Manuscript revision

The manuscript has been corrected according each referee's suggestions, see the final author's response below. In particular, Figures 3, 4, 6, 7, 8, 9, B1, C1 have been modified; Figure B1 has been moved to

5 the results section as Figure 10; Table S2 from supplementary material has been moved to Appendix B as Table B1; sections 2.6, 4.2.2 were rewritten and section 3.4.1 was added to the manuscript; Appendices A and B were rewritten; a figure showing data availability and a text further detailing data pre-processing were added to supplementary information.

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On behalf of all authors, I thank the two referees for a thorough evaluation of the manuscript with relevant and constructive comments. Following are specific responses to each of the referees' comments.

The referees' comments are in black and the author's responses in red

5 **Response to referee comment #1**

This manuscript presents two years of methane and carbon dioxide flux measurements at a lake and a fen sites. The dataset is very interesting, the framework analysis and results discussion very comprehensive and well written. I can recommend the final publication in Biogeosciences after the following comments are properly addressed:

- 10 1) Measurements of CO₂ fluxes were done by using an open path IRGA (LI-7500). Although this is a convenient or sometimes the only option in remote sites, I would use it with caution for measuring very low fluxes like the one presented here for the lake. The effect of air density fluctuations on CO₂ flux becomes very important, and the validity of WPL correction for low CO₂ flux has been also questioned in the past (see Ono et al., 2008). The authors are aware of this and that is why efforts were made to quantify the associated uncertainties on individual fluxes but also on annual sums, and why a strict approach was chosen when filtering the flux dataset. The dataset has been thoroughly quality checked based on micrometeorological and statistical criteria, as much as it was possible with the available data and instrumentation.
- 2) I was a bit surprised to see that the high frequency corrections for CO₂ flux were so high (31% on average). Usually for open path EC system is much less. I guess this may be because of relatively large separation between the IRGA and the sonic anemometer. What was the separation? What is the value range of time lag for CO₂ and which windows have you used? There may be problems with the WPL temperature term, which in theory should be measured in (or close) the path of the IRGA? This is a valid point, indeed the separation between the two instruments is large (42 cm vertical separation, 26 cm northward separation and 35 cm eastward separation). The nominal time lag between vertical wind speed and CO₂ was set to 0 s, with a searching window from -4 s to 4 s. There was no thermistor close to the IRGA, thus the ambient temperature measured at a nearby mast was used for WPL correction and throughout the paper. The effect of the WPL correction on the CO₂ fluxes is quantified in Appendix A.

3) What would be the reason of relatively high (anti)-correlation between CO_2 flux and H during 30 wintertime (from Table 2)? And what about summer? A possible explanation for the anti-correlation between CO2 flux and H during summer is discussed in the last paragraph of section 4.2.1. We suggested that it could be due to the diffusive CO_2 flux (in this case, downward) between the surface and the atmosphere being enhanced by waterside convection (denoted by positive H), as it has been shown in other lake studies where it was associated with an evasion of CO_2 . The correlation in winter, however, remains to this date unexplained, and may be linked to instrumental issues.

4) CH₄ flux: it is not clear if H₂O was measured by LGR. If not, then I guess the H₂O fluxes

5 measured by LI-7500 were used in the WPL correction for CH₄. What EddyPro does when H₂O comes from the LI-7500? The H₂O fluctuations in the sampling cell of LGR may be quite different than the ones measured by the open path IRGA. How the authors cope with this issue? Why the compensation for the pressure term was also added? Which pressure data have been used for this? Thank you for insisting on this point, which led us to find an overlook in the flux calculation. In EddyPro version 5.2, a revision of the WPL formulation by Ibrom et al. (2007) is proposed for closed path instruments. The pressure term is an addition by EddyPro to the original formulation. Air pressure measured at a nearby

We unfortunately did not use H₂O data from the FGGA in this study due to a faulty electronic connection at the time of data collection, thus we relied on the open path LI7500 analyser for H₂O
measurements. EddyPro requires a metadata file with information on instrument model, inlet tube properties, sensor separation, etc. We therefore assumed the open-source software to proceed and correct accordingly by taking these metadata information into account. However, after further investigation motivated by the referee's enquiry, we realized that EddyPro does not seem to apply the WPL correction when H₂O measurements from the same closed path instrument are not available. This was a surprise considering what the text in the manual of the software version 5.2 implies (i.e. the application of a classic *a posteriori* WPL approach if cell data are lacking for closed path instruments). It actually appears, after recent verification, that in EddyPro a lack of available H₂O data from the same closed path instrument results in no WPL correction. This is unfortunately not clear in the EddyPro

25 matter is handled in version 5.2.

mast was input to the software for this purpose.

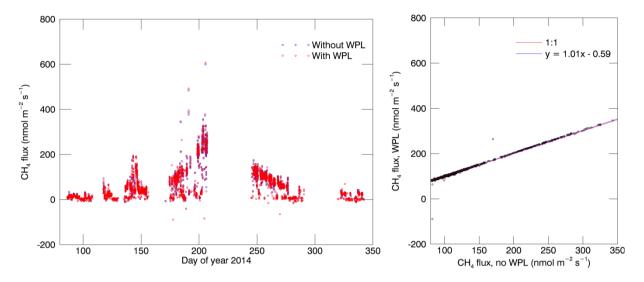
We thus performed a test on summer CH4 flux data to evaluate the magnitude of the density effects on CH₄ fluxes during the summer of 2014. We were able to recover part of the H₂O data from the FGGA using the same synchronization method as for CH₄ (cf. methods section of the paper) and used these measurements to apply the WPL correction in EddyPro using the formulation by Ibrom et al. (2007). The result is a difference of shout 1% in flux magnitude, see also Figure A of this document.

settings, where the user chooses to apply the correction or not, and to this date we are unsure on how the

30 The result is a difference of about 1% in flux magnitude - see also Figure A of this document.

The low magnitude of the WPL correction can be expected here, due to the long sampling line that attenuates significantly the H_2O signal as well as temperature and pressure fluctuations thus density effects. Hence, the correction would have likely a minimal impact on the CH₄ flux dataset.

In the revised version of the manuscript, Appendix A will be corrected to acknowledge this overlook and a quantification of the estimated (minimal) effect of density fluctuations on closed path CH_4 flux data will be added.



5 Figure A: Comparison of CH₄ fluxes with and without WPL correction for the year 2014.

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5) Besides of density fluctuations caused by H_2O , spectroscopic correction should be also applied to CH_4 flux (see Peltola et al., 2014). It seems to be some correlation between CH_4 flux and LE (table 2). Is this because of points 4 and 5? The correlation between CH_4 flux and LE is only notable during the ice-free season. It would be expected to be consistent over the year if it was due to a systematic error or bias present in the full dataset. We were not aware of the spectroscopic effect as an additional correction applied to CH_4 fluxes measured with closed path systems. Again, EddyPro offers this correction for

- applied to CH_4 fluxes measured with closed path systems. Again, EddyPro offers this correction for open path CH_4 analyzers only. That said, having to use open-path H_2O data may introduce more uncertainty than without the application of the spectroscopic effect.
- Summarizing, I know that there are limitations on including (in proper way) all these aspects, but at least it should be checked how important they are. Finally, the authors should acknowledge more clearly these aspects when discussing the uncertainty of these fluxes. We agree that those are important technical points and appreciate that the referee emphasizes it. An attempt is made in our study to quantify the importance of the density effects and spectral corrections (cf. Appendix A). In the revised version of the manuscript, these points and their potential effects on the fluxes will be made clearer. The
- 20 flux dataset has been thoroughly checked and, while acknowledging uncertainties, we remain confident

in the CH_4 flux dataset due to strong agreement with other data sources (chambers at the fen and ebullition traps at the lake); as for CO_2 fluxes, we provided the best estimates possible with the available instrumentation at the time of the study. We will emphasize more clearly the associated uncertainties in the revised manuscript.

5 Minor comments:

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Pag 6 L 11. Add also latent heat flux. OK

Pag6 L.18. "mixing ratio" usually means "dry mole fraction", but I guess this is "wet mole fraction", so without dilution correction. For calculating dry mole fraction point by point (high frequency data), simultaneous H_2O measurements are needed. This is correct and will be corrected in the text accordingly.

Pag. L.26-28. How the synchronization was done? Just using the timestamps? Raw data stored on the FGGA memory are not sampled at exactly 10Hz but at a variable frequency (11 to 12Hz). Raw CH₄ data stored on the FGGA were thus linearly interpolated on 10Hz timestamps. Additionally, to prevent mistakes due to a potentially uncalibrated clock on the FGGA, we did not use only the timestamp to synchronize the dataset. It was done in half-hour moving chunks of data by maximizing the correlation

- 15 synchronize the dataset. It was done in half-hour moving chunks of data by maximizing the correlation between logger data and FGGA data. The time showing the best correlation was chosen as a reference to adjust the clock, then CH_4 data were linearly interpolating onto the correct (logger) timestamp. Thus, when computing fluxes, the time lag for CH_4 fluxes between September 2013 and December 2014 was set to be searched within a large window that included 0s. The synchronization procedure was quality-
- 20 checked after flux computation, cf. response to referee #2, point 6.

Pag.7 L.14. FST<0.3 is quite strict criteria. What about using FST<1? Is there a relevant difference in the data coverage? A strict filtering approach was chosen due to challenging footprint conditions, to ensure that fluxes actually represented the surface of interest. Adopting FST<0.3 as a criteria resulted in removal of outliers and negative values.

25 Pag.7 L.15. Ustar threshold is taken equal to 0.1 m/s. How this was determined? The threshold was determined using the online tool available at https://www.bgc-jena.mpg.de. This precision together with a reference will be added in the text.

Pag.8 L8-10. Could you report some of these values used for the footprint calculation, e.g. roughness lengths? A dynamic roughness length was used to represent the evolution of the canopy height (and

30 presence of snow) on the fen side throughout the year, while a constant roughness length was adopted for the lake side. The values will be added in the text.

Pag.8 L13-14. Is longer footprint in winter because of more stable conditions (negative H and low wind speed)? Or why? Longer footprint in winter is most probably due to a lower roughness length (snow cover), which is a user-defined parameter, but also to more stable conditions, since H is negative during most of the winter. Indeed, the stability parameter is higher in winter increased (average of 0.10) in comparison to the summer season (average of 0.02). A sentence will be added in the text

5 comparison to the summer season (average of 0.02). A sentence will be added in the text.

Pag.8 L16. Based on what, the criterium $\sigma v < 1$ m/s was used? This criterium was used to limit lateral contamination of CO₂ fluxes into the footprint area of interest. The threshold was used after Forbrich et al (2011), who used this criterium to remove high crosswind fluctuations in their footprint study. The reference will be added.

- 10 Pag.9L.25. Why the method did not perform well for lake CO_2 flux? As shown by the density distribution in Figure C1 of the paper, lake CO_2 fluxes were very low and close to zero most of the time. Furthermore, they comprised a large amount of gaps, partly due to strict data filtering. We therefore decided to exclude the ANN results for the lake CO_2 fluxes as we found them to introduce a very high and unnecessary uncertainty.
- 15 Pag10L12-13. The random error of fluxes is usually proportional to the flux magnitude. Do you mean the relative random error (error normalized by the flux) is smaller? Yes, correct. It is proportional to the magnitude of the flux but relatively to the flux of smaller importance when fluxes are higher. This will be corrected in the new manuscript.

Pag.10 L21. How did you calculate the RE of the fluxes modeled with ANN? Each value used for gap
filling is the mean of several ANN model runs (cf. Text S1). The 25 best runs (according to r2) were averaged to output the modeled fluxes used in the gap filling. The standard deviation of these 25 model outputs was used as a quantification of the random error of each value used for gap filling. The average of these individual random errors was then computed as the mean random error for each modeled series (CH₄ fen, CH₄ lake, CO₂ fen). Information will be added to Text S1 and Table S1 in the revised manuscript.

Pag.11. L9. Is the thaw season the same as ice-out season? Yes. The term will be replaced throughout the manuscript for coherence, using "thaw season" as defined in section 2.7.

Pag.12 L15-16. Do you mean Fig. 2e and Fig 2f? Yes. This will be corrected.

Pag. 13. L15. How do you explain this fall burst of CO_2 ? The warmer summer in 2014 may have caused a thermal stratification at the end of the season not present in other years. This could result in an accumulation of CO_2 and a subsequent degassing when lake cooling in fall triggers water mixing. Pag.15 L5-10. The diel cycle of 2012 H is quite noised respect to the other years. Why? H flux data coverage in the months of June-July-August was 5% lower in 2012 as compared to 2013 and 2014, thus the seasonal diel cycle may be more sensitive to variability between days.

Pag.16 L10-20. May be some literature values can be added here for comparison. Values will be added.

5 Pag.16 L22-23. The highest correlation I can see for winter fluxes is with H. This is true, but it does not invalidate the observation made in this sentence. The correlation in winter with H is yet unexplained and could potentially be instrument-related but we have no mean of quantifying it.

Pag.16 L24. With EC it is not possible to measure advection, however You may see an increase of CO_2 mean concentration in the data, which may indicate non-turbulent transport of CO_2 from land. Thank

10 you for the suggestion. After verification, there is an increase in CO_2 concentration in winter, as compared to summer values. This will be commented in the text.

Pag.16 L.27-31. I would say that it could be important to get a rough estimate of this correction. The effect (and direction) of this correction depends on the sign of H. We will make a rough estimate of the correction and discuss it.

15 Pag.17 L.9-11. How the EBC (energy balance closure) plots look like in different years? EBC was not computed for the lake side because of the uncertainty related to the computation of heat storage in the lake with the available data. As a hint on the eddy covariance system performance, Figure B below shows the energy balance closure on the fen side on half-hourly and daily time scales (full dataset). If the allocated time to revise the manuscript allows it, we will attempt to compute an ECB on the lake 20 side too.

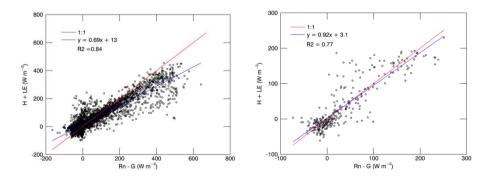


Figure B: Energy balance closure on the fen side at half-hourly (left) and daily (right) time scales.

Pag.20 chapter 4.3.2. Are there any measurements of pCO_2 for this lake during summer? Or chamber measurements? Anything that could support the sink that you measured with EC? Unfortunately, there were no coincident measurements of pCO_2 in the lake water during the study period. A new study is currently measuring pCO_2 along with chamber and EC measurements, which will be able in the future to validate or not the CO₂ sink

5 to validate or not the CO_2 sink.

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Pag.23 L17. Coordinate rotation is not really a correction. True, this will be corrected.

Pag.23 L19-22. Please report the values of time windows and time lag. Was the lag maximization applied also for sonic temperature? The lag between w and Ts should be 0. The lag maximization was not applied for sonic temperature (time lag set at 0s). The sentence will be modified for clarity and time lag windows reported.

