2).

$$CH_{4\text{ebullition}_n} = \sum_{i=1}^n (CH_{4\text{total}} - CH_{4\text{diffusion}}) \quad (2)$$

Since no emergent macrophytes were present below the automatic chambers, plant-mediated transport of CH_4 was assumed to be zero. The same accounts for negative estimates of CH_4 released through ebullition. The used R-script, a manual and test dataset are available at <https://zenodo.org/record/53168>.”.



- My second main suggestion is that the authors should include more details about the validation of the method and a more thorough discussion of the potential errors. The reader is, at the moment, unable to judge whether this automated data analysis will always lead to accurate results or when it might fail.

We agree and restructured and rewrote section 3 according to the suggestions made by both reviewers. The section was divided into the subsection 3.1 “Verification of the flux separation algorithm” (including results of the performed laboratory experiment and their discussion), 3.2 “Application to an exemplary field study” (including results and their discussion gained through field measurements) and 3.3 “Overall performance” (including a more thoroughly discussion of advantages and limitations/potential error sources of the presented flux separation algorithm). In addition we refer to eddy covariance measurements made at the same study site during the same period (Franz et al. (2015)), which resulted in total CH₄ fluxes being comparable in magnitude and seasonal development to fluxes measured by the chamber system and calculated by the presented data processing algorithm.

“CH₄_{total} fluxes observed by the AC system and calculated with the presented algorithm were comparable to CH₄ emissions measured during the study period by a nearby eddy covariance system (Franz et al. 2015).”

3. Also, it is unclear whether this automated data analysis yields better or worse results than manual data analysis, whereas each measurement is looked at and fitted individually.

We think that automatization and standardization is a prerequisite for scientific studies, helping to produce traceable, reproducible and comparable results/data. This is most often hard to achieve by manual data analysis which is not only time consuming (the presented data set contains more than 14.000 flux measurements), but also somehow subjective (due to individual expert knowledge and decision making). During recent studies, we compared manually calculated CO₂-fluxes with an automatic and standardized calculation tool using R (Hoffmann et al. 2015). Whereas the automatic calculation did not differ between multiple calculations using the same user defined parameter setups and data sets, the manual calculations made by different researchers were characterized by a rather high variability. The same accounts for repetitive calculations of the same data set by one researcher.

4. It is also unclear how the algorithm handles data gaps, disturbances and artifacts.

The algorithm requires consecutive concentration records during single chamber measurement. In case of missing measurements (such as during end of July and beginning of August) no gap filling was performed. Based on revealed flux component specific temporal dynamics and dependencies, empirical modelling might be included into the algorithm. In case of measurement gaps within one single measurement, the measurement is divided by the algorithm into two measurements (one before and one after the gap). However, such data gaps did not occur during this study. If the divided measurement are too short for the algorithm (smaller than the minimum moving window size) they are discarded. A number of filters try to discard disturbances and artifacts from calculated fluxes. To better address this issue we rewrote and extended section 2.2 (please see answer to 1. comment reviewer #1).

5. Would it work with other automated chamber systems as well or only with yours?

In principle the algorithm is supposed to work with all kind of automatic or manual closed chamber systems, as long as they deliver consecutive records for CH₄ concentrations and air temperature. This means that the only requirement is a proper data format (csv-file), which is used by the R-algorithm as source data to calculate/separate the CH₄ measurements into its components.

6. Finally, plant-mediated transport is important. Does your algorithm only work at sites where plant-mediated transport can be excluded or is there potential to further develop the algorithm in the future so that plant-mediated transport could be included?

To date, the presented algorithm separates only diffusion and ebullition, and will thus only work on open-water systems where the plant-mediated transport can be neglected. We, however, are

working on an approach to also include plant mediated transport into the calculation and separation algorithm. Therefore, below surface (sediment/water) measurement devices to record the CH₄ concentration are needed. Based on this, diffusive CH₄ emissions could be calculated via a gradient approach, while the chamber still measures the total CH₄ flux. By subtracting the diffusive from the total flux the sum of plant-mediated and ebullition flux could theoretically be derived. Both flux components could be subsequently separated, using the presented approach, assuming plant-mediated transport being more continuous than ebullition. This however is not implemented yet, since we still work on the measurement device. The advantage however, is clear since we would be thus able to measure all three flux components on the same measurement plot, without a bias due to spatial heterogeneity.

Specific Comments

7. p. 2, line 43 The validation of the method in the lab should be mentioned in the Abstract.

We agree and added the validation using the laboratory experiment to the abstract.

“The algorithm was validated by performing a laboratory experiment and tested using flux measurement data (July to September 2013) from a former fen grassland site, which converted into a shallow lake as a result of rewetting.”

8. p.3, line 67 I would mention here that CH₄ is a greenhouse gas, which explains the relevance of CH₄ flux measurements in the very beginning of the paper.

We agree and changed the first sentence.

“Wetlands and freshwaters are among the main sources for methane (CH₄), which is one of the major greenhouse gases (Dengel et al. 2013; Bastviken et al. 2011; IPCC 2013).”

9. p. 5, line 140 I am wondering if the order of the subsections could be changed. The way it is structured, most emphasis is on the field measurements, whereas I have the feeling that the focus should be on the algorithm and its validation, and then on the field measurements. Not sure if it is possible to change the order though, because obviously information about the chamber system is needed before the algorithm can be introduced.

We agree. To better emphasize the calculation algorithm and its validations using the laboratory experiment and field study respectively, we changed the order of subsections.

