

Dear Dr. Lutz Merbold,

On behalf of all co-authors, I hereby submit the revised version of the manuscript 'Aquatic greenhouse gas emissions unaffected by forest harvesting', by Marcus Klaus, Erik Geibrink, Anders Jonsson, Ann-Kristin Bergström, David Bastviken, Hjalmar Laudon, Jonatan Klaminder and Jan Karlsson, intended as a *full paper* contribution to *Biogeosciences*. Below we give a detailed response to all comments from the reviewers. We have addressed all requested changes, have included a response to each of the comments, list the changes to the manuscript as appropriate (blue text) and refer to respective line numbers in the revised version of the manuscript.

Thank you again for considering this manuscript for publication in the Journal of *Biogeosciences*. We would also like to thank the reviewers for taking the time to review this manuscript and providing constructive feedback that have helped improve the manuscript. If you require additional information or clarification, please do not hesitate to contact me at marcus.klaus@posteo.net.

Sincerely,

Marcus Klaus

Marcus Klaus

## Reviewer 1 (RC1)

General comments:

**Comment 1.0** This paper discusses the impact of forest harvesting on greenhouse gas emissions of boreal inland waters. This is done by analyzing four catchment sites, two of which were affected by forest clear cutting. Overall, the approach of the "Before-After/Control-Impact"-analysis is sound and, in general, the methodological approach is described adequately. However, in some cases more detailed information is necessary as pointed out below ('specific comments'). The study shows the impact of forestry activity on groundwater GHG concentrations and reveals the importance of the role of the riparian buffer zone-stream continuum although no clear conclusion on the mechanistic role can be drawn.

*Reply: Thank you. We do not draw any clear conclusion on the mechanism that acts to buffer the increase in groundwater*  $CO_2$  *and*  $CH_4$  *concentrations, because at this stage, we regard different mechanisms (e.g. in-stream processing, riparian processing) to be equally likely. More detailed studies targeting these mechanisms are needed as we point out in the discussion (p. 9, L. 35-37).* 

*Change:* We have added results on BACI effects on gas concentrations in streams and lakes (p. 8, L. 6-10) and now refer to a new Table (S6). These results have helped us to narrow down the discussion of the mechanisms (p. 9, L. 24-32; p. 10 L. 2; see our response to comment 1.19 and 2.0).



We have provided more detailed information as pointed out in our replies to specific comments below.

Specific comments:

Comment 1.1 P2, L30: specify the measurement period more precisely (Jun – September?)

Reply: We agree.

*Change: We have specified the sampling months (June – September; p. 2, L. 30) and also deleted "throughout the whole open water period" on page 3, L. 20 to avoid redundancy.* 

Comment 1.2 P2, L33: what is 'normal' precipitation? Better: close to the long-term average of xx mm

Reply: We agree.

Change: We have rephrased the text following the reviewer's suggestion (p. 2, L. 33).

**Comment 1.3** P3, L16: 'water chemistry' is not the right term here. Maybe merge paragraph 2.3 and 2.4 under 'Water sampling and physicochemical analysis'.

Reply: We agree.

*Change: We have restructured the text following the reviewer's suggestion (p. 3 L. 19 – p. 4, L. 15).* 

**Comment 1.4** P3, L17-18: '... and the deepest point of the lake (Fig. 2) as described in S2.' (Consider also to reorder this sentence so that the described sampling activities match with the description in the supplement because the next sentence refers to S1, while the following paragraph refers to S2 again.)

Reply: We agree.

*Change: We have restructured the text following the reviewer's suggestion (p. 3, L. 20-22).* 

Comment 1.5 P3, L21: spatial variability in CO2 and CH4 concentrations within streams

Reply: We agree.

Change: We have rephrased the text following the reviewer's suggestion (p. 3, L. 25).

**Comment 1.6** P4, L3: 'Filtered water samples' also from streams and groundwater wells? Maybe specify here again, since in the first sentence you write 'To characterize lake color and this could lead to the impression that you are talking about lake water samples only in the second sentence.

Reply: Thank you for pointing out this typo. Color was determined for lake and stream water.



*Change: We have clearly pointed out what type of analysis was done in what type of system (p. 4, L. 7-12).* 

## Comment 1.7 P4, L7: you measured TP but never mentioned in the results. Why?

*Reply: Thank you for pointing out this inconsistency. We did not include TP in this manuscript because, phosphorous is typically less responsive to clearcutting relative to nitrogen and primary production in our lakes is nitrogen and not phosphorous limited. This is clearly described in the introduction (P. 2, L. 4-11).* 

Change: We have deleted the methods description for TP as we don't show any TP data.

**Comment 1.8** P4, L24-33 in Figure S3 you indicate that you also used bootstrapping when modelling the k600 for lakes, but you never mention this in the text where you describe how you obtained the gas transfer velocity

Reply: Thank you for pointing out this inconsistency.

Change: In the main text we now refer to the detailed description in Text S6 on how we accounted for uncertainties in k<sub>600</sub> estimates (p. 4 L. 39-40). We have also moved details on error propagation procedures for k<sub>600</sub> estimates for streams to Text S6 to improve text flow and now have all details on error propagation condensed in one place. Finally, we also noticed that the error propagation procedure for gas flux calculations (Eq. 1) were not properly introduced and now refer to Fig. S3 when introducing Eq. 1 (p. 5 L. 32-33).

**Comment 1.9** P5, L17-18: you use Equ. (1) also to calculate CH4 and N2O fluxes, right? So c should be the respective gas concentration (not CO2 concentration).

Reply: Thank you for spotting this mistake. Eq. 1 was indeed used for all three gases.

*Change: We have made this clear by rephrasing the text (p. 5 L. 27).* 

**Comment 1.10** P6, L13: why did you set the 'after' period to 2013-2015? Shouldn't it be 2013-2014 if you want to analyze the clear-cut effects only (without the influence from site preparation)? Did you look at any trends/effects in the individual years after the clear-cut?

Reply: Clear-cut effects can be expected to last for more than the first two years. By first contrasting 2013-2015 with 2012 we were able to test for the general response in the first 3 years after clearcutting. Our additional analysis that contrasts 2015 with 2012, was done to test whether effects started to be visible after site preparation. We did not test for any trends, but regard the analysis of contrasting 2015 vs. 2012 and 2013-2015 vs. 2012 as a means of testing whether effects started to be visible after site preparation which may be overlooked if all three years were lumped together.



*Change: We now reason more thoroughly in the chapter on "Statistical analysis" how we defined the "after" periods and why we chose the time intervals (p. 6, L. 10-13).* 

## **Comment 1.11** P6, L6: 'paired difference' – did you do all the measurements at the different sites at exactly the same time? If not, did you account for that in the LME?

Reply: Sampling lake-, stream- and groundwater in one catchment took a whole day for us. Hence sampling at exactly the same time point was logistically impossible for us as an individual research group. However, we tried to sample Control- and impact- lake pairs as close in time as possible, typically within 2-3 days, but never more than 7 days from each other. We did not account for this minor variation in sampling dates in the LME.

*Change: We now point out more clearly in Chapter 2.3 that control and impact catchments were typically sampled within two or three days, but never more than seven days from each other (p. 3 L. 21-22).* 

## Comment 1.12 P6, L10: what were the results of the pseudo-BACI?

Reply: We are grateful for pointing out this inconsistency between methods and results. We included the pseudo-BACI analysis in an earlier version of this manuscript, but after a round of revisions, decided to not include it to not overload the paper with details and to sharpen the focus. The pseudo-BACI revealed no significant BACI effects in any of the control catchments, which gives us more confidence to state that the BACI effects found in the clear-cut catchments were due to the clear-cut treatment.

*Change: We have deleted the method description on the pseudo-BACI as we do not show any related results.* 

We realized that the caption of Table S5 was misleading in this context. We have replaced "in control and impact catchments" by "at control and impact sites in the impact catchments".

**Comment 1.13** P6, Results: in general, when you present (mean?) values, indicate that those are (multi-?)seasonal means etc. For example, on P7, L4 you write 'Whole lake temperatures (ranging from 12.8-16.5 °C) ...' – but that's the range of the mean values and not of the entire measurements, right? (also check those numbers; different from Table 2)

## Reply: Thank you for this comment!

*Change:* We now clearly state in the beginning of the results section that we refer to arithmetic mean values over each of the two time periods (before, after; p. 7 L. 8). We noticed that the numbers given in the text were wrong and replaced them by numbers given in Table 2 (p. 7 L. 14-15).

**Comment 1.14** P7, L4-5: I think the wording here is confusing because temperature did not decrease but it actually increased only more so in the control. Any idea/explanation for that?



*Reply:* We are aware of that temperatures in control lakes increased more than in impact lakes. We express this differential effect by using the formulation "decreased … relative to control lakes". However, we agree that his might be confusing.

The increase in whole lake temperatures was likely due to the higher air-temperatures in the after period relative to the before period (Table S1). As we point out on P. 10, L. 2-4, the effect size of -0.4 °C was small (Cohen's D = -0.20) and had likely no ecological or biogeochemical effects. We therefore did not speculate further on this effect in the manuscript.

*Change: We have rephrase the text according to the reviewer's suggestion (p. 7 L. 14-16). We do not know and will not attempt to speculate what could have caused the rather minor difference in temperatures.* 

**Comment 1.15** P7, L25: 'medium effect size of  $+533 \mu$ M or  $+56\%' - 533 \mu$ M is the slope of your LME, but since you included lake pair as random effect also on slopes, you should get two slopes!? Is this the mean? This also applies to all the results/tables where you present slopes/effect sizes. How do you get the 56%?

*Reply:* We indeed get two slopes and intercepts, one for each pair. We here present arithmetic mean slopes and intercepts. The relative effect size (here: 56%) is the effect size divided by the mean value in the impact system in the control year.

*Change: We now explain more clearly how slope and intercept is calculated and added a definition of the relative effect size to the methods section (p. 6 L. 18-19).* 

We realized that the relative effect size was incorrect for groundwater  $CH_4$  concentrations and have replaced 845% by the correct number (822%, p. 7 L. 39). This change had no effect on the conclusions of this study. The relative effect size for groundwater  $CO_2$  concentrations was correct.

Comment 1.16 P8, L29 ff: Discuss your results in the same order as you present the results.

Reply: We disagree with this suggestion. We think that it is most logical to start the results section with the background data (chemistry, hydrology) and slowly built up to finish off with GHG fluxes, but to start the discussion with the main finding (effects on GHG fluxes) and then relate this to findings on hydrology and chemistry.

Change: No change will be carried out.

## **Comment 1.17** P9, L4: enhanced organic matter degradation, but maybe also increased organic matter input due to forestry activity in the first place?

Reply: Enhanced organic matter degradation does not exclude the suggested mechanism of enhanced organic matter inputs from logging residues. However, we agree that logging residues should be specifically highlighted as a potential source. Logging resides indeed often increase nutrient and carbon decomposition and leaching, (e.g. Palvianen et al. 2004, Plant and Soil 263; Mäkiranta et al. 2012, Soil Biology and Biochemistry 48). Some CO<sub>2</sub> and CH<sub>4</sub> may be formed from degradation of logging residues



*in the soil and partially be emitted directly from soils to the atmosphere and partially contribute to groundwater CO2 and CH4 levels. However, the relative magnitudes of these fates are presently unclear.* 

*Change: We now mention this alternative explanation along with other potential explanations of the observed groundwater concentrations (p. 9 L. 22-24).* 

Comment 1.18 P9, L5: actually, the explanation would be the reduced CH4 oxidation

Reply: We agree with the reviewer.

Change: We now use the more specific term "oxidation" instead of "net uptake" (p. 9 L. 22).

**Comment 1.19** P9, L21: info/effects on wind speed are summarized in table 2, not table 4. Not sure if you can draw any conclusions on additional forcing on air-water gas exchange velocities, since you actually didn't measure wind speed above the lake. Also considering this, it would be interesting to see the effects on lake water GHG concentrations. Did you check this? If there are no significant effects, maybe just mention this in the first sentence of paragraph 3.3 (i.e. 'Forest clear-cuts did not affect lake water GHG concentrations (data not shown).').

*Reply: Thank you for pointing out the typo and suggesting to mention how GHG concentrations responded to the treatment! We indeed tested for BACI effects on greenhouse gas concentrations, but did not detect any significant effects, except for stream CO<sub>2</sub> concentrations (decrease). (see also reply to comment 2.0).* 

We agree that wind measured above the mires may differ from wind above the lakes. Although the wind may not be exactly same in **absolute** terms, the weather stations were installed at mire locations with wind conditions as similar as possible to the lake and we are confident that our data adequately reflect the **relative** differences between lakes and years. In the clear-cut catchments, wind was measured on open mires right next to the lakes. The mires had about the same size as the lakes and were surrounded by similar vegetation. The forest buffer zone left around the mires was similar to the forest buffer zone left around the lakes. In Lillsjölidtjärnen, the mire-buffer zone was slightly wider, while in Struptjärn, the mire-buffer zone was slightly more narrow (Fig. 2).

Change: We now refer to Table 2 instead of Table 4 (p. 9 L. 40).

We have added a brief note to Text S2 on how representative our wind speed measurements on mires were for lake conditions. In the discussion, we now acknowledge the uncertainties in wind speed estimates and toned down our interpretation that forest buffer zones effectively buffered greenhouse gas emissions from clear-cuts (p. 9 L. 39 - p. 10 L. 1).

We have added a note on the treatment effects on GHG concentrations in the results section as suggested by the reviewer (p. 8 L. 6-10), added the associated BACI results to the supplementary material (Table S6) and now refer to these results in the discussion (p. 9 L. 24-32). The decrease in stream  $CO_2$ concentrations in response to clear-cutting have given us further support for our hypotheses that riparian or in-stream processes buffered clear-cut effects.



Comment 1.20 P9, L27: however, this does not explain the results for CH4?

Reply: Indeed, this would only explain results for CO<sub>2</sub>. Enhanced in-stream methane oxidation in the sediments is likely primarily an effect of the commonly found substrate limitation of methane oxidation (e.g. Bastviken 2009; Duc et al 2010; Segers 1997), i.e. methane oxidizer communities have a higher capacity than commonly expressed and will oxidize more CH4 when concentrations increase.

*Change: We now clarify this possible explanation for CH*<sub>4</sub> *in the revised text (p. 10 L. 11-15).* 

**Comment 1.21** P9, L38-39: 'The relative pH decrease of 0.5 units...' – but the Effect size (slope) of pH in Table 2 is 0.00.

*Reply:* As indicated by "\*" and in the footnote, the model parameter estimates are based on logtransformed data (to follow best practices in calculating statistics for pH and accounting for the fact that pH is  $-log_{10}(activity of H^+)$ ). Due to rounding, the slope appears to be 0.00, but is in fact -0.0000383 (Lake Epi) and -0.000077 (Stream).

*Change: We have increased the number of decimals shown for pH in Table 2 to make the data appear correctly. We did the same for absorbance (abs420).* 

**Comment 1.22** P18 ff: check all your tables for consistency (i.e. compare with the numbers you write in your results).

Reply: Thank you for this reminder.

Change: We have checked for consistency for all numbers in the tables and text. We have corrected minor mistakes in numbers given on page 8, L. 36, 37 and p. 9 L. 2, 4). These corrections had no effect on the discussion or conclusion.

Comment 1.23 P19, Table 2ff: p-value: maybe highlight significant effects

Reply: We agree.

Change: We now highlight significant effects by bold p-values in all Tables.

## **Comment 1.24** P22, Figure 1: A)-C) not really clear what is shown in the pictures. Is A) and B) the same lake but picture taken from different angles? Is B) also before the clear-cut?

Reply: We apologize for not being clear enough.

Change: In the figure 1 caption, we have added "(A, D)" after "before" and "(B, C, E, F)" after "after". We have also added y-axis labels "Lillsjölidtjärnen" and "Struptjärn" to the figure to clarify which catchments the pictures refer to.

## **Comment 1.25** There is no dashed line in C)? Why are there pictures of only two of the four field sites?

*Reply: Thanks for pointing out the sub-optimal explanation of the dashed line.* 

We show only pictures of the clear-cut catchments here, because we want to highlight changes before and after clear-cutting. We did not include pictures from the control catchments to not overload the figure.

*Change: We now explain the dashed line more clearly in the figure 1 caption.* 

**Comment 1.26** Figure 2: Nice. Maybe exchange C) and D) to have the lakes in the same order as in Table 1

Reply: Thank you.

*Change: Change adopted. We have also modified the scale bars in all panels of Figure 2 to improve readability of numbers.* 

## Comment 1.27 P23, Figure 3: Boxplots instead of bars; also for Figure 6 and 7.

Reply: We disagree. We argue that the data visualization should reflect the statistical analysis. We are interested in treatment effects on the arithmetic means in greenhouse gas fluxes. This is what our BACI analysis tests for. To reflect this, we present bar charts of arithmetic mean fluxes ( $\pm$ standard error). Boxplots would be misleading as they would imply that we tested for differences in the distribution of the data. Boxplots would also not be suitable to express the uncertainties in mean values that we obtain from our error propagation procedure and show in Fig. 3, 6 and 7.

*Change: To prepare the reader better for the type of graphs we will show, we now point out more clearly in the introduction and methods section that we are interested in and tested for changes in the arithmetic mean gas fluxes (p. 2 L. 27; p. 6 L. 14).* 

### Comment 1.28 P24, L5 (Figure 4): what is 'minimum ice extent'?

Reply: We agree that this term might cause confusion. Our ice-in and ice-out dates were based on field observations. As we did not visit the lakes every day, this estimate is associated with uncertainties. We express these uncertainties by showing the maximum and minimum ice cover duration based on the earliest and latest possible ice-in and ice-off dates.

*Change: We have rephrased the figure 4 caption to improve clarity.* 

**Comment 1.29** P26, L14 (Figure 7): 'summarized as arithmetic means over ten bootstrap runs that take between-chamber variability into account (see Fig. S3)'. In Figure S3, bootstrapping is only indicated for the BACI statistics. From the Figure and the text it is not really obvious how you used bootstrapping and how you take between-chamber variability into account.



Reply: Thank you for pointing out this lack of clearness.

Change: We have modified Figure S3, now correctly indicating how errors were propagated for the areaweighted depth-zone specific averaging. In the figure caption, we now refer to Text S6 for details on the error propagation procedure.

**Comment 1.30** Supplement, P1, L34: how did you account for the much higher measurement height of the wind speed at Stortjärn?

*Reply: Thank you for pointing out this missing piece of information. We corrected wind speed from mast height to 10 m assuming a logarithmic wind profile following Crusius and Wanninkhof (2003, Limnology and Oceanography 48).* 

Change: We now clarify this on p. 4, L. 38-39

## Comment 1.31 Technical corrections:

In general, use passive voice ('atmospheric fluxes were quantified' instead of 'we quantified atmospheric fluxes'.

Reply: We agree to use passive voice wherever suitable.

*Change:* We now use passive voice throughout the manuscript unless active voice is needed to highlight our own thoughts or actions and distinguish from other thoughts or actions cited in the context of a sentence (e.g. on page 10, L. 8-21).

Introduce abbreviations the first time the respective spelled-out word is used and use abbreviations throughout the rest of the manuscript (i.e. for carbon (C), greenhouse gas (GHG),...).

### Reply: We agree.

*Change: Change adopted throughout the manuscript. We now also properly introduce the abbreviation for root mean square error (rmse) and total nitrogen (TN) and removed the abbreviation for gas chromatographer (GC) and use the full word instead.* 

P1, L10: 'greenhouse gas (GHG)'; use abbreviations throughout the rest of the manuscript.

Reply: We agree.

Change: Change adopted throughout the manuscript.

P1, L23: 'carbon (C) and nitrogen (N)'; use abbreviations throughout the rest of the manuscript.



Reply: We agree.

Change: Change adopted throughout the manuscript.

## P2, L10: 'oxygen (O2)'; use abbreviations throughout the rest of the manuscript.

Reply: We agree.