Pag32 L.17. Please update the reference Rannik et al. (2016). The article is now published in AMT. This will be corrected.

Caption of Table 2. Do you mean the std of lateral wind speed? Yes; this will be corrected.

Figure 2 . Please explain in the caption what are those black arrows pointing down in fig2f. Black arrows indicate the estimated time of full overturn. This will be added in the caption.

Figure 6. Please reduced the scale of CH_4 fluxes. It is very difficult to see how EC compared with ebullition data. The figure will be redrawn to improve visibility.

Please increase the font size in all figures. Font size will be increased where needed.

20 **Response to referee comment #2**

General Comments

The manuscript presents a 2.5-year CO_2 and CH_4 flux data set from a fen and lake within a subarctic peatland ecosystem. GHG fluxes from aquatic ecosystems have been identified to contribute to a large but uncertain amount to the global GHG budget.

²⁵ Thus the presented study delivers important data and a substantial contribution to scientific progress within the scope of Biogeosciences. The scientific approach and the applied methods are valid, related scientific work is amply referenced, and the results and conclusions are presented in a well-structured way and in an appropriate style. I support the publication of the manuscript after minor revision.

Specific Comments

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1. The manuscript suffers from occasional vagueness in definitions and nomenclature. For instance, "ice-out" is inconsistently used in the manuscript. On first use, in the abstract (p. 2, line 12), it denotes the point in time when the ice is completely thawed. Further into the manuscript "ice-out", "ice-out season", and "ice-out period" are used synonymously denoting the "thaw season" as defined in section 5 2.7. I suggest to stick to the nomenclature introduced and to replace "ice-out" and its variations with "thaw period" wherever this is applicable throughout the manuscript. Further, in section 2.7 it is not made very clear that the defined "thaw season" not only comprises the actual thawing of the lake ice, but also - and importantly - the subsequent initial overturning of the lake water. For clarity, the term "thaw period" will be chosen and coherently used throughout the manuscript. 10

2. Gap filling of highly variable CH_4 fluxes is notoriously challenging but at the same time crucial for determining seasonal and annual balances. Therefore, the artificial neural network (ANN) gap filling method should be well presented and discussed. I consent to the detailed description of the ANN method being given in the supplement so as to keep the text concise. However, the environmental drivers used for the ANN is important information which should be included in the main manuscript. Hence I suggest to move Table S2 to Appendix B (which in consequence could be reduced in text). Agreed, the table will be moved to Appendix B and the text adjusted accordingly. Furthermore, a paragraph on the ANN gap filling performance should be added to the results section, and Fig. B1 should be part of it (and should be improved for better legibility). Font size in figure B1 will be

increased and the performance reported in Appendix B will be moved to a short paragraph in the results 20 section.

Especially in case of CH_4 fluxes, which span 2 orders of magnitude, a discussion of how the ANN performs - both in case of the slowly varying background flux and episodic high emission events would be very interesting. This is briefly discussed when emphasizing how the performance of the

- ANN method differed between the fen CH_4 flux dataset and the lake CH_4 flux dataset. We agree that 25 ANN gap filling, as a rare method of gap filling EC fluxes and especially of lake EC fluxes (to our knowledge, we are the first to present an application of it to lake CH_4 fluxes), needs to be appropriately described and discussed. We attempted to give enough information while keeping it concise, since the gap filling itself is not the primary focus of this paper. The method used closely followed the procedure introduced in details by Dengel et al. (2013). 30

3. There was no gap-filling performed for lake CO_2 fluxes after May 2013 due to low data coverage. However, according to table S1, CO_2 flux data coverage was back to normal in 2014. Gap-filling should be resumed for 2014 data if at all possible. Please note that CO₂ fluxes from the lake were not gap filled as the rest of the dataset using a gap filling model (see response to referee #1 for further detail), but by using mean fluxes per season. This can be considered valid for normally distributed dataset, but introduces a largest bias when data coverage is lowest. The second year (June 2013 - May 2014) comprises too many gaps. Depending on the allocated time for the revision of the manuscript, we will consider extending it to the last two seasons of the dataset when data coverage increased (spring and summer 2014), but we will not compute the second annual sum.

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4. I like the statistics of measured fluxes presented in figure 4. However, with a data coverage of typically 30 %, how reliable is this information, i.e. how does the picture change when you look at the statistics of gap filled fluxes? From a statistical point of view, these results are valid, since eddy
covariance produces a lot of data points, even with 30% data coverage over a year. I would argue that computing the correlation statistics on the gap filled datasets would not be appropriate and would give bias results, since the environmental parameters tested here are some of the same variables used to develop the gap filling models. This is why we chose to compute correlation statistics on measured fluxes only. These are a statistical exploration of the available dataset; generalizing it to the whole period is indeed uncertain, considering the data coverage, and is not necessarily the intention here.

The mean and median fluxes presented in section 3.2 may have to be interpreted with a certain caution, especially concerning the transient lake fluxes. Indeed, the mean alone would not be an accurate summary of the flux magnitude, since it is affected by the occasional large degassing in the lake dataset (cf. Figure C1). Thus, we reported medians along with the means, and underlined at various stages of the manuscript (and in Figure 4) the skewed distribution of the lake flux data.

5. I think there is more potential in the ebullition flux data from bubble traps than the qualitative comparison presented in figure 6. I would like to see the attempt of a qualitative analysis in order to derive an estimate, of how much of the EC flux stems from diffusion and ebullition. Data are not available at this level of details for the present manuscript. An ongoing study will quantify the part of

25 diffusive and ebullition flux within the EC dataset by means of comparison with simultaneous chamber and bubble measurements. These results are presented in a separate study and so not included here. We hope the reviewer will accept that these studies are being kept separate.

Further, the thaw of the lake ice and the initial overturning of the lake water after the "ice-cover" period seem to be well separated in time. Hence I suggest to divide the spring emission peak into a portion

30 which originates from the escape of gas bubbles trapped in the ice, and a portion which originates from the initial overturning of the lake water. This could help to explain the large differences between the total thaw season CH_4 emissions in 2013 and 2014. This is a very good point that has actually been addressed in details in a previous study focusing specifically on the CH_4 degassing from the lake during the spring of 2013. In Jammet et al. (2015), we present a quantitative and qualitative analysis of the spring efflux in 2013 by suggesting a separation of the degassing event in three steps, which likely correspond to emissions from different gas sources (liberation of bubbles from the ice, diffusion from the water, overturn). A sentence in the discussion refers the reader to this paper for further information (section 4.2.2). Section 4.2.2 will be rephrased to make this point charger.

5 (section 4.2.2). Section 4.2.2 will be rephrased to make this point clearer.

6. How was eddy covariance raw data logged (type of data logger),

A CR1000 was used until June 2013. Occasional data loss occurred in 2012 due to the low performance of the CR1000 for heavily instrumented sites (skipping logging rows), which lead to its replacement with a CR3000 in June 2013. Data logger information will be added to the methods section.

- and what exactly was done with CH₄ raw data during August 2013 December 2014? As I understand, CH₄ concentration was taken from FGGA raw data files and had to be synchronized and combined with sonic anemometer data before being fed into EddyPro. If this is the case, have you checked if this caused any bias in the flux calculation? Please clarify. As explained in replied to referee #1 (see above), CH₄ raw data from the FGGA were synchronized with the high frequency data logged onto the data logger, then linearly interpolated to match the logger time stamp. After flux calculation half hours for
- 15 logger, then linearly interpolated to match the logger time stamp. After flux calculation, half-hours for which the synchronization procedure could not yield any reliable flux (i.e. where the method failed) were identified (lack of a significant peak in the cross-covariance function, as in Wienhold et al. (1994), Nordbo et al. (2012), Rinne et al. (2007)) and filtered out. This screening step was considered as a quality check of the synchronization procedure.
- 20 7. In addition to the maintenance-caused gap during February March 2014, there is a large gap in CH_4 flux data during December 2013 February 2014. Was all data of this period rejected by the quality screening? The same question arises for CO_2 fluxes during February March 2014. Please clarify. Thank you for raising this point. The next version of the manuscript will include (in supplementary information) a further figure visualizing the data coverage for both instruments, indicating which time
- 25 periods were flagged due to power outage or lack of data and which data were removed due to quality check procedures. During winter, measurements from the sonic anemometer and FGGA analyzer were regularly subject to multiple drop-outs and out of ranges values (filtered out during screening), most likely due to frost on the sonic and very cold air input to the analyzer, which does not perform well at low temperature.
- 30 8. The "Burba effect" seriously compromises cold season CO_2 flux data from the LI-COR Li-7500 which you used. The fact that the "Burba correction" was not applied is important information and should be given in the methods section and not as a sideline in the discussion. Agreed, this information

will be moved to methods section. To my knowledge, many researchers failed to derive a meaningful flux correction using Burba's method, in which case there is no other way than to use

the CO_2 flux data as it is. However, since you used a Los Gatos FGGA analyzer, you could use its CO_2 data to calculate another CO_2 flux data set to use during winter or to confirm winter time fluxes from

5 your Li-7500. Has this been attempted? Unfortunately, a problem with the data transfer on the CO_2 channel from the FGGA during the time of this study prohibited us to use the data for CO_2 flux computation.

9. The manuscript would benefit from focusing and shortening. Some examples are given in the next section.

10 Technical Corrections

p. 3, line 8: Change "explains" to "explain". OK

p. 3, line 8: Should read "order-of-magnitude-scale uncertainty"; consider simplifying to "large uncertainty". OK

- p. 3, line 30: Change "lake" to "lakes". OK
- 15 p. 5, line 3: Change "lake" to "lakes". OK

p. 5, line 27: "May be" sounds very weak. The cited paper must have a stronger opinion on this matter? This will be rephrased to "According to Olefeldt and Roulet (2012), the two ecosystems are hydrologically connected...".

p. 6, line 4: Change "palsa" to "palsas". OK

- 20 p. 6, lines 4-5: Change the order to "During snow melt, there is a small surface inflow feeding...". OK
 - p. 6, line 19: Add "height" after "2.50 m". OK
 - p. 8, line 11: Remove "the" between "footprint" and "model". OK

p. 9: The first paragraph and the last sentence of section 2.5 could be deleted. The first paragraph will be shortened and the last sentence will be deleted.

25 p. 9, lines 21-22: "The goodness of fit was quantified with...the absolute root mean square error (RMSE)." "Absolute" is superfluous and can be deleted. But in fact, table S2 gives the RMSE in %, and it is unclear what these percentages refer to. I strongly recommend to give the RMSE in flux units. OK

p. 9, line26: Replace "per" by "with". OK

p. 10: Section 2.6 could be shortened drastically by focusing on the reliability of the low (winter time) fluxes and on a brief outline of the error propagation method and the bias error. The section will be shortened, to the extent that it keeps the information necessary to understand the results in e.g. Table 3.

- 5 p. 11, lines 21-22: Simplify "daily energy input (upwelling > downwelling radiation)" to"mean daily net radiation". OK
 - p. 11, line 22: Wrong reference. Change Fig. 2b to Fig. 2c OK
 - p. 11, line 23: Wrong reference. Change Fig. 2c to Fig. 2d OK
 - p. 11, line 23: Change albedo from 5 % to 0.05 to be consistent with units in Fig. 2. OK
- 10 p. 12, lines 9-10: Differences in mean temperature correlate with differences in total net radiation, or more simply, mean temperatures correlate with total net radiation values. Please correct. OK

p. 12, line 14: "thermal stratification along lake depth" sounds odd. Consider changing to "thermal stratification of the lake". (Again in line 16) OK

p. 12, line 15: "... large" thermal stratification ..." Consider replacing "large" by "strong" if that is what
you mean. OK

- p. 12, lines 14-15: Replace "was repeated each year" by "was similar in both years". OK
- p. 12, lines 15, 16, 22: Wrong reference. Change Fig. 2d to Fig. 2f. OK
- p. 13, line 2: Replace "followed" by "showed". OK
- p. 13, line 12: Delete "but". OK
- 20 p. 13, lines 13-14: "The highest CO_2 uptake rates were observed during the summer of 2014, which was the warmest summer of the study period with highest solar radiation input (Table 1)." Table 1 lists only total net radiation values. As solar radiation can be expected to have a much higher explaining power for carbon fluxes (as confirmed by its inclusion in the correlation analysis, table 2), total solar radiation should be reported in table 1. Total solar radiation will be added to Table 1.
- 25 p. 14, line 15: The correlation between increases in sediment temperature and CH_4 bursts from the lake can hardly be seen – I suggest to delete this sentence. Although it is not systematic, a large part of the high CH_4 release events are preceded by an increase in surface sediment temperature and this is nice to

show. The scale of CH_4 fluxes in this figure may be too large at the moment to properly identify this; it will be changed so that the variations in fluxes are clearer to the reader. The sentence will be rephrased. The correlation with falling atmospheric pressure described in line 7 is much better visible.

p. 15, line 29: Replace "lead" by "led". OK

- 5 p. 16, line 1: I would not expect a complete ice cover at a fen dominated by vascular plants as described in the study site section. Unless the water table is very high at the onset of freezing. I suggest to rephrase this passage. The water table is always high at this site; however, the ice cover is not complete, because stems and branches are sticking out of the snow. That is what the sentence meant and it will be rephrased for clarity.
- 10 p. 16, lines 27-31: The passage on the Burba correction is pointless, because as written at the end of the paragraph - it corrects fluxes towards higher values and so cannot explain the too high fluxes during the winter 2013-2014. This sentence will be deleted and the mention of the self-heating effect will be moved to the methods section, as earlier suggested by the referee.

p. 17, lines 24-25: Mind the causal connection between temperature increase and decrease of CH₄
solubility! Rephrase, e.g. "since a seasonal increase in sediment temperature favors methanogenesis and additionally causes a decrease of CH₄ solubility..." OK

p. 18, lines 11-20: The whole paragraph seems inconclusive – how does it relate to your data? The paragraph is an attempt at explaining the (anti-)correlation observed between H and CO_2 flux during summer in our lake data. Additionally, it underlines how this relationship differs from what was shown in previous studies and suggest an explanation for this difference. The paragraph will be shortened and

- 20 in previous studies and suggest an explanation for this difference. The paragraph will be shortened and rewritten for clarity.
 - p. 19, line 2: Correct "release" to "released". OK
 - p. 19, line 23: Correct "term" to "terms". OK
 - p. 21, line 21: Change word order to "Alaskan thermokarst lakes". OK
- 25 p. 22, line 13: Change "period" to "periods". OK

Table 1: I suggest to move the dates from the figure caption to the table. Total solar radiation should be added as this is the most important driver of CO_2 fluxes during ice-free periods (in which case total net radiation could be omitted). Tables 1 and 3 should be merged into one table. Total net radiation will be replaced by total solar radiation. The dates may be added to Table 1 and Table 1 may be merged with

30 Table 3 if a wide table on a landscape layout is acceptable for the editors of Biogeosciences.

Table 2, caption: Wrong reference. Change "Table 2" to "Table 1". OK

Figure 2, caption: Add "daily means of" where applicable. Explain shaded area, "PN" and arrows. OK

Figure 3: Add grid lines, or at least y=0 lines. This helps the reader to determine if small fluxes are positive or negative or fluctuate around zero. Lines marking y = 0 will be added on Figure 3.

