Dear Editor,

We have now revised our manuscript bg-2015-263 by Guerin et al submitted for publication to Biogeosciences.

We have carefully considered all comments; we provided detailed answers in order to address them or provided clearer explanations when misunderstanding was suspected. The objective of study and the broader context of our project including the description of previously published work is now included in the introduction. Titles and subtitles were shortened, and organization and vocabulary issues were tackled.

Overall, we significantly improved the manuscript by following most of the suggestions and comments provided by four reviewers. We hope you and the reviewers will agree that our paper is now a significant contribution to the literature and deserve publication in Biogeosciences.

Kind Regards
F Guérin, on behalf of all co-authors

RC: Reviewer comment
AC: Author comment

RC: "The paper by Guerin et al. is about temporal and spatial variation in methane concentrations and fluxes in a subtropical hydroelectric reservoir. As such the topic is globally relevant and suits well the scope of BG. As a whole the paper is quite descriptive and serves more as a case study displaying results which are hard to scale up or taken as representative for subtropical reservoirs."
AC: In this paper, we show that the temporal variations of emissions are highly significant and that all estimate based on seasonal sampling (2-4 field campaigns) should be considered with caution, which is a new result. We show that in the rainy season, emissions occur mostly in inflow region due to the high water discharge and that the highest emissions during the overturn occur in remote area, which are isolated from the main water circulation (where water residence time is the longest).
We also identified a new hotspot of emissions at the turbine intake.
This is not just a case study. In addition of being one of the very first comprehensive study on a subtropical hydroelectric reservoir, this paper also gives some strong recommendations for the establishment of sampling strategy (place, time and frequency) in order to build accurate assessment of emissions from hydroelectric reservoirs.

RC: The first reviewer had pointed out that there are some problems with the topics raised in the introduction. At least some of the problems are still there. Now the very first paragraph of introduction gives e.g. the impression that it is ebullition, not diffusive fluxes which are important. In general, the results of this manuscript are not discussed properly in relation to ebullition; ebullition is briefly mentioned in Discussion and in Table 3.
It could be fruitful to formulate a hypothesis in the end of the introduction, i.e. if you think that it water intake is the key issue, please mention it here.
AC: The first paragraph lists all CH4 pathways between hydroelectric reservoirs and the atmosphere. This includes diffusive fluxes, downstream emissions, emissions from the drawdown and ebullition. Diffusive emission is probably not the major pathway in reservoirs but it is clearly not negligible and this pathway deserves more studies. This is the objective of this paper.
This paper is about diffusive fluxes. Ebullition was already described in details in another paper published last year in Biogeosciences (Deshmukh et al., 2014). The objective of this article is to describe the spatial and temporal variations of diffusive emissions and to show how these variations can affect the assessment of this emission pathway. The discussion on ebullition is out of the scope and the emissions by ebullition are given in the text and in the table 3 with the other emission pathways(downstream) to provide a comprehensive emission budget from the reservoir.
The end of our introduction was rewritten as follow in order to better describe the context and objective of our study: *"In the framework of a comprehensive project aiming at quantifying greenhouse gas emissions from the Nam Theun 2 Reservoir (NT2R), a recently flooded subtropical located in Lao PDR, we studied (1) the spatial and temporal variability of CH4 ebullitive fluxes (Deshmukh et al., 2014) and (2) the downstream CH4 emissions (Deshmukh et al., 2016). In the present study, the objective is to quantify the CH4 diffusive fluxes at the surface of NT2R and evaluate if diffusive fluxes were improperly quantified in previously published CH4 budget due to inappropriate spatial and seasonal resolution. The CH4 emissions were quantified fortnightly during three and a half year (May 2009 to December 2012) based on a monitoring of CH4 concentrations in surface water that started in June*

2009. This was performed at nine stations flooding different types of ecosystems. On the basis of these results, we discuss the spatial and temporal variations of the CH4 emissions by diffusive fluxes and the significance of hotspots and hot moments in the total emissions from the surface of the reservoir."

RC: The first reviewer also points out that more information is needed about the study site to make the life of the reader easier. The authors just reply that all the needed information can be found in earlier papers and only added the coordinates. However, I fully agree with the first reviewer that some more information must be given so that the reader of this very paper learns to know the study site and can then easily follow the text. You should also briefly explain what kind of studies you have earlier carried out in this reservoir since I was quite surprised to learn when reading the text that you have had methane EC running on the site and besides the flux studies based on EC and now on gas concentrations in the surface water in connections with some models you have also used chambers for methane flux measurements. This all was quite confusing.
AC: Our site description includes: the date of impoundment, date of commissioning, the average temperature, the rainfall, the classical meteorology, the surface of the reservoir, the volume of water, the reservoir depth, the water discharge, the type of ecosystem flooded in table 1, a map of the different sampling stations… Our first answer was to refer to previous work published, but actually, there is no need for this since description of the site is already comprehensive. Could the reviewer be more precise in terms of information requested?
The studies conducted on site and important for the understanding of the article (Deshmukh et al., 2016;Deshmukh et al., 2014) are now given in the last paragraph of the introduction. The other information related to the site description (Descloux et al., 2011;Descloux et al., 2014) are summarized in the section 2.1. The description of the calculation of diffusive fluxes and the comparison with floating chambers and eddy covariance from Deshmukh et al. (2014) is given in the section 2.6 related to the calculation of diffusive fluxes.

RC: I fully appreciate your sampling efforts over three years with a sampling frequency of once in a fortnight but I must admit that I agree with the earlier reviewer that you can hardly say that this is high frequency. It is surely higher that bulk of the scientists use, but is not high frequency. In our own projects we have sampled lakes for GHGs for several years with a frequency of once a week and we still do not call this approach high frequency measurements. Please change.
AC: After the first round of review, the mention to "high frequency" was removed following the recommendation by reviewer #1.

RC: For the reason of the protocol based on samplings once in a fortnight I am also doubtful whether you can really claim that once a month sampling is enough. You do not know what was going on in your reservoir between your samplings. Without measurements that is impossible. What I have learnt during my decades in limnology is that when you really start to measure aquatic systems continuously, you will soon be surprised by the finding how dynamic they are. How this dynamics is reflected e.g. in gas fluxes is a different matter, but you cannot just ignore the possibility of highly dynamic character of the system. Please change your text accordingly.
AC: We also performed continuous measurement at frequency of about 10 Hz continuously when deploying the eddy covariance system (Deshmukh et al., 2014), or equilibrator in other circumstances (Abril et al., 2006), and we already know how dynamic can be a reservoir or a

lake. We are fully aware for example that overturns and other sporadic events can occur in time frame of few hours or days.
However we show based on auto-correlation of time series that the stations have a memory effect of 30-40 days (L345). Saying that one month is a minimum frequency as stated L612 to perform monitoring does not imply that no event occur at a higher frequency, but simply that no annual budget of CH4 emissions from a reservoir surface should be build from measurements at a lower frequency.

RC: The authors have put a lot of effort in statistics with the result that due to large natural variation most of the findings are not statistically significant, i.e. there are no differences between the sampling points or times. This being the case the authors still use lots of space in explaining the differences in results. I found this quite confusing.
AC: Kruskal-Wallis and Mann-Whitney tests indicated no significant differences between the seasons and/or the stations. However, frequency distribution, the skewness of the dataset (third order moment), the auto-correlation of each time series and the correlation between the different stations indicated spatial and temporal differences which are commented in the section 3.4. We were able to conclude that sporadic emissions occurred mostly in the wet season at stations located in the inflow and that a monthly monitoring is the minimum required in order to reasonably estimate emissions because of the work on statistics we performed (work on the frequency distribution and work on the autocorrelation In other words, we think that these results fully justify the statistical work conducted and described here.

Some more minor comments:
RC: The title is very long; please shorten it by leaving out the name of the reservoir and perhaps the words ‘at the surface’. Subtitles in the text are also often very long and could be easily shortened.
AC: The title and a few subtitles were shorten as suggested

RC: Lines 131-134. I don’t believe that anchoring has not affected the results. it would be useful to give the information about the times/sites where and when anchoring was used e.g. in Supplement.
AC: Anchoring allowed us to sample the water column from the surface to the bottom at the same station without drifting to the shoreline or a flooded bush. We took all necessary care not to sample above the anchor and, as already explained in the manuscript, sampling of bottom water was done more than one hour after the installation of the anchor.
Is there a misunderstanding on flux measurements that should be done drifting with the water mass in running water as demonstrated by Lorke et al, 2015, biogeosciences?

RC: The phrase physico-chemical is used several times in the manuscript although it is obvious that the only parameters measured were temperature and oxygen. Please change throughout the manuscript.
AC: As noted in the manuscript, we used a multiparameter probe but decided to describe and discuss only temperature and O2 concentration.

RC: Lines 145-146. The word ‘poisoned’ sounds odd. Preserved could be better. You could also provide information about the time the samples were stored before analysed.
AC: modified accordingly

RC: Line 162. Is NTPC really the original reference for the method?
AC: NTPC, stands for Nam Theun Power Company and corresponds to the report were the hypsometric curve is given.

RC: Lines 207-208. I found the visual inspection of vortexes dubious.
AC: We do not understand whether that means that the reviewer does not believe that we observed vortexes or on the fact that vortexes look similar at the two sites.

RC: The paragraph 2.5. is very short. Although I appreciate the idea not to repeat too much information, this methane oxidation is a very crucial part of the presented work and thus, deserves more explanation.
AC: The equation used for the calculation of the aerobic methane oxidation was added to the section

RC: In the results section the paragraphs 3.3. and 3.4 are also very short. Is this really all you want to say?
AC: the section 3.3 was merged with the section 3.2.

RC: Lines 355-363. This belongs to 'Discussion'.
Lines 382-385. This also belongs to 'Discussion'.
AC: The ranges and averages of concentrations and fluxes are given in the results whereas in the discussion we describe the dynamics, the seasonal and spatial variability. Therefore, that is much more straightforward to compare values with the literature in the result section.

RC: Lines 395-400. At least part of this belongs to 'Results'.
AC: In those lines, we combine O2 concentration, CH4 storage and CH4 oxidation which are obviously described in separate sections in the results.

RC: Lines 430-444. Also here part of text could be in 'Results'
AC: As requested by the reviewer #1, we justified the choice of the four stations for the figure 7.

RC: Line 488. The lake is Pääjärvi.
AC: Corrected

RC: Lines 545-548. Again some 'Results' here.
AC: The values in mmol m-2 d-1 are given to remind our definition of extreme fluxes. In this section, emissions are discussed in Gg(CH4) y-1 in order to compare diffusive fluxes with other emission pathways from other studies (Deshmukh et al, 2014 and 2016). This cannot be done in the result section where fluxes are given in mmol m-2 d-1.

