

**Answers to anonymous referee # 1:**

Review of “Technical Note: A simple calculation algorithm to separate high-resolution CH<sub>4</sub> flux measurements into ebullition and diffusion-derived components”. The study deals with CH<sub>4</sub> emissions from a flooded fen grassland and aims at providing a simple algorithm to allow the separation of total CH<sub>4</sub> emissions measured by automated chambers into diffusive fluxes and ebullition (according to the title and the sentence P12926 L28 to P12927L3). The algorithm is actually given in another paper (Hoffman et al., 2015) and the changes are not explicitly stated so it is difficult for the reviewer to evaluate what is new in this algorithm.

The article “Automated modeling of ecosystem CO<sub>2</sub> fluxes based on closed chamber measurements: A standardized conceptual and practical approach” from Hoffmann et al. (2015) deals with Flux calculation and modelling of CO<sub>2</sub> fluxes not CH<sub>4</sub> fluxes. Both fluxes differ regarding production as well as transport/exchange processes, wherefore differences in flux calculation and evaluation are inherent. In the current paper, a substantial extension of the basic algorithm is thus presented for CH<sub>4</sub> flux calculation. The implemented changes in user-defined parameter setups are given in section 2.2. The extended algorithm allows on the one hand for CH<sub>4</sub> flux calculation, but on the other hand also introduces a new variable filter (IQR-criteria) for separating high-resolution CH<sub>4</sub> flux data into diffusion and ebullition components. As this is the main topic of this MS, it was submitted as a technical note, presenting this substantially changed and extended programming code. Changes within the R-script in terms of user-defined parameter setups are given on P 12929-12930 (i.e. death band of 25%, starting variable moving window size of 5 consecutive points, diminishing of negative CH<sub>4</sub> fluxes). The innovative nature of the presented approach is moreover stated in P 12926-12927 “Based on high-resolution CH<sub>4</sub> flux data from an AC measurement system, this study presents a new calculation algorithm for separating open-water CH<sub>4</sub> fluxes into its ebullition and diffusion-derived components based on ebullition-related sudden concentration changes during chamber closure. A variable ebullition filter is applied, using the lower and upper quartile and the interquartile range (IQR).”

The introduction is imprecise in considering related work on controlling factors of ebullition and diffusion and the methods for their determination (see detail comments). In addition, there is no distinction between results obtained in peatland and results obtained in inland water although the

controlling factors and techniques are not the same. The differences should be stressed since the studied ecosystem is a flooded peatland.

In our study, the measurement site acts as an example, based on which the presented flux separation approach is introduced, tested and results are shown. The data set itself (except for differences between the three derived flux components (total, diffusion and ebullition) is not aimed to be subject of a detailed content-related analysis (please see answers to detailed comments) since this is beyond the scope of a purely methodically focused “Technical note”. The differences in controlling variables/factors of CH<sub>4</sub> production and in measurement techniques between inland waters and peatlands mentioned by the reviewer do not affect the reliability of the presented flux separation approach; therefore no distinction was made between different ecosystems. On the one hand, the presented flux separation approach is based on and thus only applicable to a high-resolution closed chamber measurement technique, hence, other measurement techniques are not discussed in this MS. On the other hand, differences in CH<sub>4</sub> production may influence the magnitude of the obtained flux components, but not the effectiveness of the presented flux separation approach itself. However, as addressed by reviewer 2, not only the presented data processing approach, but also the presented measurement technique might be also used for peatland measurements of methane emissions after some simple modification (e.g. implementation of solid frames to assure airtight closure in case of water level below soil surface).

In the results and discussion section, the results from the table 1 & figure 3, 5 are very poorly described.

In accordance with the BG authors guide, which suggests avoiding redundant text, detailed descriptions for Tab. 1 as well as Fig. 3 and 5 are given within the table/figure captions. The content of Tab. 1 is additionally described from P12 L25 on. Fig. 3 belongs to the method section and is referred to there (P12928 L23), used to support the explanation of the IQR-criterion within flux separation approach (P12929 L21-25).

According to discussion, emissions follow a two different diurnal pattern in spring and summer. Figures showing these patterns are required since it is not visible on the Figure 5. The authors do not illustrate the dependency of the emissions with temperature although it is said that relationships exist.