5 Figs. 4, 6, 8, 9, B1: The axis labels are too small. Font size will be increased where needed.

Figure 6: Remove temperature plots. The suggested correlation between sediment temperature and CH4 flux can hardly be seen anyway. As mentioned earlier, Figure 6 will be modified to improve visibility. This will hopefully address this comment.

Figure 7: One of the two graphs can be omitted, as they show the same data. Figure 7b will be removed.

- Figure 8: I suggest to remove the data of single years. The great variability makes it difficult to extract the important information from the graphs. Add grid lines, or at least y=0 lines. Single years are present to show that there is variability between years in terms of magnitude, yet the pattern is similar between years. To improve clarity, lines showing y = 0 will be added and single years will be suppressed from the figure.
- 15 Figure C1: This figure could be deleted. There is no real gain of information compared to figure 4. Figure C1 is used to show the scale of the whole flux dataset and their distribution. In Figure 4, data are split between seasons. Figure C_1 is referred to when commenting on the distribution of the dataset and to show the scale of the lake CO_2 fluxes in comparison with the other time series, which explains in part why we could not perform ANN gap filling.
- Table S2: RMSE is given in % of what? Please use flux units. OK What is the mean random error given in the last table row? Please explain. The mean random error given for each modeled series (CH₄ fen, CH₄ lake, CO₂ fen) is the mean of the standard deviation for each individual value used for gap filling. Please see our response to referee #1 for a more detailed explanation. The information will be added in the text and mentioned in the Table caption.

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Year-round CH₄ and CO₂ flux dynamics in two contrasting freshwater ecosystems of the subarctic

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Abstract. Lakes and wetlands, common ecosystems of the high northern latitudes, exchange large amounts of the climateforcing gases methane (CH₄) and carbon dioxide (CO₂) with the atmosphere. The magnitude of these fluxes and the processes driving them are still uncertain, particularly for subarctic and Arctic lakes where direct measurements of CH₄ and CO₂ emissions are often of low temporal resolution and are rarely sustained throughout the entire year.

- 5 Using the eddy covariance method, we measured surface-atmosphere exchange of CH_4 and CO_2 during 2.5 years in a thawed fen and a shallow lake of a subarctic peatland complex. Gas exchange at the fen exhibited the expected seasonality of a subarctic wetland with maximum CH_4 emissions and CO_2 uptake in summer, as well as low but continuous emissions of CH_4 and CO_2 throughout the snow-covered winter. The seasonality of lake fluxes differed, with maximum CO_2 and CH_4 flux rates recorded at spring thaw. During the ice-free seasons, we could identify surface CH_4 emissions as mostly ebullition
- 10 events with a seasonal trend in the magnitude of the release, while a net CO_2 flux indicated photosynthetic activity. We found correlations between surface CH_4 emissions and surface sediment temperature, as well as between diel CO_2 uptake and diel solar input. At ice outDuring spring, the breakdown of thermal stratification following ice thaw triggered the degassing of both CH_4 and CO_2 . This spring burst was observed in two consecutive years for both gases, with a large inter-annual variability in the magnitude of the CH_4 degassing.
- On the annual scale, spring emissions converted the lake from a small CO₂ sink to a CO₂ source. 80 % of total annual carbon emissions from the lake were emitted as CO₂. The annual total carbon exchange per unit area was highest at the fen, which was an annual sink of carbon with respect to the atmosphere. Continuous respiration during the winter partly counteracted the fen summer sink by accounting for, as both CH₄ and CO₂, 33 % of annual carbon exchange. Our study <u>underlines shows</u> (1) the importance of overturn periods (spring or fall) for the annual CH₄ and CO₂ emissions of northern lakes, (2) the
- 20 significance of lakes as atmospheric carbon sources in subarctic landscapes while fens can be strong carbon sink and (3) the potential for ecosystem-scale eddy covariance measurements to improve the understanding of short-term processes driving lake-atmosphere exchange of CH₄ and CO₂.

Keywords: eddy covariance, methane, carbon dioxide, lake, wetland, artificial neural networks, carbon budget, subarctic

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1. Introduction

Lakes and wetlands are linked to the atmospheric carbon pool via the exchange of methane (CH_4) and carbon dioxide (CO_2) , which are two important climate-forcing gases (Myhre et al., 2013). While wetlands have been a focus of study due to their high CH_4 source function (Christensen et al., 2003; Crill et al., 1988; Olefeldt et al., 2013) and carbon sequestration capacity

- 5 (Kayranli et al., 2009; Whiting and Chanton, 2001), lakes have only recently been incorporated together with streams as a separate source in global CH₄ budgets with an uncertain global emission rate of 8–73 Tg CH₄ yr⁻¹ (Kirschke et al., 2013). The low number of experimental studies and the variability in the magnitude of emissions across lake types (Wik et al., 2016) explains some of this order of magnitude scalelarge uncertainty. Carbon emissions from lakes outweigh part of the land carbon sink, because they emit CH₄ (Bastviken et al 2011) and because they respire as CO₂ a portion of the carbon that
- 10 is transported laterally from terrestrial soils to lakes (Algesten et al., 2004; Battin et al., 2009; Cole et al., 2007; Tranvik et al., 2009). Hence, lakes play an important role within the terrestrial carbon budget.

Wetlands and lakes are particularly abundant in the subarctic and boreal regions (Smith et al., 2007; Verpoorter et al., 2014), where climate warming is occurring at a faster pace than in the rest of the world (Serreze and Barry, 2011). In this context, freshwaters have received increasing attention over the past decade, due to the potential for lakes and particularly Arctic thermokarst lakes to exert a feedback on climate warming through large CH_4 emissions (Walter Anthony et al., 2016; Walter

- 15 thermokarst lakes to exert a feedback on climate warming through large CH_4 emissions (Walter Anthony et al., 2016; Walter et al., 2006). While non-thermokarst, post-glacial lakes emit less CH_4 per unit area (Sepulveda-Jauregui et al., 2015; Wik et al., 2016) they cover a larger area and may as a whole emit half of the total CH_4 emissions (16.5 +- 9.2 Tg CH_4 yr⁻¹) recently attributed to northern (> 50 N) lakes and ponds (Wik et al., 2016).
- Biogenic CH₄ production (methanogenesis) occurs in anoxic environments such as lake sediments and water-saturated peat
 (Cicerone and Oremland, 1988). The process is controlled by the interplay between temperature and the input of organic matter (Kelly and Chynoweth, 1981; Yvon-Durocher et al., 2014; Zeikus and Winfrey, 1976). In lakes and waterlogged wetlands, CH₄ reaches the atmosphere from its production zone via direct bubble release up to the surface (ebullition), through emergent vascular plants, or via turbulence-driven diffusion through the water column (Bastviken et al., 2004; Lai, 2009; Rudd and Hamilton, 1978). The net flux of CH₄ at the lake surface is a balance between the production in the sediments and the oxidation of CH₄ into CO₂ at oxic/anoxic boundaries within the water column (Casper, 1992). In shallow
- lakes, ebullition is a main pathway for CH_4 to reach the atmosphere while bypassing the oxidation zones (Bastviken et al., 2004). In wetlands, transport from the peat soil to the surface through vascular plants by passive diffusion or by pressurization effects depending on the plant species (Brix et al., 1992), is an efficient pathway for CH_4 to avoid oxidation in the soil and water before reaching the atmosphere (Joabsson and Christensen, 2001).
- 30 Dissolved CO₂ in lake<u>s</u> is produced throughout the water column and sediments (Casper et al., 2000) or is directly imported from the catchment (Maberly et al., 2013; Weyhenmeyer et al., 2015). *In situ* production comes from the mineralization or the photochemical oxidation of carbon (C) input from the surrounding catchment (Cory et al., 2014; Dillon and Molot, 1997;

Duarte and Prairie, 2005) and from the degradation of locally produced organic carbon. CO_2 exchange across the air-water interface is primarily via diffusive release rather than ebullition (e.g. Casper et al., 2000). Lake waters are generally observed to be supersaturated in CO_2 with respect to atmospheric values due to in-lake respiration processes outweighing rates of primary production (Duarte and Prairie, 2005; Sobek et al., 2005). Hence they are generally CO_2 sources to the atmosphere,

5 albeit nutrient-rich lakes and ponds can be small CO₂ sinks during summer months (Huotari et al., 2011) or an entire summer season (Laurion et al., 2010; Pacheco et al., 2013; Shao et al., 2015; Striegl and Michmerhuizen, 1998; Tank et al., 2009).

Near-surface atmospheric forcing is a key driver for the transport and net emissions of gases from a lake to the atmosphere. Ebullition of CH_4 in lakes is partly triggered by water level changes or drops in atmospheric pressure (Casper et al., 2000; Mattson and Likens, 1990), as a decrease in the hydrostatic pressure of the overlying water column on gas saturated

- 10 sediments favors the release of bubbles (Varadharajan and Hemond, 2012). Wind-driven turbulence is a recognized driver of diffusion-limited exchange of CO_2 and CH_4 across the lake-water interface (Sebacher et al., 1983; Wanninkhof et al., 1985). Convective mixing due to the cooling of the lake surface or following the breakdown of thermal stratification in the water column can increase advection of gas-rich water from the lake bottom thus enhancing the diffusion-limited release of gases to the atmosphere (Eugster, 2003; MacIntyre et al., 2010; Podgrajsek et al., 2015), especially if the main source of those
- 15 gases is the sediments as is the case for CH_4 .

Northern ecosystems have strong seasonal contrasts with short growing seasons and long snow-covered winters. A snow cover insulates the soil from very cold air temperature (Bubier et al., 2002), while an ice lid on a lake temporarily inhibits the exchanges of gas and heat between the water and the atmosphere (e.g. Greenbank, 1945) and greatly dampens wind-driven turbulence in the water column. In lakes, gases can accumulate at the bottom during long stratification periods, or under lake

- 20 ice. Lake overturn events most often occur in spring for seasonally ice-covered lake and/or at fall if lakes thermally stratify during summer (Kirillin et al., 2012; Wetzel, 2001). Overturn events can thus lead to the fast release of accumulated gas to the atmosphere (Jammet et al., 2015; Michmerhuizen et al., 1996; Phelps et al., 1998) but also to the input of atmospheric oxygen, thus increasing the potential for methanotrophy (Kankaala et al., 2006; Schubert et al., 2012). Extension of observations across all seasons of the year is rare in high northern latitudes, particularly for lakes, yet indispensable to reduce
- the uncertainty in the magnitude of annual carbon exchange and to improve understanding of the processes driving them.

Common methods to measure lake-atmosphere fluxes include floating chambers (e.g. Bastviken et al., 2004), gas transfer models (e.g. Cole and Caraco 1998), and bubble traps (e.g. Walter et al., 2008; Wik et al., 2013; Sepulveda-Jauregui et al., 2015). If these methods are not integrated and combined, they inherently omit part of the total surface flux. The application of the eddy covariance (EC) method (Aubinet et al., 2012; Moncrieff et al., 1997) to lake environments (e.g. Anderson et al.,

1999; Eugster, 2003; Huotari et al., 2011; Mammarella et al., 2015; Podgrajsek et al., 2014; Shao et al., 2015) offers long-term flux monitoring and is a potential methodological solution for solving the spatial and temporal issues of measuring total gas exchange in lakes. A <u>fF</u>ew studies, so far, have used eddy covariance to quantify long-term CO₂ emissions from boreal

lake<u>s</u> (Huttunen et al 2011) as well as CH_4 emissions from boreal (Podgrajsek et al., 2014) or subarctic lakes (Jammet et al., 2015). We report here one of the first year-round eddy covariance measurements of both CH_4 and CO_2 fluxes from a seasonally ice-covered lake.

Surface fluxes were monitored using the eddy covariance method in a subarctic permafrost peatland undergoing thaw, a

- 5 landscape with a high percentage of pond and lake coverage. The location of the flux tower allowed for measurements to alternate between surface fluxes from a shallow lake and from a permafrost-free, waterlogged fen-type wetland. The overall aim of this work was to quantify year-round carbon fluxes in a post-glacial lake, a type widely present around the subarctic, as compared to the adjacent fen. Specifically, the objectives were (1) to compare the seasonality of CH_4 and CO_2 fluxes from two contrasting subarctic ecosystems (lake and fen), (2) to explore the possibility of identifying short-term environmental
- 10 controls on the surface-atmosphere exchange of CH_4 and CO_2 in a lake, using high, sub-daily temporal resolution measurements covering all seasons of the year and (3) to assess and compare the annual atmospheric carbon budget of a lake and a wetland.

2. Materials and methods

15 **2.1 Study site**

Stordalen Mire is a subarctic peatland complex (68°20' N, 19°03' E) with a high lake and pond coverage, located near Abisko in Northern Sweden. Mean annual temperature in the Abisko region has been increasing and fluctuating around 0°C since the 1990's as part of an accelerated warming trend <u>-(Callaghan et al., 2010)(Callaghan et al., 2010)</u>. Permafrost, which is discontinuously present in the local mires, has been thawing at an increased rate since the 1990's in the peatlands of the

- 20 region, sometimes disappearing completely (Åkerman and Johansson, 2008). In Stordalen Mire permafrost thawing has led to changes in microtopography, which controls local hydrology, which in turn leads to vegetation shifts (Christensen, 2004; <u>Malmer et al., 2005)(Christensen, 2004; Malmer et al., 2005)</u>. This accentuates the heterogeneity of a landscape comprised of elevated palsas with permafrost, thawing lawns with thermokarst ponds, and permafrost-free, water-saturated fens. The mire is bordered by post-glacial lakes on its western, northern and eastern edges. This study focuses on the lake Villasjön
- (Fig. 1) and the adjacent fen to the west of the tower. <u>According to Olefeldt and Roulet (2012)</u>**T**, the two ecosystems may beare hydrologically connected with a directional flow from the lake to the fen-(Olefeldt and Roulet, 2012).

The wetter fen areas have expanded as a result of permafrost thaw over the past decades (Johansson et al., 2006). The water table of the fen is at or above the surface throughout the year. The dominant vegetation species are vascular plants *Carex rostrata* and *Eriophorium angustifolium*. Villasjön is the largest (0.17 km²) lake of the 15 km² wide Stordalen catchment containing 27 lakes (Lundin et al., 2013). It has a mean and maximum depth of 0.7 m and 1.3 m, respectively (Jackowicz

30 containing 27 lakes (Lundin et al., 2013). It has a mean and maximum depth of 0.7 m and 1.3 m, respectively (Jackowicz-

Korczyński et al., 2010; Wik et al., 2013). The upstream catchment of Villasjön is dominated by birch forest (Olefeldt and Roulet, 2012), while its western shore is bordered by thawing palsas. <u>During snow melt, Tthere is a vernal, small surface</u> inflow feeding Villasjön on the east <u>during snow melt</u> (Wik et al., 2013). <u>but the lake can be considered a hydrologically</u> elosed system in winter (Boereboom et al., 2012) and <u>The lake</u> usually freezes to the bottom in winter. The DOC

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concentration in the lake water has been measured to be 8.1 mg l^{-1} in 2008 (Olefeldt and Roulet, 2012). Aquatic vegetation is present in the lake, on its bottom as algae, as submerged plants within its southern arm and in low density of emergent macrophytes at its shores.