RC: List of references: Check for the names of the journals; sometimes you use abbreviations, sometimes you spell out the complete name. Also check the subscripts etc.
AC: Done

RC: Table 1. What does it mean that you give water residence time in arbitrary units? I have never heard about that.
AC: In a reservoir, especially in kind of dendritic reservoirs, water pocks isolated from the inflows and outflows, like embayments for instance, have longer residence time than water located in the thalweg of the river (former river). But this is impossible to quantify this. That

is why, in addition to the quantitative information about hydrology, we put an “index” of water residence time in arbitrary units. This was change to “water residence time index” in the table.

Abril, G., Richard, S., and Guerin, F.: In situ measurements of dissolved gases (CO(2) and CH(4)) in a wide range of concentrations in a tropical reservoir using an equilibrator, Science of the Total Environment, 354, 246-251, 10.1016/j.scitotenv.2005.12.051, 2006.
Descloux, S., Chanudet, V., Poilvé, H., and Grégoire, A.: Co-assessment of biomass and soil organic carbon stocks in a future reservoir area located in Southeast Asia, Environmental Monitoring and Assessment, 173, 723-741, 10.1007/s10661-010-1418-3, 2011.
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Deshmukh, C., Serca, D., Delon, C., Tardif, R., Demarty, M., Jarnot, C., Meyerfeld, Y., Chanudet, V., Guedant, P., Rode, W., Descloux, S., and Guerin, F.: Physical controls on CH4 emissions from a newly flooded subtropical freshwater hydroelectric reservoir: Nam Theun 2, Biogeosciences, 11, 4251-4269, 10.5194/bg-11-4251-2014, 2014.
Deshmukh, C., Guérin, F., Labat, D., Pighini, S., Vongkhamsao, A., Guédant, P., Rode, W., Godon, A., Chanudet, V., Descloux, S., and Serça, D.: Low methane (CH4) emissions downstream of a monomictic subtropical hydroelectric reservoir (Nam Theun 2, Lao PDR), Biogeosciences, 13, 1919-1932, 10.5194/bg-13-1919-2016, 2016.

Effect of sporadic destratification, seasonal overturn and artificial mixing on CH_4 emissions from a subtropical hydroelectric reservoir

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Abstract

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Inland waters in general and specifically freshwater reservoirs are recognized as source of CH_4 to the atmosphere. Although the diffusion at the air-water interface is the most studied pathway, its spatial and temporal variations are poorly documented.

We measured fortnightly temperature and O_2 and CH_4 concentrations parameters at nine stations in a subtropical monomictic reservoir which was flooded in 2008 (Nam Theun 2 Reservoir, Lao PDR). Based on these results, we quantified CH_4 storage in the water column and diffusive fluxes from June 2009 to December 2012. We compared diffusive emissions with ebullition from Deshmukh et al. (2014) and aerobic methane oxidation and downstream emissions from (Deshmukh et al., 2016).

In this monomictic reservoir, the seasonal variations of CH_4 concentration and storage were highly dependant of the thermal stratification. Hypolimnic CH_4 concentration and CH_4 storage reached their maximum in the warm dry season (WD) when the reservoir was stratified. They decreased during the warm wet (WW) season and reached its minimum after the reservoir overturned in the cool dry season (CD). The sharp decreases of the CH_4 storage were concomitant with sporadic extreme diffusive fluxes (up to 200 mmol m^{-2} d^{-1}). These hot moments of emissions occurred mostly in the inflow region in the WW season and during the overturn in the CD season in the area of the reservoir that has the highest CH_4 storage. Although they corresponded to less than 10% of the observations, these CH_4 extreme emissions (>5 mmol m^{-2} d^{-1}) contributed up to 50% of total annual emissions by diffusion.

During the transition between the WD and WW seasons, a new hotspot of emissions was identified upstream of the water intake where diffusive fluxes peaked at 600 mmol m^{-2} d^{-1} in 2010 down to 200 mmol m^{-2} d^{-1} in 2012. In the CD season, diffusive fluxes from this area were the lowest observed at the reservoir surface. Emissions from this area contributed 15-25% to total annual emissions although they occur on a surface area representative of less than 1% of the total reservoir surface. We highly recommend measurements of diffusive fluxes around water intakes in order to evaluate if such results can be generalized.

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1. Introduction

Since the 1990s, hydroelectric reservoirs are known to be source of methane (CH_4) to the atmosphere. Their contribution to total CH_4 emissions still needs refinement since the discrepancies among estimates is large, ranging from 1 to 12% of total CH_4 emissions (St Louis et al., 2000;Barros et al., 2011). These two estimates are mostly based on diffusive

fluxes at the air-water interface and they overlook emissions from the rivers downstream of the dams (Abril et al., 2005;Guerin et al., 2006;Kemenes et al., 2007;Teodoru et al., 2012;Maeck et al., 2013;Deshmukh et al., 2016), CH_4 ebullition (DelSontro et al., 2010;Deshmukh et al., 2014) and emissions from the drawdown area of reservoirs (Chen et al., 2009;Chen et al., 2011) although these pathways could largely dominate diffusion at the surface of the reservoirs.

Even if CH_4 diffusion at the surface of reservoir is the best-documented emission pathway, little information is available on spatial and temporal variability of CH_4 emissions by diffusive fluxes. In tropical amictic reservoirs, the highest diffusive fluxes are usually observed during dry periods and when the stratification weaken at the beginning of the rainy season (Guerin and Abril, 2007). A study of CH_4 emissions from a dimictic reservoir suggests a potential large outgassing of CH_4 during the reservoir overturns (Utsumi et al., 1998b) as it is the case in natural monomictic and dimictic lakes (Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al., 2012;Fernández et al., 2014). Such hot moments of emissions (McClain et al., 2003) could contribute 45-80% of CH_4 annual emissions by diffusion (Schubert et al., 2012;Fernández et al., 2014). They are rarely taken into account in carbon budgets since they can only be captured by high frequency monitoring. Spatial heterogeneity of CH_4 emissions at the surface of reservoirs is also very high. It mostly depends on the spatial variations of ebullition that is controlled by sedimentation (DelSontro et al., 2011;Sobek et al., 2012;Maeck et al., 2013). The spatial variation of diffusion appears to be low with emissions being slightly higher (1) in area where dense forest is flooded as compare to the former riverbed (Abril et al., 2005), (2) at shallow sites than at deeper ones (Zheng et al., 2011;Sturm et al., 2014) and (3) in inflow zones of reservoirs compare to the main body (Musenze et al., 2014). However, as it was shown for CO_2 emissions from a tropical hydroelectric reservoir, taking into account both spatial and temporal variability of emissions significantly affect carbon budgets and emission factors (Pacheco et al., 2015).

In the framework of a comprehensive project aiming at quantifying greenhouse gas emissions from the Nam Theun 2 Reservoir (NT2R), a recently flooded subtropical located in Lao PDR, we studied (1) the spatial and temporal variability of CH_4 ebullitive fluxes (Deshmukh et al., 2014) and (2) the downstream CH_4 emissions (Deshmukh et al., 2016). In the present study, the objective is to quantify the CH_4 diffusive fluxes at the surface of NT2R and evaluate if diffusive fluxes were improperly quantified in previously published CH_4 budget due to inappropriate spatial and seasonal resolution. The CH_4 emissions were quantified fortnightly

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during three and a half year (May 2010 to December 2012) based on a monitoring of CH_4 concentrations that started in June 2009. This was performed at nine stations flooding different types of ecosystems. On the basis of these results, we discuss the spatial and temporal variations of the CH_4 emissions by diffusive fluxes and the significance of hotspots and hot moments in the total emissions from the surface of the reservoir.

2. Material and methods

2.1. Study area

The NT2 hydroelectric reservoir (17° 59' 49" N, 104° 57' 08" E) was built on the Nam Theun River located in the subtropical region of Lao People's Democratic Republic (Lao PDR) on the Nakai Plateau. A detailed description of the study site is given in Descloux et al. (2014). The filling of the reservoir began in April 2008, the full water level was first reached in October 2009 and the power plant was commissioned in April 2010. Annually, the NT2 Reservoir receives around 7527 Mm^3 of water from the Nam Theun watershed, which is more than twice the volume of the reservoir (3908 Mm^3). A continuous flow of 2 $m^3\ s^{-1}$ (and occasionally spillway release) is discharged from the Nakai Dam (ND in Fig 1) to the Nam Theun River. The water used for electricity production is delivered from water intake (WI in Fig 1) to the powerhouse (PH in Fig 1). The powerhouse is located in the valley 200 m below the plateau.

Typical meteorological years are characterized by three seasons: warm wet (WW) (mid June-mid October), cool dry (CD) (mid October-mid February) and warm dry (WD) (mid February-mid June). Daily air temperature varies between 14°C (CD season) to 30°C (WD season). The mean annual rainfall is about 2400 mm and occurs mainly (80%) in the WW season.

During the filling of the reservoir, 489 km^2 of soils and different types of vegetation (Descloux et al., 2011) were flooded by the end of October 2008. The water level in the reservoir was nearly constant from October 2008 to April 2010. After the commissioning, during the studied period (June 2009 to December 2012) the reservoir surface varied seasonally and reached its maxima (489 km^2) and minima (168 to 176 km^2 depending on the years) during the WW and WD seasons, respectively. According to these water level variations, the average depth is 8 m for a maximum depth of 39 m.

2.2. Sampling strategy

A total of nine stations (RES1-9, Figure 1) located in the reservoir were monitored fortnightly in order to determine the vertical profiles of temperature and O_2 and CH_4 concentration in the water column. The characteristics of the stations are given in the Table 1. Basically, three stations are located on the thalweg of the former Nam Theun River (RES2, RES4, RES6) whereas four other stations are located in a small embayment in the flooded dense forest (RES3), flooded degraded forest (RES5), flooded swamp area (RES7) and flooded agricultural land (RES8). The RES1 station is located 100 m upstream of the Nakai Dam, and RES9 station is located ~1 km upstream of the water intake delivering the water to the powerhouse. All samples and in situ measurements were taken in the morning or early afternoon from an anchored boat. Most of the time, the boat was attached to a buoy at the sampling station. When no buoy was present, an anchor was used with care in order not to re-suspend surface sediments. As the sampling started from the surface, the bottom water was sampled almost an hour later and should not be influenced by the perturbation generated by the anchor.

2.3. Experimental methods

2.3.1. Vertical profiles of oxygen and temperature

Vertical profiles of O_2 and temperature were measured in situ at all sampling stations with a multi-parameter probe Quanta® (Hydrolab, Austin, Texas) since January 2009. In the reservoir, the vertical resolution was 0.5 m above the oxic–anoxic limit and 1 to 5 m in the hypolimnion.