The seasonal change in diurnal patterns of CH<sub>4</sub> fluxes is displayed in the pie charts within Fig. 5. Black slices indicate high, gray slices low fluxes, with each pie chart representing a 24h-clock. For simplification, the pie charts are only shown for biweekly pooled data. Hence, a change in patterns of displayed pie charts over time indicates the mentioned shift in diurnal variability. In accordance with reviewer 2, Fig.5 was changed to improve the understanding of this complex graph. Ebullition fluxes are now displayed in gray, whereas diffusive fluxes are black. In addition to the implemented changes, Fig. 6 was added to the MS, showing the shift in diurnal variability on the basis of monthly pooled data. The dependency of emissions on temperature (seasonal dependency) is indirectly illustrated within the time-series plot (Fig. 5) and also shown (daily dependency) within Tab. 1. To improve the understanding of Tab. 1, table caption was rewritten.

In this very short discussion, there is a lot of confusion in basic concepts of limnology, methane dynamics (See detail comments) and therefore some hypothesis are probably not evaluated correctly and some others are ignored. For instance, the effect of water level variations on the diurnal patterns was not evaluated although the water level is known to be a major controlling factor of ebullition. A careful visual observation of figure 5 suggests that ebullition and water level are anticorrelated. The atmospheric pressure changes were not evaluated neither.

The influence of the water level on ebullition due to e.g. hydrostatic pressure was mentioned on P12934 L5 (now changed due to comment by reviewer 2), however, the derived ebullition related flux components did not follow the water level. The authors disagree with the suggested negative correlation between the calculated ebullition fluxes and the water level measurements, since this is not confirmed by Fig. 5. Please note that the ebullition flux is shown in Fig. 5b not Fig. 5a (Fig. 5a (water level; dashed line) and Fig. 5b (ebullition flux; bar graph)). Additionally, the water level differed by less than 10 cm (from ~24 to ~34 cm) throughout the study period and by less than 1 cm throughout the day. Hence, an influence of the sub-daily water level variation on the obtained diurnal pattern is not likely. An analysis of water level and atmospheric pressure dependencies of the calculated daily CH<sub>4</sub> emissions was added to Tab. 1. Fig. 5 and related caption were changed (see also answers to reviewer nr. 2 comment 11).

No information is given to evaluate if this artificially flooded fen behave more like an aquatic system or like a peatland in terms of emissions.

The specific emission patterns of the flooded fen are not within the scope of this MS (as a technical note), since the obtained emissions are only used to verify the presented calculation tool, as also stated by reviewer nr. 2. If needed, requirements are given (closed chamber measurements; high temporal resolution data), and the presented flux separation approach is applicable to different ecosystems, regardless of their specific emission behavior (magnitude of total CH<sub>4</sub> emissions and flux components) (P12934-12935 L26-1). However, in terms of site history (rewetted only 8 years ago (2005; P1927 L21-23); periodically dry growing seasons) and regarding the rather low water level of <35 cm (P12931 L23), the authors assume that the site represents neither a typical peatland anymore, nor a typical lake yet, but a stable inland water (shallow polythrophic inland water, consisting of an open water area and growing reedbelt in transistion). Further studies verifying this assumption are needed, but again, not within the scope of this MS.

The topic of this study fits the scope of the journal and the quality of the dataset seems reliable and valuable.

The authors really appreciate the comment, since the data reliability of the calculated and separated CH<sub>4</sub> emissions was seen as the main issue of the MS.

However, the manuscript is not well organized (missing description of results) and reveals an approximate knowledge on the CH<sub>4</sub> cycling in inland water and the related literature.

Regarding the claimed missing description of results, please see answer to comment Nr. 3. As stated in the MS, the study site used to verify the presented calculation and flux separation approach is a rewetted former fen grassland. Hence, it is not a classical lake system, but a shallow polythrophic inland water, consisting of an open water area and growing reedbelt in transistion, which developed after flooding the former fen peatland area as also stated by the authors within the comment above. Thus, literature referring to lake or aquatic systems and its specific CH<sub>4</sub> cycling is not necessarily representative for the site conditions.

The illustration could be significantly improved by adding a figures showing the diurnal variations, the dependency of emissions on temperature (and potentially water level, wind speed, RH) and improving the figure 5 which is currently difficult to read.