2.2 Eddy covariance measurements

2.2.1 Measurement set-up

- Between June 2012 and December 2014, the surface–atmosphere exchange of CH₄, CO₂–, latent heat and sensible heat flux has been monitored nearly continuously with an eddy covariance set-up located at the shore of Villasjön (Fig. 1). Data were logged on a CR1000 (Campbell Scientific, Inc., UT, USA) until May 2013; from June 2013 it was replaced with a CR3000 (Campbell Scientific, Inc., UT, USA). The 2.92 m high mast was equipped with a 3-D sonic anemometer (R3-50, Gill Instruments Ltd.) sampling wind components and sonic temperature at 10 Hz. Throughout the study period, ambient molar
- 15 densities of CO₂ and H₂O were sampled at 10 Hz with an open path infrared gas analyzer (IRGA), model LI7500 (LICOR Environment, NE, USA) mounted on the mast at 2.50 m height. Following lightning that hit the electric grid in Stordalen Mire on 27 July 2013, the initial LI7500 ceased functioning and was replaced on 1 October 2013 by a different instrument of the same model.

From June 2012 to May 2013, ambient CH₄ mixing ratiomole fraction was sampled at 10 Hz with a closed path Fast
Greenhouse Gas Analyzer (FGGA, Los Gatos Research, CA, USA) in air that was taken from a gas inlet located at 2.50 m
height on the mast through a 6 mm inner diameter polyethylene (PE) tube using a dry scroll pump (Varian TriScroll 300). The efficient flow rate in the 95 m long sampling line was 16 L min-1, ensuring the maintenance of turbulent conditions (Reynolds number ca. 4025). On 5 June 2013, the tube was replaced with an 8 mm inner diameter PE tube, which changed the flow rate in the sampling line to 23.88 L min-1 (Reynolds number ca. 4099). On 4 August 2013, the closed path CH₄
system was renewed: the IRGA was changed to a FGGA model 911-010 (Los Gatos Research, CA, USA). Due to instrumental maintenance, the IRGA was offline between February and March 2014 and replaced on 24 March 2014 by the previous FGGA. From August 2013 to December 2014, due to a failure in the electronic connection between the gas analyzer and the data logger, the raw CH₄ data measured by the FGGA were pre-processed in order to align the timestamp and frequency of the gas analyzer recordings with the time stamp of the logger sampling the wind data. The synchronization

procedure was quality-checked after flux computation (Text S2). Each change in the CH_4 measurement set-up was taken in account in the flux calculation.

2.2.2 Flux calculation and quality-check

CO₂, CH₄, sensible and latent heat fluxes as well as atmospheric turbulence quantities were calculated and output as 30 min 5 averages using the open source software EddyPro version 5.2. (hosted by LICOR Environment, USA). Processing of the 10 Hz raw data followed standard eddy covariance procedures (Aubinet et al., 2012; Lee et al., 2005); methodological choices differed partly for CH₄ and CO₂ flux processing as detailed in Appendix A.

The 30min averaged fluxes were quality checked to detect measurement errors and to ensure the fulfillment of theoretical assumptions for the application of the eddy covariance method. Spikes were present in the CO₂ flux time series, which can

- be due to weather conditions, to fast changes in the atmosphere's turbulent conditions or to faulty instrumentation. Outliers 10 in the CO₂ flux dataset were detected using the median of absolute deviation from the median (MAD) as described in Papale et al. (2006) using a threshold z = 4 and with no distinction between day and night. -Additionally, CO₂ flux averaging periods were rejected when the number of spikes per half hour was > 100 (Mammarella et al., 2015) and when skewness and kurtosis were outside the [-2,2] and [1,8] ranges (Vickers and Mahrt, 1997), respectively. Required flux stationarity (FST)
- within the 30 min flux averaging period was ensured by rejecting each flux value when the FST criterion as defined by 15 Foken and Wichura (1996) was above 0.3, a strict criterion that was chosen, given the challenging footprint on the lake side, to ensure that fluxes represented the surface of interest. The fen part of the CO_2 flux dataset was additionally filtered for poorly developed turbulence (Mauder and Foken, 2006) and u^* -filtered (Papale et al., 2006) with a threshold determined to be 0.1 m s⁻¹ using the online tool available at https://www.bgc-jena.mpg.de/bgi/index.php/Services/REddyProcWeb
- 20 (Reichstein et al., 2005) (Papale et al., 2006). Furthermore, tThe impact of self-heating on the open path CO₂ flux measurements (Burba et al., 2008) is a potential issue when using the LI7500 model, especially for such low range fluxes on the lake side low flux environments, but could lt could not be correctly quantified in this study and was thus not applied to avoid a potentially large systematic error. A rough estimate of the correction indicated that it could increase the magnitude of the CO₂ fluxes by 0.28 µmol m⁻² s⁻¹ on average-Besides, the self heating issue of this gas analyzer model results usually in an apparent uptake rather than in an excessive degassing at very cold temperatures (Burba et al., 2008).
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Quality-check and screening of the CH₄ flux time series included rejection of averaging periods when the number of spikes per half hour was > 100. Fluxes were rejected when skewness and kurtosis were outside the pre-cited range, in winter only, due to the pulse-character of CH₄ emissions during non-winter periods that could be misinterpreted as faulty raw data due to a high skewness (Jammet et al., 2015). Flux stationarity within the averaging time period was also ensured by rejecting values when the FST criterion was above 0.3. Additionally, flux values for which a time lag was not found within the

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plausibility time lag window were rejected (Eugster et al., 2011; Wille et al., 2008). Gaps were-present initially present in the

flux dataset due to instrument malfunctioning, <u>particularly in winter</u>, and power cuts (Fig. S6). After quality-check and filtering, in total 13474 CH₄ fluxes and 11629 CO₂ fluxes were available for further analysis (Table S1, Fig. S6). Data rejection rate was high; eddy covariance studies in lake environments usually report high rejection rates from quality-check routines (Jonsson et al., 2008; Mammarella et al., 2015; Nordbo et al., 2011)

5 2.3 Flux footprint and partitioning between lake and wetland

As in Jammet et al. (2015), the bidirectional wind pattern at the Stordalen Mire was used to partition the flux dataset into two main wind sectors which crossed the two different studied ecosystems. To ensure a clear distinction between the lake and the fen fluxes and the homogeneity of each surface, the lake sector was conservatively defined as 20° —135°, and the fen sector as 210° —330° ($0^{\circ} = 360^{\circ}$ is true north). Over the study period, 46.2 % of the measured fluxes originated from the lake sector, 45.7 % of the measured fluxes originated from the fen sector, and the residual 8.1 % of the measured flux data was excluded, for being of lower wind speed and originating from mixed sources. Each season was nevertheless well represented

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in the flux dataset of each ecosystem (fen and lake) thanks to regular shifts in wind direction. The flux footprint was calculated using a 2D model developed by Kljun et al. (2015). Inputs to the model include turbulence

- quantities measured at the tower (friction velocity (u^*) , standard deviation of cross-wind velocity (σ_v) , Obukhov length, 15 horizontal wind speed), height of the boundary layer (derived from the Era-Interim reanalysis product, (Dee et al., 2011)), 16 the measurement height and surface roughness length which was separately estimated for the lake (0.001 m) and the fen 17 sectors (0.002 m in winter to a maximum of 0.10 m in summer, accounting for vegetation growth). Footprints were 18 calculated for each 30 min time step where the required data was available, and averaged for the periods of interest (summer 19 and winter).
- According to the footprint the-model, most of the flux measured at the tower (peak fetch) originated from a distance of 38 m on the fen sector and 73 m on the lake sector, on average during the ice-free season. The lake surface was within the cumulative 80 % of flux footprint during the ice-free seasons (Fig. 1). In winter, the footprint model revealed that the 80 % cumulative footprint, on the lake side, included part of the land in the middle of the lake (Fig. S1). This is likely due to the lower roughness length (snow cover) but also to more stable atmospheric conditions: the sensible heat flux H is negative.
- 25 during most of the winter and the atmospheric stability parameter indicated more stable conditions in winter as compared to summer conditions. Thus, to avoid large contamination of the land respiration in the winter lake fluxes due to an extended footprint over the lake shores, the winter fluxes from the lake sector were only kept for further analysis when the standard deviation of lateral wind velocity $\sigma_v \leq 1 \text{ m s}^{-1}$ (Forbrich et al. (2011)) in order to limit lateral contamination of CO₂ fluxes into the footprint area of interest. - This removed an additional 3.7 % of the lake CO₂ flux data set.

2.4 Ancillary measurements

Supporting environmental variables were measured at 1 Hz and averaged and logged every half hour. Temperature probes (T107, Campbell Scientific, Inc., UT, USA) were installed in the peat 5 m south-west from the tower at 5 cm, 10 cm, 25 cm, and 50 cm depths. Net radiation at the fen surface was recorded with an REBS Net Radiometer, model Q7.1 (Campbell

- 5 Scientific, Inc., UT, USA). Air temperature used in this study was measured at 2 m height on a mast located in the middle of the lake (Fig. 1) with a CS215 probe (Campbell Scientific, Inc., UT, USA). At the same mast, radiation components were measured with a CNR4 Net Radiometer (Kipp & Zonen, The Netherlands) from which net radiation at the lake surface was computed. Water temperature in the center of the lake (Fig. 1) was measured at 10 cm, 30 cm, 50 cm, and 100 cm depths with intercalibrated HOBO Water Temp Pro v2 loggers (Onset Computer Corporation, MA, USA). The loggers are
- 10 suspended on a nylon line from a mooring float, which stays at the surface throughout the year. The string assembly was designed so that the bottom sensor at 100 cm depth is in the surface sediment. Lake surface albedo was computed daily as the midday ratio (10:00 to 13:00) of the shortwave radiation components, and used in combination with temperature data to delimit the ice cover seasons. Air pressure and precipitation were measured at a weather station 640 m south from the eddy covariance mast.

15 **2.5** Gap filling of CH₄ and CO₂ flux time series

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The estimation of annual carbon exchange budgets required filling the gaps in the CH_4 and CO_2 flux time series. When working with highly skewed flux datasets <u>such as CH_4 emissions from lakes</u> (Fig. C1), integrating the mean flux over the whole time period may lead to important overestimation. High frequency CO_2 and CH_4 flux measurements in lake ecosystems are still rare. There is <u>yet</u> no published account of gap filling CH_4 fluxes measured with eddy covariance., and relationships between high resolution fluxes and environmental variables are not yet well characterized.

- In the present study, G_{g} ap filling of the CH₄ flux time series was performed separately on the lake and fen flux datasets, using artificial neural networks (ANN) (Moffat et al., 2010; Papale and Valentini, 2003). Artificial neural networks (ANNs) are multivariate, non-linear regression models that are fully empirical: the observational data are used to constrain the model's numerical relationship between the inputs (independent variables i.e. environmental drivers) and output (dependent variable i.e. fluxes) (Moffat et al., 2010). ANNs have been tested and successfully used to estimate missing values and gap
- fill CO₂ flux time series measured with eddy covariance in forests (Moffat et al., 2007; Papale and Valentini, 2003) or in urban terrain (Järvi et al., 2012) and to gap fill CH₄ emissions in wetlands (Dengel et al., 2013). <u>To our knowledge, we</u> present the first attempt of using ANNs to gap fill eddy covariance measurements of CH₄ emissions from a lake.

Three ANN models were built separately on CH_4 emissions from the fen, CH_4 emissions from the lake and CO_2 emissions 30 from the fen, on the hourly scale. Model development followed workflows introduced by Papale and Valentini (2003), Moffat et al. (2007) and Dengel et al. (2013). Environmental variables to be included as inputs were selected according to their physiological relevance to the production and transport of CH_4 and CO_2 and their transport from the surface to the atmosphere, as reported in the literature. The relevance of the drivers was confirmed by correlation analysis. Input variables used for each model are reported in Table B1. A further detailed description of ANN model development and list of input variables can be found in supplementary Text S1 and Table S2. Gaps in the measured CH_4 and CO_2 flux time series were then replaced by predicted values, and the annual sums were computed by integrating the hourly flux values over time. The performance of the ANN models was assessed by comparing the predicted values with original observed values over the whole dataset (Appendix B). The goodness of fit was quantified with the coefficient of determination r^2 and the absolute root mean square error (RMSE). The ANN gap filling was most performant on the fen CH_4 flux dataset (Appendix B).

10 The ANN method did not perform as well on the lake CO_2 flux time series, which comprised a large amount of gaps and a high signal-to-noise ratio. Thus ANN results were thus excluded for the lake CO_2 fluxes to avoid introducing a high and unnecessary uncertainty. Instead, the seasonal lake CO_2 exchange was computed by multiplying the mean flux rate during each season with per the number of days. Considering the normal distribution of the CO_2 lake fluxes (Fig. C1), this method was considered an acceptable way of filling missing values. In the case of a highly positively skewed dataset such as the CH_4

15 fluxes this would lead to a large positive bias and overestimation of the annual sum.

2.6 Uncertainty analysis

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The total random error is a composite of errors associated with instrument noise, the stochastic nature of turbulence, the instrument precision and the variation of the flux footprint (Moncrieff et al., 1996). The <u>absolute relative</u> random error increases with the magnitude of the flux (Richardson et al., 2006), while over time it decreases with increasing size of the dataset because of its random nature (Moncrieff et al., 1996). It is therefore negligible when propagated over annual sums but can be important for single flux values. The random error was calculated as the sampling error for each flux value in the flux calculation software using the method of Finkelstein and Sims (2001). Considering that the total flux error of EC gives an estimation of the value under which a flux cannot significantly be differentiated from noise (the precision of the measurements), we considered this error estimation as a detection limit of the flux measurement system.

- The mean random error of the quality-checked measured CH_4 fluxes was 2.9 ± 4.3 (mean \pm STD) nmol m⁻² s⁻¹ at the lake and 4.7 ± 3.8 nmol m⁻² s⁻¹ at the fen, which corresponds to 7.6 % and 6 % of the overall mean measured fluxes, respectively. This is in the lower end of ranges reported for CO₂ and energy fluxes. The mean random error of the individual measured lake CH₄ fluxes was highest in winter (24 %) and lowest during ice-outthe thaw season (11 %) when the largest fluxes were measured. The mean random error of the quality-checked measured CO₂ fluxes was $0.20 \pm 0.57 \mu mol m^{-2} s^{-1}$ at the lake and
- 30 $0.32 \pm 0.48 \ \mu\text{mol m}^{-2} \ \text{s}^{-1}$ at the fen, equivalent to 31 % and 13 %, respectively, of the overall mean absolute measured flux, indicating that our set-up was able to measure both CH₄ and CO₂ fluxes observable at our site. These values are comparable
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to what has been reported in vegetated (Finkelstein and Sims, 2001), urban (Järvi et al., 2012), boreal lake (Mammarella et al., 2015) and other typical eddy covariance sites (Rannik et al., 2016) (Rannik et al., 2016). In spring the random error of CO_2 fluxes at the lake was 18 % of the measured flux, however during the ice-free season it was above the absolute measured flux in 20 % of the cases. Thus air-lake CO_2 exchange rates during the summer were low and sometimes close to the dataction limit

5 the detection limit.

