Dear Editor

Find below our detailed answers to all reviewer's comments together with the marked-up MS.

Kind regards
Fred Guérin, on behalf of all co-authors

ANSWER TO REVIEWER #1

All co-author thank the reviewer for its thorough review

1-REVIEWER : In general, the structure of the manuscript is well organized and flows in a logical order. However, some important facts are revealed later in the text, and therefore the manuscript would benefit from rearranging the text. For example, it is not clear for the reader that RES1-8 seem to behave rather similarly and RES9 is an exception. This could be solved by more clearly separating these two in the text, maybe even dividing them into their own chapters. Also, the reservoir should be described more precisely in 2.1. At least reservoir depth should be described and coordinates given.
ANSWER: The difference between RES1-8 and RES9 in terms of CH4 concentrations will be stated more clearly than it is in the section 3.2 by adding: "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 two groups of stations are already described and discussed in separate sections in the sections 3.5 and 3.6 (Results) and 4.2 and 4.3 (Discussion) and already fulfil the reviewer's requirements. The required reservoir characteristics were added. The reservoir description was not extended since all information necessary for the understanding of the article is included in the site description and the reservoir was described in details in several publications (Chanudet et al., 2015;Chanudet et al., 2014;Descloux et al., 2015;Deshmukh et al., 2015;Deshmukh et al., 2014;Descloux et al., 2014)

2-REVIEWER : Many topics are mentioned in the introduction in a way that suggests that the authors will return to these points. Therefore, it seems surprising that these themes are not discussed in the Discussion or Conclusions. In the first paragraph of the Introduction, it is mentioned that rivers downstream of dams and CH4 ebullition are not considered in the estimates of CH4 effluxes from hydroelectric reservoirs, and that these are a large source of discrepancy. And yet, only diffusive fluxes from the reser- voir are considered in this manuscript. In the next paragraph, spatial heterogeneity of CH4 emissions is attributed mostly to ebullition. Seems that this study contradicts that statement, but this is not clearly discussed.
ANSWER: We agree with the reviewer on the current lack of connections between the beginning of the introduction and the content of the manuscript. We added a few sentences to mention that ebullition and downstream emissions from the Nam Theun 2 Reservoir were quantified (see Deschmukh et al, 2014, Biogeosciences and Deshmukh et al., 2015, Biogeosciences Discussion, companion paper) and that the current manuscript focus on emission by diffusive fluxes at the reservoir surface. The spatial and temporal variations at the stations RES1-8 and the temporal variations at the sta- tion RES9 are discussed in details in

the sections 4.2 and 4.3, respectively and are also clearly stated in the conclusion. In order to summarize our results on the complex seasonal and spatial variations at the stations RES1-8, a few sentences were added in the section 4.2.

3-REVIEWER : Methods section needs some improvement. Were the measurements taken from a fixed platform? If not, was the boat anchored? The time of the day, or time range, should be given when the measurements were taken in general. It has been shown that gas fluxes depend on wind speed and heat flux (e.g. MacIntyre et al., 2010), and these vary along the course of the day. It can cause bias to the results if the measurements were taken always at the same time. This is not to say that the study should have been conducted some other way, as this approach is typical in these studies with manual sampling, but just that the reader is aware of this. The possible bias should also be discussed in the text.
ANSWER: The following sentence was added to the Sampling strategy section (2.2): All samples and in situ measurements were taken in the morning or early afternoon from an anchored boat. Most of the time, the boat is attached to buoy at the station. When no buoy is present, an anchor is used, with care in order not to resuspend 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. In contrast with the results of (MacIntyre et al., 2010), Sahlé et al (2014), we show in Deshmukh et al (2014, biogeosciences) at this site during several field cam- paigns between 2009 and 2011 that there was no enhancement of the diffusive fluxes (or a negligible enhancement) during continuous measurements of CH4 emissions by eddy covariance. Only ebullition had a semi-diurnal pattern. We therefore believe that this potential bias is negligible in our case.

Minor comments:
4-REVIEWER : Page 11354, line 18: “physico-chemical parameters” seem to refer only to temperature and dissolved oxygen. For making it easier for the reader to follow, I suggest to write “. . .the vertical profiles of temperature and dissolved oxygen in the water column. . .”
ANSWER: Physico-chemical is be replaced by temperature and oxygen concentration

5-REVIEWER : Page 11355, section 2.3.2. “Surface and deep-water samples for CH4. . .”. I read this as only two samples of CH4 concentration were taken, one from the surface and one from the bottom. However, e.g. in Fig. 2 many other sampling depths between these two are presented. Please clarify the sampling strategy more clearly.
ANSWER: In the revised version, this will be rewritten as follow: Surface samples were taken with a surface water sampler (Abril et al., 2007) and other samples from the water column were taken with a Uwitec water sampler.