Diurnal variations, mainly in terms of diffusive methane emissions, throughout the whole study period are shown in Fig. 5c. The dependency of methane emissions (diffusion, ebullition as well as total) on water and sediment temperature, as well as RH and wind velocity is shown in Tab. 1. To avoid showing dependencies regarding different environmental drivers (e.g., wind velocity, RH, air temp., sediment temp., water temp.) based on only one day dataset, daily datasets were used to calculate standardized beta coefficients. Subsequently, the median of all day standardized beta coefficients was given in Tab. 1. To better express this procedure, caption of Tab. 1 were rewritten (see also comment 4). To address the dependency of methane emissions on air pressure as well as water level, the standardized beta coefficients for both factor dependencies are now included in Tab. 1

P12924, L22-26: “(i) diffusion, (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), which are all subject to variable environmental drivers and conditions such as water level, atmospheric pressure, temperature gradients, and wind velocity as well as the presence of macrophytes”. This has to be corrected since in the literature, only diffusion depends on temperature gradients and wind velocity among other parameters generating turbulence and ebullition and only ebullition depends on water level (and therefore hydrostatic pressure) and atmospheric pressure among other parameters. Plant-mediated transport depends on the presence of macrophytes but it does not make sense to mention it.

Within P12924 L22-26 it was not aimed to state that diffusion, ebullition and plant mediated transport are all depending on the same environmental drivers. This sentence is a simple list of CH<sub>4</sub>-flux components and different potential drivers. To better address this issue and the misleading connection between open-water systems and plant mediated transport, the sentence was split and changed to “In this ecosystems, CH<sub>4</sub> is released via three pathways: “(i) diffusion, (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). These pathways are all subject to different environmental drivers and conditions such as e.g. water level, atmospheric pressure, temperature gradients, or wind velocity and presence of macrophytes.”.

P12925, L1-5: Walter et al (2006) is not in the reference list. Maeck et al (2014) and Walter et al (2006) are papers on ebullition only so the relative contribution of the different pathways cannot be discussed in these papers.

The references Walter et al. (2006) and Maeck et al. (2014) were intended to highlight temporal as well as spatial variability of CH<sub>4</sub> emissions (substantially driven by erratically ebullition events). To better address this, the sentence is changed to :”Since especially ebullition varies in time and space (Maeck et al. 2014; Walters et al., 2006), total CH<sub>4</sub> emissions features an extremely high spatial and temporal variability at all scales (Koch et al. 2014; Repo et al. 2007; Bastviken et al. 2004).” Walter et al. (2006) was added to the reference list.

P12925, L5-7: Prairie and Del Giorgio (2013) is not a paper about CH<sub>4</sub> emission modelling but a paper about the evaluation of the existence of microbubbles (different from the classical ebullition).

Corrected. Prairie and Del Giorgio (2013) is now only quoted within the section regarding the possible bias due to micro bubbles.

P12925, L15-18: “However, field studies measuring emitted CH<sub>4</sub> above the water surface generally measure the total CH<sub>4</sub> emissions as a mixed signal of individual CH<sub>4</sub> emission components, released via all possible pathways (i.e. diffusion, ebullition and plant mediated transport).” This statement does not reflect the reality (see for instance the supplementary material of Bastviken et al (2011, Science)). In most of the studies. Plant mediated emissions are not often taken into account.

Bastviken et al. (2011 and 2004) are measuring on lakes using floating chambers. However, floating chambers on aquatic systems sometimes measure both diffusion and ebullition methane emissions (“These chambers collected both ebullitive and diffusive fluxes across the lake surface” (Bastviken et al. 2010, P3)). The same applies to shallow water bodies or measurements above flooded peatlands, which are characterized by rather low water levels and corresponding hydrostatic pressure to be overcome by gas bubbles. Hence, as stated, a mixed signal of diffusive and ebullition fluxes is measured by the chamber system. To better address this issue the sentence was changed to: “However, field studies measuring emitted CH<sub>4</sub> above the shallow water bodies or flooded peatlands generally measure the total CH<sub>4</sub> emissions as a mixed signal of individual CH<sub>4</sub> emission components, released via all possible pathways (i.e. diffusion, ebullition and plant

mediated transport).” Accordingly, plant mediated transport is especially measured in shallow flooded peatlands. Kankaala et al. (2005) and Juutinen et al. (2003 and 2004) performed CH<sub>4</sub> measurements within the littoral zone of aquatic systems, and hence accounted as well for plant mediated transport induced emission, even though flux separation was not computed.