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The random <u>uncertainty error</u> of the fluxes modeled with ANN (<u>Appendix B</u>) was on average 4 nmol m⁻² s⁻¹ for fen CH₄ fluxes; 0.23 µmol m⁻² s⁻¹ for fen CO2 fluxes; 11 nmol m⁻² s⁻¹ for lake CH₄ fluxes. These errors were small when propagated onto seasonal and annual sums using random error propagation principle (Moncrieff et al., 1996). The systematic bias on the annual flux due to the gap filling method, the bias error (BE), was calculated on the seasonal and annual flux sums as in Moffat et al (2007) as the sum of the difference between the predicted values p_i and the observed values o_i :

$$BE = \frac{1}{N} \sum (p_i - o_i) \tag{1}$$

where N is the number of gap filled values in the flux time series. The bias error adds up over time. It was multiplied by the number of gap filled values to obtain a total seasonal and annual offset (Moffat et al., 2007). The offset can be the largest source of uncertainty in the computation of annual budgets. It is dependent on the accuracy of the model but also on the amount of gaps in the original dataset to fill. The systematic offset due to gap filling was larger for annual CO_2 fen fluxes (Table 3), likely because of the higher noise in the measurements and higher amount of gaps during the second year. For total lake CO_2 fluxes, the annual BE was calculated in the same way, using seasonal mean as the predicted values. The resulting offset due to gap filling was close to zero (Table 3), because flux values were normally distributed, which confirms that using the mean was not inducing a significant bias for this particular dataset.

20 **2.7 Definition of seasons**

Observations started in June 2012 and were sustained nearly continuously until December 2014. We adopted a lake-centric definition of seasons based on the lake ice phenology. A full year was defined from 1 June to 31 May of the next year, so that an entire ice-cover season was included in a given year. This keeps the connected thaw period and previous ice-cover season within the same year. The year was further divided into an ice-cover season (winter), an ice-free season (summer and

25 fall) and a thaw season (spring). The thaw season represents a transitional period during which the snow cover and lake ice melt. It is separated from summer, since the hydrological and biogeochemical dynamics in seasonally ice-covered lakes differ from the rest of the open water season.

Daily air temperature was used to define seasons. The start of the ice-cover season was defined as the start of the lake freeze up, i.e. the first day on which daily mean air temperature is below zero for three consecutive days. Further, this date coincided each year (2012 to 2014) with the formation of thermal stratification in the water column (Fig. 2d) due to a rise in

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bottom water temperature right after the first day of freezing (Fig. 2a, blue line). This indicates the inhibition of direct heat exchange between the lake and the atmosphere when ice forms at the lake surface. In the first year (2012), the first day of the ice-cover season was confirmed by visual observation of ice over the whole lake surface while the following years, these combined temperature observations were used to define the start of ice cover.

- 5 The end of the ice-cover season was defined as the start of thaw, i.e. the first date on which daily mean air temperature rose above 0°C for at least three consecutive days. This date preceded by 1 to 2 days the temperature rise to 0°C in the surface (10 cm) of the lake ice. Further, the start of the thaw season could be confirmed by the increase of daily energy input (upwelling > downwelling radiation) mean daily net radiation to net positive values at the lake and the fen surfaces (Fig. 2b2c) and by a decrease in lake albedo (Fig. 2e2d) until reaching open water values (mean = 5 %0.05). We thereby defined two complete
- 10 ice-cover seasons during the study period: from 15 October 2012 to 14 April 2013 (182 days) and from 16 October 2013 to 10 April 2014 (177 days). In 2014, freeze up of the lake occurred on 10 October. The ice-cover seasons were systematically characterized by negative daily energy input at the lake and at the fen (Fig. 2). Thus our season definition based on daily air temperature was robust and reproducible each year.

The end of spring <u>(i.e. thaw season)</u> and beginning of the ice-free season was defined as the first date with a daily temperature gradient in the water column close to 0°C m⁻¹ after the spring overturn, i.e. when the lake enters its isothermal conditions after complete ice thaw. The dataset of this study covers two complete years plus an additional ice-free season, i.e. three ice-free seasons, two ice-cover seasons and two springs (ice out<u>thaw</u> seasons). Seasons dates and lengths are summarized in Table 1.

3. Results

20 **3.1** Environmental conditions and lake climatology

- Mean annual (June to May) air temperature measured at the Abisko Scientific Research Station was -0.3° C in 2012-2013 and 0.9° C in 2013-2014. The latter was significantly above the long-term average (1913-2014) of $-0.5 \pm 1^{\circ}$ C (mean \pm STD). The ice-free season was warmest in 2014 and coolest in 2012 (Table 1). This difference between years was reflected both in the mean daily peat temperature measured at 10 cm depth and in the mean daily lake water temperatures, which were warmest in
- 25 2014 (Figure 2; Table 1). The differences in-mean annual-air temperature observed over our study period coincide-correlated with an increase in total net radiation at the lake and fen surfacestotal incoming solar radiation, both annually and during the ice-free periodperiods during 2012 to 2014 (Table 1). The winter 2013-2014 was on average 2.1°C warmer than the previous one and the ice-cover season was 5 days shorter due to an earlier thaw start. The thaw period started earlier in 2014 but lasted longer (Fig. 2d, Table 1); complete lake overturn (isothermal water column) following ice thaw occurred 7 days later than in 2013.
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The development pattern of thermal stratification along lake depth at freeze up and its breakdown in spring was repeated each yearsimilar in both years (Fig. 2d). Both in spring 2013 and 2014, the lake overturn occurred after the development of large strong thermal stratification along lake depth in the lake during ice outthaw (Fig.2e-fd). Temperature at the bottom of the lake was up to 4°C warmer than the surface in spring 2013 and up to 6°C warmer in 2014 likely indicating the

5 penetration of solar radiation through thinning ice before complete ice out and full water mixing. The thermal structure at the beginning of freeze up, as well as during the period of ice thaw preceding spring overturn in 2013, has been previously described in detail for this site in a winter-focused study (Jammet et al., 2015).

During the ice-free, summer season, there was slight to no thermal stratification in the water column (Fig. 2d). The shallow lake water column reacted quickly to temperature changes (Fig. 2a,d) and the dark bottom warmed up fast since solar

- radiation can reach the sediment surface. Thus, the lake had a polymictic behavior, i.e. it was regularly mixing to the bottom during the ice-free season, which ensures isothermal conditions in the water column throughout the summer. Water temperature reached a maximum of 23.8°C in July 2014. Conversely, a strong temperature gradient formed during winter (Fig. 2f). In both winters the surface sediment temperature dropped below <u>0</u>°C by February, with minima of -1.9°C in March 2013 and -1.6°C in February 2014. This suggests that the water column froze to the bottom. The temperature in the peat soil
- 15 and at the surface sediment in the lake were de-coupled from air temperature (Fig. 2a) showing the hindrance of heat exchange with the atmosphere due to the presence of ice and snow at the surface..

3.2 Year-round CH₄ and CO₂ fluxes

CH₄ emissions from the lake <u>followed_showed_a</u> highly skewed distribution (Fig. C1a); there was a large difference between the overall mean (40 nmol m⁻² s⁻¹) and overall median (12 nmol m⁻² s⁻¹). The seasonal flux pattern was characterized by low background emissions and occasional, large degassing events (Fig. 3) with 25 % of measured data above 111 nmol m⁻² s⁻¹ in the <u>ice-outthaw</u> period, and 5 % of measured data above 75 nmol m⁻² s⁻¹ within the ice-free season (Fig. 4). Over the full measurement period, summer emission rates averaged to 26 nmol m⁻² s⁻¹ (median 12 nmol m⁻² s⁻¹), <u>ice-outspring</u> emission rates to 84 nmol m⁻² s⁻¹ (median 33 nmol m⁻² s⁻¹) and winter emissions were at 2.8 nmol m⁻² s⁻¹ (median 3.0 nmol m⁻² s⁻¹). The mean and median of the winter emissions were not significantly different from the mean random error of the fluxes. CO₂ fluxes were close to normally distributed at the lake (Fig. C1b) and the overall mean rate was 0.22 µmol CO₂ m⁻² s⁻¹ (median 0.18 µmol m⁻² s⁻¹). There was a distinctive CO₂ outgassing at the time of water overturn during the spring of both 2013 and 2014 (Fig. 3d). The mean measured CO₂ exchange at the lake was significantly negative during the ice-free seasons (one sample t-test, p < 0.001), -0.14 µmol m-2 s-1, indicating a low but-net uptake of CO₂ (Fig. 4). Negative flux rates started right after the spring CO₂ outgassing in 2013 and 2014 (Fig. 3d). The highest CO₂ uptake rates were observed during the

30 summer of 2014, which was the warmest summer of the study period with highest solar radiation input (Table 1). In fall 2014, a burst of CO_2 was measured at the lake, which was not present in previous years (Fig. 3d). This fall burst was not

observed in the CH_4 flux measurements. Wintertime CO_2 emissions from the lake were significantly above the flux random error and significantly positive (Fig. 3d, Fig. 4). There was an inter-annual variability in the magnitude of the CH_4 spring degassing from the lake between the two years (Fig. 3c). In contrast, mean CO₂ degassing was higher during the second iceout thay periodseason, 0.78 μ mol CO₂ m⁻² s⁻¹ (median 0.47 μ mol CO₂ m⁻² s⁻¹) in 2013 and 0.99 μ mol CO₂ m⁻² s⁻¹ (median 0.75 μ mol CO₂ m⁻² s⁻¹) in 2014. Both CH₄ and CO₂ emissions during <u>ice outspring</u> were significantly higher than during the following ice-free season, in both years (Fig. S2).

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The distribution of CH_4 emissions from the fen was less skewed (Fig. C1a) and the overall measured mean was 77 nmol m⁻² s^{-1} (median 58 nmol m⁻² s⁻¹). The highest CH₄ emissions at the fen occurred during the ice-free season (Fig. 3), with a mean rate of 110 nmol m⁻² s⁻¹ (median 108 nmol m⁻² s⁻¹). Sustained CH₄ emissions were measured at the fen throughout the winter (Fig. 3), with a mean rate of 25 nmol $m^{-2} s^{-1}$ (median 25 nmol $m^{-2} s^{-1}$). Flux rates during the snow-melt and ice-outthaw season averaged to 35 nmol m⁻² s⁻¹ (median 33 nmol m⁻² s⁻¹). CO₂ fluxes from the fen averaged to -1.3 μ mol m⁻² s⁻¹ (median 0.2 μ mol m⁻² s⁻¹ during the ice-free seasons. CO₂ respiration was sustained throughout winter (Fig. 3b) at a mean rate of 0.8 μ mol m⁻² s⁻¹ and the release of CO₂ during the melt season was

15 Average annual seasonality of CH_4 and CO_2 fluxes in both ecosystems is shown in Fig. 5. The lake dominated CH_4 and CO_2 emissions during spring. During the ice-free seasons, on the contrary, the lake was a lower emitter of CH₄ than the fen per unit area, and its CO_2 exchange was close to neutral with a small uptake. There was a slight seasonality in CH_4 lake emissions during summer but, annually, CH₄ and CO₂ lake fluxes peaked in spring (Fig. 5). CH₄ emissions from the fen peaked in August and net CO₂ exchange peaked in July (Fig. 5). The emission of both gases occurred at lower rates but 20

continuously in winter.

low (Fig. 4).

3.3 Variability of lake-air carbon exchange within seasons

The ice-outhaw and ice-free periods are different in terms of flux dynamics. The dataset was therefore separated into seasons to explore controls on the lake fluxes. During the ice-free seasons, half-hourly CH₄ fluxes at the lake were characterized by degassing events that coincided with drops in atmospheric pressure (Fig. 6). Daily EC flux data were 25 compared with spatially averaged, daily ebullition fluxes measured in the lake with ebullition traps located nearby or within the tower footprint. The degassing events measured with the eddy covariance system coincided in timing and in magnitude with the daily ebullition fluxes measured with the ebullition traps (Fig. 6).

Due to the high skewness of the CH_4 lake flux dataset (Fig. C1) and the presence of outliers in both CO_2 and CH_4 flux datasets from the lake, we used Spearman's ρ coefficient, which is a statistical measure of association that is robust to

outliers and applicable to skewed distribution (Kowalski, 1972), to explore bivariate associations. Among potential flux 30

drivers, the highest correlation of ice-free CH₄ emissions was found with surface sediment temperature ($\rho = 0.49$, Table 2). Degassing events occurred most of the timeoften following-after an increase in surface sediment temperature (Fig. 6). When averaging all 30min CH₄ fluxes during the three ice-free seasons per bins of 1°C, an exponential regression between lake CH₄ fluxes and surface sediment temperature could explain 82 % of the variability in CH₄ emissions (Fig. 7).

- 5 Wind speed correlated best with CH_4 lake emissions during the *ice-outthaw* period (Table 2, $\rho = 0.40$). The relationship was weaker during the open water period. We observed a few CH_4 degassing events in summer that coincided with the likely mixing of the water column following a short period of thermal stratification (0.8 to 1°C gradient between 10 cm and 100 cm depths, Fig. S3a,b) but these were not systematic (Fig. S3c,d). During the ice-free season, there was a weak anti-correlation between CO_2 exchange at the lake and air and water temperature (Table 2). Wind speed correlated weakly with CO_2 lake
- 10 exchange, while there was a strong anti-correlation with sensible heat flux at the lake surface (Table 2), and with solar radiation input (Table 2).