6-**REVIEWER** : Page 11356, line 18. “...water and air CH4 concentrations were ap- plied...”. Previously, there has been no mention of measurements of atmospheric CH4 concentra- tions. How were these obtained?
ANSWER: No atmospheric air was sampled. We used an average atmospheric con- centration of 2 ppm, well in line with the concentration measured with the Los Gatos CH4 analyzer we deployed for eddy covariance measurements. This was rewritten as follow: The CH4 concentrations in water and the average CH4 concentration in air (2 ppmv) obtained during eddy covariance deployments (Deshmukh et al., 2014) were applied in equation (1) to calculate diffusive flux.

7-**REVIEWER** : Page 11357, line 6-7. “For the determination of k600, we used the formulations of. . . MacIntyre et al. (2010)”. Please specify which formulation was used. They present more than one in their article.
ANSWER: We used the equation (7) from Guérin et al (2007) which includes the com- bined effect of wind and rain on the gas transfer velocity. From MacIntyre et al. (2010), we used the average equation which included the dependency of k600 to wind speed whatever the buoyancy fluxes (k600 = 2.25 U10 + 0.16). This now specified in the revised version

8-**REVIEWER** : Page 11357, line 13. “. . .the boat drifted quickly. . .”. Which boat are the authors re- ferring to? There is no mention of a boat before. Please describe more precise how the measurements were conducted. Using word “station” leads the reader to think of a fixed mast or platform or such.
ANSWER: As mentioned in our answer to your second general comments, sampling was performed from a boat and this is stated in the manuscript. The word “station” is commonly used in limnology.

9-**REVIEWER** : Page 11357, line 19. “. . . and buoyancy flux from. . .”. How buoyancy flux was defined or calculated? There is no mention of measurements of heat budget components.
ANSWER: The buoyancy flux cannot be calculated with the dataset included in this manuscript. Therefore, saying that we calculated the fluxes taking into account was misleading since only one equation from MacIntyre et al (2010) was used whatever the heat fluxes. So the mention to buoyancy flux is removed.

10-**REVIEWER** : Page 11357, lines 21-22. “In the regulating dam where we observed the same vortexes as in RES9,. . .”. Please clarify what is meant with this sentence. By ‘same’ is meant ‘similar’? Is this based on visual observation?
ANSWER: The sentences: “The k600 was determined in the regulating dam (Desh- mukh et al., 2015) located downstream of the turbine where we observed vortexes similar to those observed at RES9. In the regulating dam where we observed the same vortexes as in RES9, the k600 was 19cmh−1 on average for 4 measurements” are rewritten as follow: “The k600 was determined in the regulating dam (Deshmukh et al., 2015) located downstream of the turbine where we visually observed vortexes similar to those observed at RES9. In the regulating dam, the k600 was 19 cm h−1 on average for 4 measurements”

11-**REVIEWER** : Section 2.6. k is a critical component when calculating the fluxes. Some kind of error estimate should be provided when k is estimated from equations. It seems that the residence times are very short in this reservoir, giving reason to believe that there are significant currents. Gas transfer equations have no parameter for currents, even though they produce turbulence at the surface, as was noted also by the authors (page 11357, lines 14-17). For this reason, more justification would be in order to convince the reader that these equations can be used for this reservoir and for different parts of the reservoir.
ANSWER: The average residence time is 6 months ranging from 1.5 to 12 months as depicted in Figure 3 and the maximum water current velocity that was measured in the reservoir is 0.2 m s-1 (Chanudet et al, 2012) as mentioned in the manuscript. Such water current velocities were only measured around the station RES9, anywhere else in the reservoir they were below 0.01 m s-1. Therefore, the water current is unlikely to be a significant controlling factor of the k600 except at RES9 where it can increase it by a maximum of 2 cm h-1 as mentioned in the manuscript. In addition, as mentioned in the section, TBL calculations were well in line with fluxes measured by floating chambers and eddy covariance (Deshmukh et al, 2014). We believe we already provided all justifications asked by the reviewer.

However, the average water current velocity was added to the manuscript and the paragraph was improved by adding more details. We consider that the use of two different relationships for the k600 determination give a wide range of emissions and could be considered as the uncertainty of the fluxes.

12-**REVIEWER** : Section 2.8. There are no references and this is the first time I have seen this kind of approach to assess spatial and temporal variations of CH4 concen- trations and fluxes. Since this is not a standard procedure in limnological literature, more description might prove useful for other scientists to assess spatial and temporal variability of CH4 in their studies.

ANSWER: Based on Kruskal-Wallis and Mann-Whitney tests, 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 ex- treme values (over 4 orders of magnitude). In order to reduce this range, the log of the concentrations was used. The resampling at a 15 days time-step was done for com- paring time series with the same number of observations and avoiding issues related to oversampling. The main differences between the seasons and stations were the occurrence of fluxes higher than 5 mmol m-2 d-1. Therefore we used the frequency distribution and the skewness in order to discriminate the seasons and the stations. These two parameters and the correlation functions are common tools in statistical software. Based on the comments of Reviewer 1 and 2, the paragraph was rewritten as follow: "Since all tests indicated that the distribution of the data were neither normal nor lognormal at the stations RES1-8, Kruskal-Wallis and Mann-Whitney tests were performed with GraphPad Prism (GraphPad Software, Inc., v5.04). No significant dif- ferences 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 occur- rence 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 CH4 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."