- 2.1 Automatic chamber system
- 2.2 Flux calculation and separation algorithm
- 2.3 Verification of applied flux separation algorithm
- 2.4 Exemplary field study

Since a closed chamber system is the general basis for the calculation algorithm, details of the used automatic chamber system are mentioned at the beginning of the Material and Method section. After this, the flux calculation and separation algorithm as the actual focus of the paper is now mentioned, followed by its validation/verification in the lab. The subsection “*Ancillary field measurements*” was merged with “*Study site*” to the more general heading “*2.4 Exemplary field study*”.

10. p. 5, line 153 and following. Do you have references for the composition of the vegetation? Has the study site been described previously?

Details given about the vegetation are based on a monitoring performed at the study site during our project, wherefore no reference was given in the MS. However, the Study site as well as the composition of the vegetation was also described by Franz et al. (2015; Biogeosciences, doi:

10.5194/bg-13-3051-2016), Steffenhagen et al. (2012) and Hahn-Schöfl et al. (2011). We therefore added these references to the MS as well.

11. p.6, line 165 The chamber system seems to be quite sophisticated. Is it a commercial system or did you develop it? Is it described elsewhere? If yes, please add references.

The chamber system is a non-commercial system, developed by our working group. A comparable system was used for CO₂ measurements on an agricultural landscape within the study of Hoffmann et al. (2016; Biogeosciences Discuss., doi:10.5194/bg-2016-332, 2016). This study was not referred to, due to differences not only in the measured trace gas, but also regarding the used analyzer and measured ecosystem. Moreover, the article is only published as a discussion paper yet.

12. p.6, line 175 Explain overcompensation or maybe show it in the Figure.

We added a more detailed explanation for overcompensation to section 2.1.

“However, due to the large chamber volume, complete mixture of the chamber headspace took up to 30 seconds. As a result of this, most peaks due to ebullition events were directly followed by a smaller decrease in measured CH₄ concentration. This signal indicates a short term overestimation of the ebullition event (peak), which was compensated after the chamber headspace, was mixed proper (decrease), a phenomenon further on referred to as overcompensation (Fig. 3).”

In addition we now refer to overcompensation within the figure caption to Fig. 3.

“Negative ΔCH_4 values indicate an overcompensation due to (temporally) insufficient headspace mixing.”

13. Fig. 3 is referred to here, but Fig. 2 hasn't been mentioned yet. I suggest to adjust the order of the Figures.

We corrected the order of Figures throughout the MS.

14. p.6, line 179 Several questions remain open.

1) Over which time period were the measurements performed? This is mentioned in the abstract, but the information should be included here as well.

We included this information now as well in section 2.4.

“Ecosystem CH₄ exchange was measured from beginning of July to end of September 2013 at a flooded former fen grassland site, located within the Peene river valley in Mecklenburg-Western Pomerania, northeast Germany (53°52'N, 12°52'E).”

2) Were the chambers vented after each 10 min measurement?

Yes, the chambers were vented (using the internal fan) during the entire 50 min between two measurements of the same chamber. To better address this important information we added the following sentence to “2.1 Automatic chamber system”:

“Each chamber was vented using the internal fan throughout the entire 50 min between two measurements at the same chamber position.”

3) How much time passed when the system switched to the next chamber? The reason for this question is the following: If you pump the analyzed air back into the chamber, you will have contamination every time you switch (i.e. air from chamber I is still in the analyzer, you switch to chamber II, air from chamber I will thus be returned to chamber II).

We think this is a really important issue and are glad for this valuable remark. In general, the tubes connected to the sensor were vented for 3 minutes between two measurements:

- 1 minute before switching to the next chamber using the air of the reopened chamber which was measured for 10 minutes.
- 2 minutes after switching to the next chamber, using air of the open chamber, to be measured next. After this 2 minutes the chamber will be automatically deployed on the frame.

Since the 1 minute before switching will be biased by the measured performed directly before, the tubes were vented using unbiased air for 2 minutes. We therefore added the following sentence to “2.1 Automatic chamber system”:

“When switching from one chamber to another, the tubes were vented for two minutes using the air of the non-deployed chamber to be measured next.”

15. p. 6, line 181 What was the water depth of the studied system?

The water depth is shown in Fig. 5 (now 6), and ranged from 22 to 35 cm throughout the study period. In addition we added the limits to “3.3 Overall performance” as follows:

“This is in particular the case, when measuring at wetland ecosystem with a varying water level, such as at the exemplary study site (22 to 35 cm).”

16. You say that temperatures were recorded at different (i.e. multiple?) water depths, but the only water depth you give is 5 cm above the sediment surface?

We stated that we measured temperature in different water and soil depths (*“Temperatures were recorded in different water (5 cm above sediment surface) and sediment depths (2 cm, 5 cm, and 10 cm below the sediment-water interface), using thermocouples (T107, Campbell Scientific).”*). This seems to be misleading, since we actually meant that we measured temperatures in four depths underneath the chambers (one water depth (5 cm) and three different sediment depths). We therefore changed the sentence to:

“Temperatures were recorded in the water (5 cm above sediment surface) and different sediment depths (2 cm, 5 cm, and 10 cm below the sediment-water interface), using thermocouples (T107, Campbell Scientific).”