During the <u>ice-outthaw</u> seasons, half-hourly CO₂ fluxes strongly correlated with CH₄ emissions ($\rho = 0.67$, p < 0.001, Table 2) and followed the same emission pattern at the half-hourly scale, both in 2013 and 2014 (Fig. S4). This correlation was not sustained during the open water seasons, when the two flux datasets had a very different short-term variability. Both CH₄ and

15 CO₂ degassing in spring positively correlated with increasing air temperature.

The diurnal course of CO_2 , CH_4 and turbulent energy fluxes at the lake and the fen was calculated on hourly fluxes. Only days with more than 75 % of hourly data coverage were selected. The median flux of each hour was then computed across all days within each year during the open-water season, and plotted along with the 25th and 75th percentiles (Fig. 8). There was no diel cycle visible in the fen CH_4 fluxes, while net CO_2 exchange at the fen surface showed a clear peak uptake at

- 20 noon. Lake methane fluxes tended to be higher fluxes in the morning hours. A systematic diel pattern was observed in the lake CO_2 fluxes during each open water season (Fig. 8), with a slight peak in the mornings in 2012 and 2013 while CO_2 uptake peaked in the middle of the day in 2014. The 24-hour cycle of sensible heat flux (H) at the lake peaked in the late morning (Fig. 8) at ca. 10 a.m. local time (median 20 W m⁻²), while being < 10 W m⁻² in the afternoon. The diel CO_2 pattern at the lake also coincided with daily variation in water surface and air temperature and was in antiphase with the diel pattern
- of sensible heat flux. Latent heat flux (LE) at the lake peaked in the afternoon. At the hourly time scale, solar radiation could explain 88 % of the diel variability in air-lake CO_2 exchange during the summer months (Fig. 9). This light response curve resembled the one measured at the fen, although less pronounced.

3.4.1 Performance of the ANN modelling

The predictive performances of the models depended on the amount of gaps in the original dataset and on the accuracy of the choice of environmental drivers. The ANN gap filling method was most performant on the fen dataset, achieving an r² of 0.88 during the training phase, and an r² of 0.85 between measured and predicted values over the whole dataset (expressing the capacity of generalization of the model) with a relative mean square error (RMSE) of 23 % (Fig. 10). The ANN gap filling of lake CH₄ fluxes achieved an r² of 0.71 in the training phase and an r² of 0.70 between predicted and measured fluxes on the whole dataset, with an RMSE of 51 %. The lake model was most accurate for periods with the best data coverage in the measured dataset (spring seasons, r² = 0.77). The lower accuracy of the model during the ice-free seasons (r² = 0.47) is also due to the pulse-character of lake ebullition, which was not always reproduced by the model, while the background seasonal trend was present.

For the fen CH_4 fluxes, the model was most accurate during the ice-free seasons when fen CH_4 emissions are tightly linked to peat temperature and least performant during the thaw periods. Unsurprisingly, the prediction performance of the models

15 was dependent on data coverage in the original dataset but also on the accuracy of the choice of environmental drivers. On the annual scale, both fen and lake models were most performant during the first year (June–May), which had the least amount of data loss, with an r^2 of 0.88 (RMSE 23 %) on the first year for the fen model and of 0.82 (RMSE 40 %) for the lake model between predicted and measured fluxes.

The ANN modeling was likewise performant on the fen CO_2 flux dataset, achieving an r² of 0.86 (RMSE 35 %) over the

20 whole dataset between measured and predicted value (Figu. 10). The model was most performant during the ice-free seasons. This can be explained by a better data coverage but also by better constrained processes during the growing season and on the annual scale, than during the snow-cover season, when additional drivers than the one selected as model inputs may play a role in the variability of the fluxes at the hourly scale (sensitivity to footprint changes, turbulence, variation in snow depth, partial melt, etc.).

25 3.4.2 Annual and seasonal sums

We report seasonal emissions for all available seasons, and for lake CO_2 total annual emissions for the first year only (June 2012 to May 2013), due to the absence of a robust gap filling model for the second year, which had least data coverage when data coverage was lower. During the ice-free seasons, total lake CO_2 flux was negative and total lake CH_4 flux was positive (Table 3). Annually, the largest contribution to total carbon exchange at the lake was from during the ice-outspring season,

whereas the ice-free season was quantitatively the most important period for the annual carbon exchange at the fen (Table 3). On average over both years, the ice-cover season accounted for 33 % of the fen annual carbon (CH_4+CO_2) exchange per m². During the first year, the lake C-emissions equaled 70 % of the total net fen C-exchange. On a carbon mass basis, CO_2 exchange dominated the total carbon budget. Total net annual carbon exchange (CH_4+CO_2) at the fen was -38.2 g C m⁻² in the first year and -52 g C m⁻² in the second year (average -45.1 g C m⁻² yr⁻¹), while the lake total carbon exchange was positive at 26.7 g C m⁻² yr⁻¹ (first year only), of which 80 % was emitted as CO_2 .

4. Discussion

4.1 Contrasting annual seasonality of carbon fluxes between lake and fen

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The average annual seasonality of the fluxes across the study period (Fig. 5) shows that both ecosystems had different peak timings in terms of CH_4 and CO_2 exchange. CH_4 emissions from the fen followed the expected seasonality of emissions from boreal and subarctic wetlands (Hargreaves et al., 2001; Jackowicz-Korczyński et al., 2010; Rinne et al., 2007). The dense emergent vegetation cover at the waterlogged fen dominated by vascular plants, which are efficient conduits for CH_4 to reach

- the atmosphere, -lead to maximum primary production of organic carbon during the summer. The continuous emission of CH₄ and CO₂ through snow during the winter season is an important feature of the annual flux cycle. <u>Unlike the ice cover at</u> the lake, which is complete, stems and branches sticking out of the snow at the fen site allow a sustained connection with the <u>atmosphere</u>. It indicates a less than complete ice cover over the fen unlike the ice cover on the lake. This limited-limits the trapping and the buildup of CH₄ in the fen under snow during winter. A previous study showed that CH₄ emissions from the
- 20 <u>this</u> fen <u>atduring snow melt-ice out</u> are correlated with air temperature thus with daily snow melt and release of trapped gases (Jammet et al., 2015), as seen elsewhere (Friborg et al., 1997; Gažovič et al., 2010). The flux rates during the <u>ice-outthaw</u> season were however much lower than during the summer. <u>Both-On the other hand, both CH₄ and CO₂ fluxes from the lake peaked during the <u>ice-outspring</u> season. These pulses coincided with the time of complete water overturn following <u>lake-ice</u> thaw_out on the lake₇ (Fig. 2) and can be explained by the release of gases previously stored in and under lake ice. The</u>
- 25 annual seasonality was measured from 2.5 years of measurements, and further years of observations are needed to evaluate and explain inter-annual variability in the magnitude of the emissions.

The mean annual CH₄ efflux from the lake is in line with a regional estimate of ebullition flux in post-glacial and glacial lakes (32.2 mg CH₄ m⁻² d⁻¹, Wik et al. (2016)). Mean fen CH₄ fluxes agreed well with chamber measurements conducted over the same period in *Eriophorum*-dominated plots in the Stordalen Mire (P. Crill, unpublished data). The mean CH₄ flux

measured during the ice-free seasons is within the in the upper range of summer CH₄ fluxes measured in northern wetlands that are dominated by sedges (ca. 40 to 280 mg CH₄ m⁻² d⁻¹, 25th –95th percentiles, Olefeldt et al., 2013). Measured CO₂ flux rates at the fen site also agreed with previous EC studies within the wettest part of the mire (e.g. Christensen et al., 2012) and with flux rates measured with gas chambers in sedge-dominated vegetation plots in previous years (Bäckstrand et al., 2010)

5 and during the study period (P. Crill, unpublished data), at the seasonal and annual scales. The fen is thus representative of minerotrophic northern fens where high CH_4 emissions are measured, due to year-round anoxia in the soil and to the dominance of vascular plants (Olefeldt et al., 2013).

Lakes that freeze solid in winter are not expected to emit a significant amount of CO_2 at the surface, unless ice-free holes caused by strong bubble seeps are present (e.g. Sepulveda-Jauregui et al., 2015), which we do not observe in the lakes of

- 10 Stordalen (Wik et al., 2011). The observation of CO_2 fluxes from the lake during the second winter at rates that are within the magnitude of land winter respiration (Fig. 3) was thus unexpected. The high winter flux rates were coincident with strong winds, increasing air temperature and high latent heat flux. Whether these are due to a physical evasion of CO_2 through snow over the lake surface or to lateral advection of land-emitted CO_2 is unclear. Increased ambient CO_2 concentration over the lake in winter may be an indication of non-turbulent transport of CO_2 from land. Indeed, tThe extended flux footprint in
- winter, which includes lake shores, might include part of the land in the middle of the lake, leading to vegetation-like flux magnitudes. Although this effect was limited by filtering for large wind dispersion during winter periods, part of the flux could still be influenced by land respiration to an extent that we cannot quantify. Furthermore, the impact of self heating on the open path CO₂ flux measurements (Burba et al., 2008) is a potential issue, especially for such low range fluxes on the lake side, but could not be correctly quantified in this study and was thus not applied to avoid a potentially large systematic
 error. Besides, the self heating issue of this gas analyzer model results usually in an apparent uptake rather than in an

excessive degassing at very cold temperatures (Burba et al., 2008).

During the summer, the <u>small-low flux range magnitude</u> of CO_2 fluxes from the lake resulted in a low signal-to-noise ratio (high relative random error). The uncertainty linked to the inclusion of low-frequency contributions_-is a problem that has been discussed widely in the eddy covariance community, particularly for low-flux environments (e.g. Sievers et al., 2015).

- The lake studied here can be qualified as a low flux environment with respect to CO_2 exchange during the open water season. Testing CO_2 flux calculation with a new method that removes the low frequency contributions (Sievers et al., 2015) on a portion of our data in July 2012 showed nevertheless that the summer fluxes at the lake were coincident with our measurement, showing with a negative mean and diel pattern with slight uptake during the day (J. Sievers, personal communication, 2016). This confirms-indicates that despite a high noise our CO_2 lake flux measurements during summer are
- 30 trustworthy. Besides, the flux footprint was representative of each ecosystem (lake vs. fen). The diel patterns of sensible and latent heat fluxes from the lake resembled those observed in boreal lakes (Mammarella et al., 2015; Nordbo et al., 2011; Shao et al., 2015; Vesala et al., 2006), which further supports that our EC measurement from the eastern sector are

representative of the lake surface. <u>Overall, CO_2 fluxes</u> -in the context of a low flux environment such as this lake should be interpreted with care; we provide here the best estimate possible with the available instrumentation at the time of the study.

4.2 Season-dependent transport pathways of CH₄ and CO₂ from the lake to the atmosphere

5 4.2.1 Ice-free season

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Eddy covariance measures a direct flux across the surface-atmosphere interface, spatially integrating over m² to km² all emitting pathways that are responsible for the transport of gas from the ecosystem to the atmosphere (i.e., total flux). In this study, CH₄ emissions from the lake during the ice-free seasons were dominated by short, large degassing events (Fig. 3, Fig. C1) that coincided with drops in atmospheric pressure. Furthermore, daily EC observations coincided with spatially averaged ebullition fluxes measured with bubble traps (Fig. 6), which supports that CH₄ fluxes are representative of the lake surface. These observations <u>also</u> suggest that the total CH₄ efflux from the lake during the ice-free seasons was mostly due to the release of bubbles formed in the sediments (ebullition), in line with previous observations that ebullition is the main pathway for CH₄ emissions in shallow lake areas (Bastviken et al., 2004).

- A strong relationship was found between CH₄ efflux and surface sediment temperature, thus the slight seasonality in summer CH₄ emissions from the lake (Fig. 5) is likely due to the seasonal increase in temperature in the production zone. This seasonal trend also supports a bubble release mechanism, since a seasonal increase in sediment temperature favors methanogenesis <u>but alsoand additionally causes</u> a decrease of CH₄ solubility (Casper et al., 2000; Wik et al., 2013). This <u>means suggests</u> that the amount of CH₄ emitted at the lake surface is directly linked to the amount of CH₄ produced within the sediments, as it has been observed using bubble-traps (Wik et al., 2014). A significant relationship between bubble flux
- and surface sediment temperature similar to the one we reported here, was observed in the lakes of the Stordalen catchment by Wik et al. (Wik et al., 2014), who identified the threshold for ebullition in the Stordalen lakes at 6°C. Our EC system does measure fluxes for sediment temperatures under 6°C, which could be diffusive. The occasional occurrence of degassing during summer that timed up with short de-stratification events (Fig. S3) indicates that hydrodynamic transport and diffusion
- of CH_4 linked to lake mixing may happen in this lake, as it has been observed in a boreal lake (Podgrajsek et al., 2014). Water currents can also trigger bubble release by disturbing surface sediments (Joyce and Jewell, 2003).

Exchange of CO_2 across the lake-air interface is mainly diffusion-limited due to the temperature of dissolution of CO_2 in water, which does not favor the release of CO_2 in bubbles (Tranvik et al., 2009). On average, the net CO_2 flux at the surface of the lake during the ice-free seasons revealed photosynthetic activity. The strong light response curve of median diel

emissions (Fig. 9) is largely influenced by flux rates measured during the warm, sunny summer of 2014. The presence of the diel pattern in summertime CO_2 fluxes was tested for the influence of advection from the lake shore by recalculating the fluxes from the lake using a 5 min average instead of 30 min (Eugster, 2003; Podgrajsek et al., 2015; Vesala et al., 2006). The pattern persisted for 5 min averaged fluxes during the summer of 2012 (Fig. S5), which suggests that advection had a small effect on the summer CO_2 fluxes.

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In an Alaskan lake (Eugster, 2003), negative flux of CO_2 from the atmosphere to the lake surface was attributed to advection because the fluxes occurred during stable atmospheric condition and negative sensible heat flux (H) thus a warming of the lake surface. We observed an opposite relationship here; CO_2 uptake at the lake in summer was associated with unstable atmospheric condition and positive H (Table 2). The diel pattern in H flux from the lake is coherent with the observation of surface water temperature being systematically higher than air temperature during morning hours (not shown), resulting in the cooling of the lake surface. It was in antiphase with the diel patter in CO_2 fluxes (Fig. 8). Water-side convection due to cooling of the lake surface has been shown to enhance the diffusion-limited exchange of CO_2 (Eugster, 2003; Podgrajsek et al., 2015). If the lake water is under___saturated in CO_2 with respect to the atmosphere, this results in a downward CO_2 flux. The anti-correlation we observe between CO_2 flux and H during summer could thus be due to the diffusive CO_2 flux between the surface and the atmosphere being enhanced by convection near the lake surface. Diel pattern in CO_2 flux linked to lake mixing have been observed in other_other_eddy covariance studies, where it was associated with a release of CO_2 to the atmosphere (Mammarella et al., 2015; Podgrajsek et al., 2015).

Finally, the low burst of CO2 observed in fall 2014 may be the result of an accumulation of CO2 during the warmer summer

20 <u>2014</u>, when sustained warm temperatures could cause a thermal stratification of the lake at the end of the season. When lake cooling in fall triggers water mixing, accumulated gases at the lake bottom can be released to the atmosphere (e.g. Kankaala et al., 2006). In other years, regular mixing of the lake during summer may have prevented this phenomenon to occur.