2.3.2. Methane concentration in water

The evolution of CH_4 concentrations has been monitored from May 2009 to December 2012 on a fortnightly basis. Surface samples were taken with a surface water sampler (Abril et al., 2007) and other samples from the water column were taken with an Uwitec water sampler. (Abril et al., 2007). Water samples were stored in serum glass vials, capped with butyl stoppers, sealed with aluminium crimps and preserved with $HgCl_2$ (Guerin and Abril, 2007). Samples were analysed within 15 days. Before gas chromatography analysis for CH_4 concentration, a N_2 headspace was created and the vials were vigorously shaken to ensure an

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equilibration between the liquid and gas phases. The concentration in the water was calculated using the solubility coefficient of Yamamoto et al. (1976).

2.3.3. Gas chromatography

Analysis of CH_4 concentrations were performed by gas chromatography (SRI 8610C gas chromatograph, Torrance, CA, USA) equipped with a flame ionization detector. A subsample of 0.5 ml from the headspace of water sample vials was injected. Commercial gas standards (10, 100 and 1010 ppmv, Air Liquid "crystal" standards) were injected after analysis of every 10 samples for calibration. Duplicate injection of samples showed reproducibility better than 5%.

2.4. Water column CH_4 storage

Between two sampling depth of the vertical profiles of CH_4 concentrations, the CH_4 concentrations were assumed to change linearly in order to calculate the concentration in each 1-m layer of water. The volume of water in each layer was calculated using the volume-capacity curve (NTPC, 2005). The CH_4 storage was calculated by multiplying the average CH_4 concentrations of each layer by the volume of the layer and summing-up the amount of CH_4 for all depth intervals.

2.5. Aerobic CH_4 oxidation

The depth-integrated CH_4 oxidation rates at each station were calculated on the basis of the specific oxidation rates (d^{-1}) determined at NT2 (Deshmukh et al., 2016) and the vertical profiles of CH_4 and O_2 concentrations in the water column as already described in (Guerin and Abril, 2007). The depth-integrated CH_4 oxidation rates at each station were estimated only from January 2010 since the vertical resolution of the vertical profiles of O_2 and CH_4 was not high enough in 2009.

As the aerobic methane oxidation rates we obtained were potential, $CH_{4\text{-}ox}$ were corrected for two limiting factors, the oxygen availability and the light inhibition as described in Guerin and Abril (2007). The final equation to compute in situ oxidation rates ($CH_{4\text{-}ox}$, mmol m_a^{-2} d_a^{-1}) is:

$$CH_{4\text{-}ox} = C_{CH4} \cdot S_{CH4\text{-}ox} \cdot C_{O2} / (C_{O2} + K_{m\,(O2)}) \cdot d \cdot I(z)$$

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with C_{CH4}, the CH_4 concentration; $S_{CH4\text{-}ox}$, the specific $CH_{4\text{-}ox}$ from Deshmukh et al. (2016); C_{O2}, the oxygen concentration; $K_{m(O2)}$, the Km of O_2 for CH_4 oxidation, d, depth of the water layer and I(z), the inhibition of methanotrophic activity by light as defined by Dumestre et al. (1999) at the Petit Saut Reservoir. Finally, the CH_4 oxidation rates were integrated in the oxic water column, from the water surface to the limit of penetration of oxygen.

2.6. Diffusive fluxes from surface concentrations

The diffusive CH_4 fluxes were calculated from the fortnightly monitoring of surface concentrations with the thin boundary layer (TBL) equation at all stations in the reservoir (RES1-9). The CH_4 surface concentrations in water and the average CH_4 concentration in air (1.9 ppmv) obtained during eddy covariance deployments (Deshmukh et al., 2014) were applied in equation (1) to calculate diffusive flux:

$$F = k_T \times \Delta C \quad (1)$$

where F, the diffusive flux at water-air interface; k_T, the gas transfer velocity at a given temperature (T); $\Delta C = C_w - C_a$, the concentration gradient between the water (C_w) and the concentration at equilibrium with the overlying atmosphere (C_a). Afterward, the k_T was computed from k_{600} with the following equation:

$$k_T = k_{600} \times (600/Sc_T)^n \quad (2)$$

with Sc_T, the Schmidt number of CH_4 at a given temperature (T) (Wanninkhof, 1992); n, a number that is either 2/3 for low wind speed (< 3.7 m s^{-1}) or 1/2 for higher wind speed and turbulent water (Jahne et al., 1987).

For the determination of k_{600} at the stations RES1-8, we used both the formulations from Guerin et al. (2007) which includes the cumulative effect of wind (U_{10}) and rain (R) on k_{600} ($k_{600} = 1{:}66e^{0{:}26U10} + 0{:}66R$), and the average formulation of MacIntyre et al. (2010) (($k_{600} = 2.25\ U_{10} + 0.16$) whatever the buoyancy fluxes. As shown by (Deshmukh et al., 2014), the average of the fluxes obtained from these two relationships compared well with fluxes measured by floating chambers at the reservoir surface and no enhancement of the CH_4 fluxes could have been attributed to the variations of buoyancy fluxes when the eddy covariance system was deployed. Since the water current velocities were lower than 1 cm s^{-1} in most of

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the reservoir (Chanudet et al., 2012), the effect of water current on k_{600} was not included. For calculation purpose, wind speed (at 10 m height) and rainfall from two adjacent meteorological stations located at Nakai Village (close to RES9 station) and at the Ban Thalang Bridge (close to RES4 station, Figure 1) were used. At these stations, the average k_{600} was 6.5 cm h^{-1} over the course of the year.

At the water intake (RES9) where the hydrology and hydrodynamics is different from the other stations, it was impossible to quantify the k_{600} since the boat drifted quickly to the shoreline because of water currents in the narrow channel. According to Chanudet et al. (2012), water current velocity in this area of the reservoir is about 0.2 m s^{-1}. After Borges et al. (2004), the contribution of such water currents in a water body with depth ranging from 9 to 20 m is 2.0±0.5 cm h^{-1} which should be summed up with the contribution of wind and rainfall from Guerin et al. (2007) and MacIntyre et al. (2010). It gives an average of 9 cm h^{-1}. The k_{600} was determined in the regulating dam (Deshmukh et al., 2014) located downstream of the turbine where we visually observed vortexes similar to those observed at RES9. In the regulating dam, the k_{600} was 19 cm h^{-1} on average for 4 measurements (not show). In order to be conservative for the estimation of emissions from the water intake, we considered a constant value of k_{600} (10 cm h^{-1}) which is in the lower range of (1) the k_{600} calculated from (Guerin et al., 2007), MacIntyre et al. (2010) and Borges et al. (2004), and (2) k_{600} values determined in area with comparable hydrology/hydrodynamics.

2.7. Total emissions by diffusive fluxes

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Based on physical modelling (Chanudet et al., 2012), it has been showed that the station RES9 located at the water intake is representative of an area of ~3 km^2 (i.e. 0.6% of reservoir water surface), whatever the season. This 3-km² area was used to extrapolate specific diffusive fluxes from RES9. The embayment where RES3 is located represents a surface area of 5-6% of the total surface area of the reservoir whatever the season (maximum 28 km^2), to which were attributed the specific diffusive fluxes from RES3. The diffusive fluxes calculated for RES1, RES2, RES4, RES5, RES6, RES7 and RES8 stations were attributed to the water surface area representative for each station, taking into account the seasonal variation of the reservoir water surface from the surface-capacity curve (NTPC, 2005).

2.8. Statistical and correlation analysis

Statistical tests were performed to assess the spatial and temporal variations in the surface CH_4 concentrations and diffusive fluxes at all stations in the reservoir. Normality of the concentration and diffusive datasets was tested with R software (R Development Core Team, 2008) and the Nortest package (Gross and Ligges, 2015). The data distribution was tested with the Fitdistrplus package (Delignette-Muller et al., 2015).

Since all tests indicated that the distribution of the data were neither normal nor lognormal, Kruskal-Wallis and Mann-Whitney tests were performed with GraphPad Prism (GraphPad Software, Inc., v5.04). No significant differences were found between the seasons and/or the stations. These test results were attributed to the very large range of surface concentrations due to the sporadic occurrence of extreme values (over 4 orders of magnitude). In order to reduce this range, the log of the concentrations was used. For each station, the time series of the log of the CH_4 surface concentrations were linearly interpolated and re-sampled every 15 days in order to compare time series with the same number of observations. The log of the concentrations was used to determine the frequency distribution, the skewness of the dataset (third order moment), the auto-correlation of each time series and the correlation between the different stations. All analyses were performed using Matlab.

3. Results

3.1. Temperature and O_2 dynamics in the reservoir

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During the three and half year of monitoring at the stations RES1-8, the NT2R was thermally stratified with a thermocline at 4.5 ± 2.6 m depth in the WD (Feb-Jun) season as revealed by the vertical profiles of temperature (Figure 2). In the WW season, the temperature vertical profiles at the stations RES1-8 either showed a thermocline (RES7 and RES8 in 2010 and 2011, Figure 2) whereas in some occasions, the temperature decreased regularly from the surface to the bottom during sporadic destratification (RES1-3, Figure 2). On average during the WW season, a thermocline was located at 5.8 ± 4.8 m depth. During the CD season, the reservoir overturned as already mentioned by Chanudet et al. (2012) and the temperature was constant from the surface to the bottom (Figure 2) in the different years. In order to illustrate the destratification, a stratification index (ΔT) which corresponds to the difference between the surface and bottom water temperature was defined. During the periods of stratification in

the WD seasons, ΔT was up to 10°C higher than during reservoir overturn in the CD season with ΔT close to zero (Figure 3a). During the WW season, the ΔT decreased gradually.

During the WD season at the stations RES1-8, an oxicline was most of the time located at a depth concomitant with the depth of the thermocline whereas oxygen penetrated deeper in the WW season (Figure 2). During these two seasons, the epilimnion was always well oxygenated with O_2 concentrations higher than 200 µmol L^{-1}. In the WD season, the hypolimnion was completely anoxic whereas O_2 reached occasionally the hypolimnion during the sporadic destratification events in the WW season (29±54 µmol L^{-1}, Figure 2 and 3b). During the CD season (reservoir overturn), the water column was often oxygenated from the top to the bottom of the reservoir (Figure 2). On average over the whole reservoir, the lowest hypolimnic oxygen concentration was observed in 2010 before the reservoir was commissioned (Figure 3b).

After the commissioning of the reservoir (April 2010), the water column located near the water intake (RES9) got totally mixed as revealed by the homogeneous temperature and oxygen profiles from the surface to the bottom whatever the season (Figure 2). The water column at RES9 was always well oxygenated (163 ± 62 µmol L^{-1}, Figure 2).