17. p. 6, line 191

I have a general question regarding the data analysis. Shouldn't you discard data after each ebullition event? The reason is the following: Let's say the chamber is closed and you have diffusive emissions in the beginning. They are driven by the gradient between water CH₄ concentration and chamber CH₄ concentration. After an ebullition event, the CH₄ concentration in the chamber is enhanced over the normal boundary layer concentration, therefore, you will have reduced diffusive emission. Isn't that a systematic error?

In general, every new concentration record during one chamber measurements will be influenced by the concentration records before, as long as these records alter the concentration gradient. This is a well-known (e.g. Hoffmann et al. 2015), general limitation of closed chamber measurement system, and most often handled by reducing the measurement length or enhancing the chamber volume, which minimize the change within the concentration gradient. Regarding the specific measurement site we assume that the influence, however, will be negligible. The reason therefore is the rather high CH₄ concentration within the sediment as measured by Hahn-Schöfl et al. (2011). They reported CH₄ concentrations within organic sediment probes taken at the same site in 2007 for an incubation experiment ranging up to 500.000 ppm. As a result, even during bigger ebullition events, which might enhance the chamber air CH₄-Concentration up to 30 ppm, the actual gradient between sediment and atmospheric CH₄-concentration remains high. However, the

diffusion from sediment to water to atmosphere might be delayed, resulting in reduced diffusive CH₄ emissions after bigger ebullition events

18. Can you estimate the magnitude of this error?

The magnitude of this error can be estimated by calculating the diffusive flux twice, using the presented tool and a variation within the user defined parameters: once disregarding the last e.g. 50% of each measurement, and once disregarding the first e.g. 50% of each measurement during the calculation. By comparing the resulting fluxes of both calculations, a systematic error and its magnitude would occur in terms of significantly lower fluxes during the second calculation. However, a significant difference between these two calculations was not obtained for the presented data set.

19. p. 6, line 192 Even though the script has been described elsewhere, I'd suggest you give a brief summary of the data processing nevertheless. Otherwise it will be hard for the reader to follow.

We rewrote section 2.2 and added a flow chart (see flow chart above) to further explain the developed algorithm

20. p.7, line 199 You should list which values were used for the user-defined parameters (maybe as a Table)

We agree and added user defined parameters and used values to section 2.2 of the MS and to flow-chart respectively.

21. p. 7, line 207 I think it would be good to include a flow chart to support your explanation of how the algorithm works. It would make it easier to follow. In general, the description of how the algorithm works could be a bit more extensive and possibly be supported by graphics (e.g. flow chart, example data)

We added a flow-chart to the MS (please see answer to 1. Comment reviewer #1) explaining how the flux calculation and separation algorithm is working.

22. p.7, line 216

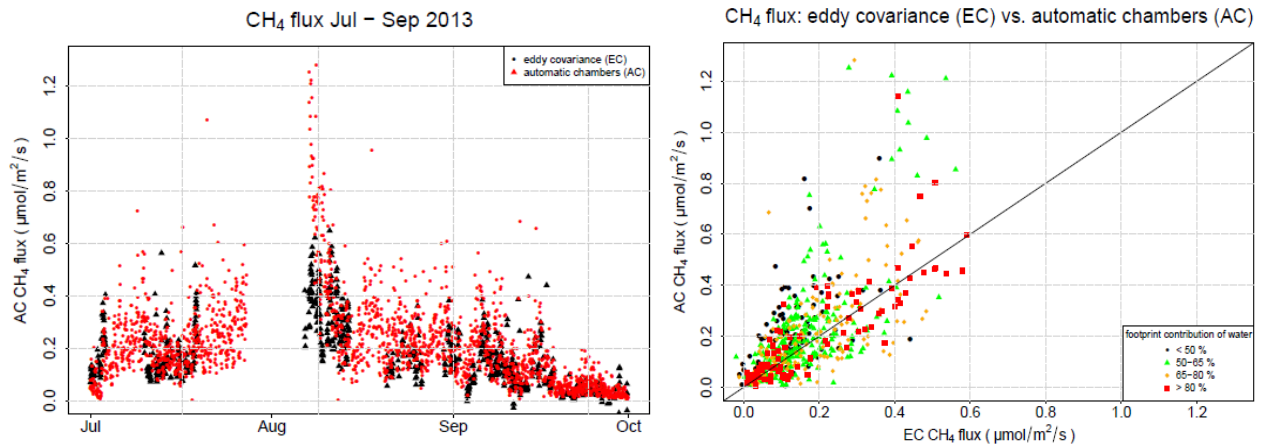
“To exclude measurement artifacts triggered by the process of closing...” This information should appear earlier in the Section, you should describe first which data is discarded and then how fluxes are derived from the remaining data.

We rewrote section 2.2. As a result the information about discarded concentration records is appearing earlier now.

23. p. 7, line 222

This is a nice way to validate the algorithm for ebullition events. Was the algorithm also somehow verified for the diffusive flux? Maybe previously? This would be an important information.

No direct verification of the presented algorithm to calculate diffusive CH₄ emissions was made. However, the total CH₄-flux measured at the exemplary study site was verified (see figure below) by measurements of a nearby (>10 m distance to chambers) eddy covariance system performed during the same period. Both measurement devices (eddy covariance and automatic chambers) yielded in comparable results regarding the dynamics and magnitude of obtained total CH₄-emissions. Thus, ebullition (lab experiment) and total CH₄-emission (eddy covariance measurements) were verified. To better address this issue we added the following sentence to the MS:



“CH₄_{total} fluxes observed by the AC system and calculated with the presented algorithm were comparable to CH₄ emissions measured during the study period by a nearby eddy covariance system (Franz et al. 2015).”