Change: Change adopted throughout the manuscript.

P2, L25: 'site preparation' (be consistent with the use of hyphen)

Reply: We agree.

Change: Change adopted (p. 2 L. 25).

P2, L26: 'CO2, CH4 and N2'

Reply: We agree.

Change: Change adopted (p. 2 L. 27).

P2, L32: '1-3 °C'

Reply: We agree.

Change: Change adopted throughout the manuscript.

P3, L32-33: 'At the deepest point of each lake, at the stream master site and at the groundwater wells...'

Reply: We agree.

Change: Change adopted (p. 3 L. 38).

P4, L24: For both lakes and streams gas transfer velocities (k), the water column depth that equilibrates with the atmosphere per unit time, were obtained as described in the following. (Use passive voice, no comma after "streams", no hyphen in "gas transfer")

Reply: We agree.

*Change: Change adopted (p. 4 L. 21). We also now use italic letter for the coefficients k and k\_{600} throughout the manuscript and supplementary material.* 

P4, L26: 'wind speed'



Reply: We agree.

Change: Change adopted (p. 4 L. 31).

P4, L37: delete 'respectively'

Reply: We agree.

Change: Change adopted (p. 5 L. 5).

P5, L2: 'sub-reach'

Reply: We agree.

Change: Change adopted (p. 5 L. 9).

P5, L20-21: 'Atmospheric CO2 and N2O concentrations were 425 ppm and 350 ppb (median of biweekly in-situ measurements), respectively, and atmospheric

Reply: We agree.

Change: Change adopted. We now also specify the method of these in-situ measurements (p. 5 L. 28-29).

P5, L40: '...were the arithmetic mean flux of all chambers located at the respective depth.'

Reply: We agree.

Change: Change adopted (p. 6 L. 8).

P6, L3: 'site preparation'

Reply: We agree.

Change: Change adopted (p. 6 L. 12).

P6, L9: 'soil sampling' – before you just talking about groundwater sampling so try to be consistent with the wording. See also P7, L7.

Reply: We agree.

*Change: We are now consistent with the wording throughout the manuscript and supplementary material.* 



P6, L12: (Pinheiro et al., 2015) is the citation for the R package so put it after "'lme' function"; also give citation for the program R and mention which version you used.

## Reply: We agree.

*Change:* Change adopted (p. 6 L. 21). We now introduce in Chapter 2.3 that "All data analysis described in the following were done using the statistical program R 3.2.2 (R Development Core Team, 2015), if not declared otherwise." (p. 4 L. 14-15).

## P7, L4: '16.5 °C'

## Reply: We agree.

Change: Change adopted (p. 7 L. 15). Note that the number changed (see response to comment 1.13)

## P7, L8: delete 'Here'.

Reply: We agree.

Change: Change adopted (p. 7 L. 19).

## P8, L1: the symbol for mole is 'mol' not 'M', i.e. 99 mmol m-2 d-1. See also L4, L5, L10.

Reply: We agree.

Change: Change adopted (p. 8 L. 18).

## P8, L6: delete 'clear' (it's double)

Reply: We agree.

Change: Change adopted (p. 8 L. 24).

## P8, L14: 'mmol m-2 d-1'

Reply: We agree.

Change: Change adopted throughout the whole manuscript.



P8, L16: 'varied from 1.2 to 1.3 mmol m-2 d-1 in the control stream and from 0.07 to 0.18 mmol m-2 d-1 in the impact streams'

Reply: We agree.

Change: Change adopted (p. 8 L. 34).

P8, L22: delete 'linear mixed-effects models' or just use abbreviation

Reply: We agree.

Change: Change adopted (p. 8 L. 40).

P8, L26 and L28: '\_mol m-2 d-1'

Reply: We agree.

Change: Change adopted (p. 9 L. 2, 4 and even p. 8 L. 32-34)

P9, L2: 'However, aquatic GHG emissions are also fueled by direct catchment inputs of the respective dissolved gases'

Reply: We agree.

Change: Change adopted (p. 9 L. 19).

P9, L8: replace 'in average' with 'on average'

Reply: We agree.

Change: Change adopted (p. 9 L. 26).

P20, Table 3: 'Effect size of forest clear-cutting on DIC and CH4 concentrations (\_M) in groundwater in the impact catchments.'

Reply: We agree.

Change: Change adopted (Table 3).

P25, L4 (Figure 5): replace 'lakes' with 'streams'



Reply: Thank you for spotting this typo!

Change: Change adopted (Figure 5).

## P25 f Figure 6 and Figure 8: delete 'dissolved'

Reply: We agree.

*Change: Change adopted (Figure 6, 8).* 

Supplement, P2, L17: 'dissolved inorganic carbon (DIC)' – you already use the abbreviation before (e.g. in L8 and in the main text)

Reply: We agree.

Change: Change adopted (Text S1, p. 1 L. 26).

Supplement, P14, Table S4: check numbers! "Before" should have the same values as in Table 2, right?

Reply: The mean values reported in Table 2 and Table S4 are indeed not exactly the same for some variables (e.g. lux). For these variables, we report bootstrapped mean values that take uncertainty due to gap filling into account. Hence, slight differences in reported mean values can occur, but note that these values are similar in a statistical sense (within the limits of their standard error).

Change: No change will be carried out.

## Reviewer 2 (RC2)

**Comment 2.0** Klaus et al. studied greenhouse gas emissions (CO2, CH4, and N2O) from lakes and streams in catchments that underwent forest harvesting. Using a BACI design in four boreal catchments, they found very little change in greenhouse gas emissions after harvesting. The study was well designed and well executed. The manuscript is well written. I have some minor comments and suggestions for improvements. The only major comment I have is that as far as I can tell the authors don't report the differences in CO2, CH4, and N2O concentrations in surface water in lakes and streams, they just report the fluxes. The only significant difference they found is in concentrations of the greenhouse gases in ground water, but what about concentrations in surface water? If there is a lack of difference in concentrations, that might help reduce the number of potential explanations for the lack of responses in fluxes. If there were no differences in concentrations, the authors should state that.

*Reply: Thank you for suggesting to mention how GHG concentrations responded to the treatment! We indeed tested for clear-cutting and site preparation effects on greenhouse gas concentrations, but did not* 



detect any significant effects, except for stream  $CO_2$  concentrations (decrease) (see also reply to comment 1.19).

Change: We now briefly mention the treatment effects on greenhouse gas concentrations in streams and lakes in the results section (p. 8 L. 6-10), add associated BACI statistics to the suppl. material (Table S6) and refer to these results in the discussion (p. 9 L. 24-31). The decrease in stream  $CO_2$  concentrations in response to clear-cutting gave us further support for our hypotheses that riparian or in-stream processes buffered clear-cut effects.

## Below I provide specific comments.

**Comment 2.1** Page 1, lines 11-14- I would separate into two sentences after the word Catchments. It is a very long sentence!

Reply: We agree.

Change: We separated the sentence into two (p. 1 L. 11-14).

**Comment 2.2** Page 5, line 4- seems like low agreement between k600 measurements and estimates. Is this common in the literature?

Reply: Our  $k_{600}$  estimates indeed show relatively poor agreement between methods. However, low agreement across different methods is common in running waters (Lorke et al. 2015, Biogeosciences 12; Hall and Madinger 2018, Biogeosciences 15) just as it is in lakes (Gålfalk et al. 2013; Journal of Geophysical Research 118), mainly because there can be extensive high-resolution variability in k in both time and space so differences also between nearby locations and over short distances are to be expected.

Change: No change will be carried out.

## Comment 2.3 Page 5, line 25- add: after modifications

Reply: We agree.

Change: Change adopted (p. 5 L. 35).

**Comment 2.4** Page 7 line 35- why are concentrations of CO2, CH4, and N2O in lake and stream water not reported?

*Reply: Thank you for this question! We realized (also per comment 2.6) that reporting concentrations of greenhouse gases in lakes and streams would provide valuable context to the flux estimates given, and facilitate comparisons to results given for groundwater concentrations.* 



*Change: We now report greenhouse gas concentrations and set changes in groundwater into the context of stream and lake water concentrations in the discussion (see response to comment 2.0).* 

**Comment 2.5** Page 8, line 41- N2O does not result from bacterial decomposition of inorganic N. It results from incomplete denitrification and nitrification. I would reword this sentence.

Reply: Thanks for pointing out this inaccuracy!

*Change:* We reworded the sentence as suggested and also modified the following sentence and add "DIN" in addition to DOC derived from catchment soils as a potential driver of aquatic greenhouse gas fluxes (p. 9 L. 16-18).

**Comment 2.6** Page 9 lines 9-12- I don't follow the percent increase in CO2 and CH4 calculations. Is the 8.45 fold increase, the equivalent of an 845% increase? Also, I am a little confused because these are calculations for changes of concentrations, but you never provide the concentrations changes for lake and stream water, just the fluxes.

*Reply: The relative effect size (here: 56%) is the effect size divided by the mean value in the impact system in the control year (see also our reply to comment 1.15).* 

*Change:* We now explain more clearly in the methods section how slope and intercept is calculated (p.6 L. 18-19). We also now express more clearly that the effect size here is given relative to the value at the impact site in the control year (p. 7 L. 36, 40).

We realized that the relative effect size was incorrect for groundwater  $CH_4$  concentrations: we have replaced 845% by 822% (p. 7 L. 39). This had no effect on the conclusions of this study. The relative effect size for groundwater  $CO_2$  concentrations was correct.

Comment 2.7 Page 9 line 20- I think the word "remain" should be changed to "retain"

Reply: Thank you for spotting this typo.

Change: Change adopted (p. 9 L. 39).

**Comment 2.8** Table 2- why do the Control and Impacts have such different discharges (27-40 L/s versus 3-4 L/s).

*Reply: The streams included in our study are all representative for headwater streams in the Swedish Boreal forested landscape. The control stream had higher discharge than the impact streams because of its larger catchment area (catchment-area specific discharge was similar in all catchments (1.0, 1.4 and 1.5 mm d<sup>-1</sup> in Struptjärn, Lillsjölidtjärnen and Övre Björntjärn in June-September 2012) and well within the range of what has been measured previously in our study region (c.f. Lyon et al. 2012, Water Resources Research 48). Our study focusing on greenhouse gas emissions from streams and lakes is only* 



one of many that are about to result from this project (see e.g. Deininger et al., accepted in "Ecological Applications", focusing on clear-cut effects on in-lake basal productivity). The different interests in this experiment made it particularly challenging to find control and impact catchments that were similar in all variables of interest.

*Change: We have added specific discharge data as background information to the study site description* (*p. 3, L. 2*) *and to the results section* (*p. 7 L. 10*).

**Comment 2.9** Figure 3- why 37.5-42.5 and then 5-105cm depth? It seems strange to have a shallow and then the whole soil column together? Why not separate shallow vs deep?

*Reply: We agree that this figure legend was confusing and suggest a clearer description of the soil water sampling.* 

Depth specific groundwater sampling was done to target depths that are hydrologically most strongly connected to stream water (Leith et al. 2015). Depth integrated sampling was done to characterize the whole soil profile.

Change: We have altered the figure 3 legend and instead write "Concentrations of DIC and dissolved  $CH_4$  in groundwater at depth specific locations (37.5-42.5 cm; panel A-B) and depth integrated locations (5-105 cm; C-D)...". We now introduce the terms "depth-specific" and "depth integrated" in Chapter 2.3 and clarify why we sampled at a specific depth and integrated across the whole soil profile (p. 3 L. 32-34). This implied moving some of the details given in Text S2 to the main text.

We are now more consistent throughout the manuscript and only speak of "groundwater" instead of "soil water" as done partly in the previous version of the manuscript. We also now specify more clearly in the captions of Table 2 and S4 that groundwater refers to the depth-integrated locations.

**Comment 2.10** Figure 5- it would make easier to compare across sites if all panels had the same scale on the y-axes.

Reply: We agree.

Change: We have modified Figure 5. Now, the y-axis scale is the same in all panels that show  $CO_2$  fluxes in the three study streams.

## Further changes done

We noticed slight inaccuracies and missing details in the methods description of the gas transfer velocity measurements (Text S4). We have correct these mistakes and add some details.

We realized that the first paragraph of the introduction focused on carbon cycling only while our manuscript also includes nitrogen cycling ( $N_2O$ ). To make this consistent, we now also refer to nitrogen



cycling and included two additional references (Sponseller et al. 2016, Seitzinger and Kroeze 1998), replacing one of the carbon-related references (Jonsson et al. 2007) (p. 1 L. 32-33).

We have added a brief clarification in chapter 2.2 that treated catchments and sites are referred to as 'impact' and untreated ones as 'control' in the remainder of the manuscript (p. 3 L. 17-18).

We realized that we overlooked a statistically significant treatment effect on stream DIN concentrations. We now mention this effect in the results (p. 7 L. 30) and discussion section (p. 11 L. 8).

In the submitted manuscript, we cited Deininger et al. (unpublished data). This data is now accepted for publication in the Journal *Ecological Applications*. We now refer to this reference instead (p. 8 L. 11).

In the submitted manuscript, we used the term "emission" in some occasions. Since streams and lakes are not necessarily consistently oversaturated in greenhouse gases relative to the atmosphere (which would imply emission), we now consistently use the term "air-water fluxes" throughout the whole manuscript.

We noticed that we refer to two different references by Schelker et al. (2013) but did not distinguish in the main text which of the two we refer to. We now clearly indicate by letters "a" and "b" which reference we refer to (p. 9 L. 22; p. 10 L. 37).

The name of Lillsjölidtjärnen was misspelled in Fig. S2. We have corrected this.

We added background information on the sign of measured GHG fluxes in the abstract (p. 1 L. 15-16) and results (p. 8 L. 12).

We noticed that discharge given for the inlet of Övre Björntjärn was inconsistent between Table 1 and 2 (41.7 vs. 40.9 L s<sup>-1</sup>). We corrected the value in Table 1 to be consistent with Table 2).

We corrected a minor mistake in a value given in Table S5 (992 instead of 991).

Due to methodological concerns in Weyhenmeyer et al. 2015, we replaced this reference by an equivalent one (Bogard and del Giorgio 2016) (p. 1 L. 37).

We did some minor editing of the text to improve text flow and correct spelling and grammar mistakes.

# Greenhouse gas emissions from boreal inland waters unchanged after forest harvesting

Marcus Klaus<sup>1\*</sup>, Erik Geibrink<sup>1</sup>, Anders Jonsson<sup>1</sup>, Ann-Kristin Bergström<sup>1</sup>, David Bastviken<sup>2</sup>, Hjalmar Laudon<sup>3</sup>, Jonatan Klaminder<sup>1</sup>, Jan Karlsson<sup>1</sup>

<sup>1</sup>Department of Ecology and Environmental Science, Umeå University, SE-90187 Umeå, Sweden <sup>2</sup>The Department of Thematic Studies - Environmental Change, Linköping University, SE-58183 Linköping, Sweden <sup>3</sup>Department of Forest Ecology and Management, Swedish University of Agricultural Science, SE-90183 Umeå, Sweden *Correspondence to: Marcus Klaus (marcus.klaus@posteo.net)* 

Abstract. Forestry practices <u>often</u> result in an increased export of carbon and nitrogen to downstream aquatic systems.
 Although these losses affect the greenhouse gas (<u>GHG</u>) budget of managed forests, it is unknown if they modify <u>GHG</u> emissions of recipient aquatic systems. To assess this question, <u>hir-water</u> fluxes of carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) were quantified <u>for</u> humic lakes and their inlet streams in four boreal catchments <u>using a Before/After-Control/Impact-experiment</u>. <u>Two catchments were</u> treated with forest clear-cuts followed by site preparation (18% and 44% of the catchment area), <u>GHG</u> fluxes and hydrological and physicochemical water characteristics <u>were measured</u> at multiple

- 15 locations in lakes and streams at high temporal resolution throughout the summer season over a four year period. Both lakes and streams evaded all GHGs, implying impact of terrestrial export on carbon cycling in the recipient waters. Yet, the treatment did not significantly change <u>GHG fluxes in streams or lakes within three years after</u> the treatment, despite significant increases of CO<sub>2</sub> and CH<sub>4</sub> concentrations in hillslope groundwater. Our results highlight the importance of the riparian zone-stream continuum as effective biogeochemical buffers and wind shelters to prevent <u>GHG</u> leaching from forest clear-cuts and evasion
- 20 via downstream inland waters. These findings are representative for low productive forests located in relatively flat landscapes where forestry practices cause only a limited initial impact on catchment hydrology and biogeochemistry.

#### **1** Introduction

Land use activities have greatly enhanced inputs of carbon (C) and nitrogen (N) from terrestrial or atmospheric sources to the aquatic environment, reducing the terrestrial C sink function and aggravating global climate change (Dawson and Smith, 2007;
Regnier et al., 2013; Vitousek et al., 1997). The terrestrial C sink is largely determined by forest ecosystems which contribute to a net uptake of greenhouse gases (GHG) from the atmosphere (Goodale et al., 2002; Myneni et al., 2001). This net uptake can be further increased by well-informed forest harvesting strategies (Kaipainen et al., 2004; Liski et al., 2001). Hence, forest management is a widely used instrument to fulfill GHG budget commitments under the Kyoto Protocol (IGBP Terrestrial Carbon Working Group, 1998). Yet, mitigation measures neglect that a significant part of terrestrial C and N taken up by forests is exported to aquatic systems (Battin et al., 2009; Öquist et al., 2014; Sponseller et al., 2016). These exports are sensitive to logging activity (Nieminen 2004; Schelker et al. 2012; Lamontagne et al. 2000) and a large proportion is processed in inland waters and emitted back to the atmosphere as GHGs such as carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>) and nitrous oxide (N<sub>2</sub>O) (Cole et al., 2007; Seitzinger and Kroeze, 1998). Revealing potential changes in the GHG budget of the aquatic environment downstream forest clear-cuts is therefore crucial to evaluate the overall potential of forestry to mitigate climate

35 warming

Forestry effects on aquatic <u>GHG</u> emissions are largely unknown and difficult to predict due to multiple processes involved. In boreal headwaters, stream and lake CO<sub>2</sub> and CH<sub>4</sub> originate largely from soils (Bogard and del Giorgio, 2016; Hotchkiss et al., 2015; Rasilo et al., 2017), These soil-derived inputs typically increase after forest clear-cutting because of increased soil respiration (Bond-Lamberty et al. 2004; Kowalski et al. 2003) and discharge (Andréassian, 2004; Martin et al.,

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2000). Forest clear-cutting often also increase dissolved organic carbon (DOC) export to streams and lakes (Schelker et al. 2012; Nieminen 2004; France et al. 2000) where it stimulates respiration and reduces light penetration, lake primary production and net  $CO_2$  uptake (Ask et al. 2012; Lapierre et al. 2013). Therefore, any elevated terrestrial <u>C</u> inputs due to forest clear-cutting may further increase net heterotrophy and  $CO_2$  emissions (Ouellet et al., 2012) or stimulate methanogenic bacterial

- 5 activity in lakes (Huttunen et al., 2003). Forest clear-cuts also often enhances nutrient exports, with less pronounced changes for phosphorous, but large increases for N<sub>e</sub> especially for nitrate (Nieminen 2004; Palviainen et al. 2014; Schelker et al. 2016). Nitrate leakage affect <u>GHG</u> cycling in boreal inland waters, yet predictions on the direction of net effects are difficult. Nitrate inputs may suppress (Liikanen et al., 2003) or stimulate (Bogard et al., 2014) CH<sub>4</sub> production, enhance CH<sub>4</sub> oxidation (Deutzmann et al., 2014) and promote denitrification and N<sub>2</sub>O emissions (McCrackin and Elser, 2010; Seitzinger and Nixon,
- 10 1985). Nitrate inputs to N limited boreal aquatic systems stimulate phytoplankton production and thereby enhance  $CO_2$  uptake and oxygen ( $O_2$ ) production (Bergström and Jansson, 2006). Increases in DOC would, however, consume  $O_2$  (Houser et al., 2003). Changes in  $O_2$  concentrations would influence the balance between methanogenesis and methanotrophy (Bastviken et al. 2008), as well as nitrification and denitrification (Mengis et al. 1997). Removal of riparian vegetation may increase littoral light availability and water temperature (Steedman et al. 2001, Moore 2005), with potential effects on net  $CO_2$  and  $CH_4$
- production (Wik et al., 2014; Yvon-Durocher et al., 2012, 2014). Forest clear-cuts <u>could</u> also increase wind exposure (Tanentzap et al., 2008; Xenopoulos and Schindler, 2001) and thus result in increased gas transfer velocities as indicated by the wind based relationships found in lakes (Cole and Caraco, 1998). Likewise, enhanced discharge may affect turbulence and gas transfer velocities in streams (Raymond et al., 2012). Clear-cut effects on hydrology and biogeochemistry can be further amplified by site preparation, the trenching of soils before replanting (Schelker et al. 2012; Palviainen et al. 2014).
- Even though spatial surveys indicate that changes in vegetation (Maberly et al., 2013; Urabe et al., 2011), forest fires (Marchand et al. 2009) and forestry activities (Ouellet et al., 2012) affect the <u>GHG</u> balance of inland waters, mechanistic evidence from whole-catchment forest manipulation experiments is lacking. Here, the impact of forest clear-cuts and site preparation on summer season means of air-water CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O fluxes was experimentally assessed for streams and lakes in four boreal headwater catchments. A whole-catchment manipulation experiment was performed using a Before-After/Control-Impact (BACI) design. Two "impact" catchments received a forest clear-cut and site preparation following one year of pre-treatment sampling. Two "control" catchments were left untreated throughout the whole study period of four years. We hypothesized an increase in aquatic CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O emissions in response to forest clear-cuts and site preparation.