3.2. Seasonal dynamics of the CH_4 concentration in the reservoir

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At the station RES1-8, when the water column is thermically stratified with a steep oxicline in the WD and often in the WW seasons, CH_4 concentrations are in average ~150 times higher in the reservoir hypolimnion (246 ± 234 µmol L^{-1}) than in the epilimnion (1.6 ± 7.7 µmol L^{-1}) (Figure 2). The gradient of CH_4 concentration at the thermocline/oxicline was steeper during the WD season than during the WW season (Figure 2). During the CD season, the average CH_4 concentration in the reservoir bottom water lowered by a factor of three compare to the WD and the WW seasons. However, the reservoir overturn increased the average CH_4 concentrations in the epilimnion by a factor of two (3.4 ± 14.8 µmol L^{-1}) in comparison with the WD and WW seasons. After the commissioning, the CH_4 vertical profiles of concentration before turbine intake (RES9) were homogeneous from the surface to the bottom. The average CH_4 concentration from the surface to the bottom peaked up to 215 µmol L^{-1} with averages of 39.8 ± 48.8, 29.9 ± 55.4 and 1.9 ± 4.3 µmol L^{-1} during the WD, WW and CD seasons, respectively (Figure 2). The concentrations at RES9 were up to 10 times lower than the maximum bottom concentrations at the other stations for a given season. Since the station

RES9 behaved differently from the other stations, results from this station will be treated separately.

The overall bottom CH_4 concentration (Figure 3c) and dissolved CH_4 stock in the reservoir (Figure 3d) increased at the beginning of the WD season. The higher bottom CH_4 concentration and storage in the reservoir are concomitant with both the establishment of anoxia in the hypolimnion and the reservoir thermal stratification (Figure 3). Hypolimnic CH_4 concentration and storage reached their maxima (up to 508 ± 254 µmol L^{-1} and 4.7 ± 0.5 $Gg(CH_4)$, Figure 3c,d) at the end of the WD-beginning of the WW season when the residence time of water in the reservoir was the lowest (40 days, Figure 3d). Along the WW season, the thermal stratification weakened (Figure 3a) and the CH_4 concentration and dissolved CH_4 stock decreased (Figure 3c,d) while the residence time of water increased (Figure 3d). In the CD season, the reservoir overturns as evidenced by the low ΔT and the penetration of O_2 to the hypolimnion (Figure 3a,b). During CD season, the bottom CH_4 concentration and the storage reached their minima (down to 1.3 ± 4.5 µmol L^{-1} and 0.01 ± 0.001 $Gg(CH_4)$, Figure 3c,d) when the residence time of water was the longest (Figure 3d). The sharp decrease of CH_4 storage and concentration in the transition from the WW to the CD seasons is concomitant with a sharp increase of O_2 concentration at the bottom (up to 160 ± 89 µmol L^{-1}, Figure 3).

During the three and a half years of monitoring, the same seasonal pattern as described above is observed although the annual CH_4 bottom concentration and storage was threefold higher in 2009 and 2010 than in the year 2011 (Figure 3c,d). In the dry year 2012, the reservoir bottom CH_4 concentration and storage was almost twice higher than in wet year 2011.

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3.3. Aerobic CH_4 oxidation in the reservoir

Between 2010 and 2012, the depth integrated aerobic CH_4 oxidation rates ranged between 0.05 and 380 mmol m^{-2} d^{-1} at the stations RES1-RES8 (Figure 4). On average, aerobic oxidation was higher in the WW season (55±63 mmol m^{-2} d^{-1}) than in the CD (30±46 mmol m^{-2} d^{-1}) and WD (36±32 mmol m^{-2} d^{-1}) seasons and it was not statistically different for the three years. In the WD season, aerobic CH_4 oxidation was on average twice higher in 2010 than for the two following years whereas in the CD season, the highest aerobic oxidation rate was observed in 2012.

3.4. Spatial and seasonal variability of surface CH_4 concentration and diffusive fluxes at the reservoir surface (RES1-RES8)

The surface concentrations at the stations RES1-8 ranged from 0.02 to 150 μmol L^{-1} and were 2.0±10.5 μmol L^{-1} (median = 0.9), 1.5±5.5 μmol L^{-1} (median = 0.4) and 3.4±14.7 μmol L^{-1} (median = 0.2) on average for the CD, WD and WW season, respectively. The surface concentration followed a loglogistic distribution, which indicates the existence of extremely high values. This is confirmed by the fact that the skewness of the time series of the log of the CH_4 concentrations for all stations is positive (Figure S3), especially at the stations RES1, RES3 and RES7 for which the skewness is >1. Over the course of the three and a half year of survey, the surface concentrations were not statistically different between all stations and no statistically significant seasonal variations were observed because of the occurrence of sporadic events at all season (Figure S2a). The normalized distribution of concentrations (in log) according to seasons (Figure 5) indicates that these high concentrations were observed without any clear seasonal trend at the station RES1, RES5 and RES6 (<1 up to 150 μmol L^{-1}). At the stations RES2 and RES3, the concentrations up to 128 μmol L^{-1} were mostly observed in the CD season when the reservoir overturns. At the station RES4 located at the Nam Xot and Nam Theun confluence and at the stations RES7 and RES8 both located in the inflow region of the Nam Theun River, the high surface concentrations (up to 64.60 μmol L^{-1}) were mostly observed during the WW season when the reservoir undergoes sporadic destratification. The auto-correlation function of the time series of the log of the surface CH_4 concentrations and diffusive fluxes at the stations RES1-8 indicated that at all stations (except RES1) have a memory effect of 30 to 40 days (Figure S1). This implies that with a sampling frequency of 15 days, we captured most of the changes in the surface CH_4 concentrations. At the station RES1, the changes in CH_4 concentrations are faster than at other stations and would have deserved a monitoring with a frequency higher than 15 days.

During the monitoring at RES1-RES8 stations, the average diffusive flux was 2.8 ± 12.2 mmol m^{-2} d^{-1} ranging from 0.01 to 201.86 mmol m^{-2} d^{-1} without any clear interannual and seasonal trends (Figure S2b). As for the concentrations, flux data followed a loglogistic distribution. The median flux in the WD season is 40 to 80% higher than the median in the WW and CD season, respectively. However, the average fluxes in the WW and CD season are 30% higher than in the WD season (Table 2). This confirms the presence of extremely high values during WD and CD seasons, as expected from the surface concentrations. All seasons

386 together, around 7% of the diffusive fluxes that we observed were higher than 5 mmol m^{-2} d^{-1}
387 which corresponds to extremely high diffusive fluxes in comparison with data from the
388 literature for reservoirs and lakes (Bastviken et al., 2008;Barros et al., 2011). The median and
389 average of these extreme fluxes higher than 5 mmol m^{-2} d^{-1} were 2 times higher in the WW
390 and CD seasons than in the WD season (Table 2).

391 At NT2, diffusive CH_4 fluxes covered the whole range of fluxes reported for tropical
392 reservoirs, depending on the season. Most of the fluxes at the NT2R Reservoir were around
393 one order of magnitude lower than the ones at Petit Saut Reservoir (French Guiana) just after
394 the impoundment (Galy-Lacaux et al., 1997), and in the same order of magnitude as reported
395 for reservoirs older by 10 to 18 years (Abril et al., 2005;Guerin et al., 2006;Kemenes et al.,
396 2007;Chanudet et al., 2011). However, some diffusive fluxes at the stations RES1-8 in the
397 WW and the CD seasons (up to 202 mmol m^{-2} d^{-1}) are among the highest ever reported at the
398 surface of a hydroelectric reservoir or a lake (Bastviken et al., 2011;Barros et al., 2011) and
399 rivers downstream of dams (Abril et al., 2005;Guerin et al., 2006;Deshmukh et al., 2016).

400 **3.5. Surface methane concentrations and diffusive fluxes at the water intake (RES9)**

401 After the commissioning of the reservoir (Julian day 450), the concentrations at the stations
402 RES9 (Figure 6a) located at the water intake were up to 30 times higher than at any other
403 stations that is 36.6±35.8 μmol L^{-1} (median = 24.3), 37.6±67.0 μmol L^{-1} (median = 0.9) and
404 1.0±1.7 μmol L^{-1} (median = 0.3) in the WD, WW and CD season, respectively. The surface
405 concentrations at RES9 were significantly higher in the WD and WW seasons than in the WW
406 and CD seasons ($p = 0.0002$ and Figure 6a). The highest concentration was observed each
407 year at the end of the WD season-beginning of the WW season in between June and August.
408 These maxima decreased from 215 μmol L^{-1} in August 2010 to 87 μmol L^{-1} in June 2012.

409 The diffusive fluxes ranged between 0.03 and 605.38 mmol m^{-2} d^{-1} (Figure 6b and Table 2).
410 On average, the CH_4 diffusive fluxes at RES9 were two to forty times higher than at the other
411 stations in the CD, WD and WW season. Diffusive fluxes at this station are usually higher
412 than 10 mmol m^{-2} d^{-1} from April to July that corresponds to the WD season and the very
413 beginning of the WW season. In 2010, diffusive fluxes were on average 241 ± 219 and 239 ±
414 228 mmol m^{-2} d^{-1} respectively for the WD and WW seasons. In 2011 and 2012, the fluxes
415 dropped down by a factor of two in the WD season (112 ± 110 mmol m^{-2} d^{-1}) and almost by a

factor of forty in the WW season (6.8 ± 14.4 mmol m^{-2} d^{-1}). Overall, emissions at RES9 decreased by a factor of two between 2010 and 2012.

At the water intake, CH_4 diffusive fluxes during the transition between the WD and WW seasons (up to 600 mmol m^{-2} d^{-1}) are the highest reported at the surface of an aquatic ecosystem (Abril et al., 2005;Guerin et al., 2006;Bastviken et al., 2011;Barros et al., 2011;Deshmukh et al., 2016).

4. Discussion

4.1. CH_4 dynamic in the reservoir water column

The gradual decrease of the CH_4 concentration from the anoxic bottom water column to the metalimnion and the sharp decrease around the oxicline in the metalimnion (Figure 2) is typical in reservoirs and lakes where CH_4 is produced in anoxic sediments and flooded soils (Guerin et al., 2008;Sobek et al., 2012;Maeck et al., 2013), and where most of it is oxidized at the oxic-anoxic interface (Bedard and Knowles, 1997;Bastviken et al., 2002;Guerin and Abril, 2007;Deshmukh et al., 2016).