4.2.2 Ice-out Thaw season

- The strong correlation of CH_4 and CO_2 emissions at the lake during both <u>ice-outspring</u> periods suggests that the gases were emitted to the atmosphere via the same mechanism, i.e. by turbulence-driven release of gases that have been accumulating in the lake. Conversely, the correlation is very low during the ice-free periods, when ebullition is the dominant process of CH_4 release. The outgassing pattern at <u>ice-outthaw</u> coincided both years with the breakdown of thermal stratification in the water column after complete ice disappearance. Bubbles trapped in the winter ice of lake Villasjön contain both CH_4 and CO_2
- 30 (Boereboom et al., 2012). CO₂ stored in lake ice during winter can originate from benthic respiration which can occur under ice while dead plants from the previous summer are decomposing (Karlsson et al., 2008). Methanotrophy can be important

during overturn events in lakes (Kankaala et al., 2006; Schubert et al., 2012) thus CO₂ may also be produced in lake water during ice thaw, which lasts several days, as an output of CH_4 oxidation. Dissolved CO_2 could also enter the lake as catchment input via lateral meltwater run-off before complete overturn (Denfeld et al., 2015).

The processes underlying CH₄ degassing during ice-out that in 2013 have been discussed in details in a previous study

- 5 (Jammet et al., 2015), suggesting that the spring burst is the combination of different gas sources, i.e. liberation of bubbles from the ice, diffusion of gases from the water to the air and release of stored gases from the bottom of the lake during complete overturn, and iAs CO₂ fluxes covariate closely with CH₄ emissions from the lake during spring (Fig. S4), it is likely that CO₂ was released via the same physical mechanisms. The degassing pattern observed in 2013 was repeated in 2014, with a mean and median CH_4 flux rate smaller than the previous year, but still significantly higher than the CH_4
- 10 emissions of the ice-free season (Fig. S2). In 2014 the ice-out-thaw period started earlier but was longer (Figure 2), and our measurement system may have missed part of the degassing due to instrument failure. As ice thaws, gases contained in bubbles can migrate to the water (Greene et al., 2014) and be released to the atmosphere when thermal stratification gradually breaks because of the warming up of the water column. We can speculate that a delay in the timing of overturn following ice thaw may favor oxidation of CH_4 within the water column when it is already partly mixing, which would raise the concentration of dissolved CO_2 in the water and could contribute to a smaller burst of CH_4 during complete overturn.

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4.3 -Annual atmospheric carbon budget

4.3.1 Carbon function

The fen was an annual sink of carbon with respect to the atmosphere, while the lake was an annual source, at a magnitude representing 70 % of the fen sink. The total annual C-emission from the lake is within the range of annual C-emissions 20 (CH₄+CO₂) from lakes of subarctic Sweden (5 to 54 g C m⁻² yr⁻¹, Lundin et al., 2015) estimated mostly using water grab sampling.

At the fen, we report a stronger summer sink of CO₂ (three ice-free seasons average -206.8 g C m⁻², Table 3) compared to earlier studies in the inner fens of the Stordalen mire (-133 g C m⁻², years 2001-2008, Christensen et al. 2012), but annually a

- net CO₂ uptake (-58.5 to -79.1, average -66.3 g C m⁻² yr⁻¹) that is similar to the 2001-2008 average (-66 g C-CO₂ m⁻² yr⁻¹, 25 Christensen et al 2012) and smaller than the average for years 2006-2008, which were warm years (-90 g C-CO₂ m⁻² yr⁻¹, Christensen et al. 2012). The difference is due to the higher CO₂ respiration we measured in winter, which equaled 54 % of the summer sink on average during the measuring period. The annual CH_4 emissions of 21.2 C- CH_4 m⁻² yr⁻¹ (Table 3) is very close to what has been reported for the internal fens of Stordalen in an eddy covariance study where winter emissions were
- estimated with a temperature relationship (Jackowicz-Korczyński et al., 2010). This highlights the stability of the fen in 30

terms of CH_4 emissions but also the low sensitivity of the annual sum to the choice of gap filling method for the fen CH_4 flux dataset, which is tightly linked to temperature.

To determine whether an ecosystem is a net source or sink of carbon within the landscape carbon cycling, a full net ecosystem carbon balance (NECB) must take into account both vertical carbon exchange but also lateral carbon transport, in

- 5 and out of the system (Chapin et al., 2006). In 2008, net DOC export at the fen was 8.1 g C m⁻² yr⁻¹ and net POC export was 0.6 g C m⁻² yr⁻¹ (Olefeldt and Roulet, 2012). Combined with our annual atmospheric carbon budget (Table 3), this results in a fen NECB of -29.5 g C m⁻² yr⁻¹ in the first year and -43.3 g C m⁻² yr⁻¹ in the second year. These numbers are marginally smaller than the long-term carbon accumulation of -51 g C m⁻² yr⁻¹ inferred from the analysis of a peat cored in Stordalen and attributed to a period when the mire was dominated by graminoids (Kokfelt et al., 2010). We are not aware of existing
- 10 data on net export of DOC and POC through the lake to make a similar estimate.

In term of radiative forcing, considering the 28-fold stronger global warming potential of atmospheric CH_4 vs. atmospheric CO_2 over 100 years (GWP100, Myhre et al. 2013), vertical carbon exchange has a warming impact on the atmosphere at both ecosystems through their net annual emissions of CH_4 . Annual estimates that disregard winter and transitional seasons are likely missing part of the annual carbon emissions from seasonally freezing lakes and wetlands.

15 **4.3.2** The lake as a summer CO₂ sink

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Because of dynamic external and internal factors governing the consumption and production of CO_2 in surface waters, the CO_2 function of a lake can vary seasonally (Maberly, 1996; Shao et al., 2015). Lake Villasjön was an annual source of CO_2 due to the spring outgassing, but it was a small sink of CO_2 in the open-water period. While flux rates in summer 2012 and 2013 were negative but close to the noise level, the uptake was larger and significant in 2014 when the summer was hotter

- and sunnier. Averaged estimates from water sampling measurements in the lakes of the Abisko area indicate the lakes to be mainly CO_2 sources during the summer, except for a few lakes that were seasonal CO_2 sinks during the ice-free season (Karlsson et al., 2013). In the few eddy covariance studies available from Arctic and boreal sites, lakes are reported as CO_2 sources during the ice-free season (Lohila et al., 2015; Mammarella et al., 2015; Podgrajsek et al., 2015) and occasional CO_2 sinks during the warm summer months, while being sources on the seasonal scale (Anderson et al., 1999; Eugster, 2003;
- 25 Huotari et al., 2011; Jonsson et al., 2008). No coincident measurement of pCO2 in the lake water is available for the study period. A future study combining pCO2 with EC will help further defining the direction of the flux observed at the ecosystem-scale.

Although Villasjön is representative of a widespread postglacial lake type across subarctic and Arctic latitudes, it differs from most lakes studied in the northern lakes literature due to its particularly shallow depth, which results in the lack of longterm stratification during the open-water season. Lakes that are similarly shallow are often thermokarst lakes or peatland ponds (Vonk et al., 2015). Summer CO₂ uptake at the level of what we report here has been observed in highly productive lakes (Pacheco et al., 2013) or in thaw ponds colonized by submerged plants and microbial mats (Laurion et al., 2010; Tank et al., 2009). Estimates of air-lake carbon exchange using water sampling and floating chambers (Karlsson et al., 2013) showed that a minority of lakes in subarctic Sweden were CO₂ sinks in summer with a total seasonal CO₂ exchange from - 3.8 to -10 g C m⁻² yr⁻¹, while being large sources at ice-out, offsetting the summer sink.

Lakes having poor hydrological connections with their upstream catchment have been reported in previous studies to be net CO₂ sink in summer, e.g. in Minnesota (Striegl and Michmerhuizen, 1998) or in thaw ponds of the Canadian Arctic (Tank et al., 2009). In the latter study, within-lake DOC was proposed to occur as a byproduct of macrophyte photosynthesis, showing that net CO₂ uptake in lakes is not always associated with low DOC concentrations. In large and shallow lakes

- 10 surrounded by peatlands, vegetation develops on the sediment surface thanks to the presence of humic acids supplied by the peaty shores and a well-illuminated bottom (Banaś et al., 2012). The analysis of peat and lake sediment records in Stordalen suggested that a significant amount of peat is exported from the mire to lake Villasjön during periods of mire erosion, likely due to permafrost thaw (Kokfelt et al., 2010). This lake may have high nutrient content due to peat input at the shores, organic-rich sediments and autochtonous vegetation. Lakes that do not stratify tend to be more productive because of the
- 15 more regular mixing of nutrients (Tranvik et al., 2009; Wetzel, 2001).

4.3.3 Influence of overturn on annual C-emissions

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On an annual scale, the ice-outthaw period accounted for 50 % of annual carbon exchange (CH₄ + CO₂) at lake Villasjön and turned the lake from a summer CO₂ sink to an annual source. In other, deeper lakes that stratify in summer and don't fully mix in spring, fall overturn led to the highest emissions of CH₄ or CO₂ of the year (Kankaala et al., 2006; Schubert et al., 2012), accounting for a large part of the annual CO₂ flux (Huotari et al., 2011). Other seasonally ice-covered lakes emitted large amounts of CH₄ and CO₂ following ice-out (Anderson et al. 1999, Karlsson et al. 2013), while high concentrations of dissolved CO₂ and CH₄ under lake ice has been measured in Northern American lakes (Striegl and Michmerhuizen, 1998), across lakes of the Swedish subarctic (Karlsson et al., 2013), Alaskan thermokarst lakes (Sepulveda-Jauregui et al., 2015) or in thaw ponds in Canada (Tank et al. 2009). Accumulation of CH₄ and CO₂ under the ice is thus a general feature of lakes with an anoxic hypolimnion or sediment in winter, but studies reporting direct measurement of the outgassing of CH₄ and CO₂ at the lake surface right after ice-outthaw are scarce because it is a rapid and variable phenomenon that is seldom included in direct flux measurements.

The large impact of CO_2 and CH_4 release during spring has been observed in lakes of the Abisko area where water samples before and after ice-out were used to estimate the thaw release, which accounted on average for 45 % of annual emissions

30 (Karlsson et al., 2013; Lundin et al., 2013). A few regional studies reported on a lesser importance of the spring season on
 annual carbon emissions from lakes. Sepulveda-Jauregui et al. (2015) sampled 40 <u>Alaskan</u> thermokarst <u>Alaskan</u>-lakes where

the maximum emission of CH_4 and CO_2 were measured in summer. Many of these lakes were thermokarst lakes that continuously emitted CH₄ in winter through open holes, which we don't observe in lake Villasjön. Thermokarst lakes are usually stronger CH₄ emitters than post-glacial lakes on a per unit area basis, yet post-glacial lakes seem to be a larger overall source because they cover a larger area in the high northern latitudes (Wik et al., 2016). A recent review by Wik et al.

5 (2016) compiled CH₄ emissions from several types of lakes. Over the total of 733 sites, ice outthe than period was estimated to contribute ~23 % of annual emissions of lakes and ponds. Only four sites were measured with EC and those four only comprised ice-free season (July-August) measurements. Thus the comparison to our results is limited by differences in temporal and spatial scale of the methods.

Our study underlines the high significance of shoulder seasons (more precisely, overturn periods following periods of gas

- 10 storage) for the biogeochemistry of lakes and the emission of CO_2 and CH_4 to the atmosphere. The relative importance of these periods on the annual emissions depends on the extent of the overturn (Huotari et al., 2011; Kankaala et al., 2006), the extent of methanotrophy during and before full lake mixing (Kankaala et al., 2006; Schubert et al., 2012), as well as the amount of degradable organic matter in the hypolimnion. Although these overturn periods cover only a few weeks or days, they are important for both CH₄ and CO₂ emissions in lakes and should be included in measurement campaigns when
- 15 feasible.

5. Conclusions

The waterlogged fen and the shallow lake showed contrasting annual cycles in term of CH₄ and CO₂ exchange with the atmosphere. This difference is explained, first, by the presence of an ice lid over the lake surface which led to the storage of gases in winter and large subsequent emissions in spring, while evasion of CH_4 and CO_2 to the atmosphere from the fen in the wintertime limits the importance of emissions during ice and snow melt. Second, the dense cover of vascular plants at the

fen leads to high CH₄ emissions and CO₂ uptake in summer.

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Annually, the fen was a net carbon sink with respect to the atmosphere, while the lake was a source of carbon due to the degassing in spring that outweighed the <u>apparent</u> uptake of CO_2 in summer. This study confirms the importance of overturn periods in lakes for both CH4 and CO2 annual emissions. The magnitude of the degassing at ice outduring the thaw season may depend greatly on lake type, morphometry and productivity status. The lake studied represents a common type of shallow postglacial lake across the subarctic latitudes. Further direct measurements of surface fluxes covering several years and different lake types are needed to evaluate the inter-annual variability in the magnitude of the degassing in shoulder seasons as well as its importance for the annual emissions of northern lakes in general.

Finally, ebullition was identified as the main transport pathway for CH4 emissions in the shallow subarctic lake and a net CO2 sink in summer indicated large-photosynthetic activity. Turbulence-driven diffusive release of CO2 and CH4 was predominant during spring overturn following ice-out. These results show the potential of the EC method in lake environments for a better understanding of flux processes and annual seasonality in the understudied but abundant postglacial lakes and ponds.

Data availability

5 The eddy covariance data and meteorological data used in this study are available upon request to the lead author. A version of the flux dataset before flux source partitioning between lake and fen is available on the FLUXNET database with ancillary data.

Authors contributions

TF, MJ, PC designed the study. MJ collected, analyzed and interpreted the data. SD developed and performed the gap filling

10 modeling. EK pre-processed part of the eddy covariance raw data. FJWP performed the 2D footprint modeling and drew the footprint figure. MW provided methane ebullition data. MJ wrote the manuscript, figures and all authors commented on it.

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Appendices

Appendix A: Details in eddy covariance flux calculation

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Processing of the raw eddy covariance data for flux calculation included despiking (Vickers and Mahrt, 1997), angle of attack correction on raw wind components (Nakai et al., 2006), 2-D axis-rotation_correction (Wilczak et al., 2001) on wind speed components, and detrending of 30 min. raw data intervals by block averaging the vertical wind speed and scalar signals (Moncrieff et al., 2004). The time delay between vertical wind speed and each scalar gas concentration measurements (CO₂, CH₄, H₂O, sonic temperature) was removed by finding the maximum of the cross covariance function of vertical wind

speed and each scalar (Fan et al., 1990). The time-window search was set to ± 4 s for CO₂ and H₂O; the median time lag was 0 s. For CH₄, the time-window search was adjusted for each gas and for each period when a change in the set-up occurred; the median time lag between vertical wind speed and CH_4 concentration measurements varied between 9 and 18 s.