P12925, L19-23: “Measurements of CH<sub>4</sub> ebullition can be performed using manual or automatic gas traps, as well as optical and hydroacoustic methods (Wik et al., 2011, 2013; Maeck et al., 2013; Walter et al., 2008; Ostrovsky et al., 2008; Huttunen et al., 2001; Chanton and Whiting, 1995), often requiring a certain degree of manipulation of the studied system.” What are the optical methods the authors are referring to? What kind of ecosystem manipulations are necessary for the measurement of ebullition?

Wik et al. (2011) evaluated (methane) bubbles trapped in ice of Siberian lakes. This was done via “optical analyses” of photographic images. A camera was used by Ostrovsky et al. (2008) to monitor and optically evaluate ebullition events (both quoted within this section). This sentence was changed to “often requiring a certain degree of needed instrumentation **within** the studied system”. Concerning the required instrumentation and thus manipulation of the system, Bastviken et al. (2010) give a detailed explanation of instrumentation necessary for the use of bubble shields. Maeck et al. (2014) mentioned anchor weights of bubble traps, needed to be placed in a distance to avoid disturbed ebullition measurements. Hence, both systems require instrumentation, which might change system conditions, and thus measured methane emissions.

P12925, L23-27: “Diffusive CH<sub>4</sub> fluxes are commonly either derived indirectly as the difference between total CH<sub>4</sub> emissions and measured ebullition, or directly obtained based on the use of bubble shields or gradient measurements of CH<sub>4</sub> concentration differences\_(DelSontro et al., 2011; Bastviken et al., 2010, 2004).” Only the cited research groups use bubble shield chambers and it is used only in ecosystems characterized by extreme ebullition which avoid the measurement of diffusion only. In all other study, diffusion is measured with classical floating chamber (without shield) or calculated from the concentration gradient.

The exemplarily measured system within our study is characterized by high methane emissions and regular ebullition events, comparable to conditions at study sites described e.g. in Bastviken et al. (2010). The comment regarding the use of floating chambers for diffusive methane

emission measurements seems to be contradictory to statement number 13, were it is stated that in “most [...] studies, only diffusion is measured or evaluated, and when “total emission” is measured, ebullition and diffusion are measured separately”. This is only possible with floating chambers, if either ebullition is absent (thus requiring an algorithm for flux separation into ebullition and diffusion), or with bubble shielded chambers.

P12926, L9-12: “CH<sub>4</sub> flux separation approaches based on manual chamber measurements with rather low temporal resolution fail to capture the rapidly changing absolute and relative contributions of the pathway-associated flux components both in time and space (Maeck et al., 2013; Walter et al., 2006).” Walter et al (2006) is not in the reference list. Maeck et al (2014) and Walter et al (2006) are papers on ebullition only so the relative contribution of the different pathways cannot be discussed in these papers.

Please see answer to comment 11.

P12926, L12-15 “Improvements in measurement techniques, particularly by using micrometeorological methods (e.g., Eddy Covariance (EC)), however, allowed for high temporal resolution records of CH<sub>4</sub> emissions (Juszczak and Augustin, 2013; Schrier-Uijl et al., 2011; Cole et al., 2010; Wille et al., 2008).” Cole et al. (2010) did not use eddy covariance.

It is true that Cole et al. (2010) did not use an EC measurement system. However, they refer to possible near future advantageous use of ADV systems (acoustic doppler velocimeters) which was referred to as one of the “improvements in measurement techniques” within the MS. However, to avoid a misleading understanding, the reference was deleted.

All references here are from peatland studies. Deshmukh et al, 2014, Schubert et al. (2012, ES&T), Eugster et al (2011, biogeosciences) and Repo et al. (2014) should be cited for aquatic ecosystems.

Since the study site is a rewetted former fen grassland site, mainly peatland references were cited. However, Deshmuk et al. (2014), as well as Repo et al. (2007) are already cited.

12926, L15-17: For automatic chambers, Ramos et al (2006, Geophysical Letters) could be cited.



Regarding the deployment of automatic chambers, Koskinen et al. (2014) and Lai et al. (2014) are cited, which are both referring on peatlands, comparable to the studied ecosystem. To better represent aquatic systems, Ramos et al. (2006) was added.

P12928, L19-21: Are the chambers equipped with a vent in order to avoid over pressure in the headspace when chamber are lifted down?

The automatic chambers are not equipped with a vent. Over-pressure in the chamber headspace is avoided by very slowly lowering the chamber onto the water surface. Moreover, with a total volume of 1.5 m<sup>3</sup>, the chamber headspace is very large, thus also reducing the risk of over-pressure within the chamber. Finally, possible disturbances due to chamber closure at the beginning of each measurement are diminished by the applied death band (25%) (Fig. 3b and d; P8L11).