CH_4 concentrations and storage increase concomitantly with the surface water temperature and the establishment of the thermal stratification during the WD season and peak at the end of the WD season-beginning of the WW season (Figure 2 and 3). During the WW season, CH_4 concentrations and storage decrease slowly (Figure 3) while aerobic methane oxidation reaches its maximum (Figure 4). When the reservoir overturns at the beginning of the CD season, the CH_4 hypolimnic concentrations and storage reach their minima (Figure 3). The overturn favours the penetration of oxygen down to the bottom (Figure 2 and 3b). The sharp decrease of the CH_4 concentrations and CH_4 storage during this period is expected to result from sudden outgassing (Section 4.2) together with an enhancement of the aerobic CH_4 oxidation as already observed in lakes that overturn (Utsumi et al., 1998b;Utsumi et al., 1998a;Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al., 2012;Fernández et al., 2014). A large increase of the aerobic methane oxidation was only observed in the CD season in the dry year 2012 (Figure 4) because the amount of hypolimnic CH_4 to be oxidized at the beginning of the CD season was still high in the water column (Figure 3c,d).

As the reservoir overturns during the period over which the water residence time is the longest in the reservoir, the temporal evolution of the concentrations is anti-correlated with the residence time (Figure 3c,d). The seasonal dynamics of the CH_4 in the monomictic NT2R differs from permanently stratified reservoirs like Petit Saut Reservoir where CH_4 concentration increased with retention time (Abril et al., 2005). However, at the annual scale the water residence time has a strong influence on CH_4 concentration and storage in the reservoir. Before the reservoir was commissioned (April 2010), the water residence time was up to 4 years and the CH_4 storage was up to four times higher than in 2011 and 2012 (Figure 3d). Although a decrease of concentration and storage with the age of the reservoir was expected (Abril et al., 2005), the storage in the dry year 2012 was twice higher than in the wet year 2011 due to an increase of the water residence time by 25% between 2011 and 2012. In wet years like 2011, the thermal stratification is weaker than in dry years since the warming of surface water is less efficient and the high water inputs alters the stability of the reservoir thermal stratification as shown by the sharper decrease and the larger range of ΔT in 2011 than in 2012 (Figure 3a). As a consequence, the oxygen diffusion to the hypolimnion was higher in 2011 than in 2012 (Figure 3b) and it enhanced aerobic methane oxidation by 20% in the water column in the WW season in 2011 as compared to 2012 (Figure 4). It therefore suggests that the hydrology affects both the thermal stratification and the hypolimnic storage of CH_4 in reservoirs, indirectly controls aerobic methane oxidation, and ultimately influences emissions.

4.2. Hot moments of emissions during sporadic destratification and reservoir overturn

The figure 7 illustrates the evolution of the diffusive fluxes, the stratification index (ΔT), the CH_4 storage and the aerobic CH_4 oxidation at the stations RES1, RES3, RES7 and RES8. These four stations were selected for their contrasting skewness (Figure S3) which gives an indication on the occurrence of extreme events and the facts that they are representative for all station characteristics (Table 1). It shows that the large bursts of CH_4 (from 5 up to 200 mmol m^{-2} d^{-1}) always occurred when ΔT decreased sharply (>4°C, Figure 7a,d,g,j) and are usually followed by a sharp decrease of the CH_4 storage in the water column (Figure 7b,e,h,k). These hot moments of emissions occurred mostly in the CD at the stations RES1 and RES3 whereas it was in the WW season at the stations RES7 and RES8 (Figure 7). In the WD season, diffusive fluxes gradually increased together with the CH_4 storage in the water column (Figure 7a,d,g,j) and they remained always lower than 20 mmol m^{-2} d^{-1}. These sporadic high

477 fluxes occurred in the WD season at RES3, RES7 and RES8 (Figure 7d,g,j). They are usually
478 associated with ΔT variations lower than 2°C and the CH_4 storage decrease that is associated
479 with these fluxes is not as sharp as the one observed in the CD and WW season (Figure
480 7e,h,k).

481 We therefore confirm the occurrence of hot moments of emissions during the reservoir
482 overturn in the CD season as already observed in lakes that overturn in temperate regions
483 (Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al.,
484 2012;Fernández et al., 2014). The highest emissions determined at NT2R are one order of
485 magnitude higher than previously reported outgassing during overturn and they occur mostly
486 in the section of the reservoir that has the longest water residence time (RES1-3, Table 1) and
487 the largest CH_4 storage (Figure 7b,e,h,k). This suggests that the impact of reservoir overturn
488 can be very critical for the whole-reservoir CH_4 budget in tropical hydroelectric reservoirs and
489 especially in young ones where hypolimnic concentration could reach up to 1000 μmol L^{-1}.
490 Hot moments of emissions also occur during sporadic destratifications in the WW season in
491 the inflow region (RES4 and RES6-8) where the inflow of cool water from the watershed
492 might disrupt the thermal stratification in reservoirs. This is contrasting with the observations
493 in older reservoir than NT2R where high emissions from the inflow region were recently
494 attributed to an enhancement of CH_4 production fuelled by the sedimentation of organic
495 matter from the watershed (Musenze et al., 2014). The high emissions in the WD seasons
496 were associated with early rains and associated high winds that occur sometimes in the last
497 fifteen days of May. This shows that a moderate erosion of the stratification when hypolimnic
498 CH_4 concentrations are high could enhance vertical transport of CH_4 toward the surface and
499 emissions to the atmosphere. Basically, this intense monitoring shows that spatial and
500 temporal variations of CH_4 emissions are largely controlled by the hydrodynamics of the
501 reservoir with extreme emissions occurring mostly in the inflow region during the wet season
502 and mostly in area remotely located from the inflow zone and the riverbed during reservoir
503 overturns in the CD season. Even if less frequent, moderate erosion of the stable and steep
504 thermal stratification during warm seasons, could also lead to high emissions.

505 The evolution of depth-integrated aerobic CH_4 oxidation is not clearly related with the
506 reservoir overturns and the CH_4 burst (Figure 7). Significant increases in the aerobic CH_4
507 oxidation occurred mostly during the first half of the WD season when the stratification was
508 unstable and at the very beginning of the destratification in the WW, when ΔT started to

509 decrease. The oxidation could reach high values (up to 380 mmol m^{-2} d^{-1}) during these two
510 periods since the yield of CH_4 in the water column to sustain the activity of methanotrophs is
511 higher than in the CD season when the reservoir overturns. It shows that in reservoirs or lakes
512 like NT2R that destratify progressively before the overturn, there is no substantial increase of
513 the CH_4 oxidation when the water body overturns as it could be observed in lakes that
514 overturn within a few days (Kankaala et al., 2007). In addition, the contribution of CH_4
515 oxidation to the total loss of CH_4 (sum of diffusion and oxidation) in the WD and WW
516 seasons was 90-95% during the entire monitoring whereas it was 85% in the CD season.
517 During overturns, a significant amount of CH_4 is oxidized (Utsumi et al., 1998a;Utsumi et al.,
518 1998b;Kankaala et al., 2007;Schubert et al., 2012) but it also indicates that the removal of
519 CH_4 during overturn is not as efficient as during seasons with a well established thermal
520 stratification.

521 During the periods with major loss in the CH_4 storage with concomitant CH_4 burst, we
522 compared the change in the yield of CH_4 with the sum of emissions and oxidation. Most of
523 the time, the emissions alone and/or the sum of emissions and oxidation were significantly
524 higher than the amount of CH_4 that was lost from the water column. At the Pääjärvi Lake in
525 Finland (López Bellido et al., 2009), the fact that measured or calculated emissions exceed the
526 loss of CH_4 in the water column was attributed to a probable underestimation of the CH_4
527 storage in the lake by under-sampling the shallow area of the lake. In this study, emissions,
528 storage and oxidation were estimated at the same stations, avoiding such sampling artefacts.
529 Therefore, it suggests that CH_4 is provided by lateral transport or by production in the flooded
530 soil and biomass (Guerin et al., 2008) at a higher rate than the total loss of CH_4 from the water
531 column by emissions and oxidation. This hypothesis could only be verified by a full CH_4
532 mass balance including production and total emissions from the reservoir, which is beyond
533 the scope of this article.

534 **4.3. Hot spot of emissions at the water intake (RES9)**

535 After the commissioning of the reservoir, the temperature and the oxygen and CH_4
536 concentrations were constant from the surface to the bottom of the reservoir at the vicinity of
537 the water intake. On the basis of physical modelling and measurements of water current
538 velocities (Chanudet et al., 2012), the vertical mixing at this station was attributed to the water
539 withdrawal at the intake generating turbulence and water currents over a surface area of 3
540 km^2. At this station, CH_4-rich water from the reservoir hypolimnion reached the surface and

led to diffusive fluxes up to 600 mmol m^{-2} d^{-1} in the WD-WW seasons (Figure 6b) whereas fluxes are 3 orders of magnitude lower in the CD season. To the best of our knowledge, this is the first time that a hotspot of emissions is reported upstream of a dam or an intake bringing water to the turbines. At NT2, the intake is located at the bottom of a narrow and shallow channel (depth =9-20 m) on the side of the reservoir. This design enhances horizontal water current velocities, the vertical mixing and therefore the emissions. The existence of such a hotspot at other reservoirs might be highly dependant on the design of the water intake (depth among other parameters) and its effect on the hydrodynamics of the reservoir water column.

4.4. Estimation of total diffusive fluxes from the reservoir

Yearly emissions by diffusive fluxes peaked at more than 9 Gg(CH_4) in 2010 when the reservoir was commissioned and they decreased down to ≈ 5 Gg(CH_4) in 2011 and 2012 (Figure 8a and Table 3). Yearly integrated at the whole reservoir surface, these emissions correspond to diffusive fluxes of 1.5 to 4 mmol m^{-2} d^{-1}. These emissions are significantly lower than diffusive fluxes measured at the Petit Saut Reservoir during the first two years after flooding but similar to those determined in the following years (Abril et al., 2005) and values reported for diffusive fluxes from tropical reservoirs in Barros et al. (2011). In absence of the extreme emissions (both hotspots and hot moments), diffusive emissions from NT2R would have been one order of magnitude lower than emissions from tropical reservoirs as expected from the lower flooded biomass compare to Amazonian reservoirs (Descloux et al., 2011). Due to the specific dynamic of diffusive fluxes at NT2R, diffusion at the reservoir surface contribute 18 to 27% of total emissions (Table 3) that is significantly higher than at other reservoirs tropical reservoirs where it was measured (See Deshmukh et al., 2016 for a detailed discussion).