24. p.8, line 235

At present, the Results and Discussion Section is not very well structured and it is easy to mix up the different experiments. It has to be made clear that what you did was a two-step approach: First you validated the algorithm by testing it under lab conditions, second you applied the algorithm to field data. The reader could be under the impression that you’re validating your method with field data, but of course it is the lab measurements that support your theory. The field data is to show how useful your algorithm is for the quantification and interpretation of fluxes. Therefore, I would like to suggest to structure the Results and Discussion Section into 3.1 Validation of the algorithm through laboratory measurements, 3.2 Application of the algorithm to field data, 3.3 Overall performance of the algorithm. That would help the reader distinguish the different experiments.

We restructured and rewrote section 3. and subdivided it into:

- 3.1 Verification of the flux separation algorithm
- 3.2 Application to an exemplary field study
- 3.3 Overall performance

25. I think that the lab measurements need more discussion – it is the evidence that your method works for ebullition events. But you should also discuss potential errors.

We agree and extended this section, adding potential error sources which might occur during the lab experiments, but also problems which might occur when applying the algorithm under field conditions.

26. In 3.3 you could evaluate the overall performance of the algorithm, the advantages it has, but also include a discussion of potential errors. In what I suggested to be Section 3.3, I would recommend that you also include a short outlook as to which further developments the algorithm requires and what its potential is to be used as a general tool for automated chamber measurements (kind of what you’re doing in your conclusions).

We extended the discussion by mention advantages compared to other direct or indirect flux separation approaches (“3.3 Overall performances”). Furthermore, disadvantages/potential error sources of the presented algorithm are more thoroughly discussed now (“3.3 Overall performance”).

“Compared to direct measurements of diffusion or ebullition, as reported by e.g. Bastviken et al. (2010), the presented calculation algorithm in combination with the used AC system, features two major advantages. On the one hand it allows deriving the ebullition and diffusion flux components based on the same measurement and for the same spatial entity, which prevents an interfering influence of spatial heterogeneity on observed flux components. This is not the case for flux separation based on a combination of different measurement devices, such as automatic chambers and bubble traps, which need a sufficient number of repetitions and degree in data aggregation to reduce the bias, emerging from the spatiotemporal heterogeneity of erratically occurring ebullition events. On the other hand, the solely data processing based flux separations approach allows for an application, when the use of direct measurement systems for either ebullition (gas traps, funnels) or diffusion (bubble shields) might be limited. This is in particular the case, when measuring at wetland ecosystem with a varying water level, such as at the exemplary study site (22 to 35 cm). During the summer month 2009 and 2016 the water level dropped substantially, being either next to or even below the surface (data not shown). This limited the theoretical use of bubble traps and shields, despite of potential ebullition from the water saturated sediment, to periods with a sufficient water level, resulting in larger measurement gaps. Similar to that, parallel measurements of different trace gases (e.g., CO₂ and CH₄) are not affected by the presented flux separation algorithm.

However, flux separation using the presented algorithm might be biased by steady ebullition of micro bubbles and frequently occurring strong ebullition events. Steady ebullition of micro bubbles, results in an overestimation of CH₄_{diffusion} and underestimation of CH₄_{ebullition}, an effect, which might be reduced by enhancing the measurement frequency and thus the sensitivity of the variable IQR-filter. Frequently occurring strong ebullition events, however, might disable the calculation of CH₄_{diffusion}, which hampers flux separation for the corresponding measurement. Out of 14.828 valid automatic chamber measurements, the algorithm failed to calculate CH₄_{diffusion} during 170 measurements. This equals 1.15 % of all measurements. Taken into account that the presented measurement site is characterized by rather large CH₄ emissions (Franz et al. 2015) and frequently occurring ebullition events, this limitation seems to be negligible.

Compared to other data processing based approaches for CH₄ flux separation (e.g. Goodrich et al. 2011; Miller and Oremland 1988), the integration of the ebullition component into measurements rather than the calculation of single ebullition events ensure a reliable flux separation, despite of potential measurement artefacts such as overcompensation or incomplete ebullition records. 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 CH₄ 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 is stated.”

27. Do you think it is possible to integrate plant-mediated fluxes in the future or is your algorithm only applicable in systems where these can be neglected?

As stated in the MS, to date the algorithm is only applicable to open water areas of wetland ecosystems, where plant-mediated transport can be neglected. However, as mentioned above (general comments), we added an outlook to section “4. Conclusions”, which states that by using additional measurement devices to obtain CH₄ concentrations in the sediment and overlying water column, an extension of the presented algorithm to also include a separation of plant-mediated transport, might be theoretically possible.

“During future studies, the possibility to implement the separation of CH₄ released through plant mediated transport into the presented algorithm should be addressed. This might be possible by

complete chamber CH₄ concentration measurements with CH₄ concentrations measured in different water and/or sediment depth. This might allow to directly deriving CH₄diffusion fluxes, whereas the remaining plant mediated transport and ebullition flux components could be separated using the algorithm.”

28. You should also answer the question under which circumstances the performance of the algorithm might be poor, and which errors can be expected. Could just anyone who measured a chamber time series use your algorithm and get reliable results?