#### 2 Methods

#### 2.1 Study sites

30	Sampling was carried out during June-September 2012-2015 in four headwater lakes and three lake inlet streams (one lake
	lacks an inlet stream) in the catchments (220-400 m a.s.l.) of Övre Björntjärn, Stortjärn, Struptjärn and Lillsjölidtjärnen,
	northern Sweden (Table 1, Fig. 1). During the experimental period, mean annual temperature in the region was 1-3_°C higher
	than the long-term average (1960-1990) of 1.0 °C, while annual precipitation was <u>close to the long-term average of 500-600</u>
	mm in all years, except for 2012 (800 mm) (http://www.smhi.se/klimatdata/meteorologi). In the study catchments, mean
35	summer air temperatures and precipitation sums (June-September) varied between 11.1_°C and 342 mm in 2012 and 12.8_°C
	and 245 mm in 2014, respectively (Table S1). Catchment soils were typically well drained and characterized by podzol

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developed on locally-derived glacial till and granitic bedrock. The catchments were mainly (>85%) covered by managed coniferous forest (*Picea abies*, *Pinus sylvestris*) with scattered birch trees (*Betula* sp.) and minerogenic oligotrophic mires (<15%). Site quality class was rather low with timber productivities of 2-3 m<sup>3</sup> ha<sup>-1</sup> yr<sup>-1</sup> (SLU, 2005). The catchments were

drained by a hand dug ditch network established in the early 20th century to improve the forest productivity. The riparian zone was about 2-6 m wide and characterized by organic rich peat soils. The regional hydrology is characterized by pronounced

snow melt episodes in April/May, summer and winter low flow periods and autumn storms. Drainage channels all culminate in single lake inlets with specific discharges of 1.0, 1.4 and 1.5 mm d<sup>-1</sup> in Struptjärn, Lillsjölidtjärnen and Övre Björntjärn in June-September 2012. The study lakes were shallow, small, humic and dimictic with a seasonal mixed layer depth of 0.5-2 m during summer stratification lasting from late May to Mid-September. Lake ice was present from late October to Mid-May during the study period.

#### 2.2 Forest clear-cutting and site preparation procedure

Forest clear-cutting was carried out on snow-covered (~60cm) frozen soil in February 2013 in the catchments of Struptjärn and Lillsjölidtjärnen by national or private forest companies according to "common practice" methods of whole-tree harvesting where about 30% of tops, twiggs and needles were left on-site (Fig. 1). The forests cut were coniferous forest with an age of about 90-120 years. In early November 2014, clear-cuts were site-prepared by disc trenching, a common soil scarification

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method to improve planting conditions (Fig. 1C). Clear-cut areas were defined by the forest companies, 14 ha and 11 ha in size, and corresponded to 18% and 44% of total lake watershed areas, respectively (Table 1). Clear-cuts covered 40% and 60% of the stream reaches of the inlets of Struptjärn and Lillsjölidtjärnen, respectively. Along the inlet stream of Lillsjölidtjärnen, 10-70 m wide stream buffer strips were left and remained intact throughout the study period. Buffer strips along the inlet

15 stream of Struptjärn were <10 m (Fig. 1E) and damaged by a wind throw event in winter 2014/15, where 70% of trees within the buffer strip fell along half of the clear-cut affected reach, causing bank collapse and soil erosion (Fig. 1F). Lake buffer strips were 15-60 m wide in both catchments and stayed largely intact throughout the study period. <u>Hereafter, treated</u> catchments and sites are referred to as "impact" and untreated ones as "control".

#### 2.3 Water sampling and physicochemical analysis

- 20 Surface water was sampled biweekly for dissolved CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O concentrations at a stream-site close to the lake inlet (hereafter referred to as "master" stream site) and the deepest point of the lake (Fig. 2) as described in Text S1. Control and impact catchments were typically sampled within two or three days, but never more than seven days from each other. To account for temporal variability, surface water CO<sub>2</sub> concentrations was also monitored at the deepest point of the lakes and the master stream sites at 2-hour intervals using non-dispersive infra-red CO<sub>2</sub> sensors (see Text <u>\$2</u> for details). To account for
- spatial variability in CO<sub>2</sub> and CH<sub>4</sub> concentrations <u>within streams</u>, 300 m long stream transects <u>were sampled</u> at five sites chosen to represent the variability in riparian vegetation and turbulence patterns of the catchment stream. Spatial variability within lakes was accounted for by biweekly sampling of CO<sub>2</sub> concentrations at additional four near-shore locations (Fig. 2). Average near-shore concentrations did not differ from concentrations at the deepest point (linear regression with insignificant intercept and slope= $0.97\pm0.01$ , p<0.001, R<sup>2</sup>=0.99, n=130). Therefore, only data from the deepest point was used for the remainder of
- 30 this work. Within-lake variability in CH<sub>4</sub> concentrations was accounted for by floating chamber deployments as described further below. In impact catchments, groundwater was sampled biweekly for dissolved inorganic carbon (DIC) and CH<sub>4</sub> concentrations from wells at depth integrated locations (5-105 cm) and depth specific locations (37.5-42.5 cm). These depths were chosen to separate responses in the whole soil profile and in shallow groundwater that is tightly connected to streams in our study region (Leith et al., 2015). Wells were located at two forested hillslope sites, one affected by forest clear-cutting and
- 35 one serving as an untreated reference, (Fig. 2, Text S1).
  Profiles of dissolved O<sub>2</sub> concentrations and photosynthetic active radiation (PAR) were measured biweekly at the deepest point in each lake using handheld probes (ProODO, YSI Inc., Yellow Springs, OH, USA; LI-193 Spherical Quantum

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Sensor, LI-COR Biosciences, Lincoln, NB, USA). At the deepest point of each lake, <u>at</u> the stream master site and <u>at the</u> groundwater wells additional water samples were collected biweekly in acid washed plastic bottles for physicochemical

analysis. At the master stream sites, samples were taken daily by an automatic water sampler (ISCO 6712 Full-Size Portable

Sampler, Teledyne Inc., Lincoln, NE, USA). At each field visit, 2-4 of these samples were chosen based on the recorded hydrograph to represent the variability of the flow conditions during the past two-week period.

We also monitored lake water temperature profiles, stream temperature and discharge and weather conditions including wind speed, air temperature, precipitation, air pressure, relative humidity and light intensity at 5-60 min intervals using logger systems described in detail in Text <u>\$2</u>. Between 3 and 12% of logger data was missing and filled using multiple imputation, linear regression or linear interpolation methods (see Text \$3 and Table \$2),

 $\sqrt{10}$  characterize <u>water</u> color, spectral absorbance at a wavelengths of 420 nm (a<sub>420</sub>) was measured on filtered <u>lake and</u> stream water (acid washed Whatman GF/F 0.7µm) using a spectrophotometer (V-560 UV-VIS, Jasco Inc., Easton, MD, USA). Filtered water <u>from lake-</u>, stream- and depth integrated groundwater sampling was acidified with 500 µl of 1.2 M HCl per 50

10 ml of sample prior to analysis <u>for DOC and total N (TN) analysis</u> on a total organic <u>C</u> analyzer (IL 500 TOC-TN analyzer, Hach Lange, CO, USA). For dissolved inorganic nitrogen (DIN =  $NO_2^{-} + NO_3^{-} + NH_4^{+}$ ) analysis, <u>water from lake-</u>, <u>stream-</u> and depth integrated groundwater sampling were filtered through 0.45 µm cellulose acetate filters prior to freezing and analyzed using an automated flow injection analyzer (FIA star 5000, FOSS, Denmark). <u>All chemical analyses were performed</u> at the Department of Ecology and Environmental Science (EMG), Umeå University. <u>All data analysis described in the</u>

15 following were done using the statistical program R 3.2.2 (R Development Core Team, 2015), if not declared otherwise.

#### 2.4 Lake physics calculations

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Lake thermal characteristics were calculated based on 5 min temperature profile data using functions provided by the <u>R-package</u> 'rLakeAnalyzer' (Read et al., 2011). This included epilimnion, hypolimnion and whole lake mean temperatures, Schmidt stability, the depths of the actively mixing layer (z<sub>mix</sub>) and the upper and lower boundary of the metalimnion (z<sub>upr</sub> and

- 20  $z_{lwr}$ , respectively). For  $z_{mix}$ , we chose a density gradient threshold value of 0.1 kg m<sup>-3</sup> per meter. Mean O<sub>2</sub> concentrations for the epilimnion (water surface to  $z_{upr}$ ), hypolimnion ( $z_{lwr}$  to lake bottom) and the whole lake were then calculated by weighting O<sub>2</sub> concentrations by the areal proportion of the depth stratum they represent and integrating these numbers through all depths, following the whole-lake depth-integrated approach by Sadro et al. (2011). Stratum-specific areas were derived from hypsographic curves, established from bathymetric data. Bathymetry data was collected using an echo sounder with internal
- 25 GPS antenna (Lowrance HDS-5 Gen2), and interpolated by ordinary kriging (root mean square error (rmse)=0.3 m) using the geostatistical analysis package in ArcMap 10.1 (ESRI, U.S.). Light extinction coefficients (k<sub>d</sub>) were calculated as the slope of the linear regression between natural logarithm of photosynthetic active radiation and depth.

#### 2.5 Gas transfer velocity estimates

For both lakes and streams gas transfer velocities (k), the water column depth that equilibrates with the atmosphere per unit
 time, were expressed as keep representing CO<sub>2</sub> transfer at 20 °C water temperature. For lakes, three published keep models were used to account for prediction uncertainties, including two wind speed based models calibrated for small sheltered lakes (Cole and Caraco, 1998) and boreal lakes of various sizes (Vachon and Prairie, 2013), and a surface renewal model calibrated for small boreal lakes (Heiskanen et al., 2014). Calculations were based on scripts provided by the <u>R-package</u> 'LakeMetabolizer' (Winslow et al., 2016). Measured input variables included wind speed, wind mast height, latitude, lake area, air pressure, air

- temperature, relative humidity and surface water temperature. Modelled input variables included k<sub>d</sub>, z<sub>mix</sub>, incoming shortwave radiation (sw=lux/244.2, following Kalff (2002)) and net longwave radiation (calculated from measured input variables using the 'calc.lw.net.base' function in the <u>R-package</u> 'rLakeAnalyzer' (Read et al., 2011). To match temporal resolutions, biweekly k<sub>d</sub> values were interpolated linearly to 10 min resolution. Wind speed was corrected from mast height to 10 m above ground assuming a logarithmic wind profile following Crusius and Wanninkhof (2003). To account for uncertainties in <u>k<sub>600</sub> estimates</u>
   for lakes due to gap filling of input variables, we applied a bootstrapping approach (Text S6). For streams, <u>k<sub>600</sub> was estimated</u>
- separately for the four sub-reaches that are bound by the five stream sampling sites. Estimations were based on a total of 23

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propane injection experiments and 282 triplicate gas flux chamber measurements carried out at 3-5 representative sites per sub-reach (Fig. S1). Propane injection experiments and flux chamber measurements were repeated 5-10 times per sub-reach, respectively during autumn 2013-spring 2015, to cover a wide range of flow conditions (0.01-0.95th, 0.10-0.99th and 0.25-0.99th percentile of discharge measurements during 1 June - 30 September in 2012-2015 in Övre Björntjärn, Struptjärn and Lillsjölidtjärnen). Details on gas transfer measurements in streams are given in Text S4.

Eux chamber measurements and propane injection experiments were used to establish predictive models of k600 based on stream discharge. Stream discharge was used instead of the more mechanistically relevant variable of flow velocity because both variables were highly correlated (marginal R<sup>2</sup>=0.71) but only discharge was available at hourly intervals. Hence, hourly k<sub>600</sub> was computed for each sub-reach in four steps. First, the arithmetic mean k<sub>600</sub> of site-specific flux chamber measurements

- 10 was calculated for each sub-reach. These sub-reach specific k<sub>600</sub> agreed relatively well with k<sub>600</sub> from propane-injection experiments (R<sup>2</sup>=0.58, Fig. S2). However, flux chamber measurements were restricted to relatively smooth water surfaces, excluding waterfalls and rapids and therefore underestimated reach-scale k600 by a factor of 0.61. Second, flux chamber-derived  $k_{600}$  was corrected using linear relationships (median R<sup>2</sup>=0.90) with propane-derived  $k_{600}$  whenever they were statistically significant or  $R^2$  was >90%. Third, corrected flux-chamber derived  $k_{600}$  was combined with propane-derived  $k_{600}$  values to
- 15 establish sub-reach specific linear regression models that predict  $k_{600}$  based on local discharge (R<sup>2</sup>=0.56-0.94, Table S3). Whenever the best linear model had a negative intercept, the model was refitted, constraining the intercept to zero to avoid negative predicted  $k_{600}$ . Fourth, the  $k_{600}$ -discharge models were used to predict  $k_{600}$  based on hourly time series of discharge measured at the master stream site and scaled to the respective sub-reach using the mean discharge ratio measured at both sites during propane injection experiments. Throughout these experiments, discharge ratios varied by 5±2%. To account for uncertainties in k<sub>600</sub> estimates for streams we propagated errors from flux chamber and propane injection experiments and

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#### 2.6 Gas flux estimates

discharge rating curves (Text S6).

The diffusive gas flux (F) across the lake or stream water interface was calculated using Fick's law

 $F = a(c_{wat} - c_{ea})k$ 

25 where  $c_{wat}$  is the measured <u>gas</u> concentration of the water,  $c_{eq}$  is the <u>gas</u> concentration of water if it was in equilibrium with ambient air calculated from measured air concentration and water temperature using Henry's constant, and a is the chemical enhancement factor (for CO2 transfer only) set to 1, as enhancement is negligible if pH < 8 (Wanninkhof and Knox, 1996). Atmospheric CO2 and N2O concentrations were 425 ppm and 350 ppb (median of biweekly in-situ measurements using gas chromatography, Text S1)), respectively, and atmospheric CH<sub>4</sub> concentrations were below the detection limit of our gas 30 chromatographer (~3 ppm) and assumed to be 1.893 ppm (http://cdiac.ornl.gov/pns/current\_ghg.html). The coefficient k was calculated from kooo following Jähne et al. (1987), with the Schmidt coefficient, set to -0.5 and using gas-specific parameterizations of Schmidt numbers for in situ water temperature according to Wanninkhof (1992). Errors in F were

propagated from standard errors in  $c_{wat}$  and k (Fig. S3).

- Fluxes of CH<sub>4</sub> were measured in 2012 and 2014 using floating chambers according to Bastviken et al. (2010) with 35 the following modifications: 26-32 chambers were placed in each lake to cover five depth zones (water depth 0-1 m, 1-2 m, 2-3 m, 3-4 m and >4 m) with one chamber placed at the deepest point and the remainder arranged along depth transects of 3-4 chambers (Fig. 2). Depth transects were chosen to represent the typical shore-line characteristics (inlets, mires, forests). A volume of 50 ml of gas was sampled weekly from June to August from each floating chamber before and after an accumulation period of 24 hours using polyethylene syringes. A volume of 30 ml of sampled gas was injected to glass vials (22 ml;
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PerkinElmer Inc., U.S.) sealed with natural pink rubber stoppers (Wheaton 224100-171) and filled with saturated NaCl solution. During gas transfer, the vials were held upside down to let the excess solution escape through an open syringe needle until around 2 mL solution was left in the vial. To minimize leakage, vials were stored upside down until analysis with a gas chromatograph (7890A, Agilent 70 Technologies, U.S.A.) with a Supelco Porapak Q 80/100 column, a 71 Flame Ionization Detector (FID) and a Thermal Conductivity Detector (TCD) at the Department of Thematic Studies - Environmental Change, Linköping University, Sweden. In addition to chamber sampling, surface water was sampled at the beginning and the end of each 24 h accumulation period in the middle of each transect and analyzed them for dissolved CH4 as described above. Chamber-specific total CH<sub>4</sub> fluxes were calculated and separated into diffusion and ebullition components using the statistical

approach described in Bastviken et al. (2004). Whole-lake fluxes were calculated as the area-weighted mean of depth-zone

specific fluxes which in turn were the arithmetic mean flux of all chambers located at the respective depth.

#### 2.7 Statistical analysis

- 10 Clear-cut and site preparation effects were assessed following the paired BACI approach of Stewart-Oaten et al. (1986). To test for 'clear-cut' effects, i.e. the general response in the first three years after clear-cutting, the 'after' period was set to the years 2013-2015. To evaluate whether effects after site preparation differed from general responses, we also tested for 'site preparation' effects by setting the 'after' period to the year 2015. The 'before' period was always the year 2012. Treatment effects were analyzed in terms of effect sizes (ES), which is defined as the arithmetic mean change (after-before) in the
- 15 sampling specific differences between groundwater-, stream or lake pairs (impact-control). The significance of the ES was tested using a linear mixed-effects model (LME) with "paired difference" as the dependent variable and "Time" (Before/After) as a fixed effect. The factor "pair" was included as a random effect on both slopes and intercepts to account for potential natural variability in responses across the two impact catchments. Reported ES, slopes and intercepts are arithmetic means over the pairs. Each impact lake was paired with the arithmetic mean of the control lakes, each impact stream with the control
- 20 stream and each impact groundwater sampling site with the respective control site in the impact catchments. All LMEs were analyzed by means of the "Ime" function in the R-package "nIme" (Pinheiro et al., 2015) using the restricted maximum likelihood approach after BACI model assumptions were evaluated (Text S5). Whenever temporal autocorrelation was significant (Text S5), a first-order autocorrelation term (corAR1, for time series of biweekly observations) or an autoregressive moving-average correlation structure (corARMA, for time series of daily means derived from hourly discharge or 2-hourly
- 25 CO2 flux data) was included.