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Most of the increase of CH_4 emissions by diffusive fluxes from 4 to 9 Gg(CH_4) between 2009 and 2010 is due to very significant emissions of 2-3 Gg(CH_4) at the water intake (Figure 8a). This outgassing of CH_4 was triggered by the vertical mixing generated by the withdrawal of water from the reservoir to the turbines. Although the area under the influence of the water intake is less than 1% of the total area of the reservoir, emissions at the water intake contributed between 13 and 25% of total diffusive emissions and 4 to 10 % if considering both ebullition and diffusion (Table 3). It is worth to note that emissions at this site are only significant within 3-5 month per year at the end of the WD season-beginning of the WW season when the storage of CH_4 reach its maximum in the reservoir (Figure 8b). This new

hotspot equals 20 to 40% of downstream emissions and contributes between 4 and 7% of total emissions from the NT2 reservoir surface when including ebullition and downstream emissions (Table 3 and (Deshmukh et al., 2016)). Very localized perturbation of the hydrodynamics, especially in lakes or reservoirs with CH_4-rich hypolimnion, can generate hotspots of emissions contributing significantly to the total emissions from a given ecosystem. These hotspots could be found upstream of dams and water intake in reservoirs but also around aeration stations based on air injection or artificial mixing that could be used for improving water quality in water bodies (Wüest et al., 1992).

The contribution of extreme diffusive fluxes (> 5 up to 200 mmol m^{-2} d^{-1}) to total emission by diffusion range from 30 to 50% on a yearly basis (Figure 8a) and from 40 up to 70% on a monthly basis (Figure 8b) although these hot moments represent less than 10% of the observations during the monitoring. In the literature, the statistical distribution of CH_4 emissions dataset always follows heavy-tailed and right skewed distribution like the log-normal, the Generalized Pareto Distribution (Windsor et al., 1992;Czepiel et al., 1993;Ramos et al., 2006;DelSontro et al., 2011) or loglogistic (this study) which indicates that CH_4 emissions are always characterized by high episodic fluxes. The quantification of emissions thus requires the highest spatial and temporal resolutions in order to capture as many hot moments as possible. At a single station, extreme emission events never lasted more than 2 months (3 consecutive sampling dates) and probably lasted less than 15 days most of the time (Figure 7). The auto-correlation function of the concentration time series indicate that a minimum sampling frequency of 1 month is required in this monomictic reservoirs for an accurate description of the change in the surface concentrations and estimation of the emissions (Figure S1). A lower temporal resolution can significantly affect (positively or negatively) the emissions factors of non-permanently stratified freshwater reservoirs. This is particularly critical in the inflow regions when water inputs from the watershed increase in the rainy season in all reservoirs and at the beginning of the overturn in regions of the world where reservoirs are not permanently stratified like in Asia (Chanudet et al., 2011) which concentrate 60% of the worldwide hydroelectric reservoirs (Kumar et al., 2011).

5. Conclusion

The fortnightly monitoring of CH_4 diffusive emissions at nine stations revealed complex temporal and spatial variations that could hardly been characterized by seasonal sampling. The highest emissions occur sporadically during hot moments in the rainy season and when

606 the reservoir overturns. In the rainy season, they mostly occur in the inflow region because the
607 increase of the discharge of cool water from the reservoir tributaries contributes to sporadic
608 thermal destratification. During the reservoir overturn, extreme emissions occur mostly in
609 area remotely located from the inflows and outflows that are supposed to have the highest
610 water residence time. It shows that diffusive emissions can be sporadically as high as
611 ebullition and that these hot moments could contribute very significantly to the total emissions
612 from natural aquatic ecosystems and reservoirs. Our results showing that a monthly frequency
613 monitoring is the minimum required to capture all emissions is probably not applicable to
614 every aquatic ecosystem. However, it suggests that quantification of emissions based on 2-4
615 campaigns in a year might significantly affect emissions factors and carbon budgets of
616 ecosystems under study.

617 We also identified a new hotspot of emissions upstream of the water intake resulting from the
618 artificial destratification of the water column due to horizontal and vertical mixing generated
619 by the water withdrawal. In the case of the NT2R, emissions from this site contribute up to
620 25% of total diffusive emissions over less than 1% of the total reservoir area. We highly
621 recommend measurements of diffusive fluxes around water intakes (immediately upstream of
622 dams, typically) in order to evaluate if such results can be generalized.

623 **Acknowledgements**

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630

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Table 1: Characteristics of the nine monitoring stations in the Nam Theun 2 Reservoir

Station	Flooded ecosystem[1]	Hydrology	Water residence time index[2]
RES1	Dense forest	100 m upstream of the Nakai Dam	**
RES2	Dense forest	Thalweg of the Nam Theun River	**
RES3	Dense forest	Embayment	***
RES4	Degraded forest	Confluence Nam Theun-Nam Xot Rivers	**
RES5	Degraded forest	Aside from the main stream	**
RES6	Degraded forest	Thalweg of the Nam Theun River	*
RES7	Swamp	Between inflows and water intake	*
RES8	Agricultural soils	Between inflows and water intake	*
RES9	Civil construction	Water intake	*

[1]Descloux et al. (2011)

[2]Water renewal index in arbitrary units, (***) stands for longer residence time, (**) for average residence times and (*) for shorter residence times than average for the whole reservoir

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Frédéric Guérin 31/3/y 11:05
Supprimé: time[2]

Frédéric Guérin 31/3/y 10:52
Supprimé: residence

Frédéric Guérin 31/3/y 11:05
Supprimé: time

Frédéric Guérin 31/3/y 16:14
Supprimé: long

Frédéric Guérin 31/3/y 10:52
Supprimé: residence

Frédéric Guérin 31/3/y 16:17
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Supprimé: residence

Table 2 : Median, average, ranges and proportion of diffusive fluxes (F_{CH4}) < 1 and > 5 mmol m^{-1} d^{-1} for three seasons

Station		Warm Dry (WD)	Warm Wet (WW)	Cool Dry (CD)
RES1-RES8	n	212	252	217
	range	0.01-102.59	0.01-201.86	0.01-94.64
	median	1.08	0.64	0.20
	Average ± SD	2.23±7.37	3.12±14.58	3.04±12.89
	% F_{CH4} < 1	48%	63%	86%
	% F_{CH4} > 5	6.6%	7.5%	7.4%
	Mediane F > 5	10.67	13.80	23.75
	Average F > 5	16.69±25.04	30.23±45.99	36.45±33.19
RES9	n	39	45	36
	range	0.24-342.00	0.03-605.38	0.07-17.62
	median	40.81	1.23	0.48
	average ± SD	83.33±15.57	78.58±24.73	2.21±0.69

819 Table 3: Methane emissions from the Nam Theun 2 Reservoir between 2009 and 2012.

$Gg(CH_4)\ year^{-1}$	2009	2010	2011	2012
Emission from reservoir				
Diffusion at RES9 only	0.02±0.01	2.33±0.21	0.86±0.12	0.66±0.11
Total diffusion	4.45±1.01	9.34±2.32	3.71±0.81	4.95±1.09
Contribution of RES9 to diffusion (%)	0.4	24.9	23.2	13.3
Ebullition[1]	11.21±0.16	14.39±0.11	14.68±0.10	12.29±0.09
Total emissions from reservoir	15.66±1.02	23.73±2.32	18.39±0.82	17.25±1.09
Contribution of RES9 (%)	0.1	9.8	4.7	3.8
Total downstream emissions[2]	7.79±0.90	10.73±0.83	2.29±0.41	2.00±0.32
Total emissions (reservoir + downstream)	23.45±1.36	34.46±2.46	20.67±0.92	19.24±1.14
Contribution of diffusion to total emission	19%	27%	18%	26%
Contribution of RES9 to total (%)	<0.1	6.8	4.2	3.4

820 [1]Deshmukh et al. (2014)
821 [2](Deshmukh et al., 2016)
822
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825 Figure captions
826
827 Figure 1: Map of the sampling stations and civil structures at the Nam Theun 2 Reservoir
828 (Lao PDR).
829
830 Figure 2: Vertical profiles of temperature (°C), oxygen (μmol L^{-1}) and methane (μmol L^{-1}) at
831 the stations RES1, RES3, RES7, RES8 and RES9 in the Nam Theun 2 Reservoir.
832 Representative profile of the years 2010 (circle), 2011 (square) and 2012 (triangle) are given
833 for each seasons: cool dry in blue, warm dry in red, and warm wet in grey.
834
835 Figure 3: (a) Stratification index (ΔT, see text), (b) O_2 concentration in the hypolimnion
836 (μmol L^{-1}), (c) CH_4 concentration in the hypolimnion (μmol L^{-1}) and (d) CH_4 storage in the
837 water column ($Gg(CH_4)$ $month^{-1}$, bars) and water residence time (days, black line with circles)
838 in the Nam Theun 2 Reservoir (Lao PDR) between 2009 and 2012. The red, grey and blue
839 colours indicate the warm dry (WD), warm wet (WW) and cool dry (CD) seasons,
840 respectively. For the panels (a), (b) and (c), the boxes show the median and the interquartile
841 range, the whiskers denote the full range of values and the plus sign (+) denotes the mean.
842
843 Figure 4: Seasonal variations between 2010 and 2012 of the depth-integrated aerobic CH_4
844 oxidation (mmol m^{-2} d^{-1}) at the stations RES1-RES8 calculated from the aerobic oxidation
845 rates determined by (Deshmukh et al., 2016). WD stands for warm dry (in red), WW for
846 warm wet (in grey) and CD for cool dry (in blue). The boxes show the median and the
847 interquartile range, the whiskers denote the full range of values and the plus sign (+) denotes
848 the mean.
849
850 Figure 5: Frequency distribution of the log of CH_4 concentrations (μmol L^{-1}) at the nine
851 monitoring stations of the Nam Theun 2 Reservoir. The red, grey and blue colours indicate the
852 warm dry (WD), warm wet (WW) and cool dry (CD) seasons, respectively.
853
854 Figure 6: (a) Surface concentrations and (b) diffusive fluxes between June 2009 and
855 December 2012 at the station RES9 located at the water intake. Julian day 0 is 1st of January,
856 2009. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool
857 dry (CD) seasons, respectively.

Figure 7: (a, d, g, j) stratification index (ΔT, red line, see text) and diffusive fluxes, (b,e,h,k) CH_4 storage and (c,f,i,l) depth-integrated aerobic CH_4 oxidation (mmol m^{-2} d^{-1}, black line) calculated from the aerobic oxidation rates determined by (Deshmukh et al., 2016) and ΔT (red line) between June 2009 and December 2012 at the stations RES1, RES3, RES7 and RES8 at the Nam Theun 2 Reservoir. Julian day 0 is 1st of January, 2009. The red, grey and blue colour dots indicate the warm dry (WD), warm wet (WW) and cold dry (CD) seasons, respectively.