We added sentences regarding the reliability of the presented approach to section 3.1 (micro bubbles) and 3.3 (frequent strong ebullition events). In principle the algorithm is supposed to work with all kind of automatic or manual closed chamber systems, as long as these systems deliver consecutive records for CH₄ concentrations and air temperature. The reliability however, is not only a question of the presented flux calculation and separation algorithm, but also of the underlying measurement data. The quality of the data depends on the used chamber designs (e.g. airtight sealing, ventilated, pressure equilibration), measurement settings (e.g. frequency of concentration records) as well as the measured ecosystem (does steady ebullition through micro bubbles occur?). To better address this important issue we added the following to “3.3 Overall performance”.

“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 CH₄ 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.”

29. Do the flux estimates derived with your algorithm have a robust error propagation estimation?

Errors can be only calculated for the diffusive flux component, based on the underlying linear regression fit. Since the total flux is calculated using the difference between the start and end CH₄ concentration, an error estimation for single measurements/fluxes is not possible. This also prevents an error calculation for the ebullition flux component of a measurement. Thus errors are not given for all flux components of single measurements.

30. p.9, lines 267-270 Your reasoning is: In the literature, it has been shown that CH₄ production is related to temperature. Therefore, our measurements show a pattern that relates CH₄ to temperature. But actually the reasoning is the other way around: You find in your data that CH₄ is related to temperature. This is in accordance with the literature.

We agree and rewrote this paragraph.

“Measured total CH₄ emissions showed distinct seasonal patterns following the temperature regime at 10 cm sediment depth. This is in accordance with Christensen et al. (2005) and Bastviken et al. (2004), who showed that biochemical processes driving CH₄ production are closely related to temperature regimes, which determine the CH₄ production within the sediment.”

31. p.9 lines 282-286 You do have the data to support this theory (you mention that you measured the water temperature at different depths). I suggest to use your data to prove your theory.

We stated that we measured temperature in different water and soil depths (“Temperatures were recorded in different water (5 cm above sediment surface) and sediment depths (2 cm, 5 cm, and 10 cm below the sediment-water interface), using thermocouples (T107, Campbell Scientific).”). This seems to be misleading, since we actually meant that we measured temperatures in four depths underneath the chambers (one water depth (5 cm) and three different sediment depths). We therefore changed the sentence to:

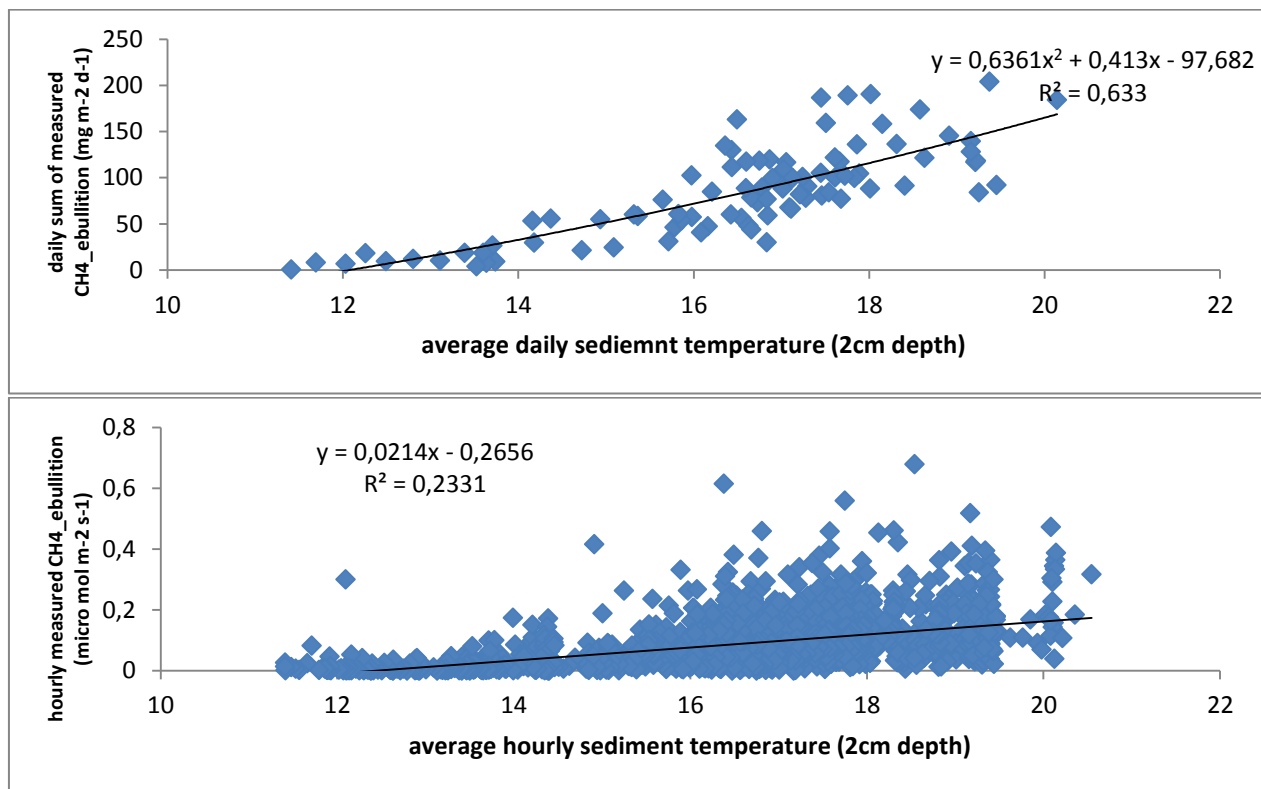
“Temperatures were recorded in the water (5 cm above sediment surface) and different sediment depths (2 cm, 5 cm, and 10 cm below the sediment-water interface), using thermocouples (T107, Campbell Scientific).”.