To guarantee homoscedasticity and normality of model residuals, the dependent variables were log+n-transformed if necessary prior to model fitting, where n is the smallest value that when added leads to normal data. To assess the statistical and biogeochemical significance of clear-cutting effects we used the p-value and slope of the LMEs (as an estimate of ES) and Cohen's D, defined as D=ES/2s, where s is the standard deviation of paired differences in the before period (Osenberg and Schmitt, 1996). Cohen's D were "small" if D<0.2, "medium" if 0.2≤D<0.8, and "large" if 0.8≤D. Uncertainties in BACI statistics for gas fluxes and gap-filled logger data were accounted for by combining standard methods of error propagation a

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	<b>Deleted:</b> In addition, we ran pseudo-BACI analyses on soil sampling sites in the control catchments to assess whether differential site-specific changes may have happened within catchments that were unrelated to forest clear-cut effects.
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statistics for gas fluxes and gap-filled logger d	lata were accounted for by combining standard methods of error propagation	on and			
bootstrapping (see Text S6 and Fig. S3).					
Clear-cut effects on CO <sub>2</sub> and CH <sub>4</sub> flux	xes were also investigated on potential differences along the stream reaches	s (Fig.	-[1	Deleted: We also investigated if c	
2) depending on the site-specific percentage	of the drainage area affected by forest clear-cutting. First, stream-site-sp	ecific		Deleted: emissions	
drainage areas were delineated from a 2 m c	digital elevation model (DEM) derived from airborne laser scanning (Sw	vedish		Deleted: differed	
National Land Survey, 2015) using Hydrolog	y tools in ArcGIS 10.1 (ESRI, Redlands, CA). Modelled flow direction in	some		Deleted: To do so, we f	
ditches were not well represented by the model	l compared to field observations. In this case, the DEMs were manually com	rected		Deleted: delineated	
(elevation $\pm 20$ cm) to emphasize observed di	itch flow directions. <u>Next</u> , BACI analyses were performed as described	above		Deleted: we manually corrected	
where each impact stream site was paired wi	th the respective site in the control stream, with respect to the order rega	arding		Deleted: We then performed	

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their distance from the lake inlet. In addition, tests were carried out on linear relationships between the effect size (weighted

by SE) of each stream site and the respective percentage of forest clear-cut using an LME with "stream" as random effects on

both slopes and intercepts. Here, the dependence of sites within streams was accounted for by setting the alpha-level of the statistical analysis to 0.01. Alpha levels of all other statistical analyses were set to 0.05.

#### **3 Results**

#### 3.1 Hydrological and physicochemical response

- 5 Forest clear-cuts did not affect riparian groundwater levels or stream discharge (Table 2). Instead, these hydrological characteristics were more regulated by inter- and intra-annual variability in precipitation and snow melt water inputs. Groundwater levels decreased from 34-35 cm depth in the relatively wet control year to 40-42 cm depth in the relatively drier impact years (here and hereafter, we refer to arithmetic mean values over each time period). At the same time, stream discharge decreased from 41 to 27 L s<sup>-1</sup> in the control catchment and from 4 to 3 L s<sup>-1</sup> in the impact catchments, corresponding to a
- 10 decrease in specific discharge from 1.5 to 1.0 mm d<sup>-1</sup> and 1.1 to 0.9 mm d<sup>-1</sup>. Other physical parameters such as wind speed, light intensity, epilimnion and hypolimnion temperature and Schmidt stability also remained largely unaffected. Light intensities tripled in impact streams (from 3402 to 9969 lux, corresponding to about 14 to 41 W m<sup>-2</sup>, Kalff 2002) and showed a large effect size. This effect was, however, not significant because of high variability across impact streams (large effect in Struptjärn, no change in Lillsjölidtjärnen; stream-specific data not shown). Whole lake temperatures increased from 10.7 to
- 15 11.5 °C in impact lakes, but this increase was 0.4 °C less than the increase in control lakes, Mixing depth decreased from 1.7 to 1.5 m in impact lakes while it remained at 1.8 m in control lakes. These thermal effects were significant but of small size (Table 2).

Forest clear-cuts did not affect concentrations of O2, DOC and DIN in ground-, stream- or lake water. Epilimnion and hypolimnion O2 concentrations were around 8 and 1-2 mg L<sup>-1,</sup> respectively (Table 2). Hypolimnetic water did quickly turn anoxic during summer stratification (Fig. S4). The DOC concentrations ranged from 63 to 77 mg L<sup>-1</sup> in groundwater, 25 to 29 mg L<sup>-1</sup> in streams and 18 to 21 mg L<sup>-1</sup> in lakes (Table 2). Concentrations of DIN ranged from 467 to 538  $\mu$ g L<sup>-1</sup> in groundwater, 21 to 32  $\mu$ g L<sup>1</sup> in stream water and 14 to 20  $\mu$ g L<sup>1</sup> in lake water. Concentrations of TN decreased in impact streams from 595

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- to 505 µg L<sup>-1</sup>, a significant medium size effect relative to the increase in control streams from 498 to 531 µg L<sup>-1</sup>. However, TN remained unaffected in groundwater (1572-1958 µg L<sup>-1</sup>) and lake water (367 to 446 µg L<sup>-1</sup>). Spectral absorbance at 420 nm ranged from 12 to 15 m<sup>-1</sup> in streams and 9 to 13 m<sup>-1</sup> in lakes and was not affected by clear-cutting, However, pH showed a significant BACI effect and increased more in control systems compared to impact systems: from 3.9 to 4.8 in the control stream and from 4.4 to 4.6 in the impact streams, and from 4.2 to 5 in control lakes and from 5.1 to 5.4 in the impact lakes (Table 2).
- Most hydrological and physicochemical parameters remained unaffected by the treatment even after site preparation 30 (Table S4). The only significant BACI effects concerned medium size decreases in stream pH and increases in stream DIN concentrations, and small or medium size decreases in hypolimnetic and whole-lake temperatures and mixing depths and increases in Schmidt stability.

#### 3.2 Response of GHG concentrations

Groundwater DIC and CH4 concentrations increased in response to forest clear-cutting. Specifically, in shallow groundwater 35 (37.5-42.5 cm), DIC concentrations increased from 992 to 1345  $\mu$ M at control sites but from 957 to 1846  $\mu$ M at impact sites, a significant medium effect size of +533 µM or +56% relative to the impact site in the control year (Fig. 3A, Table 3). Wholesoil profile (5-105 cm) DIC concentrations increased at similar rates (medium effect size of + 458 µM), yet this change was not statistically significant (Fig. 3C, Table 3). CH<sub>4</sub> concentrations in shallow groundwater decreased from 24 to 16  $\mu$ M in control sites but increased from 11 to 94  $\mu$ M at impact sites, a significant *large* effect size of +93  $\mu$ M or +822% relative to the impact site in the control year (Fig. 3B, Table 3). Whole-soil profile CH4 concentrations increased at even larger absolute rates

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(+ 139 µM), but this change was only of *medium* size and not statistically significant due to high variability (Fig. 3D, Table 3).

Site preparation did not cause any additional effects on groundwater DIC and CH<sub>4</sub> concentrations (Table S5). However, effect sizes remained at medium (+518 to +799 µM) and large levels (+69 to +208 µM), respectively, and DIC in shallow groundwater was still significantly elevated relative to reference conditions.

In streams and lakes, CO2 concentrations were between 269 and 349 µM and 95 and 109 µM, CH4 concentrations between 0.2 and 3.4 µM and 0.3 to 1.1 µM and N2O concentrations between 16 and 22 nM and between 12 and 17 nM, respectively (Table S6). Stream and lake water GHG concentrations did not respond to forest clear-cutting or site preparation, except for stream CO<sub>2</sub> that increased less in impact streams (from 314 uM to 349 uM after clear-cutting and 329 uM after site preparation) relative to the control stream (from 269 µM to 347 µM and 314 µM, respectively).

#### 3.3 Response of GHG emissions from streams and lakes

Lakes and streams evaded CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O to, the atmosphere but the fluxes did not respond to forest clear-cutting. For CO<sub>2</sub> fluxes, this observation is based on daily averages of 2-hourly time series shown in Fig. 4 and 5. CO<sub>2</sub> fluxes varied synchronously across all lakes at daily and seasonal time scales with emission events during storms and a general increase 15 towards autumn (Fig. 4). Daily means of 2-hourly estimates were validated by estimates based on biweekly spot measurements (LME, slope=0.97±0.03, p<0.001, marginal R<sup>2</sup>=0.87, residual standard error (rse)=9.9 mmol m<sup>-2</sup> d<sup>-1</sup>, n=180). Time series of the differences between impact and control lakes do not reveal any systematic change in offset or seasonality between the before and after period. Depending on the k model chosen, seasonal mean CO2 fluxes varied between 41 and 99 mmol m<sup>-2</sup> d <sup>1</sup>. However, consistent for all models, there was no significant BACI effect associated with forest clear-cuts (Fig. 6A, Table 4) or site preparation (Table S7).

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In streams, 2-hourly time series revealed pronounced emission peaks during storm events (Fig. 5). These emission peaks were strikingly synchronous between streams, but peak amplitudes varied from around 200 mmol  $m^{-2}$  d<sup>-1</sup> to up to 2000 mmol m<sup>-2</sup> d<sup>-1</sup> (Fig. 5). Between-stream differences did not change in the after period relative to the before period, indicated by non-significant BACI effects associated to forest clear-cutting (Table 4) and site preparation (Table S7). Daily means of 2-hourly emission estimates were validated by estimates based on biweekly spot measurements with excellent agreement (LME, slope=1.05±0.01, p<0.001, marginal R<sup>2</sup>=0.97, rse=28.3 mmol m<sup>-2</sup> d<sup>-1</sup>, n=180).

Seasonal means of diffusive CH<sub>4</sub> fluxes across the lake-atmosphere interface also varied depending on the k model chosen (between 0.17 and 0.81 mmol m<sup>-2</sup> d<sup>-1</sup>), but regardless of model choice there was no significant BACI effect associated with forest clear-cuts (Fig. 6B, Table 4) or site preparation (Table S7). This result, derived from spot measurements during June-September at the deepest point of the lake, was confirmed also for total CH4 fluxes (including ebullition) by independent weekly measurements using floating chambers deployed across the whole lake during mid-June to late-August (Fig. 7, Table 4). Accordingly, total CH<sub>4</sub> fluxes integrated over the whole lake surface varied from 0.22 to 0.52 mmol  $m_{-2}^2 d^{-1}$  of which 72-82% was due to ebullition and the remainder due to diffusion. Diffusive CH4 fluxes across the stream-atmosphere interface varied from  $1.2 \text{ to } 1.3 \text{ mmol } \text{m}^2 \text{d}^{-1}$  in the control stream and from 0.07 to 0.18 mmol  $\text{m}^2 \text{d}^{-1}$  in the impact streams (Fig. 6D) and remained unaffected by forest clear-cutting or site preparation (Table 4, Table S7).

Across five sites sampled along 300 m long stream reaches, CO2 and CH4 fluxes varied from 43 to 465 mmol m<sup>-2</sup> d<sup>-1</sup> and from -0.02 to 543 mmol m<sup>-2</sup> d<sup>-1</sup>, respectively (Fig. 8A, C). BACI effect sizes were *small* but had a large variability ranging from -53 to 295 mmol m<sup>-2</sup> d<sup>-1</sup> and -4.32 to 0.27 mmol m<sup>-2</sup> d<sup>-1</sup> (Fig. 8B, D, Table S8). These effect sizes were non-significant across the whole length of both impact stream reaches and did not vary across the clear-cut gradient with a five-fold increase in the areal proportion of the stream reach drainage area affected by forest clear-cutting (LME, slope=10.9±5.3 mmol CO2 m <sup>2</sup> d<sup>-1</sup>%clear-cut<sup>-1</sup>, t=2.06, p=0.08, marginal R<sup>2</sup>=0.34 and 0.002±0.003 mmol CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>%clear-cut<sup>-1</sup>, t=0.54, p=0.61, marginal R<sup>2</sup>=0.03, respectively).

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Seasonal means of diffusive N<sub>2</sub>O fluxes across the lake-atmosphere interface varied, depending on the <u>k model</u> chosen, between 0.4 to  $3_{\frac{5}{2}} \mu \text{mol m}_{k}^{22} d^{-1}$ . Consistent for all <u>k</u> models, there was no significant BACI effect associated with forest clear-cuts (Fig. 6C, Table 4). The same was true for diffusive N<sub>2</sub>O fluxes across the stream-atmosphere interface, ranging from <u>1.7</u> to <u>3.5</u>  $\mu$ mol m<sub>k</sub><sup>2</sup> d<sup>-1</sup> (Fig. 6F, Table 4).

#### 5 4 Discussion

This study is to our knowledge the first experimental assessment of forest clear-cut and site preparation effects on <u>GHG fluxes</u> <u>between</u> inland waters <u>and the atmosphere</u> and expands on previous forest clear-cutting experiments that primarily have focused on effects on <u>hydrology</u> or water <u>chemistry</u>. Our whole-catchment BACI experiment showed no significant initial effects of forest clear-cutting and site preparation on <u>GHG fluxes in</u> streams or lakes despite enhanced potential <u>GHG</u> supply

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from hillslope groundwater. This suggests that the generally strong effects of clear-cut forestry on terrestrial C and nutrient cycling are not necessarily translated to major effects in <u>GHG fluxes</u> in recipient downstream aquatic ecosystems. Our results are representative for low-productive boreal forest systems ( $<3 \text{ m}^3 \text{ ha}^{-1} \text{ yr}^{-1}$ ) in relatively flat landscapes, which represent the dominant forest type subject to clear-cut forestry in the boreal biome (Zheng et al. 2004; SFA 2014).

- What caused the contrasting response in <u>GHGs</u> between groundwater and open water? Open water CO<sub>2</sub>, CH<sub>4</sub> and
   N<sub>2</sub>O can result from bacterial <u>degradation of organic matter</u> (Bogard and del Giorgio, 2016; Hotchkiss et al., 2015; Peura et al., 2014) and <u>incomplete denitrification and nitrification</u> (McCrackin and Elser, 2010; Seitzinger, 1988), respectively. <u>These</u> processes are connected to DOC and DIN dynamics. The lack of initial responses in <u>catchment-derived DOC and DIN could</u>, <u>therefore</u>, explain the lack of responses in aquatic <u>GHG fluxes</u>. However, aquatic <u>GHG fluxes</u> are also fueled by direct catchment inputs of the respective dissolved gases (Rasilo et al. 2017; Striegl and Michmerhuizen 1998; Öquist et al. 2009).
- 20 Groundwater CO<sub>2</sub> and CH<sub>4</sub> concentration increased in response to the clear-cut treatment (Fig. 3), potentially as a consequence of enhanced organic matter degradation due to enhanced post-clear-cut soil temperatures (Bond-Lamberty et al. 2004; Schelker et al. 2013<u>a</u>), or reduced <u>CH<sub>4</sub> oxidation (Bradford et al., 2000; Kulmala et al., 2014). Some CO<sub>2</sub> and CH<sub>4</sub> may be formed from degradation of logging residues and litter in the soil (Mäkiranta et al., 2012; Palviainen et al., 2004). However, it is presently unclear whether this source has contributed to enhanced groundwater CO<sub>2</sub> and CH<sub>4</sub> concentrations. Concentration increases</u>
- 25 were most pronounced in shallow groundwater, the hotspot for riparian <u>GHG</u> export to headwater streams in our study region (Leith et al., 2015). Considering that clear-cut areas covered <u>on</u> average ~30% of the stream and lake catchments, but ~80% of the sub-catchments of the groundwater sampling sites, the 56%-increase in <u>groundwater CO<sub>2</sub></u> concentrations <u>could have</u> caused an increase of at most 21% (0.3/0.8\*0.56) in CO<sub>2</sub> concentrations in the impact streams and lakes. Part of the lack of a response could be due to difficulties in detecting such subtle changes <u>given the relatively high natural variability (Table S6</u>).
- 30 However, the 8<sub>±</sub><u>fold\_increase in groundwater CH<sub>4</sub> concentrations could have supported at most 3-fold increases (0.3/0.8\*8) in CH<sub>4</sub><u>concentrations\_in streams and lakes, much larger than observed in our study (Table S6)</u>. This mismatch suggests the following three alternative explanations:</u>

*First*, groundwater-derived <u>GHG</u>s were transport-limited and, hence, only a minor source for <u>GHG</u> fluxes in our lakes and streams. Even though external sources often dominate CO<sub>2</sub> and CH<sub>4</sub> emissions in headwater streams (Hotchkiss et al. 2015; Öquist et al. 2009; Jones and Mulholland 1998), soil-derived gases may only be a minor source for <u>GHGs in headwaters</u> during summer low flow conditions (Dinsmore et al. 2009; Rasilo et al. 2017). Such conditions were present over extended parts during the dry post-treatment period (Table S1, Table 4).

Second, the riparian zone effectively buffered potential clear-cut and site preparation effects on aquatic <u>GHG fluxes</u>.
 In part, this <u>could be</u> because the riparian buffer zones <u>were</u> wide enough to <u>retain</u> their wind sheltering function. Wind speeds
 indeed remained unaffected by clear-cutting on nearby mires (Table 2). <u>This would indicate that riparian buffer zones may</u> have prevented additional forcing on air-water gas exchange velocities, assuming that relative changes in wind speeds on the

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mires were representative for lake conditions (Text S2). In addition, riparian zones may have acted as efficient reactors of <u>GHGs</u> and <u>reduced</u> their concentration during transport from the hillslope to the open water (Leith et al. 2015; Rasilo et al. 2017, Rasilo et al. 2012). This applies especially to methane which can be efficiently oxidized in the large redox gradients in riparian zones, similar to inorganic <u>N</u>(Blackburn et al., 2017).

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- *Third*, in-stream processing effectively buffered potential clear-cut and site preparation effects on aquatic <u>GHG fluxes</u>. In boreal headwater streams, metabolism can strongly regulate  $CO_2$  emissions at summer low flow conditions (Rasilo et al., 2017). Therefore, additional  $CO_2$  leaking from clear-cut soils could have been taken up by algae stimulated in growth by increased light intensities (Kiffney et al. 2003; Clapcott and Barmuta 2010). We indeed observed strong algae blooms in the inlet stream of Struptjärn in response to a tripling in light intensities after forest clear-cutting (Fig. S5). Increased algal <u>CO</u><sub>2</sub>
- 10 and N uptake could explain the observed decrease in stream CO<sub>2</sub> and TN concentrations. In the experimental lakes, however, we did not observe any change in primary production in response to the treatment (Deininger et al., accepted), Additional CH<sub>4</sub> transported from clear-cut soils could have been efficiently oxidized. Enhanced in-stream CH<sub>4</sub> oxidation in the sediments is likely primarily an effect of the commonly found substrate limitation of CH<sub>4</sub> oxidation (e.g. Bastviken, 2009; Duc et al., 2010; Segers, 1998), i.e. CH<sub>4</sub> oxidizer communities have a higher capacity than commonly expressed and will oxidize more CH<sub>4</sub>
- 15 when concentrations increase. Despite lacking mechanistic understanding of the biogeochemical function of the riparian zones and headwater streams in our catchments, we can conclude from groundwater, stream and lake observations that they must have effectively prevented the potential increase in aquatic <u>GHG</u> emissions. In addition, the riparian buffer vegetation left aside could have acted as wind shelters that prevented potential increases in emissions due to enhanced near-surface turbulence. However, the biogeochemical processing of <u>GHG</u>s in the riparian zone-stream continuum should be given special attention in future clear-cut experiments to resolve the mismatch between responses in hillslope groundwater and receiving streams and
- lakes.

Our experiment revealed statistically significant BACI effects on pH and lake thermal conditions. The relative pH decrease of 0.5 units in impact relative to control systems is a common clear-cut effect in northern forests (Martin et al., 2000; Tremblay et al., 2009). However, most relevant for the scope of this paper, this change did not bias CO<sub>2</sub> concentrations, because shifts in the bicarbonate buffer system are minor ( $\leq 2\%$ ) at the observed pH levels of  $\leq 5$  (Stumm and Morgan, 1995). Likewise, pH is not a major control on aquatic CH<sub>4</sub> cycling (Stanley et al., 2016). This applies even to N<sub>2</sub>O here, because we did not

observe any increase in N<sub>2</sub>O emissions in the post-clear-cut period that would be expected from the positive effect of higher pH levels on nitrification (Soued et al. 2016). Whole-lake temperatures and mixing depths decreased significantly in impact lakes relative to control lakes. However, these effects were small in absolute terms (-0.4 °C, -0.2 m, respectively) and associated
 with relative epilimnion volume changes of about 10%. Such subtle changes are unlikely to have had major effects on metabolism and lake-internal vertical exchange processes as a driver of <u>GHG fluxes</u>.