Figure 8: (a) Total emissions by diffusive fluxes in 2009, 2010, 2011 and 2012, and (b) monthly emissions by diffusive fluxes between May 2009 and December 2012. Emissions from RES9 (water intake) are shown in black, emissions resulting from diffusive fluxes lower than 5 mmol m^{-2} d^{-1} from the stations RES1 to RES8 are shown in white and emissions resulting from diffusive fluxes higher than 5 mmol m^{-2} d^{-1} from the stations RES1-RES8 are shown in grey.

875
876 Figure 1
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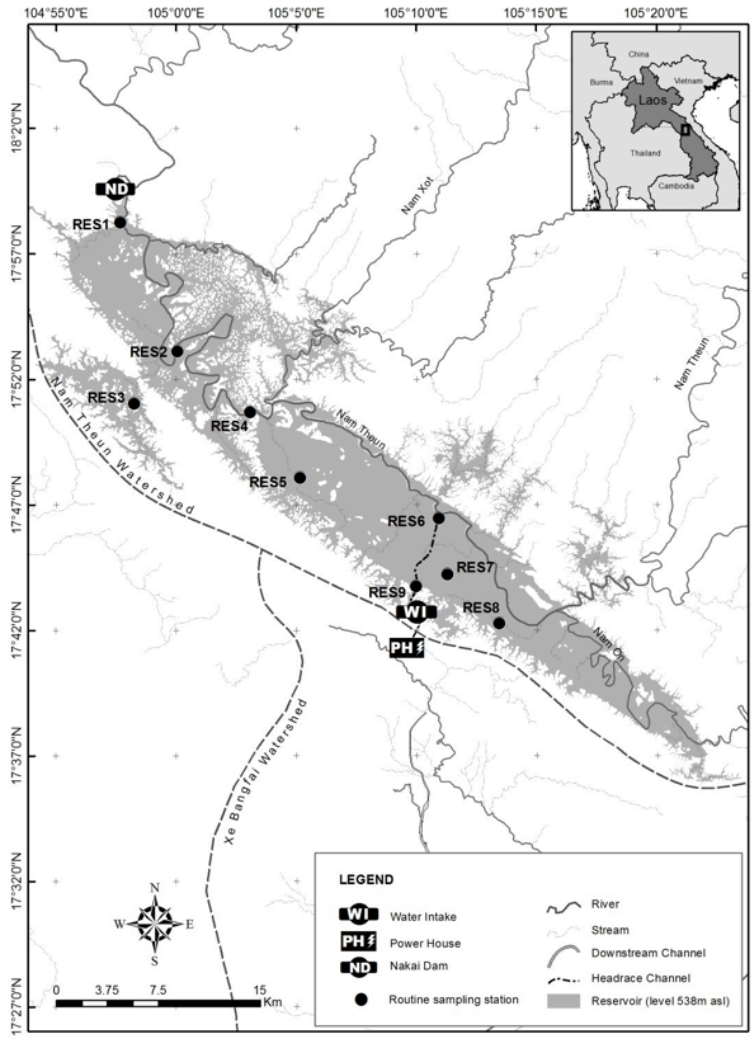

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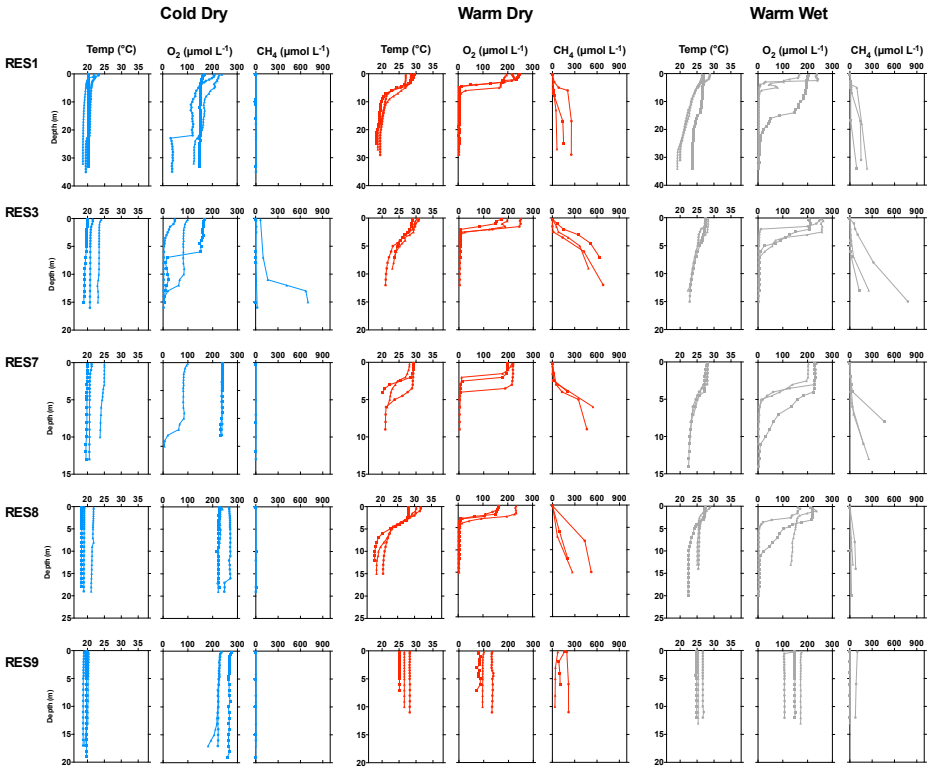

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886

Figure 3

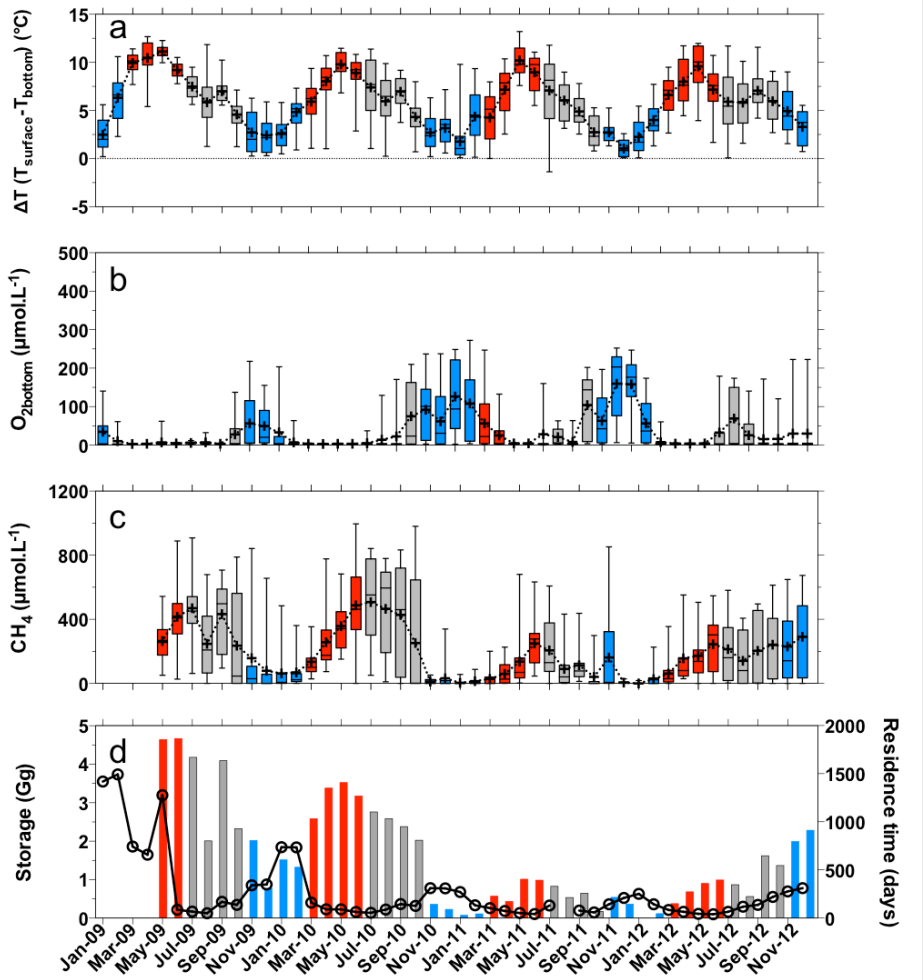

a
b
c
d
ΔT (T surface-T bottom) (°C)
O2bottom (µmol.L-1)
CH4 (µmol.L-1)
Storage (Gg)
Residence time (days)
Jan-09
Mar-09
May-09
Jul-09
Sep-09
Nov-09
Jan-10
Mar-10
May-10
Jul-10
Sep-10
Nov-10
Jan-11
Mar-11
May-11
Jul-11
Sep-11
Nov-11
Jan-12
Mar-12
May-12
Jul-12
Sep-12
Nov-12

Figure 4

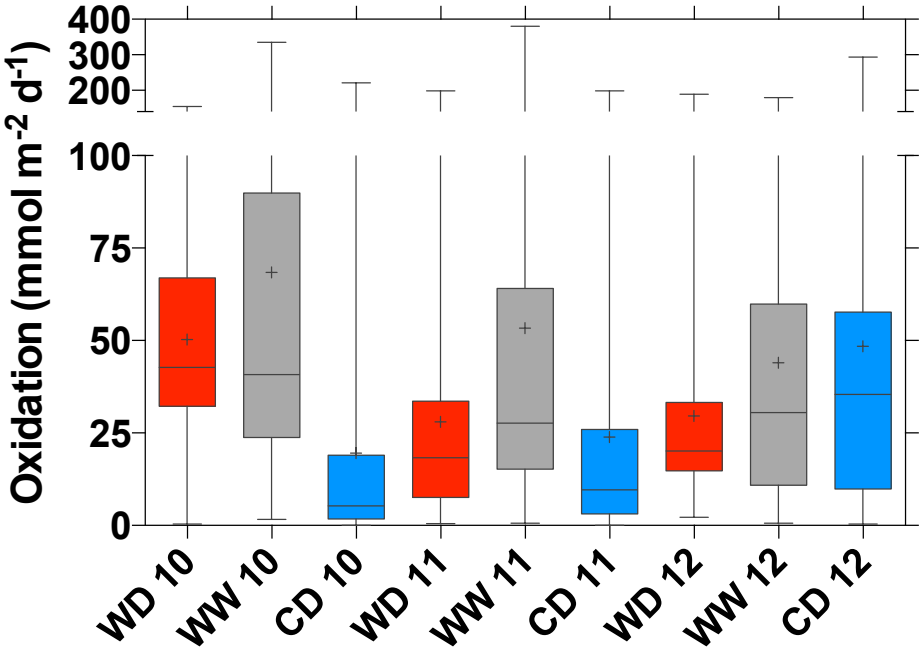

Oxidation (mmol m-2 d-1)
400
300
200
100
75
50
25
0
WD 10
WW 10
CD 10
WD 11
WW 11
CD 11
WD 12
WW 12
CD 12