The effect of density fluctuations on CO₂ fluxes was corrected (Webb et al., 1980). The correction lowered the amplitude of

- 5 the CO₂ flux dataset on average by 53% (slope of the linear regression between non-density corrected CO₂ fluxes and final corrected fluxes = 0.47, $r^2 = 0.73$). Due to the unavailability of H₂O concentration measurements from the methane analyzer during most of the study period, WPL correction was not applied by the flux software on CH_4 fluxes. Applying the correction on part of the data using the available H₂O concentration from the methane analyzer showed a difference of flux magnitude of about 1 % with the non-corrected dataset. The low magnitude of the WPL correction can be expected for this
- 10 setup, due to the long sampling line that attenuates significantly the H_2O signal as well as temperature and pressure fluctuations, thus density effects. For CH₄-fluxes, density fluctuations were compensated after flux calculation using the formulation of Ibrom et al. (2007a), including the pressure induced fluctuation term. Comparing CH₄ fluxes with and without density correction showed a difference of less than 5% in the flux magnitude. Turbulent fluxes calculated with the eddy covariance method are affected by spectral losses due to the instrumental setup and the limited time response of the
- instruments. Losses in the low frequency range due to the finite flux averaging time were corrected analytically after 15 Moncrieff et al. (2004). CO₂ flux loss in the high frequency range was also corrected analytically (Moncrieff et al., 1997), while CH₄ fluxes derived from the closed-path system required an *in situ* assessment of the system's cut-off frequency (Ibrom et al., 2007)(Ibrom et al., 2007b) due to the long sampling line. This assessment was done separately for each period with a continuous instrumental set-up and the associated flux attenuation was calculated and compensated following the
- formulation by Horst (1997). The magnitude of the spectral loss hence of the total spectral correction was on average 31% 20 for CO₂ fluxes and 37% for CH₄ fluxes.

Appendix B: Performance of the Computing the performance of the ANN models

The ANN gap filling method was most performant on the fen dataset, achieving an r^2 of 0.88 during the training phase, and 25 an r^2 of 0.85 between measured and predicted values over the whole dataset (expressing the capacity of generalization of the model) with a relative mean square error (RMSE) of 23 % (Figure B1). The ANN gap filling of lake CH₄ fluxes achieved an r^2 of 0.71 in the training phase and an r^2 of 0.70 between predicted and measured fluxes on the whole dataset, with an RMSE of 51 %. The lake model was most accurate for periods with the best data coverage in the measured dataset (spring seasons, $r^2 = 0.77$). The lower accuracy of the model during the ice free seasons ($r^2 = 0.47$) is also due to the pulse character of lake ebullition, which was not always reproduced by the model, while the background seasonal trend was present.

For the fen CH_4 fluxes, the model was most accurate during the ice-free seasons when fen CH_4 -emissions are tightly linked to peat temperature and least performant during the ice out periods. Unsurprisingly, the prediction performance of the models was dependent on data coverage in the original dataset. On the annual scale, both fen and lake models were most performant during the first year (June May), which had the least amount of data loss, with an r^2 of 0.88 (RMSE 23 %) on the first year for the fen model and of 0.82 (RMSE 40 %) for the lake model between predicted and measured fluxes.

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The ANN modeling was likewise performant on the fen CO_2 flux dataset, achieving an r^2 of 0.86 (RMSE 35 %) over the whole dataset between measured and predicted value (Figure B1). The model was most performant during the ice free seasons. This can be explained by a better data coverage but also by better constrained processes during the growing season and on the annual scale, than during the snow cover season, when additional drivers than the one selected as model inputs may play a role in the variability of the fluxes at the half hourly scale (sensitivity to footprint changes, turbulence, variation

- 10 may play a role in the variability of the fluxes at the half hourly scale (sensitivity to footprint changes, turbulence, variation in snow depth, partial melt, etc.). The performance of the ANN models was assessed by comparing the predicted values with original observed values of the entire dataset (Table B1). This represents the actual ability of the ANN to generalize (Papale and Valentini 2003) to untrained conditions. The goodness of fit was quantified with the coefficient of determination r^2 and the root mean square error (RMSE). Additionally, the mean random error of the predicted flux values was calculated as the
- 15 mean of the standard deviation around each individual value used for gap filling. In other words: each modeled value used for gap filling is the mean of several ANN model runs. The 25 best runs (according to r^2) were averaged to obtain the modeled fluxes used in the gap filling. The standard deviation of these 25 model outputs was thus used as a quantification of the random error of each modeled flux value. The average of these individual random errors was then computed as the mean random error of each modeled flux series (CH₄ fen, CH₄ lake, CO₂ fen), reported in Table B1.

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Appendix C

Figure C1.

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Tables

5

Table 1: Climatic conditions per season: <u>mean_air temperature measured at the Abisko Scientific Research</u> Station, peat temperature at 10 cm depth in the fen, surface sediment temperature in the lake (1 m depth)<u>and</u>, total <u>net radiation at the fen surface and at the lake surfaceincoming solar radiation (S_i)</u>. Season delimitations (see <u>text for a definition</u>) are: 1 June 2012 <u>14 Oct 2012</u>; 26 May 2013 <u>15 Oct 2013</u>; 2 June 2014 <u>9 Oct 2014</u>; <u>15 Oct 2012 <u>14 Apr 2013</u>; 16 Oct 2013 <u>10 Apr 2014</u>; 15 Apr 2013 <u>25 May 2013</u>; 11 Apr 2014 <u>1 June</u> <u>2014</u>; 1 June 2012 <u>31 May 2013</u>; 1 June 2013 <u>31 May 2014are reported for each year (see text for a</u> <u>definition</u>).</u>

Season	Year	<u>Dates</u>	Length (days)	Air T (°C)	Peat T at 10 cm (°C)	Surf. sed. T (°C)	Total Rn, f en <u>S</u>i (10³ Wm⁻²)
Ice-free	2012	<u>1 June – 14 Oct.</u>	136	7.8	8.9	10.2	<u>808</u> 375
	2013	<u> 26 May – 15 Oct.</u>	143	10.2	9.9	12.0	<u>902</u> 467
	2014	<u>2 June – 9 Oct.</u>	130	10.4	10.5	12.8	<u>1019</u> 545
Ice-cover	2012-2013	<u> 15 Oct. – 14 Apr.</u>	182	-7.9	0.0	0.7	<u>279</u> - 168
	2013-2014	<u> 16 Oct. – 10 Apr.</u>	177	-6.5	0.2	0.8	<u>239-151</u>
Ice- out<u>Thaw</u>	2013	<u> 15 Apr. – 25 May</u>	41	4.3	0.3	1.9	<u>250</u> 112
	2014	<u>11 Apr. – 1 June</u>	52	2.2	1	1.9	<u>502</u> 117
Annual	2012-2013	<u>1 June – 31 May</u>	365	-0.3	3.5	4.7	<u>1424</u> 374
	2013-2014	<u>1 June – 31 May</u>	365	0.9	4	5.1	<u>1543</u> 377.2

Table 2: Statistical exploration of the lake flux dataset: Spearman's rank correlation coefficient (ρ) showing shows the degree of association between half-hourly lake CH₄ flux and lake CO₂ flux with potential drivers of variability. All data were grouped per season. See Table 2–1 for the limitation of the seasons. Lake fluxes are filtered in winter for high standard deviation of lateral wind speed-in winter.

	La	ke CH4 flu	ux (Spearmai	n's ρ)	Lake CO ₂ flux (Spearman's ρ)				
	Annual	Ice free	Ice- out<u>Thaw</u>	Ice cover	Annual	Ice free	Ice- out<u>Thaw</u>	Ice cover	
Air T	0.36***	0.40***	0.26***	0.14**	- 0.21***	- 0.22***	0.38***	0.16***	
T water surface	0.27***	0.48***	0.37***	0.18*	- 0.21***	- 0.21***	0.27***	0.09***	
T bottom	0.24***	0.49***	0.49***	0.10*	- 0.26***	- 0.21***	0.40***	-0.10***	
Wind speed	0.36***	0.32***	0.40***	0.27***	0.26***	0.13***	0.38***	0.36***	
H flux	- 0.15***	0.12***	-0.18***	-0.12*	- 0.62***	- 0.67***	-0.38***	-0.50***	
LE flux	0.24***	0.38***	0.16***	0.08	- 0.29***	- 0.49***	0.05	-0.14***	
Solar radiation	0.27***	0.13***	-0.02	-0.11*	- 0.09***	- 0.52***	0.02	-0.22***	
CO ₂ flux	0.41***	- 0.13***	0.67***	0.21**					

5 *** p-value < 0.001; ** p-value < 0.01; * p-value < 0.1

Table 3: Seasonal and annual sums of CO₂ and CH₄ fluxes after gap-filling, in g C m⁻² y⁻¹. Sum BE is the bias due to the gap filling model (Eq. (1)), scaled to annual flux units and multiplied by the number of gaps in the flux dataset over the season or year. Sum of CO₂ fluxes from the lake is only reported for the first year.

		Fen CH	n CH4 flux Lake CH4 flux		Fen CO ₂ flux		Lake CO ₂ flux		
Season	Year	Total flux	Sum BE	Total flux	Sum BE	Total flux	Sum BE	Total flux	Sum BE
Ice-free	2012	14.8	0.1	2.5	-0.1	-179.4	-14.8	-24.4	< 0.001
	2013	16.2	-0.1	2.8	-0.1	-201.3	13.2	nc*	nc
	2014	16.8	0.09	3.0	-0.5	-239.6	-5.1	nc	nc
Ice- cover	2012-2013	4.1	-0.2	0.3	-0.3	109.6	-8.8	12.4	< 0.001
	2013-2014	4.2	0.7	0.4	-0.4	115.5	20.7	nc	nc
Ice- out Thaw	2013	1.4	-0.1	2.5	-0.2	11.3	-0.4	33.3	< 0.001
our <u>inaw</u>	2014	1.7	-0.07	1.3	0.5	11.7	1.1	nc	nc
Annual	2012-2013	20.3	0.2	5.3	-1.9	-58.5	-26.1	21.5	< 0.001
	2013-2014	22.1	-0.1	4.4	0.6	-74.1	97.4	nc	nc
Annual	Average ± STD	21.2 ± 1.3	-0.2	4.9 ± 0.6	-0.7	-66.3 ± 11	35.7	21.5	< 0.001

*nc = not computed

Table B1: Characteristics of the artificial neural networks that were developed for gap filling fen and lake CH_4 fluxes and fen CO_2 fluxes. All networks were developed with one hidden layer and with four fuzzy datasets as additional input to force seasonality. The mean random error is the average of the standard deviation around the modeled flux values for each series (cf. Text S1).

	Fen CH ₄	Fen CO ₂	Lake CH ₄
Input variables	<u>Tair</u>	Photosynthetic active	Tair
	<u>Tpeat at 10 cm</u>	radiation (PAR)	T water at 10 cm
	Wind speed,	<u>Tair</u>	T in sediment surface (100
	Air pressure,	Vapour pressure deficit	<u>cm)</u>
	Net radiation (fen)	<u>(VPD)</u>	Wind speed
	Incoming solar radiation	Net radiation (fen)	Air pressure
			Net radiation (lake)
			Incoming solar radiation
Number of neurons	<u>6</u>	<u>6</u>	<u>9</u>
<u>R2 (all predicted vs. obs.)</u>	<u>0.85</u>	<u>0.86</u>	<u>0.70</u>
<u>RMSE (all predicted vs.</u> obs., μmol m ⁻² s ⁻¹)	<u>0.022</u>	<u>35%</u>	<u>0.043</u>
Mean random error of all	<u>0.004</u>	<u>1.3</u>	<u>0.011</u>
$\frac{\text{predicted values (µmol m})}{\frac{2}{\text{s}^{-1}}}$			

Figure captions

Figure 1: Location of the study site (a) and flux footprint of the flux tower in summer averaged over all years (b). The color scale indicates the extent of the fractional contribution from the source area to the fluxes measured at the tower. The location of the flux tower is indicated along with the location of the main environmental data sources.

- Figure 2: <u>Daily mean of Aa</u>ir, peat and surface sediment temperature (a)-; cumulative daily precipitation (b)-; <u>daily mean net</u> radiation input at the fen and the lake surfaces (c)-; albedo of the lake surface (d)-; <u>daily</u> temperature gradient in the lake water column defined as (T_{w,10cm} T_{w,100cm}) divided by the depth difference Δz (e); water temperature profile in the lake center derived from continuous <u>daily</u> temperature measurements at depths 10 cm, 30 cm, 50 cm and 100 cm (= bottom) (f). The shaded areas indicate the periods of ice-cover at the lake. PN stands for polar night, i.e. near zero solar input. The black arrows indicate the estimated time of complete lake overturn after ice-out.
 - **Figure 3**: Measured CH_4 and CO_2 fluxes at the fen (a,b) and the lake (c,d) over the full study period. Light grey dots are half-hourly values and black dots show a 5-days running mean. Note on panel (c): 9 flux values above 800 nmol m⁻² s⁻¹ are not displayed for visibility.

Figure 4: Measured flux rates of CH₄ (left panel) and CO₂ (right panel) per season and per ecosystem. The central line of the

15 boxplots shows the median, box edges show 25th and 75th percentiles, and whiskers show 5th and 95th percentiles. The black dots indicate the mean flux rate. Outliers are not displayed.

Figure 5: Averaged seasonality of CO_2 and CH_4 fluxes at both ecosystems. Dots show daily means across the whole measurement period, lines are a smoothing filter of the daily mean with a 30 days window. Dashed lines show the standard deviation around the daily means, smoothed with a 30 days window.

Figure 6: CH₄ emissions at the lake during the ice-free seasons of 2012, 2013 and 2014, from day 152 do day 290. Grey dots are half-hourly eddy covariance observations, open black dots are daily means of the eddy covariance fluxes and red dots show spatially-averaged daily CH₄ ebullition measured in the lake with bubble traps. Water temperature at depths 10 cm (red line) and 100 cm (sediment surface, blue line), as well as atmospheric pressure (black line) are also shown.

Figure 7: Relationship between lake CH₄ emissions and surface sediment temperature <u>during summer</u>, using <u>all half-hourly</u> 25 CH₄ flux rates averaged by bins of 1°C across the three ice-free seasons (a) and arrhenius plot of the natural logarithm of the flux versus the inverse surface sediment temperature in Kelvin (b). Error bars show the standard deviation of the CH₄ flux rates around the means within each averaging bin. The red line iss are regression fits, with $r^2 = 0.82$ (*p*<0.001).

Figure 8: Diel medians, mean and $25^{\text{th}}-75^{\text{th}}$ percentiles of CH₄ fluxes, CO₂ fluxes, sensible heat flux (H) measured at the lake (upper panels) and at the fen (lower panels), from June to August of each year. Grey lines show the hourly median values for each year and the black line is a median of the three ice-free seasons.

Figure 9: Diel median of the net CO_2 exchange at the fen (red) and at the lake (blue) versus diel median solar radiation,

5 between June and August. Each dot is the hourly median flux of the combined three ice-free seasons. Note that the two yaxis have different scales.

Figure <u>10</u>**B1**: Evaluation of the artificial neural network models: measured vs. predicted hourly flux values for fen CH_4 fluxes (a), lake CH_4 fluxes (b) and fen CO_2 fluxes (c). In red is the 1:1 line.

Figure C1: Probability density functions of (a) the measured CH_4 fluxes (bin size 10 nmol m⁻² s⁻¹) and (b) the measured CO_2 fluxes (bin size 0.2 µmol m⁻² s⁻¹) from the fen and from the lake during the entire measurement period.