In contrast to many previous boreal forest clear-cut experiments (Schelker et al. 2012; Nieminen 2004; Lamontagne et al. 2000; Winkler et al. 2009; Bertolo and Magnan 2007; Palviainen et al. 2014), hydrology and water chemistry remained largely unaffected by our treatments. The absence of effects is no absolute evidence of an absence of impacts, but the response is low relative to natural variability and restricted to initial responses within three years after clear-cutting. *First*, hydrological responses may have been masked or delayed given that the post-clear-cut period was much dryer than the pre-clear-cut period (Buttle and Metcalfe 2000; Schelker et al. 2013<u>b</u>; Kreutzweiser et al. 2008). During the post-clear-cut period, groundwater levels may have fallen below a threshold level in both control and impact catchments where any minor clear-cut induced increase in water levels would not have translated into comparable increases in stream discharge. This is because stream discharge largely depends on the transmissivity which typically decreases exponentially with depth in Swedish boreal <u>soils</u> (Bishop et al., 2011). *Second*, the proportion of clear-cuts in our catchments (18-44%) was just around the threshold level (~30%) above which significant effects on hydrology and water chemistry can be expected in our study region (Ide et al., 2013; Palviainen et al., 2014; Schelker et al., 2014). These threshold values can however vary and are highly site-specific

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(Kreutzweiser et al. 2008; Palviainen et al. 2015). For example, the relatively high baseline DOC concentrations in our streams and lakes (20 and 29 mg L<sup>-1</sup>, respectively) are potentially less likely to be further enhanced by forest clear-cuts. Relatively wide riparian buffer strips and gentle catchment slopes (Table 1) may have further dampened these effects (Kreutzweiser et al. 2008). Third, the time it takes for the system to respond may have exceeded the experimental period. For example, it can

- 5 take four to 10 years for groundwater nitrate concentrations to respond to clear-cutting in low-productive forest ecosystems due to tight terrestrial N cycling (Futter et al., 2010). Similar delays have been found for responses in boreal stream and lake water chemistry, often triggered by site preparation (Schelker et al. 2012; Palviainen et al. 2014). In the first year after site preparation, only stream DIN concentrations started to increase, but not DOC or GHG concentrations. However, the absence of initial effects does not necessarily imply absence of longer-term effects. On decadal time scales, forestry may change soil
- 10 Cycling (Diochon et al. 2009), leading to enhanced terrestrial organic matter exports and lake CO<sub>2</sub> emissions (Ouellet et al. 2012). Clearly, future work should explore how universal our results are across different hydrological conditions, other types of systems and longer time scales.
  - The particular complexity and multiple controls of catchment-scale GHG fluxes emphasize the need of large scale experiments to assess treatment responses in realistic natural settings (Schindler, 1998). We addressed this challenge by sampling at high spatial and temporal resolution. However, logistical challenges forced us to restrict the analysis to 1 June to 30 September, the period for which we were able to collect consistent data in all years and all catchments. Hence, we do not account for potential clear-cut effects on stream-atmosphere fluxes during snow melt (April-May) or late autumn storms,
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### (October-November), when a large proportion of GHGs in streams can be supplied from catchment soils (Leith et al. 2015; Dinsmore et al. 2013). Similarly, we do not account for potential clear-cut effects on lake-atmosphere fluxes during ice-breakup 20 (Mjd-May) which can be fueled by gases directly derived from catchment inputs or as a result of degadation of catchmentderived organic matter during winter (Denfeld et al. 2015; Vachon et al. 2017). Peak flow conditions during spring or late autumn are hot moments of clear-cut effects on C and N export to aquatic systems (Schelker et al. 2016; Laudon et al. 2009; Ide et al. 2013). Spring can also contribute disproportionally to annual GHG fluxes of boreal headwater streams (Dinsmore et al. 2013; Natchimuthu et al. 2017) and lakes (Huotari et al., 2009; Karlsson et al., 2013). Strong seasonality in CO2 fluxes was also apparent in our systems (Fig. 4, 5). Hence, future investigations of clear-cut effects should be based on whole-year

sampling.

#### 5 Conclusions

In summary, our experiment shows for the first time that air-water GHG fluxes in lakes and streams during the summer season do not respond initially to catchment forest clear-cutting and site preparation, despite increases in the potential supply of CO<sub>2</sub> 30 and CH4 from clear-cut-affected catchment soils. These results suggest that the riparian buffer zone-stream continuum likely acted as a biogeochemical reactor or wind shelter and by that effectively prevented treatment-induced increases in aquatic GHG emissions. Our findings apply to initial effects (3 years) in low-productive boreal forest systems with relatively flat terrain where a modest but realistic treatment (18-44% of lake catchments clear-cut) caused only limited effects on catchment hydrology and biogeochemistry.

#### 35 Data availability

All data shown in Figures in the main document are provided by supplementary files (Files S1-S5).

### Appendices

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Available supplementary information contains extended methods (Texts S1-S6), <u>eight</u> tables (Tables S1-<u>S8</u>) and five figures (Figures S1-S5).

#### Author contribution

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JK and AB designed the study with contributions from JKl, DB and HL. MK, EG and AJ performed the fieldwork. MK analyzed the data with contributions from EG. MK wrote the manuscript. All co-authors revised the manuscript. Author abbreviations: MK: Marcus Klaus, AB: Ann-Kristin Bergström, AJ: Anders Jonsson, DB: David Bastviken, EG: Erik Geibrink, HJ: Hjalmar Laudon, JKl: Jonatan Klaminder, JK: Jan Karlsson.

#### **Competing interests**

The authors have no conflict of interest to declare.

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20 [grant no. 2012-2085] and the European Research Council [grant no. 725546] with grants awarded to David Bastviken.

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Table 1: Morphological and physicochemical characteristics of the lakes and streams of the experimental catchments. Temporally variable parameters are given as mean values of the pre-treatment period (June-September 2012). Abbreviations: a20=spectral absorbance at 420 nm, DOC=Dissolved organic carbon concentration, TP=total phosphorous concentration, TN=total nitrogen concentration.

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Lak ient Si leng				<b>v</b>	•						catchment cover	er er	
82 284 79 25 233	ea Mean m depth [m]	Lake water residence time [d]	Stream discharge [ <u></u> [s s <sup>-1</sup> ]	a <sub>420</sub>	펍	DOC DOC	TP [µg L <sup>-1</sup> ]	TN [µg L <sup>-1</sup> ]	Catchment / stream slope [%]	Buffer strip width [m]	Forest Mire [%] [%]	Mire [%]	Proportion of forest clear-cut [%]
284 79 25 233		<u> 30</u>	•	13	4.5	20	13	403	6.2		88	12	
79 25 233	4.0	<u>64</u>	•	12	4.0	22	22	398	8.3		84	16	
25 233	3.8	387	•	10	4.9	19	24	367	7.4	50	96	4	18
233	3.8	115		~	5.6	15	19	345	13.1	20	98	2	44
	0.3		4 <u>0.9</u>	12	3.9	28	26	503	1.9		06	10	
Struptjärn* 46 1.4	0.2		5.5	10	4.2	36	24	762	1.9	9	94	9	17
Lillsjölidtjärnen* 19 0.6	0.1		3.0	2	4.9	21	15	829	4.0	35	100	0	51

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Variable System Wind speed Open mire* Discharge Stream*	tem																		
Variable Syst Wind speed Open I Discharge Stree	tem	1	Contro	trol	Impact	act		Control	0	Impact	act			Effe	Effect size (Slope)	(Slope	(6		
Wind speed Open I Discharge Stree		Unit	mean	Se	mean	se	c	mean	se	mean	se	c	mean	se	đ	÷	٩	bse	Cohen's D
		m s¹	1.8	0.1	1.0	0.1	244	2.0	0.0	0.9	0.0	732	0.0	0.1	973	-0.8	0.42	0.03	-0.12
	am*	L s <sup>-1</sup>	40.9	3.4	4.2	0.4	244	27.0	1.8	3.3	0.2	732	0.2	0.2	973	1.0	0.32	0.01	0.13
Water level Groundwater	<u>dwater</u>	cm	34.5	1.8	34.6	1.8	17	42.1	1.9	40.4	1.8	55	-25.1	51.9	69	-0.5	0.63		-0.22
ыдл. intensity Lake	ę,	lux	31199	1096	22408	898	244	34371	598	23230	554	732	-2379	2308	973	-1.0	0.30	0.01	-0.11
Stream*	am*		5907	248	3402	169	244	6408	104	6966	357	732	0.2	0.2	973	1.1	0.27	0.01	0.84
Temperature Stream	am	ů	8.6	0.1	8.1	0.1	244	9.0	0.1	8.4	0.1	732	-0.1	0.4	973	-0.2	0.84	0.00	-0.02
Lake Epi	Epi		14.4	0.2	14.8	0.2	245	15.9	0.1	16.2	0.1	732	-0.1	0.2	974	-0.9	0.39	0.02	-0.08
Lake Hypo	Hypo		7.0	0.0	6.0	0.1	227	7.5	0.1	6.5	0.0	716	-0.1	0.2	940	-0.6	0.58	0.01	-0.05
Whole Lake <sup>‡</sup>	Lake <sup>‡</sup>		11.1	0.1	10.7	0.1	245	12.3	0.1	11.5	0.1	732	-0.4	0.1	974	-2.8	0.01	0.00	-0.20
Mixing depth Lake* Schmidt	<e*< td=""><td>E</td><td>1.8</td><td>0.1</td><td>1.7</td><td>0.1</td><td>227</td><td>1.8</td><td>0.0</td><td>1.5</td><td>0.0</td><td>716</td><td>-0.2</td><td>0.1</td><td>940</td><td>-2.4</td><td>0.02</td><td>0.00</td><td>-0.15</td></e*<>	E	1.8	0.1	1.7	0.1	227	1.8	0.0	1.5	0.0	716	-0.2	0.1	940	-2.4	0.02	0.00	-0.15
Stability Lake		J m²²	13.3	0.6	12.8	0.5	245	16.5	0.4	15.4	0.3	731	-0.7	0.5	973	-1.4	0.17	0.00	-0.09
Lake	Epi	mg L <sup>-1</sup>	8.2	0.1	8.1	0.2	20	8.2	0.1	8.1	0.1	58	-0.1	0.2	75	-0.4	0.66		-0.06
Lake Hypo <sup>‡</sup>	Hypo‡		2.2	0.5	0.8	0.4	17	2.4	0.3	0.7	0.2	53	0.2	1.0	67	0.2	0.85		0.05
Whole	Lake		6.4	0.3	5.1	0.3	20	6.2	0.2	5.1	0.2	58	0.1	0.3	75	0.6	0.56		0.07

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-	Lake Epi Stream Groundwate Lake Epi Stream <sup>†</sup>	Groundwater Lake Epi Streae Epi Coroundwater Lake Epi†* Lake Epi Lake Epi Stream	ased on log-l constancy of non-additivity stimates basi	
Table 2 continued.	DOC TN	DIN 10 DIN 84 PIN	"tME estimates based on log-transformed data #Assumption on constancy of paired differences in before-period not met *Assumption on non-additivity of paired differences in before-period not met "mean and LME estimates based on H+ concentrations, se based on pH value "mean and LME estimates based on H+ concentrations, se based on pH value	
Ta			יד ד <sup>ב</sup> ד ד ד ד	

Table 3: Effect size of forest clear-cutting on DIC and CH<sub>4</sub> concentrations [<u>nM</u>] in groundwater in the impact catchments as shown in Figure 3, Given are linear mixed-effects model slope estimates (mean), their standard errors (se), degrees of freedom (df), t- and p-values and Cohen'D. <u>Statistically significant p-values (<0.05) are highlighted bold.</u>

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				Effect size	(Slope	e)		
Figure	Substance	Soil depth [cm]	mean	se	df	t	р	Cohen's D
3A)	DIC	37.5 - 42.5	533.3	175.7	68	3.0	0.00	0.63
3C)	DIC	5 - 105	458.0	605.8	69	0.8	0.45	0.30
3B)	CH <sub>4</sub>	37.5 - 42.5	93.4	44.4	66	2.1	0.04	1.62
3D)	CH <sub>4</sub>	5 - 105	139.0	182.2	69	0.8	0.45	0.71

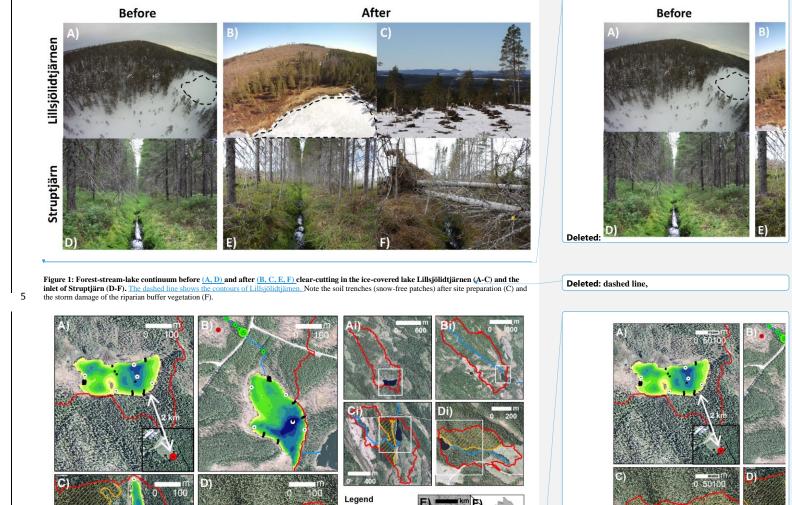
Table 4: Effect size of forest clear-cutting on fluxes of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O across the interface between lakes or streams and the atmosphere as shown in Figure 6 and 7. Shown are linear mixed-effects model slope estimates, their standard errors (se), degrees of freedom (df), t- and p-values and Cohen'D, summarized as arithmetic means over ten bootstrap runs that take uncertainty from gap filling and gas flux models into account (see Fig. S3). This uncertainty is expressed as bootstrap standard errors (be) of p-values. For lake-atmosphere fluxes, estimates based on three different & models are shown. Note that parameter estimates are based on log+n transformed data, where n is the smallest number that, when added, leads to positive normal values. Abbreviations: "Logger"=Daily mean of 2-hourly Vaisala measurement, "Chamber"=Floating chamber, "Spot"=Spot measurement.

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							Effect	size (	Slope)			_
Figure	Gas	Flux type	System	Method	k model	mean	se	df	t	р	bse	Cohen's D
6A)	CO <sub>2</sub>	Diffusion	Lake	Logger	Cole	0.13	0.11	965	1.25	0.23	0.03	0.02
-	$\rm CO_2$	Diffusion	Lake	Logger	Vachon	0.15	0.10	965	1.45	0.17	0.03	0.02
-	$CO_2$	Diffusion	Lake	Logger	Heiskanen	0.09	0.07	965	1.33	0.31	0.06	-0.03
6D)	$CO_2$	Diffusion	Stream	Logger	This study	0.16	0.23	982	0.77	0.47	0.07	0.08
6B)	$CH_4^\dagger$	Diffusion	Lake	Spot	Cole	0.00	0.21	72	0.00	0.93	0.02	0.14
-	$CH_4^\dagger$	Diffusion	Lake	Spot	Vachon	-0.01	0.22	72	-0.07	0.91	0.02	0.16
-	CH <sub>4</sub>	Diffusion	Lake	Spot	Heiskanen	-0.01	0.22	72	-0.06	0.88	0.03	0.31
7	CH₄†	Diffusion+Ebullition	Lake	Chamber	-	-0.02	0.24	33	-0.20	0.49	0.09	-0.13
6E)	CH₄†	Diffusion	Stream	Spot	This study	0.04	0.08	74	0.51	0.62	0.05	0.07
6C)	N <sub>2</sub> O	Diffusion	Lake	Spot	Cole	-0.08	0.05	48	-1.45	0.17	0.02	-0.03
-	N <sub>2</sub> O	Diffusion	Lake	Spot	Vachon	-0.09	0.06	48	-1.45	0.16	0.01	-0.04
-	$N_2O^\dagger$	Diffusion	Lake	Spot	Heiskanen	-0.11	0.07	48	-1.56	0.13	0.02	-0.03
6F)	$N_2O^\dagger$	Diffusion	Stream	Spot	This study	-0.01	0.10	47	-0.05	0.87	0.03	-0.07

<sup>†</sup>Assumption on non-additivity of paired differences in before-period not met

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Depth [m]

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CH4 Chamber Lake sampling Stream sampling

Groundwater

Stream Catchment Clear-cut O Master site

Weather station

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0 1 2 3 4 5 6 7 8 Figure 2: Maps of the experimental lakes and streams (A-D), their catchments (Ai-Di) and their location in Sweden (E-F). Detailed maps show the lake bathymetry, the main channel of the inlet stream and the location of gas concentration sampling sites in lakes, streams, and hillslope <u>groundwater</u>, floating CH<sub>4</sub> chambers and weather stations. White frames or dots in smaller-scale maps illustrate the extent or location of corresponding larger-scale maps, respectively. Panel labelling is consistent across all map scales and as follows: A) Stortjärn, B), Övre Björntjärn, C) <u>Struptjärn</u> and D) <u>Lillsjölidtjärnen</u>

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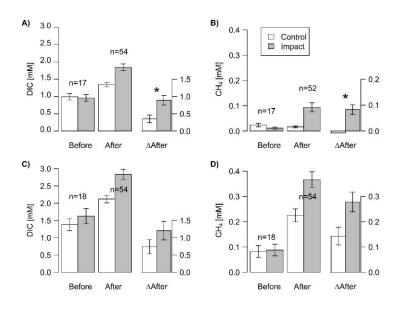


 Figure 3: Concentrations of DIC and dissolved CH<sub>4</sub> in groundwater at depth specific locations (37.5-42.5 cm; A-B) and depth integrated locations (5-105 cm; C-D) before and after clear-cutting at impact sites, and the respective differences between before and after (AAfter, shown in the same units). Each bar represents mean values (±propagated standard errors) of repeated observations over time. Significant (p<0.05) effect sizes are marked by "\*". Abbreviations: n=number of observations.</td>

**Deleted:** at 37.5-42.5 cm depth (A-B) and 5-105 cm depth (C-D)

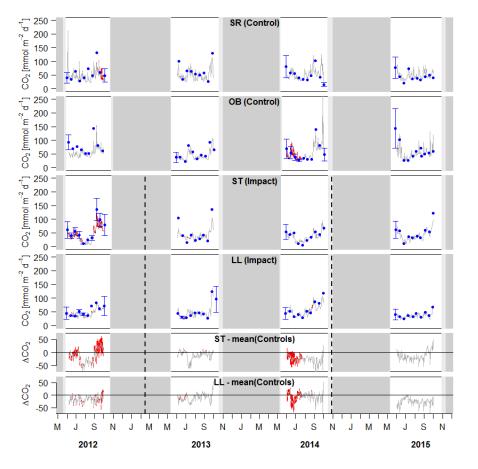
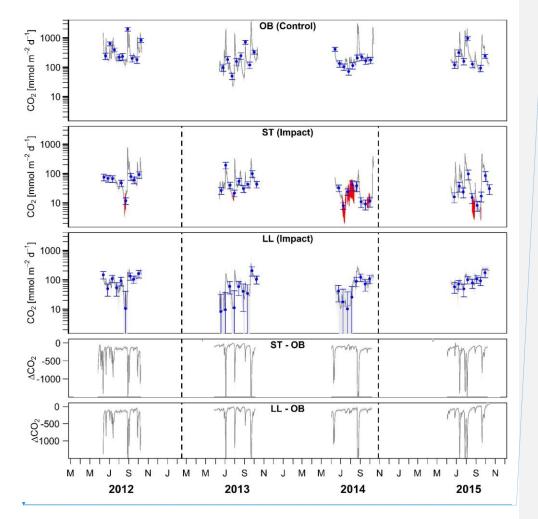


Figure 4: Time series of lake-atmosphere CO<sub>2</sub> fluxes based on daily means of 2-hourly concentration measurements (grey lines) and biweekly spot measurements (blue dots) and the <u>k</u> model by Cole and Caraco (1998). Given are absolute fluxes and differences ( $\Delta CO_2$ ) between impact and control lakes. Shadings and error bars show propagated standard errors (see Fig. S3 <u>and Text S6</u>). Gap-filled data is colored in red. Bars show the <u>lake ice cover duration with uncertainties expressed by grey scale (dark\_minimum duration, light</u> <u>=maximum duration</u>, <u>Dashed lines mark the timing of forest clear-cutting (2013) and site preparation (2014). Units are consistent across</u> all panels. Abbreviations: SR=Stortjärn, OB=Övre Björntjärn, ST=Struptjärn, LL=Lillsjölidtjärnen.