32. p. 10, line 297 What exactly is the correlation between temperature and ebullition fluxes? I'd suggest to either give a correlation coefficient here or to include a Figure.

We agree and added the coefficients of determination to the MS.

“This is confirmed by a distinct correlation between daily mean sediment temperatures and corresponding sums of measured ebullition fluxes (r^2 : 2 cm = 0.63; 5 cm = 0.63; 10 cm = 0.62).”

The figures show the distinct correlation between the average daily sediment temperature (2 cm depth) and daily sums of calculated ebullition fluxes (upper figure) and the correlation of hourly sediment temperatures with ebullition fluxes calculated for single measurements (figure below). The difference in r^2 between both correlations confirms the statement that ebullition events occur erratically and that “periods characterized by more pronounced ebullition seemed to roughly follow the sediment temperature-driven CH_4 production within the sediment as e.g. reported by Bastviken et al. 2004 (Fig. 5 (now 6)).” as stated within the MS.

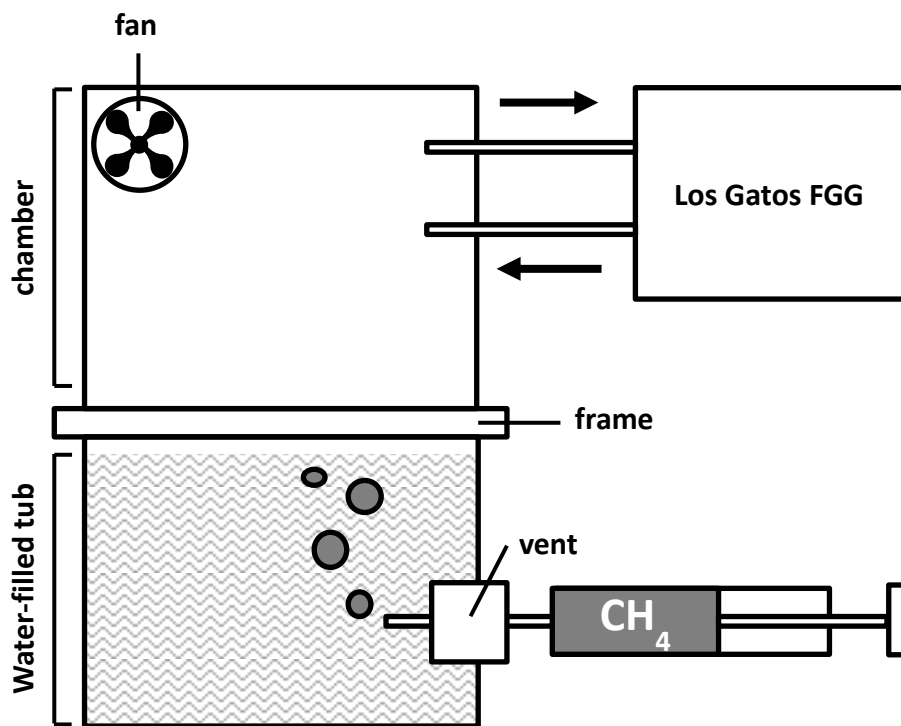


33. p.10, line 308 Does the contribution of ebullition to the total flux (in %) also exhibit a diurnal pattern?

Yes. However, as shown in Fig. 5b (now 6b), ebullition itself did not show any clear systematic or diurnal trend. The contribution of ebullition (in %) to total (hourly) CH_4 emissions only shows diurnal patterns, because of diffusive CH_4 fluxes (evidencing diurnal patterns) being subtracted from the total CH_4 flux. Hence, the diurnal trend in the contribution (in %) of ebullition to total CH_4 fluxes is solely due to dynamics found for the diffusive flux component. Absolute ebullition fluxes do not show any diurnal trend.

34. Fig. 2 This Figure would benefit from annotations (e.g. the fan, the chamber, water tub). “Injections of gaseous mixture amounted to ...” - this information is not relevant in the caption and is already given in the text.

We agree and added annotations for the fan, the chamber, the water-filled tub, the sealed frame and vent (see below). The sentence “*Injections of gaseous mixture amounted to ...*” was removed from the figure caption.