Figure 5

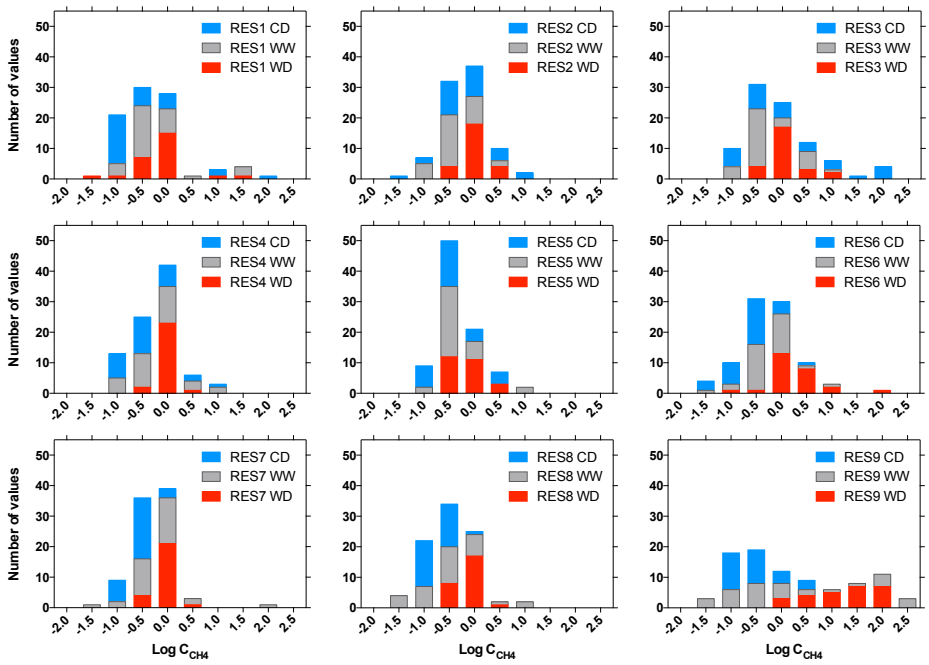

RES1 CD
RES1 WW
RES1 WD
RES2 CD
RES2 WW
RES2 WD
RES3 CD
RES3 WW
RES3 WD
RES4 CD
RES4 WW
RES4 WD
RES5 CD
RES5 WW
RES5 WD
RES6 CD
RES6 WW
RES6 WD
RES7 CD
RES7 WW
RES7 WD
RES8 CD
RES8 WW
RES8 WD
RES9 CD
RES9 WW
RES9 WD
Number of values
Log C_{CH4}

Figure 6

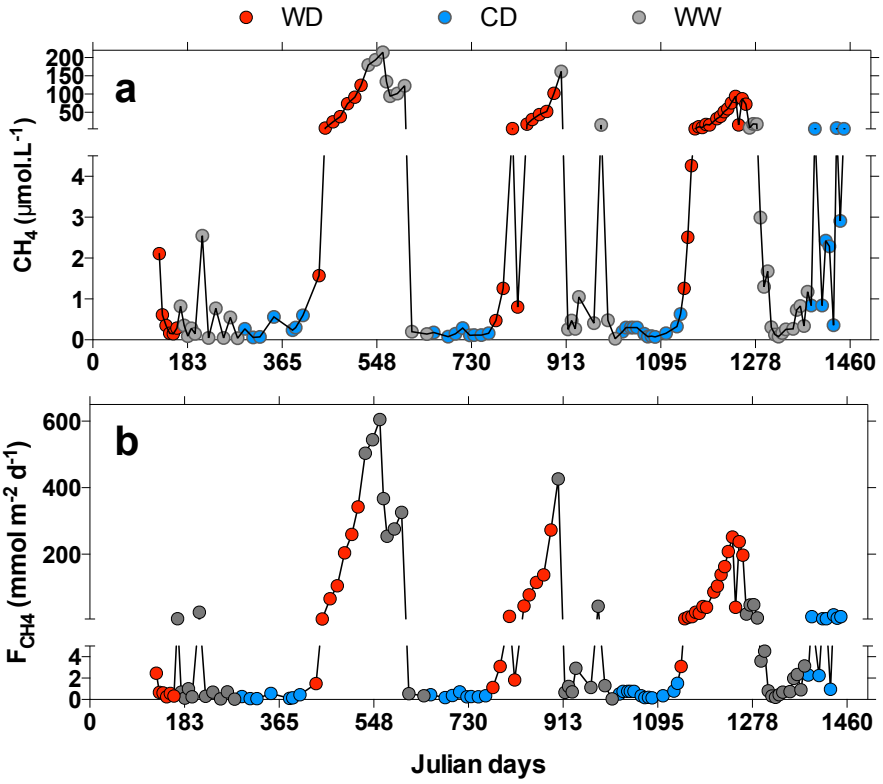
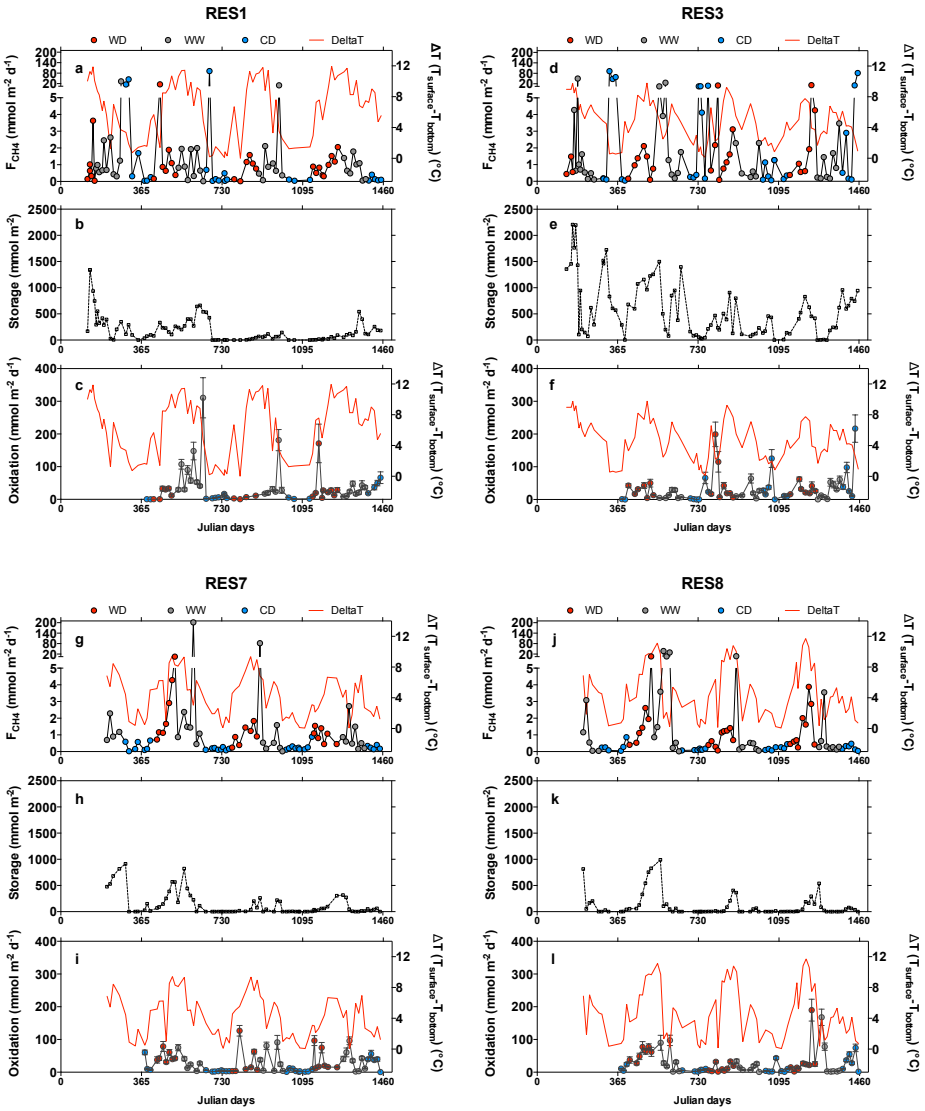
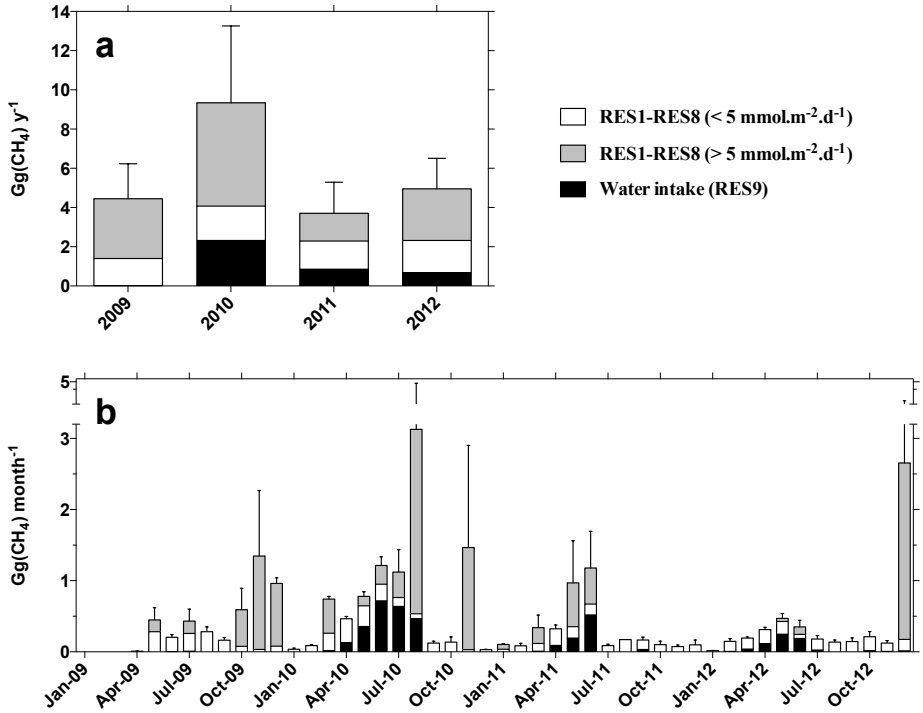

WD
CD
WW
a
b
CH_4 (µmol.L^{-1})
F_{CH4} (mmol m^{-2} d^{-1})
Julian days
0
183
365
548
730
913
1095
1278
1460

Figure 7


RES1
RES3
RES7
RES8
WD
WW
CD
DeltaT
a
b
c
d
e
f
g
h
i
j
k
l
Julian days
365
730
1095
1460

917
918 Figure 8
919



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