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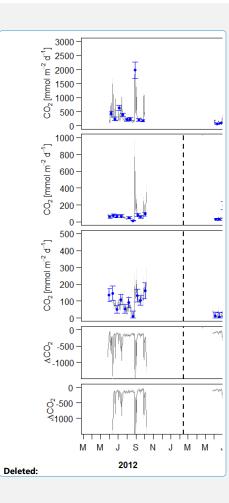


Figure 5: Time series of stream-atmosphere CO<sub>2</sub> fluxes based on daily means of 2-hourly concentration measurements (dark grey lines) and biweekly spot measurements  $\pm$  standard errors (blue dots and error bars). Given are absolute fluxes and differences ( $\Delta$ CO<sub>2</sub>) between impact and control streams, shadings and error bars show propagated standard errors (see Fig. S3 and Text S6). Gap-filled data is colored in red. Dashed lines mark the timing of forest clear-cutting (2013) and site preparation (2014). Units are consistent across all panels. Abbreviations: SR=Stortjärn, OB=Övre Björntjärn, ST=Struptjärn, LL=Lillsjölidtjärnen.

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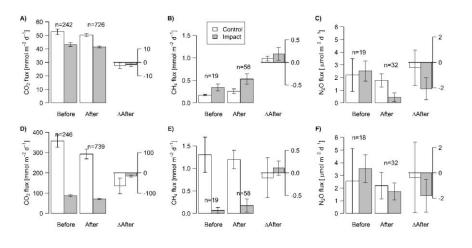
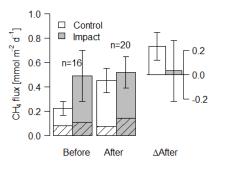


Figure 6: Fluxes of CO2, CH4 and N2O across the interface between lakes (A-C) or streams (D-F) and the atmosphere in control and impact catchments before and after forest clear-cutting, and the respective differences between before and after (AAfter, shown in the same units). Each bar represents mean values (±propagated standard errors) of repeated observations over time, summarized as arithmetic means over ten bootstrap runs that take uncertainty from gap filling and gas flux models into account (see Fig. S3 and Text S6). Data is based on daily means of 2-hourly measurements (CO<sub>2</sub>) or biweekly (CH<sub>4</sub> and N<sub>2</sub>O) concentration measurements. Lakeatmosphere fluxes are here calculated using the k model by Cole and Caraco (1998). Abbreviations: n=number of observations.



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10 Figure 7: Fluxes of CH4 by diffusion (shaded) and ebullition (non-shaded) across the lake-atmosphere interface in control and impact catchments before and after forest clear-cutting, and the respective differences between before and after (AAfter, shown in the same units). Fluxes were measured by the use of flux chambers (e.g. independent approach compared to fluxes calculated from concentrations in Fig. 6). Each bar represents mean values (±propagated standard errors) of whole-lake fluxes measured weekly from Mid-June to Mid-August 2012 and 2014, summarized as arithmetic means over ten bootstrate runs thate between-chamber variability into account (see Fig. S3 and Text S6). Whole-lake fluxes are the area-weighted mean of depth-zone specific fluxes. Abbreviations: n=number

15 of observations.

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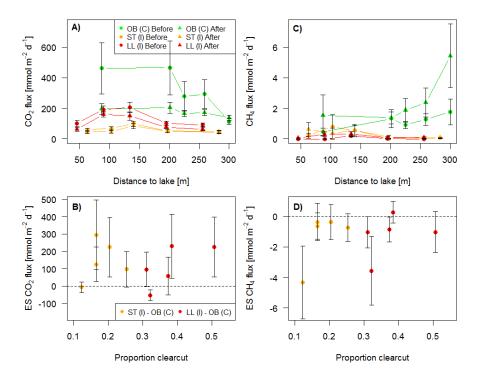


Figure 8: Fluxes of A) CO<sub>2</sub> and C) CH<sub>4</sub> across the stream-atmosphere interface along stream transects in the control catchment (C) and two impact catchments (I) before and after forest clear-cutting (OB=Övre Björntjärn, ST=Struptjärn, LL=Lillsjölidtjärnen,). Effect sizes (ES) defined as the before-after change in the difference between control- and impact streams are shown in panel B) and D). Each point represents seasonal mean values (±standard errors) of biweekly observations, summarized as arithmetic means over ten bootstrap runs that take uncertainty from gap filling and gas flux models into account (see Fig. S3 and Text S6).

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## Supplementary information for

# Greenhouse gas emissions from boreal inland waters unchanged after forest harvesting

Marcus Klaus<sup>1\*</sup>, Erik Geibrink<sup>1</sup>, Anders Jonsson<sup>1</sup>, Ann-Kristin Bergström<sup>1</sup>, David Bastviken<sup>2</sup>, Hjalmar Laudon<sup>3</sup>, Jonatan Klaminder<sup>1</sup>, Jan Karlsson<sup>1</sup>

<sup>1</sup>Department of Ecology and Environmental Science, Umeå University, SE-90187 Umeå, Sweden <sup>2</sup> The Department of Thematic Studies - Environmental Change, Linköping University, SE-58183 Linköping, Sweden <sup>3</sup>Department of Forest Ecology and Management, Swedish University of Agricultural Science, SE-90183 Umeå, Sweden *Correspondence to: Marcus Klaus (marcus.klaus@posteo.net)* 

### 10 Text S1 Sampling and analysis of dissolved gases

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Partial pressure of CO<sub>2</sub> in stream and lake surface waters was measured by a hand held non-dispersive infra-red CO<sub>2</sub> sensor (GM70 Carbon dioxide meter, Vaisala Inc. Helsinki, Finland) or an infrared gas analyzer (IRGA EMG-4, PP-Systems Inc., Amesbury, MA, U.S.) coupled to a gas equilibrator (MINIMODULE 1.7 x 5.5 G542, Membrana Liqui-Cel Inc., Wuppertal, Germany) through which sample water was transferred by a peristaltic pump (Master-Flex 7518-12, Cole-Parmer Instrument

- 15 Company, East Bunker Ct Vernon-Hills, IL, USA). Both CO<sub>2</sub> sensors were calibrated monthly against reference gas mixtures (AGA, Linde AG). Molar CO<sub>2</sub> concentrations were derived from Henry's law constants using water temperatureparameterizations in Wanninkhof (1992). For DIC and CH<sub>4</sub> sampling, 4 ml of water was injected into gas-tight 22 ml glass vials (Perkin Elmer Inc., Waltham, MA, USA) containing 50 µl 1.2M HCl, sealed with 20 mm natural pink rubber stoppers flushed with N<sub>2</sub> gas prior to sampling. For N<sub>2</sub>O sampling we used the headspace equilibration technique where 50 ml of
- headspace gas (air taken in upwind direction 2 m aboveground) was equilibrated with 540 ml of surface water by vigorous shaking for 1 min and then transferred to a glass vial (as described above, but here without HCl) allowing any overpressure to be released during gas injection. CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O concentrations in the vial headspace were analyzed using a gas chromatograph (GC, Clarus 500, Perkin Elmer Inc., Waltham, MA, USA) equipped with a methanizer and flame ionization detector (for CH<sub>4</sub> analysis) and an electron capture detector (for N<sub>2</sub>O analysis). Headspace concentrations were converted to molar concentrations by means of the ideal gas law using Bunsen solubility coefficients given in Wanninkhof (1992).

Groundwater was sampled biweekly for nutrient, DIC and CH<sub>4</sub> concentrations from PVC groundwater wells located at two forested hillslope sites (inclination 1-5%), 10-70 m from the impact lakes. The two wells had an inner diameter of 18 mm and were 100 and 110 cm deep with openings across a depth range of 37.5-42.5 cm and 5-105 cm to separate responses in surficial groundwater (depth specific sampling) from general responses in the whole profile (depth integrated sampling). A
 proportion of 65%-85% of the drainage areas of one of the hillslope sites were affected by forest clear-cutting while the forest

- in the drainage area of the other sites were left intact (Fig. 2). At each site, groundwater levels were measured and groundwater collected from two wells using a peristaltic pump (Master-Flex 7518-12, Cole-Parmer Instrument Company, East Bunker Ct Vernon-Hills, IL, USA) by carefully transferring it to acid washed plastic bottles. Subsamples for chemical analysis were taken from these bottles immediately after sampling. Parallel sampling of groundwater for DIC and CO<sub>2</sub> concentrations (n=11 per groundwater sampling site) showed that 92±11% (mean±standard deviation) of the DIC pool was CO<sub>2</sub> due to the low pH of
- <5 (data not shown). Hence, we use DIC as a proxy for groundwater CO<sub>2</sub> concentrations.

### Text S2 Logger systems

Surface water CO<sub>2</sub> observations were measured 2-hourly at 10 cm water depth at the deepest point of the lakes and the master stream sites using non-dispersive infra-red CO2 sensors (CARBOCAP GMP 222, Vaisala Inc., Helsinki, Finland) enclosed in a semi permeable PTFE membrane, coupled to Vaisala GMT 220 transmitters (Johnson et al., 2010) and connected to a data logger (CR200X, Campbell Scientific Inc., Logan, UT, USA). Times series were corrected for sensitivities to temperature and

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Lake water temperature was measured at 5 min intervals at every 0.5 m (0-3 m depth) or 1-2 m (below 3 m) at the deepest point of the lake using temperature loggers (Hobo TidbiT V2, Onset Inc., Bourne, MA, USA). Stream water height and temperature was measured hourly with a water height data logger (WT-HR 100, Trutrack Inc.), placed at well-defined reaches 10 at the master stream sites (Fig. 2). Water height logger readings were drift-corrected based on biweekly manual water height measurements. Discharge was measured occasionally throughout the whole study period using slug injections based on salt in solution following Moore (2005). Measured discharge was related to water height using rating curves, i.e. piecewise power type equations with one or two segments and segment-specific normalized root mean square errors of 0.10-0.23 at Övre Björntjärn (n=30), 0.31 at Lillsjölidtjärnen (n=33) and 0.19-0.33 at Struptjärn (n=42). The rating curves were used to calculate

pressure following Johnson et al. (2010) and for linear drifts based on probe-calibrations before and after the field season.

15 hourly discharge from logged water height.

> Wind speed, relative humidity, air temperature, precipitation and air pressure were measured every 5-10 min at 2.5 m above open mires 100-300 m from Övre Björntjärn, Lillsjölidtjärnen and Struptjärn, respectively (Fig. 2). The mires had about the same size as the lakes and were surrounded by similar vegetation with similar clear-cut buffer zones left aside. Hence, weather measurements on the mires can be regarded to be representative for lake conditions, at least in terms of the relative

20 differences between lakes and years. At Övre Björntjärn and Lillsjölidtjärnen we used mobile weather stations (Hobo U30-NRC, Onset Inc., Bourne, MA, USA). At Struptjärn, wind speed and precipitation was measured using a propeller wane (RM Young wind monitor, R.M. Young Company, MI, USA) and a tipping bucket rain gauge (ARG100, EML Inc., North Shields, UK), respectively, connected to a data logger (CR10, Campbell Scientific Inc., Logan, UT, USA). Air pressure and air temperature was measured every 10 min using a water level logger (Hobo U20 001-01-Ti, Onset Inc., Bourne, MA, USA), 25 Relative humidity was assumed to be the same as at Övre Björntjärn. Weather data with 10 min intervals for Stortjärn was derived from the reference climate monitoring program at Svartberget experimental forest, Vindeln, Sweden, 2 km from Stortjärn (Laudon et al., 2013). Here, wind speed was measured at 16 m above dense spruce forest using a propeller wane. Relative humidity and air temperature was measured at 1.7 m above ground in an open area. Air pressure was scaled from observations at Struptjärn using the barometric formula (Iribarne and Godson, 1981). Light intensity was measured every

10 min using lux meters (Hobo UA-002-64, Onset Inc., Bourne, MA, USA) placed 1 m above ground within 30 m from each

lake in an open environment and within 1 m from four of the five stream sampling sites.

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### Text S3 Gap filling of logger data

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Continuously logged data showed occasional gaps (Table S2). For lake CO<sub>2</sub> concentrations, we occasionally observed diel cycles which were greatly exaggerated by biofouling. We identified and gap-filled erroneous patterns based on independently measured covariates (water temperature, lux, wind speed) following a multivariate outlier detection and multiple imputation approach using 10 bootstrap runs, described in detail by Klaus et al. (2017). Stream CO<sub>2</sub> concentrations peaked above the detection limit of the probes (10000 ppm) during extreme summer low-flow in Struptjärn. To avoid extrapolation, we filled these gaps by linear interpolation of spot measurements, assuming an error of ±50%. Gap-filled data totaled 7% of CO2 measurements in lakes and 3% in streams (Table S2). Missing wind speed data (12%) were gap-filled using a multiple imputation model with variable squared time effects (Honaker et al. 2011) trained for each year with wind speed observations from all other weather stations and carried out using 10 bootstrap runs. Gaps in air temperature, air pressure, relative humidity

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# Deleted: Text S2 Sampling and analysis of dissolved

Partial pressure of CO<sub>2</sub> in stream and lake surface waters was measured by a hand held non-dispersive infra-red CO<sub>2</sub> sensor (GM70 Carbon dioxide meter, Vaisala Inc. Helsinki, Finland) or an infrared gas analyzer (IRGA EMG-4, PP-Systems Inc., Amesbury, MA, U.S.) coupled to a gas equilibrator (MINIMODULE 1.7 x 5.5 G542, Membrana Liqui-Cel Inc., Wuppertal, Germany) through which sample water was transferred by a peristaltic pump (Master-Flex 7518-12, Cole-Parmer Instrument Company, East Bunker Ct Vernon-Hills, IL, USA). Both CO2 sensors were calibrated monthly against reference gas mixtures (AGA, Linde AG). Molar CO2 concentrations were derived from Henry's law constants using water temperature-parameterizations in Wanninkhof (1992). For DIC and CH4 sampling, 4 ml of water was injected into gas-tight 22 ml glass vials (Perkin Elmer Inc., Waltham, MA, USA) containing 50 µl 1.2M HCl, sealed with 20 mm natural pink rubber stoppers flushed with  $N_{\rm 2}$  gas prior to sampling. For  $N_2O$  sampling we used the headspace equilibration technique where 50 ml of headspace gas (air taken in upwind direction 2 m aboveground) was equilibrated with 540 ml of surface water by vigorous shaking for 1 min and then transferred to a glass vial (as described above, but here without HCl) allowing any overpressure to be released during gas injection. CO2, CH4 and N2O concentrations in the vial headspace were analyzed using a gas chromatograph (GC, Clarus 500, Perkin Elmer Inc., Waltham, MA, USA) equipped with a methanizer and flame ionization detector (for CH4 analysis) and an electron capture detector (for N2O analysis). Headspace concentrations were converted to molar concentrations by means of the ideal gas law using Bunsen solubility coefficients given in Wanninkhof (1992). Groundwater was sampled biweekly for nutrient, dissolved inorganic carbon (DIC) and CH4 concentrations from PVC groundwater wells located at two forested hillslope sites (inclination 1-5%), 10-70 m from the impact lakes. The two wells had an inner diameter of 18 mm and were 100 and 110 cm deep with openings across a depth range of 37.5-42.5 cm and 5-105 cm to separate responses in surficial groundwater from general responses in the whole profile. A proportion of 65%-85% of the drainage areas of one of the hillslope sites were affected by forest clear-cutting while the forest in the drainage area of the other sites were left intact (Fig. 2). At each site, groundwater levels were measured and groundwater collected from two wells using a peristaltic pump (Master-Flex 7518-12, Cole-Parmer Instrument Company, East Bunker Ct Vernon-Hills, IL, USA) by carefully transferring it to acid washed plastic bottles. Subsamples for chemical analysis were taken from these bottles immediately after sampling. Parallel sampling of groundwater for DIC and CO2 concentrations (n=11 per soil site) showed that 92±11% (mean±standard deviation) of the DIC pool was CO2 due to the low pH of <5 (data not shown). Hence, we use DIC as a proxy for groundwater CO2 concentrations.¶

and lux data (4-11% of the total record) were filled using linear regression models trained with data from the nearest logger from the respective year ( $R^2$ =0.63-0.99, Table S2). Here, time series were subsampled to 8 hour intervals to avoid problems of serial autocorrelation (Breusch-Godfrey test, p>0.05). Gaps in time series of lake thermal characteristics (4% of the total record) were filled using linear regression models trained with data collected in the other replicate lake in the respective year

5 (R<sup>2</sup>=0.60-0.96). Here, time series were subsampled to 10 day intervals to avoid problems of serial autocorrelation (Breusch-Godfrey test, p>0.05).

### Text S4 Gas transfer velocity measurements in streams

Air-water gas transfer rates in streams were measured using a static polymethylmethacrylate gas flux chamber (60 x 20 x 23 cm<sup>3</sup>) with a hexagonal base and rounded edges to minimize chamber induced turbulence (Fig. S1). For each gas flux measurement, the chamber was mounted to a tripod and placed onto the water surface centered in the stream with the main axis oriented in flow direction and the side walls extending 2 cm into the water. CO<sub>2</sub> concentrations in the chamber were measured using a CO<sub>2</sub> logger (CO2 Engine® ELG, SenseAir AB, Delsbo, Sweden, Accuracy = ± 30 ppm ± 3 % of measured value, response time <25 s) that was off-set calibrated against N<sub>2</sub> gas before each field visit. The CO<sub>2</sub> logger was mounted on top of the chamber and connected to a pump (SP 270 EC-LC 12VDC, Schwarzer Precision GmbH + Co. KG, Essen, Germany)
that circulated air at a rate of 600 cm<sup>3</sup> min<sup>-1</sup> through a Nafion membrane tube (ME-110-03-12, Perma Pure Inc., Lakewood, NJ, U.S.A.) enclosed within a box of silica gel, CO<sub>2</sub> measurements consisted of a 30 s cycle during which air was pumped for 18 s and measurements were taken for 12 s with mean CO<sub>2</sub> concentrations logged. After 4-8 minutes, the flux chamber was

lifted to reset inside-air CO<sub>2</sub> concentrations to ambient levels. This procedure was repeated 3 times for each sampling site yielding three linear regression slopes that describe the rise in CO<sub>2</sub> concentration over time. The mean(±standard deviation)
 coefficient of determination (R<sup>2</sup>) of 846 individual measurements of the linear regression was 93±11%. Average coefficient of determination among triplicate slope measurements was 11±9%. The gas transfer velocity (k) was calculated using Fick's law of diffusion

$$k = \frac{F}{a \left( c_{wat} - c_{eq} \right)}$$

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where F is the CO<sub>2</sub> flux as estimated by the linear regression slopes, C<sub>wat</sub> is the CO<sub>2</sub> concentration in water, C<sub>eq</sub> is the CO<sub>2</sub>
concentration of water if it was in equilibrium with ambient air calculated from measured air concentration and water temperature using Henry's constant and *a* is the chemical enhancement factor set to 1, as enhancement is negligible if pH < 8 (Wanninkhof and Knox, 1996). Molar CO<sub>2</sub> concentrations were derived from Bunsen solubility coefficients using water temperature-parameterizations in Weiss (1974). In-situ water temperature specific *k* values were normalized to 20°C to yield *koop* following Jähne et al. (1987) using Schmidt number parameterizations for water temperature according to Wanninkhof
(1992). Reported errors in *koop* were the standard errors of regression slopes of CO<sub>2</sub> concentration increases over time

propagated for triplicate measurements at three sites per sub-reach (Fig. S3).