P12931, L19-22: In what extent diffusive fluxes could be affected since the occurrence of ebullition modifies significantly the concentration gradient?

As shown in Fig. 3, the concentration of CH<sub>4</sub> inside the chamber increases due to ebullition events. With a total volume of 1.5 m<sup>3</sup>, the chamber headspace is, however, comparable huge, which, together with a rather short measurement time of 10min, is limiting the absolute concentration change within the chamber headspace, and thus the influence on the concentration gradient. Additionally, the (chamber) measurement system itself (and its potential shortcomings) is not seen as a substantial part of the presented approach, and therefor beyond the scope of this paper. The new and important aspect is seen within the calculation approach/data processing algorithm (which is assessable on different chamber design as pointed out in L: 224-226).

P12932, L9-13: Is there any correlation between fluxes and temperature at the seasonal scale?

Yes. As stated in P12932 L9, the “measured total CH<sub>4</sub> emissions showed clear seasonal patterns, following the temperature regime at 10 cm soil depth”. This is also visible when comparing Fig. 5, where time series of methane emissions, as well as sediment and water temperature are shown.

P12932, L13-17: Is there any correlation, within single days, between fluxes and temperature

(air, water or sediments) that could explain the diurnal variations?

Yes (see also comment 9). Tab. 1 shows the median of the calculated standardized (beta) coefficients (based on daily data subsets), and thus the correlation between sediment/water/water-air gradient/air temperature and CH<sub>4</sub> fluxes (and its components). Especially the air and water-air temperature gradients correlated well with the calculated diffusive fluxes, whereas the total CH<sub>4</sub> emission is only slightly correlated with air temperature (Tab. 1). As stated in the MS (P12932 L23-25), this was seen as an indication of turbulent mixing being responsible for the recorded diurnal trends of methane emissions.

P12932, L25: The potential role of thermal stratification in the water column is discussed although the maximum depth of about 35 cm avoids any establishment of thermocline in this wetland. Thermocline usually establish at depth of 4-10m in lakes.

The establishment of a thermocline is not stated within the MS. The stated differences in turbulent mixing due to different water temperature gradients (or air-water temperatures gradients) are also mentioned by Koebisch et al. (2015).

P12933, L9-17: The relationships between fluxes and temperature, wind speed, RH must be shown in order that some explanations are given for the interpretation of the observed trends.

Based on daily datasets, median standardized beta coefficients are given in tab. 1 to show the dependency of methane emission and its components on RH, wind velocity and temperature, Air pressure and water level dependencies have been added to the revised version of the MS.

P12934, L5-12 The different emissions rates obtained with the four different chambers were attributed to depth but depths are not given to validate this hypothesis.

In accordance with the comment of Reviewer 2 on this issue, P12934 L5-12 was changed (please see comments on reviewer 2).

Until the end of the discussion, there is confusion between the controlling factors of ebullition (water depth and therefore hydrostatic pressure in the literature) and diffusive fluxes (concentrations gradient and gas transfer velocity in the literature). In the manuscript, diffusive fluxes and gas transfer velocities are supposed to depend on the hydrostatic pressure.

There is not such a statement within the MS. If the reviewer refers to P12934 L5, it is stated that the shown “trends might be a result of the increasing water column further from the shore, causing a reduced gas transfer across the air–water interface as a result of e.g. higher hydrostatic pressure to be overcome by gas bubbles on the one hand and an increased diffusive CH<sub>4</sub> flux based on enhanced CH<sub>4</sub> gradients on the other hand”. The enhanced CH<sub>4</sub> gradient between sediment and water/atmosphere is assumed to be caused by suppressed ebullition events and not related to hydrostatic pressure. However, in accordance to an issue raised by reviewer 2, this section was rewritten (please see answers to reviewer 2).

Again the references cited here are not adequate since Wik et al (2011) and Walter et al (2008) presented only ebullition and could not compare it with diffusion.

Both references refer to “highly variable ebullition events”. Hence, the sentence was changed to “Thus, the detected spatial variability of total CH<sub>4</sub> emissions was dominated by highly variable ebullition events (Wik et al., 2011; Walter et al., 2008) rather than by systematic differences in diffusive CH<sub>4</sub> emissions”.