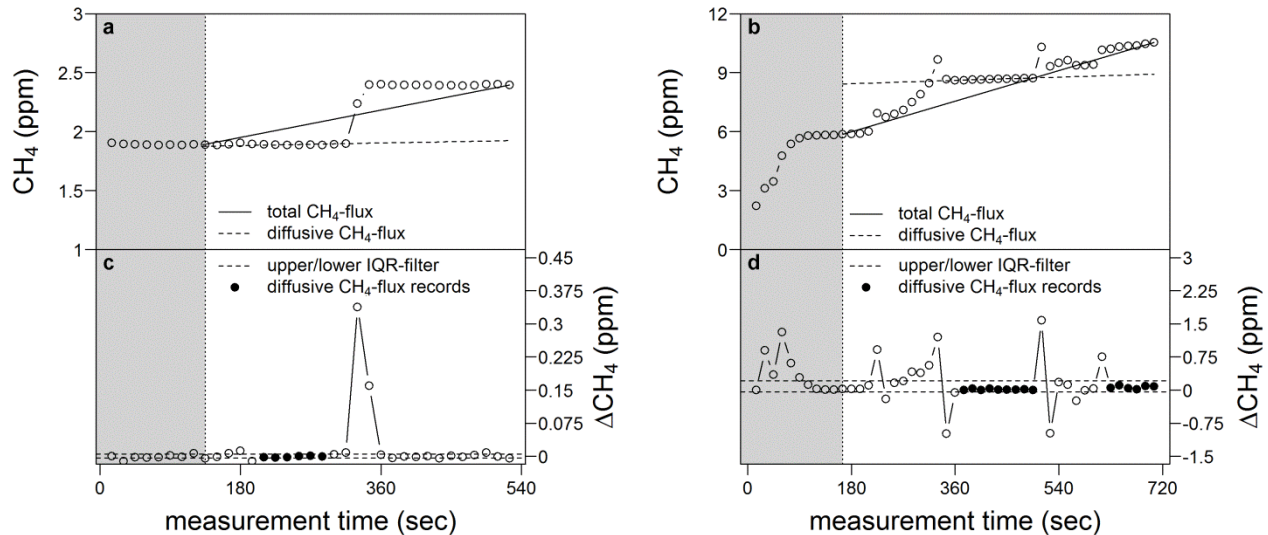


35. Fig. 4 This Figure is not very readable and very complex. To make it easier for the reader to understand the Figure, I suggest the following changes: Data points should be bigger, it is almost impossible to distinguish open and black circles. Axis labels should be bigger. The Figure needs a legend that allows the reader to see what the dashed/solid lines and open/black circles denote without having to read the caption. At the same time, if this legend is included, you can remove the extensive and somewhat complicated descriptions of dashed/solid lines, open/black circles in the caption. Why was no death band applied in a and c?

We agree and enhanced the size of the data points and axis labels. In addition we included a legend explaining the dashed and solid line in Fig. 3a/3b and the filled dots and dashed lines in Fig. 3c/3d. We also shortened the figure caption. Furthermore, we included the death band and the data points removed due to the death band to a and c as well (please see figure below).

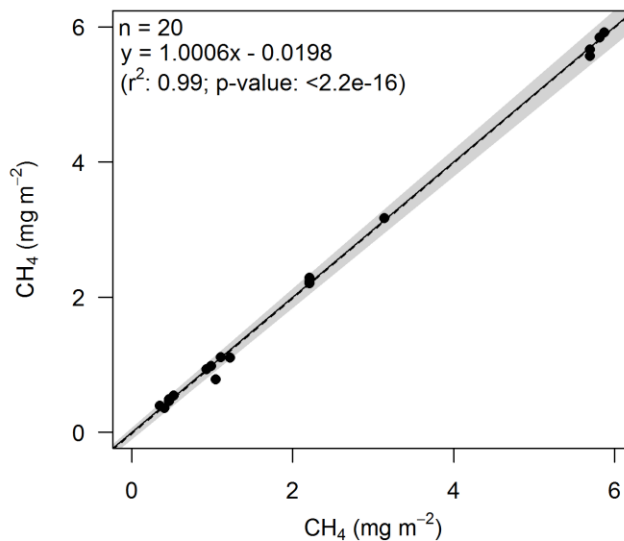
” Time series plot of recorded concentrations (ppm) within the chamber headspace for (a) a simulated ebullition event and (b) an exemplary field study CH_4 measurement. Time spans dominated by diffusive CH_4 release are marked by (c-d) black dots, enclosed by the 25 % and 75 % quantiles ± 0.25 IQR of obtained concentration changes, shown as black dashed lines. Unfilled dots outside the dashed lines display ebullition events (see also Goodrich et al. 2011; Miller and

Oremland 1988). Gray shaded areas indicate the applied deathband at the beginning of each measurement (25%). Negative ΔCH_4 values indicate a overcompensation due to (temporally) insufficient headspace mixing.”