Propane injection experiments followed principles described in Wallin et al. (2011), with the following modifications. At a distance of 10-20 m upstream the uppermost sub-reach, liquefied petroleum gas (PC10, AGA gas AB, Luleå, Sweden) was injected into the stream at constant rates (2-8 l min<sup>-1</sup>) and pressures (0.5-1 bar) using 2-3 aquarium gas diffusion stones (length=10 cm, diameter=5 cm). Flow rates were set by a propane regulator (Unicontrol 500, AGA gas AB, Luleå, Sweden)

and monitored using a gas flow meter (ZYIA LZM, Yuyao Kingtai Instrument Co., Ltd., Zhejiang, China). Parallel to propane diffusion, we continuously injected a saturated sodium chloride (NaCl) solution using a peristaltic pump (FMI Lab Pump QBG, Fluid metering Inc<sub>2</sub>, Syosset, NY, USA) and measured electrical conductivity every 10 s using conductivity loggers (HOBO U24, Onset Computer Corporation, Bourne, MA, U.S.A.) at five sites downstream, marking the upper and lower end of each

of the sub-reaches (Fig. S1). Once conductivity reached a stable plateau (after 0.3-8 h), indicating propane saturation across

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the whole stream reach, we took triplicate samples (50 ml) of bubble-free stream water at each site starting at the uppermost site using plastic syringes closed gas tight with three-way stopcocks. During extreme summer low flow, we varied propane injection sites in Lillsjölidtjärnen and Struptjärn to cover specific sub-reaches only, because travel times were too long for meaningful whole-reach injections. Within 4 hours after propane sampling, we replaced 20 ml of water by 20 ml of N2 gas,

- shook syringes vigorously for 1 min and transferred the headspace gas to glass vials (22 ml; PerkinElmer Inc., Waltham, MA, 5 U.S.A.) capped with butyl rubber stoppers (27232, Supelco Analytical, Bellefonte, PA, U.S.A.) by simultaneously withdrawing 20 ml of inside air. Within 24 hours after transfer to vials, the vial gas was analyzed on propane concentration by a gas chromatographer (Clarus 500, Perkin Elmer Inc., Waltham, MA, U.S.A.) equipped with a flame ionization detector. Propane concentrations in triplicate samples varied by 5±4% (mean±sd).
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To account for dilution of propane by lateral water inputs, we estimated stream discharge for each sub-reach as Q = $\frac{s}{cal(c_s-c_b)}$ , where s is the NaCl injection rate,  $c_s$  is the electrical conductivity when it has reached a stable plateau,  $c_b$  is the background electrical conductivity before salt injection and cal is a coefficient derived from in-situ calibrations linking electrical conductivity to NaCl mass. Stream discharge was constant along the reach in Övre Björntjärn but increased by a factor of 2.5 and 3 in Struptjärn and Lillsjölidtjärnen, respectively, from the upper- to the lowermost sub-reaches. The gas

- 15 transfer coefficient (d<sup>-1</sup>), the proportion of gas evaded over a specific reach per unit time, was calculated for propane using the log-ratio of discharge-corrected propane concentrations at the up- and downstream end of each sub-reach and converted to normalized gas transfer coefficients koog following equations given in Wallin et al. (2011). Gas transfer coefficients were multiplied by the average sub-reach depth to obtain gas transfer velocities (m  $d^{-1}$ ). Average sub-reach depth was obtained from six stream depth measurements taken at each of the three sites per sub-reach, where one measurement was taken at each of the
- 20 six flux chamber edges (Fig. S1). Reported errors in  $k_{600}$  were propagated from standard errors of triplicate propane measurements and standard errors of the 18 stream depth measurements per sub-reach

### Text S5 Testing assumptions of the BACI analysis

To assess potential biases in the paired-BACI analysis of atmospheric gas fluxes due to pretreatment trends and autocorrelation, we tested the following assumptions (Stewart-Oaten et al. 1986): (1) Constancy of inter-lake differences in the before period, tested by linear mixed-effects models with "inter-lake difference" as the dependent variable, "sampling occasion" (Day of year) as a fixed effect and "pair" as a random effect on both slopes and intercepts; (2) Absence of significant positive firstorder autocorrelation, tested by a Durbin-Watson test with the residuals of the model in (1) as the dependent variable and "sampling occasion" (Day of year) as the independent variable using the "dwtest" function of the R package "Imtest" (Zeileis and Hothorn, 2002); (3) Additivity of inter-lake differences in the before period, tested by linear mixed-effects models with "inter-lake differences" as the dependent variable, "inter-lake sums" as a fixed effect and "DOC-level" as a random effect on both slopes and intercepts.

### **Text S6 Error propagation**

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We accounted for uncertainties in BACI statistics for gas fluxes and gap-filled logger data by combining standard methods of error propagation and bootstrapping (Fig. S3). BACI analyses were run 10 times, with each observation sampled from a normal distribution defined by its mean estimate and propagated standard error. Standard errors were estimated as follows:

(1) For diffusive gas fluxes in streams, standard errors were propagated from errors due to gap filling of gas concentrations (standard error of 10 imputations, Text S3), the root-mean-square-error (rmse) of discharge rating curves and variability in k600 within sub-reaches and across triplicate flux chamber and propane injection experiments (Fig. S3A). Specifically, in corrections of flux chamber-derived k600 based on linear relationships

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temperature

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In-situ water temperature and -specific gas transfer coefficients from propane injection experiments were normalized to  $20^{\circ}$ C to yield k<sub>600</sub> following Jähne et al. (1987) using Schmidt number parameterizations for wate temperature according to Wanninkhof (1992). For propane injections,

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Moved up [1]: For flux chamber measurements, errors in k600 were the standard errors of regression slopes of CO2 concentration increases over time propagated for triplicate measurements at three sites per sub-reach (Fig. S3).

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with propane-derived  $k_{600}$ , observations were weighted by the root mean square of the standard error of triplicate flux chamber and propane injection experiments. In predictions of  $k_{600}$  based on discharge, observations were weighted by the root mean square of the standard error of  $k_{600}$  and the rmse of discharge rating curves, where each error term was normalized to the respective mean estimate.

(2) For diffusive gas fluxes in lakes, standard errors were propagated from errors due to gap filling of gas concentrations (standard error of 10 imputations, Text S3) and from errors in modelled <u>keee</u> (Fig. S3B). <u>Errors</u> in modelled <u>keee</u> were derived from a separate bootstrap algorithm run 10 times, where all input variables were sampled from a normal distribution defined by the mean estimate and standard error derived from gap filling (as described in Text S3).

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- (3) For total CH<sub>4</sub> fluxes in lakes, errors were estimated from the area-weighted standard errors of depth-zone specific error estimates which in turn were the standard errors of fluxes of all chambers located therein (Fig. S3C).
- (4) For weather variables and lake thermal characteristics standard errors were propagated from prediction errors of linear regressions used for gap filling (Fig. S3D).

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**Deleted:** wind speed (standard error of 10 imputations), and other weather variables and lake thermal characteristics used as inputs to the  $k_{600}$  model (prediction error of linear regressions)

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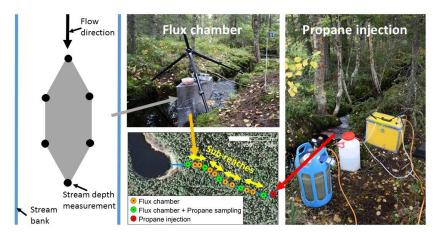
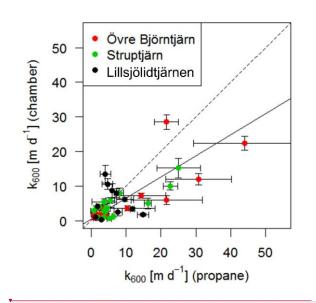


Figure S1: Field setup of gas transfer velocity measurements in streams by static flux chambers and propane injection experiments. The stream reach is divided into four sub-reaches above and below which propane concentrations were measured. Within each sub-reach, flux chamber measurements were done at three sites. At each site, stream depth was measured at the six edges of the flux chamber centered in the stream and aligned along flow direction.



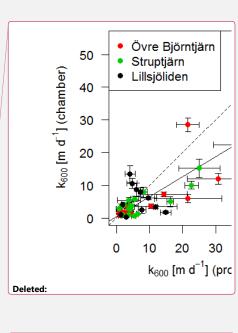
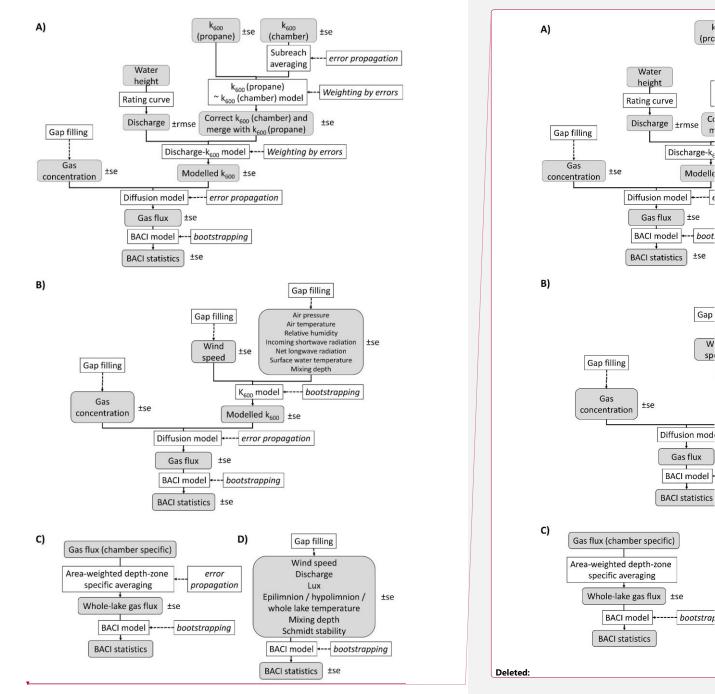


Figure S2: Sub-reach specific  $k_{600}$  derived from propane injection experiments (x) compared to the mean  $k_{600}$  of flux chamber measurements performed at three sites per sub-reach (y), given for three streams. The solid line shows the regression line of a linear mixed-effects model with "sub-reach" nested in "stream" as random effects on both slopes and intercepts where observations were weighted by the root mean square of measurement errors (se also Fig. S3A) expressed by error bars (y=0.61x±0.13+0.53±0.43, p-value of slope <0.001, R<sup>2</sup>=0.58, n=46). The dotted line shows a hypothetical 1:1 relationship.

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Figure S3: Overview of the errors associated with estimates of diffusive gas fluxes across the air-water interface in A) streams and **B**) lakes, C) total lake CH<sub>4</sub> flux and D) other continuously measured physical parameters. Errors were propagated following standard rules and bootstrapping (<u>Text S6</u>). For gap-filling procedures, see Table S2. Abbreviations: "BACI"=Before/After-Control/Impact, " $k_{600}$ " = gas-transfer velocity for CO<sub>2</sub> at 20°C. "se"=standard error, "rmse" = root mean square error.

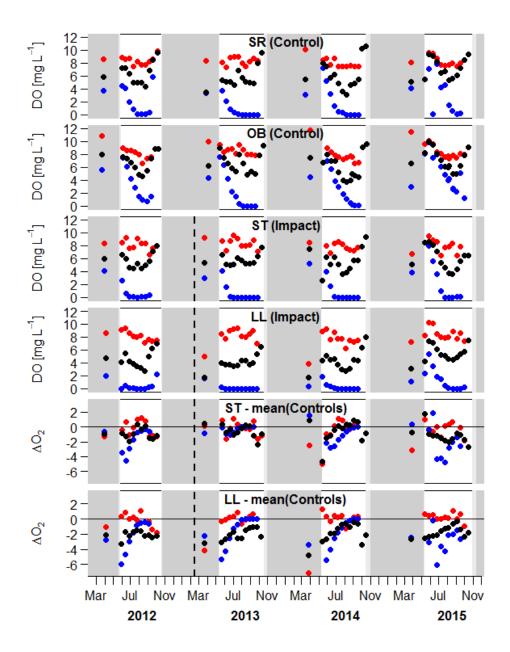


Figure S4: Time series of depth-integrated  $O_2$  concentrations based on biweekly profile measurements (red=epilimnion, blue=hypolimnion, black=whole lake). Given are absolute values and differences ( $\Delta O_2$ ) between impact and control lakes. Bars show the minimum (dark grey) and maximum (light grey) lake ice extent. The vertical dashed line marks the timing of forest clear-cutting. Units are consistent across all panels. Abbreviations: SR=Stortjärn, OB=Övre Björntjärn, ST=Struptjärn, LL=Lillsjölidtjärnen.



Figure S5: Algae bloom observed in July 2013 in the inlet stream of Struptjärn after forest clear-cutting.

Tables

Table S1: Air temperatures and precipitation sums during the study period (June-September 2012-2015), given as means±sd across the study catchments.

Variable	Unit	2012	2013	2014	2015
Air temperature	°C	11.1±0.3	12.7±0.3	12.8±0.3	11.6±0.3
Precipitation	mm	342±12	321±26	245±42	274±29

		_	gap lengt	n [d]			
Parameter	System	Proportion of gaps	mean	sd	Gap filling method	Training data	R <sup>2</sup>
CO <sub>2</sub>	Lake	0.07	8	7	Multivariate imputation#	Co-variates*	-
CO <sub>2</sub>	Stream	0.03	6	8	Linear interpolation	Spot measurements	-
Wind speed	Open mire	0.12	39	10	Multivariate imputation#	All other weather stations	-
Air temperature	Open mire	0.11	41	47	Linear regression	Nearest weather station	0.96
Air pressure <sup>†</sup>	Open mire	0.10	32	46	Linear regression	Nearest weather station	0.99
Relative humidity <sup>‡</sup>	Open mire	0.09	85	50	Linear regression	Nearest weather station	0.83
Lux	Lake	0.04	25	27	Linear regression	Nearest lux logger	0.83
Lux	Stream	0.11	23	8	Linear regression	Nearest lux logger	0.63
Surface Temperature	Lake	0.04	27	20	Linear regression	Replicate lake	0.93
Epilimnion temperature	Lake	0.04	27	20	Linear regression	Replicate lake	0.96
Hypolimnion temperature	Lake	0.04	27	20	Linear regression	Replicate lake	0.89
Whole lake temperature	Lake	0.04	27	20	Linear regression	Replicate lake	0.93
Schmidt Stability	Lake	0.04	27	20	Linear regression	Replicate lake	0.94
Mixing depth	Lake	0.04	27	20	Linear regression	Replicate lake	0.60

Table S2: Details on the extent and filling of gaps in continuously logged data. Given are the length of gaps as a proportion of the total data set, the mean $\pm$ sd gap length, the gap filling method, the training data used for gap filling and the mean  $\mathbb{R}^2$  value of the linear regressions used for gap filling. Training data included all data collected in the year the gap occurred.

<sup>†</sup>not measured in Stortjärn; modelled using bathymetric formula

<sup>‡</sup>not measured in Struptjärn; mean and error estimates assumed to be the same as in Övre Björntjärn

#Honaker et al. 2011

\*see Klaus et al. (2017)

			-		Interce	ept			Slop	be		-			
Catchment	Sub-reach	Distance to lake [m]	n	mean	se	t	р	mean	se	t	р	R <sup>2</sup>	rse		
Övre Björntjärn	1	87	19	1.470	1.297	1.13	0.27	0.339	0.062	5.46	0.00	0.64	4.24	 	Formatted: Font: Bold
Övre Björntjärn*	2	201	18	0.000	NA	NA	NA	0.262	0.027	9.65	0.00	0.85	1.77		Formatted: Font: Bold
Övre Björntjärn	3	225	17	0.956	0.514	1.86	0.08	0.136	0.028	4.84	0.00	0.61	1.68		
Övre Björntjärn†	4	259	17	0.956	0.514	1.86	0.08	0.136	0.028	4.84	0.00	0.61	1.68		Formatted: Font: Bold
Övre Björntjärn‡	5	300	2	1.635	0.548	NA	NA	NA	NA	NA	NA	NA	NA		Formatted: Font: Bold
Struptjärn*	1	63	13	0.000	NA	NA	NA	0.343	0.056	6.10	0.00	0.76	2.38	 	Formatted: Font: Bold
Struptjärn	2	103	15	1.591	0.288	5.53	0.00	0.187	0.034	5.44	0.00	0.69	1.13		Formatted: Font: Bold
Struptjärn	3	140	13	0.432	0.698	0.62	0.55	0.796	0.061	12.97	0.00	0.94	1.63		
Struptjärn	4	197	13	0.720	0.754	0.95	0.36	0.332	0.088	3.78	0.00	0.56	2.17		Formatted: Font: Bold
Struptjärn**	5	283	13	0.720	0.754	0.95	0.36	0.332	0.088	3.78	0.00	0.56	2.17		Formatted: Font: Bold
Lillsjölidtjärnen	1	46	10	1.536	1.284	1.20	0.27	0.591	0.237	2.50	0.04	0.44	2.28		<b>Formatted:</b> Font: Bold
Lillsjölidtjärnen	2	90	10	1.475	0.691	2.13	0.07	1.120	0.185	6.07	0.00	0.82	1.85		Formatted: Font: Bold
Lillsjölidtjärnen*	3	134	9	0.000	NA	NA	NA	1.966	0.171	11.50	0.00	0.94	2.84		Formatted: Font: Bold
Lillsjölidtjärnen	4	195	10	0.578	0.300	1.93	0.09	1.405	0.135	10.37	0.00	0.93	1.80		Formatted: Font: Bold
Lillsjölidtjärnen <sup>†</sup>	5	256	10	0.578	0.300	1.93	0.09	1.405	0.135	10.37	0.00	0.93	1.80		Formatted: Font: Bold

Table S3: Parameter estimates of sub-reach-specific linear regression models that predict *k*<sub>600</sub> (m d<sup>-1</sup>) based on discharge (L s<sup>-1</sup>). Each observation was weighted by the root mean square of their standard errors. Abbreviations: se=standard error, t=t-value, p=p-value, NA=Not available, n=number of observations. Statistically significant p-values (<0.05) are highlighted bold.

intercept constrained to zero

<sup>†</sup>model of n-th subreach assumed to be the same as model of (n-1)th subreach, motivated by their similar morphology

\*k600 assumed to be constant (mean of 6 flux chamber measurements), because discharge variations were negligible and greatly buffer ed at this mire site

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Table S4: Physicochemical characteristics of lake-, stream-, and groundwater at control and impact sites before and after site preparation (year 2012 and 2015, respectively). Given are also the estimated effect size (linear mixed effects model slope), its standard errors (se), degrees of freedom (df), t- and p-values and Cohen'D, summarized as arithmetic means over ten bootstrap runs that account for uncertainty from gap filling. This uncertainty is expressed as bootstrap standard errors (bsc) of p-values. Statistically significant p-values (<0.05) are highlighted bold. Water levels [cm] are relative to the soil surface. Groundwater data refers to depth-integrated locations (5-105 cm). Abbreviations: Epi=Epilimnion, Hypo=hypolimnion,  $z_{mix}$ = mixing depth [m], DOC=Dissolved organic carbon concentration [mg L<sup>-1</sup>], TN=total nitrogen concentration [µg L<sup>-1</sup>], DIN=dissolved inorganic nitrogen concentration [µg L<sup>-1</sup>], at 20 minimum concentration [µg L<sup>-1</sup>].

[m<sup>-1</sup>].

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**Deleted:** refer to the upper 5-105 cm of the soil profile

					Before					After									4	 Formatted Table
			Con	trol	Imp	act		Con	trol	Imp	act			Ef	fect siz	e (Slop	e)		_	
	_																		Cohen's	5
Variable	System	Unit	mean	se	mean	se	n	mean	se	mean	se	n	mean	se	df	t	р	bse	<ul><li>■ D</li></ul>	Formatted Table
Wind speed	Open mire*	m s <sup>-1</sup>	1.8	0.1	1.0	0.1	244	2.0	0.1	0.9	0.0	244	-0.1	0.1	485	-1.3	0.24	0.04	-0.16	
Discharge	Stream*	L s <sup>-1</sup>	40.9	3.4	4.2	0.4	244	32.9	2.6	4.7	0.4	244	0.2	0.3	485	0.5	0.64	0.02	0.08	
Water level	Groundwater	cm	34.5	1.8	34.6	1.8	17	42.4	3.0	39.1	2.9	18	-57.4	51.6	32	-1.1	0.27	-	-0.49	Deleted: Soil
Lux	Lake	lux	31199	1096	22426	897	244	33677	1013	22393	888	244	-2518	2166	485	-1.2	0.25	0.00	-0.11	
	Stream*		5952	251	3398	168	244	6199	192	10572	614	244	0.2	0.2	485	1.2	0.25	0.01	0.96	
Temperature	Stream	°C	8.6	0.1	8.1	0.1	244	8.2	0.1	7.9	0.1	244	0.2	0.2	485	1.3	0.19	0.00	0.07	
	Lake Epi		14.4	0.2	14.8	0.2	245	14.8	0.2	15.2	0.2	244	0.0	0.2	486	-0.2	0.81	0.00	-0.03	
	Lake Hypo		7.0	0.0	6.0	0.1	227	9.3	0.0	7.1	0.1	238	-1.3	0.4	462	-3.5	0.00	0.00	-0.68	Formatted: Font: Bold
	Whole Lake <sup>‡</sup>		11.1	0.1	10.7	0.1	245	12.2	0.1	11.4	0.1	244	-0.4	0.0	486	-9.8	0.00	0.00	-0.24	Formatted: Font: Bold
Zmix	Lake*	m	1.8	0.1	1.7	0.1	227	2.0	0.1	1.6	0.1	238	-0.4	0.1	462	-4.1	0.00	0.00	-0.26	Formatted: Font: Bold
Schmidt Stability	Lake		13.3	0.6	12.8	0.5	245	11.2	0.5	12.6	0.4	244	1.8	0.6	486	3.0	0.00	0.00	0.25	Formatted: Font: Bold
Oxygen	Lake Epi	mg L <sup>-1</sup>	8.2	0.1	8.1	0.2	20	8.3	0.2	8.2	0.2	20	-0.1	0.4	37	-0.2	0.82	-	-0.05	Formatted: Font. Bold
	Lake Hypo <sup>‡</sup>	0	2.2	0.5	0.8	0.4	17	4.3	0.5	1.7	0.5	18	-0.7	1.1	32	-0.7	0.52	-	-0.18	
	Whole Lake		6.4	0.3	5.1	0.3	20	7.2	0.4	5.7	0.3	20	-0.2	0.5	37	-0.3	0.77	-	-0.08	
DOC	Lake Epi	mg L <sup>-1</sup>	22	1.0	18	0.9	20	22	0.9	19	1.2	20	1.2	2.9	37	0.4	0.67	-	0.18	
	Stream	0	29	0.9	28	1.4	59	25	1.1	24	1.6	75	1.3	1.8	131	0.7	0.47	-	0.07	
	Groundwater,		67	3.0	77	2.4	14	55	5.7	66	6.7	10	1.9	8.5	21	0.2	0.82	-	0.06	Deleted: Soil
TN	Lake Epi	µg L⁻¹	409	15.7	367	14.3	20	427	13.7	401	19.6	20	16.2	48.3	37	0.3	0.74	-	0.15	
	Stream <sup>†</sup>		498	13.5	595	35.3	58	450	17.0	486	23.6	75	-49.6	52.1	130	-1.0	0.34	-	-0.10	
	Groundwater,		1572	180.4	1798	83.8	14	1288	167.2	1575	150.9	10	58.3	256.5	21	0.2	0.82	-	0.05	Deleted: Soil
DIN	Lake Epi	μg L-1	20	1.6	19	2.0	20	15	1.3	10	1.4	20	-4.7	3.3	37	-1.4	0.17	-	-0.26	

	Stream		21	2.0	23	2.2	57	16	0.8	30	1.9	73	10.3	4.4	127	2.4	0.02	-	0.25	Formatted: Font: Bold
	Groundwater,†		467	98.9	523	42.4	13	411	95.3	501	67.5	7	4.7	166.3	17	0.0	0.98	-	0.01	Deleted: Soil
pН	Lake Epi <sup>†*</sup>		4.2	0.1	5.1	0.1	20	4.9	0.0	5.4	0.0	20	0.00	0.00	37	1.0	0.31	-	0.25	Deleted: Soli
	Stream*		3.9	0.1	4.4	0.1	20	4.6	0.0	4.6	0.0	20	0.00	0.00	37	3.1	0.00	-	0.36	
	olioani		0.0	0.1		0.1	20	1.0	0.0	1.0	0.0	20	0.00	0.00	0,	0.1	2.00		0.00	Formatted: Font: Bold
<b>a</b> <sub>420</sub>	Lake Epi	m <sup>-1</sup>	12.4	0.4	9.3	0.6	20	12.4	0.3	10.0	0.7	20	0.01	0.02	37	0.4	0.70	-	0.14	
	Stream		15.1	0.4	13.6	0.7	53	12.5	0.3	12.1	0.7	76	0.02	0.01	126	1.7	0.08	-	0.16	

\*LME estimates based on log-transformed data

‡Assumption on constancy of paired differences in before-period not met

<sup>†</sup>Assumption on non-additivity of paired differences in before-period not met

#mean and LME estimates based on H+ concentrations, se based on pH value

 Table \$5: Seasonal mean(±se) concentrations [μM] of DIC and CH4 in ground water in control and impact catchments before and after site preparation in impact catchments (years 2012 vs. 2015). Given are also linear mixed effects model slope estimates of the effects of site preparation (mean), their standard errors (se), degrees of freedom (df), t- and p-values and Cohen'D. Statistically significant p-values (<0.05) are highlighted bold.</th>

	-		В	efore					After								
	<u> </u>	Con	trol	Imp	bact	_	Con	trol	Imp	Impact			Effect	size (Sl	ope)		_
Substance	Soil depth [cm]	mean	se	mean	se	n	mean	se	mean	se	n	mean	se	df	t	р	Cohen's D
DIC	37.5 - 42.5	99 <mark>2,</mark>	90	957	99	17	1446	138	1949	153	17	518	249	31	2.1	0.046	0.61
DIC	5 - 105	1380	172	1624	221	18	2062	196	3072	285	19	799	971	34	0.8	0.42	0.52
CH <sub>4</sub>	37.5 - 42.5	23.7	7.0	11.4	4.2	17	6.8	1.8	69.1	21.5	16	68.5	69.1	30	1.0	0.33	1.19
CH <sub>4</sub>	5 - 105	82.0	23.4	88.0	22.8	18	207.6	41.1	406.8	62.1	19	207.9	279.2	34	0.7	0.46	1.07

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Table S6: Seasonal mean( $\pm$ se) concentrations of dissolved CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O in stream and lake water in control and impact catchments before and after clear-cutting (years 2012 vs. 2013-2015) and site preparation (years 2012 vs. 2015). Given is also the estimated effect size (linear mixed-effects model slope), its standard error (se), degrees of freedom (df), t- and p-values and Cohen'D. Statistically significant p-values (<0.05) are highlighted bold. Note that parameter estimates are based on log+n transformed data, where n is the smallest number that leads to positive normal values. Abbreviations: Logger=Daily mean of 2-hourly measurement, Spot=Biweekly spot measurement.

		_	-	_	_		Before		_			After										Formatted: English (United States)
					Con	trol	Impa	act		Conti	rol	Impa	act		E	Effect s	size (S	lope)				
Gas Ur	oit M	lethod	System	Treatment	mean	50	mean	se	n	mean	se	mean	se	n	mean	se	df	t	n	- Cohen's D		Formatted Table
									0.40					700				4 74	2 0 0 0			Formatted Table
<u>СО₂</u> ± <u>µ</u> М		ogger	Lake	Clear-cut	<u>103.0</u>		<u>109.2</u>		<u>242</u>	<u>95.0</u>	<u>1.5</u>	<u>104.3</u>	<u>1.8</u>	<u>726</u>	<u>0.27</u>	<u>0.15</u>	<u>965</u>	<u>1.74</u>	<u>0.08</u>	<u>0.05</u>		
				Site preparation	<u>103.0</u>	<u>2.2</u>	<u>109.2</u>	<u>3.2</u>	<u>242</u>	<u>96.8</u>	<u>1.8</u>	<u>97.0</u>	<u>2.2</u>	<u>242</u>	<u>0.02</u>	<u>0.33</u>	<u>481</u>	<u>0.07</u>	<u>0.95</u>	<u>-0.10</u>		
<u>CO2</u> µI	<u>M Lo</u>	ogger	<u>Stream</u>	Clear-cut	<u>269.2</u>	<u>6.9</u>	<u>314.4</u>	<u>6.6</u>	<u>246</u>	<u>346.6</u>	<u>4.9</u>	<u>349.4</u>	<u>3.1</u>	<u>739</u>	<u>-0.13</u>	0.02	<u>982</u>	<u>-5.85</u>	<u>0.00</u>	<u>-0.28</u>		
				Site preparation	<u>269.2</u>	<u>6.9</u>	<u>314.4</u>	<u>6.6</u>	<u>246</u>	<u>313.9</u>	<u>6.7</u>	<u>328.8</u>	<u>4.8</u>	<u>247</u>	<u>-0.10</u>	<u>0.03</u>	<u>490</u>	<u>-3.70</u>	<u>0.00</u>	<u>-0.20</u>		
<u>СН₄± µМ</u>	<u>M s</u>	<u>Spot</u>	Lake	Clear-cut	<u>0.3</u>	<u>0.0</u>	<u>0.8</u>	<u>0.2</u>	<u>19</u>	<u>0.4</u>	<u>0.1</u>	<u>1.1</u>	<u>0.2</u>	<u>56</u>	<u>0.09</u>	<u>0.30</u>	<u>72</u>	<u>0.30</u>	<u>0.76</u>	<u>0.17</u>		
				Site preparation	<u>0.3</u>	<u>0.0</u>	<u>0.8</u>	<u>0.2</u>	<u>19</u>	<u>0.3</u>	<u>0.1</u>	<u>0.8</u>	<u>0.2</u>	<u>20</u>	0.02	<u>0.41</u>	<u>36</u>	<u>0.05</u>	<u>0.96</u>	<u>-0.01</u>		
<u>СН₄<sup>±,#</sup> µМ</u>	<u>M s</u>	<u>Spot</u>	<u>Stream</u>	Clear-cut	<u>0.8</u>	<u>0.1</u>	<u>0.2</u>	<u>0.0</u>	<u>18</u>	<u>3.4</u>	<u>1.0</u>	<u>3.1</u>	<u>2.6</u>	<u>58</u>	<u>-0.17</u>	<u>0.26</u>	<u>73</u>	-0.65	<u>0.52</u>	<u>0.01</u>		
				Site preparation	<u>0.8</u>	<u>0.1</u>	<u>0.2</u>	<u>0.0</u>	<u>18</u>	<u>1.2</u>	<u>0.2</u>	<u>0.1</u>	<u>0.1</u>	<u>20</u>	<u>-0.18</u>	<u>0.12</u>	<u>35</u>	<u>-1.48</u>	<u>0.15</u>	<u>-0.01</u>		
<u>N2O nM</u>	<u>M s</u>	<u>Spot</u>	Lake	Clear-cut	<u>15.2</u>	<u>1.1</u>	<u>16.7</u>	<u>1.4</u>	<u>19</u>	<u>13.3</u>	<u>0.8</u>	<u>12.2</u>	<u>0.7</u>	<u>32</u>	<u>-0.15</u>	<u>0.12</u>	<u>48</u>	<u>-1.26</u>	<u>0.21</u>	<u>-0.04</u>		
<u>N2O nN</u>	<u>M 5</u>	<u>Spot</u>	<u>Stream</u>	Clear-cut	<u>20.0</u>	<u>1.9</u>	22.2	<u>2.6</u>	<u>18</u>	<u>15.7</u>	<u>1.2</u>	<u>18.2</u>	<u>1.4</u>	<u>32</u>	0.18	<u>0.20</u>	<u>47</u>	<u>0.87</u>	<u>0.39</u>	<u>0.01</u>		
+Assumpt	tion or	n non-a	dditivity o	f paired difference	<u>s in bef</u>	ore-per	riod not m	<u>iet</u>														Formatted: English (United States)
#Assumpt	tion or	n consta	ancy of pa	aired differences in	before	-period	not met															Formatted: English (United States)
#one outlie	er ren	noved																				Tormatted. English (Onited States)

Table \$7: Seasonal mean (±se) fluxes [mmol m<sup>-2</sup> d<sup>-1</sup>] of dissolved CO<sub>2</sub> and CH<sub>4</sub> across the interface between lakes or streams and the atmosphere in control and impact catchments before and after site preparation (years 2012 vs. 2015). Given is the estimated effect size of site-preparation (linear mixed-effects model slope), its standard error, (se), degrees of freedom (df), t- and p-values and Cohen'D, summarized as arithmetic means over ten bootstrap runs that account for uncertainty from gap filling and gas flux models (see Fig. S3). This uncertainty is expressed as bootstrap standard errors (bse) of p-values. For lake-atmosphere fluxes, estimates based on three different k models are shown. Note that parameter estimates are based on log+n transformed data, where n is the smallest number that leads to positive normal values. Abbreviations: Logger=Daily mean of 2-hourly measurement, Spot=Biweekly spot measurement. Cole=Cole and Caraco

(1998), Vachon=Vachon and Prairie (2013), Heiskanen=Heiskanen et al. (2014).

						Before					After									
				Con	trol	Imp	act	_	Cor	trol	Imp	act	_		Effec	ct size				
Gas	System	Method	k model	mean	se	mean	se	n	mean	se	mean	se	n	mean	se	df	t	р	bse	Cohen's D
CO <sub>2</sub>	Lake	Logger	Cole	52.8	2.0	43.0	1.4	242	50.4	1.4	37.2	0.9	242	-0.11	0.30	481	-0.4	0.73	0.02	-0.08
CO <sub>2</sub>	Lake	Logger	Vachon	75.6	2.3	58.8	1.9	242	71.7	1.8	51.1	1.3	242	-0.09	0.31	481	-0.3	0.78	0.02	-0.07
$\text{CO}_2^\dagger$	Lake	Logger	Heiskanen	98.1	4.0	76.2	3.3	242	98.5	3.5	69.2	2.5	242	-0.06	0.23	481	-0.3	0.79	0.04	-0.08
CO <sub>2</sub>	Stream	Logger	This study	352.8	30.5	86.8	4.6	246	313.8	32.6	93.5	5.7	247	0.10	0.23	490	0.5	0.66	0.07	0.07
$CH_4^{\dagger}$	Lake	Spot	Cole	0.18	0.02	0.33	0.07	19	0.24	0.13	0.33	0.14	20	-0.09	0.30	36	-0.3	0.76	0.02	-0.10
$CH_4^{\dagger}$	Lake	Spot	Vachon	0.24	0.02	0.48	0.11	19	0.31	0.17	0.45	0.21	20	-0.13	0.32	36	-0.4	0.68	0.02	-0.09
CH <sub>4</sub>	Lake	Spot	Heiskanen	0.28	0.04	0.39	0.10	19	0.45	0.26	0.40	0.27	20	-0.15	0.29	36	-0.5	0.62	0.05	-0.17
$CH_4^{\dagger}$	Stream	Spot	This study	1.26	0.38	0.07	0.07	19	0.76	0.19	-0.05	0.03	20	0.08	0.14	36	0.6	0.56	0.08	0.12

†Assumption on non-additivity of paired differences in before-period not met

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Table 58: Linear mixed effects model estimates of the effects of forest clear-cutting on CO<sub>2</sub> and CH<sub>4</sub> fluxes across the stream-atmosphere interface as shown in Figure 8. Given are the estimated effect size (model slope), its standard error, (se), degrees of freedom (df), t- and p-values and Cohen'D, as arithmetic means over ten bootstrap runs that account for uncertainty from gap filling and gas flux models (see Fig. S3). Uncertainty is expressed as bootstrap standard errors (bse) of p-values. Parameter estimates are based on log+n transformed data, where n is the smallest number that leads to positive normal values.

					Effe	ct size (	Slope)			
Figure	Gas	Catchment	Distance to lake [m]	mean	se	df	t	р	bse	Cohen's D
8B)	CO <sub>2</sub>	Lillsjölidtjärnen	46	0.21	0.56	28.00	0.39	0.70	0.07	0.20
8B)	$\text{CO}_2^\dagger$	Lillsjölidtjärnen	90	0.34	0.44	28.00	0.79	0.45	0.05	0.19
8B)	CO <sub>2</sub>	Lillsjölidtjärnen	134	-0.01	0.44	28.00	-0.01	0.78	0.05	0.07
8B)	CO <sub>2</sub>	Lillsjölidtjärnen	195	0.11	0.37	28.00	0.29	0.77	0.05	0.15
8B)	CO <sub>2</sub>	Lillsjölidtjärnen	256	-0.37	0.38	28.00	-0.98	0.38	0.07	-0.36
8B)	$CO_2^{\dagger}$	Struptjärn	63	0.47	0.50	27.00	0.96	0.36	0.04	0.23
8B)	CO <sub>2</sub>	Struptjärn	103	0.33	0.51	27.00	0.63	0.56	0.07	0.21
8B)	CO <sub>2</sub>	Struptjärn	140	0.13	0.49	27.00	0.31	0.72	0.07	0.14
8B)	CO <sub>2</sub>	Struptjärn	197	0.23	0.41	27.00	0.58	0.58	0.06	0.21
8B)	CO <sub>2</sub>	Struptjärn	283	-0.10	0.38	27.00	-0.21	0.71	0.04	-0.05
8D)	$CH_4^\dagger$	Lillsjölidtjärnen	46	-0.04	0.32	26.00	-0.14	0.88	0.02	-0.66
8D)	$CH_4^\dagger$	Lillsjölidtjärnen	90	0.07	0.23	25.00	0.23	0.81	0.04	0.07
8D)	CH <sub>4</sub>	Lillsjölidtjärnen	134	-0.15	0.32	25.00	-0.47	0.64	0.03	-0.57
8D)	$CH_4^\dagger$	Lillsjölidtjärnen	195	-0.13	0.34	25.00	-0.40	0.70	0.02	-0.50
8D)	$CH_4^\dagger$	Lillsjölidtjärnen	256	-0.31	0.65	26.00	-0.48	0.64	0.02	-0.76
8D)	CH <sub>4</sub>	Struptjärn	63	0.07	0.19	24.00	0.33	0.69	0.06	-0.35
8D)	CH <sub>4</sub>	Struptjärn	103	-0.05	0.56	24.00	-0.09	0.69	0.05	-0.11
8D)	CH <sub>4</sub>	Struptjärn	140	-0.07	0.27	24.00	-0.26	0.71	0.06	-0.72
8D)	$CH_4^{\dagger}$	Struptjärn	197	-0.03	0.26	24.00	-0.02	0.81	0.05	-0.35
8D)	CH₄†	Struptjärn	283	-0.39	0.68	24.00	-0.59	0.56	0.03	-0.83

†Assumption on non-additivity of paired differences in before-period not met

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