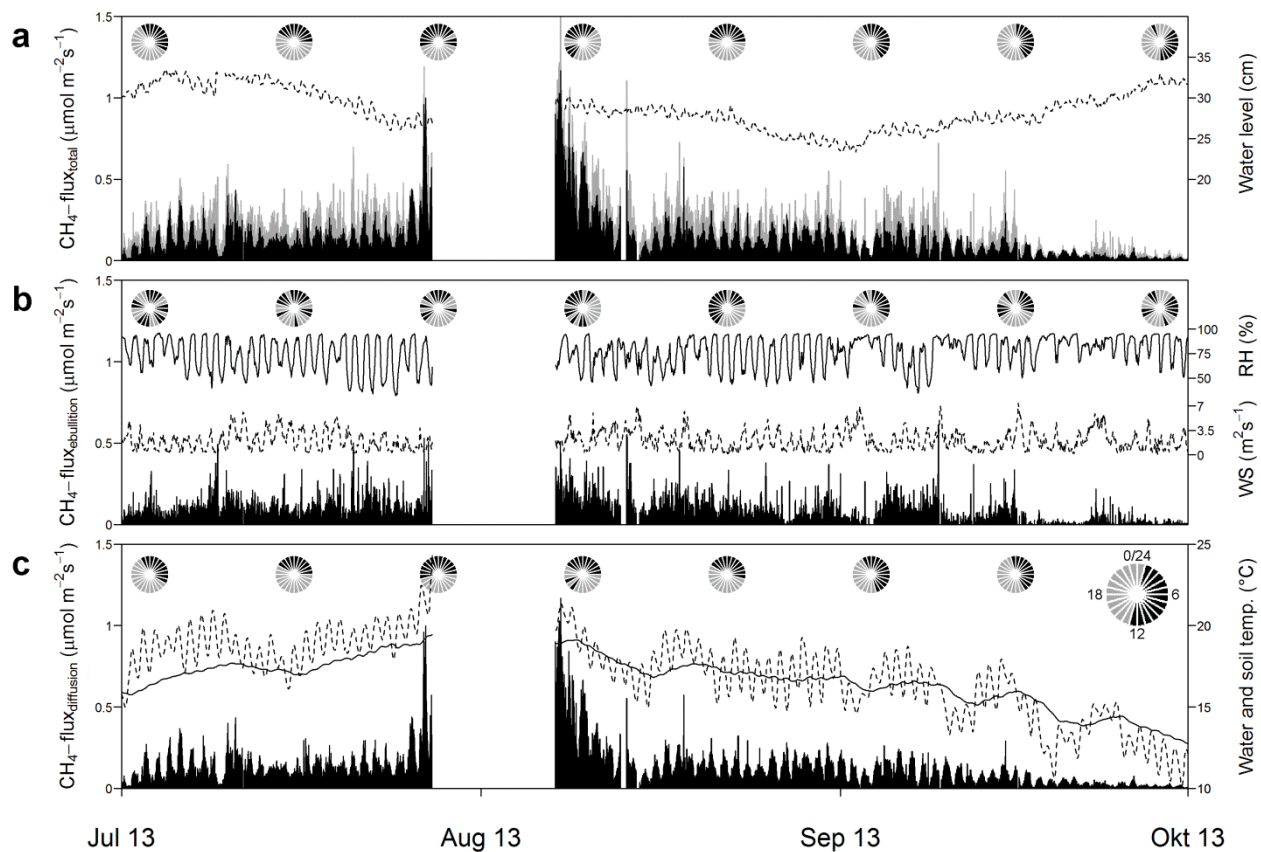
36. Fig. 3 The data points should have error bars. The axis limit could be reduced to 7. If r^2 is shown, I'd suggest to also show p and the calculated slope and intercept of the regression line.

We reduced the axis limits to 6 and added p -value, slope and intercept (please see figure below). Errors can be only calculated for the diffusive flux component, but not the total flux, which is calculated using the difference between the start and end CH₄ concentration. This prevents an error calculation for single ebullition events. As a result of this no error bars can be given for calculated ebullition events within this figure.



37. Fig. 5 This is a very interesting way to present your data. However, similar to Fig. 3, the Figure is very complex and not easily readable. I would like to suggest bigger labels, and a legend like I said in my comment above. A general question, does the bottom slice of the pie (i.e. 6 o'clock on a normal clock) correspond to 12 o'clock noon? If this is correct, then maybe it is good to warn the reader that what he normally perceives as 6 o'clock is not 6 o'clock in this Figure. I think it would be a good idea to have an "example clock-pie" with the actual hours (Let's say, 0:00, 6:00, 12:00, 18:00) next to the Figure so that it is easier to understand the clock-concept at first glance, otherwise the clock-concept might be a bit misleading.

We agree and enhanced the size of all labels and the axis titles. We decided to not include a legend, because the figure is already quiet complex. Yes, the bottom slice of the pie (i.e. 6 o'clock on a normal clock) corresponds to 12 o'clock noon. We really appreciate the remark and included a bigger slice at the top left of figure c, were the concept of the 24-hours-clock is shown now (please see figure below).



Technical corrections

38. p. 2, line 46 change to "given in the literature"

Done.

39. p.3, line 77 What does "at all scales" refer to?

We removed this part of the sentence. It was intended to make clear that ebullition events occur erratically during short (seconds to minutes) but also longer periods (hours to days or even more), as also stated by Anthony and Anthony (2013) who wrote that "*ebullition is episodic*" with "*frequencies of several minutes to weeks depending on the seep type, atmospheric pressure*

dynamic, and season of year” and that the size of bubbles emitted to the atmosphere during a ebullition event is different.

40. p.5, line 148 change to “in the beginning”

Done.

41. p.5, line 150 change to “were reported”

Done.

42. p.5, line 156 “below the chambers”

Done.

43. p.7, line 203 “outlier”

Done.

44. p. 7, line 222 I am unsure about “reasonable controlled conditions”. I’d suggest to delete “reasonable”?

Done.

45. p. 8, line 232 change to “were calculated”

Done.

46. p. 8, line 262 “explanatory approaches could be addressed” - I think the wording needs to be changed here.

We removed this sentence from the MS due to made changes within section 3.

47. p. 9, line 286 “This dynamics are ...” should be changed to “These dynamics are”

Done.

48. p. 9, line 291 daytime and nighttime are sometimes written as day time and night time throughout the text (here it is just most obvious because there are two different versions in the same sentence). Please check the article for consistent spelling of those terms.

Changed to “*daytime*” and “*nighttime*” throughout the entire MS.

49. p.13, line 393 I think the title of that publication should be “Automated modeling of ecosystem CO₂ fluxes based on periodic closed chamber measurements: ...”

We corrected the reference title.

References (not in the MS):

- Anthony and Anthony, K. M. W., Anthony, P.: Constraining spatial variability of methane ebullition seeps in thermokarst lakes using point process models. *Journal of Geophysical Research: Biogeosciences* 118, 1015-1034, 2013.
- Cohen , J.: A power primer. *Psychological Bulletin* 112(1), 155-159, 1992.
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- Fisher, R. A.: *Statistical methods and statistical inference*. 504p., 1956.
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