13-**REVIEWER** : Page 11360, lines 13-18. During WD and WW, the overall water column CH4 concentrations seem to be rather high compared to other sampling sites, especially since the oxidation rate of CH4 and k are estimated high at this location. Could the authors provide a reason or guess why the concentrations keep up so high?

ANSWER: The concentrations at RES9 from the surface to the bottom are always lower than the maximum concentration in the hypolimnion at other stations. This fol- lowing sentence was added: "The concentrations at RES9 are up to 10 times lower than the maximum bottom concentrations at the other stations for a given season."

14- **REVIEWER** : Page 11361, lines 12-13. "In the dry year 2012, the reservoir bottom CH4 concentration and storage was almost twice higher than in wet year 2011." Could the authors provide any explanation for this?

ANSWER : This section is the result section and expla- nation requires taking into account aerobic oxidation, hydrology and water residence time so explanation are all given in the discussion. See from L23 P11365 to L7 P11364 of the submitted manuscript and the answer to the comment 16.

15-**REVIEWER** : Page 11362, lines 14-16. “The surface concentrations were not statistically different. . .”. I read this so that the surface water CH4 concentrations and fluxes varied independent of the season. However, there is, per visual observation, an evident pattern in both CH4 concentrations and fluxes in Fig. S2. Also, later in Dis- cussion, 4.1., the significance of stratification and overturn to gas concentrations and fluxes are described. Could the authors elaborate this paradox?

ANSWER: We agree that the baseline of the temporal evolution of diffusive fluxes and concentration depict a pattern with higher fluxes in the WD season. However, due to the occurrence of high fluxes and concentrations without clear seasonal patterns at all stations, there was no statistical difference between the seasons while using classical statistical tests as now explicitly mentioned in the MS (see answer to comment 12). The occurrence of extreme values precludes statistical tests to give the “expected results” based on visual observations of the graphs. In the section 4.1, there is a description of the seasonal dynamic in the water column, mostly based on bottom concentration and storage, not on the surface concentrations and fluxes. The surface concentrations and fluxes are described in the section 4.2 and in the figure 7 and it is said that high fluxes occur mostly in the WW season in the inflow zone and mostly in the CD in the rest of the reservoir.

16-**REVIEWER** : Page 11366, lines 6-7. “It therefore suggests that the residence time.”. I think the authors have a nice idea here, but the statement is perhaps too simplified. The reason seems to be that higher water inflow and outflow rates (with appropriate characteristics, like colder T than in the reservoir) affect the stratification behavior in the reservoir, which results in changes in methane oxidation rate. Residence time itself gives no information of how the water body stratifies or not.

ANSWER: We agree with the reviewer that we should focus more on the destratification due to high water inputs. The text was modified as follow: “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 CH4 in reservoirs, indirectly controls aerobic methane oxidation and ultimately emissions.”

17-**REVIEWER** : Page 11366, lines 10-12. Could the authors provide a reason why sites RES1,3,7 and 8 were chosen? In general, the choice of which sites are discussed seems arbitrary.

ANSWER: These stations were not selected arbitrary. RES1 was chosen because of its highest skewness indicating that extreme events are more frequent at this station than at all other stations in the reservoir; they occur in both the CD and WW season. RES3 was chosen because overturn occurs mostly in the CD season during lake over- turn. RES7 and RES8 were selected as they are located in the inflow zone with high and intermediate skewness, respectively. The following sentence was added: “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 representa- tive for all station characteristics (Table 1).”

18-**REVIEWER** : Page 11366, lines 19-22. Could the authors clarify these lines. Do they suggest that during WD season at RES3,7 and 8 the reason for these high fluxes were

overturn, as in CD season? What would be the cause for destratification during this season? Also, if there would be data available to validate these causes, it would be interesting to see.
ANSWER: Actually, these high emissions in the WD seasons were associated with early rains and associated high winds that occur sometimes in the last fifteen days of May. Due to the very high hypolimnic CH4 concentrations at this period of the year, a sporadic destratification due to wind and rain enhance vertical transport of CH4 toward the surface and diffusive fluxes. This was added in the manuscript.

19-**REVIEWER** : Page 11368, lines 24-25. "This design enhances. . .". This is a good finding. I would assume that it also increases lateral transport of hypolimnic waters, which in turn bring more CH4 to the area of strong vertical mixing. Therefore, this spot has even larger spatial impact causing outgassing of CH4 from large area.
ANSWER: It increases lateral and vertical transport and the concentration at this site is close to the average of the concentration in the whole reservoir. The physical modelling and the measurements of vertical and horizontal water current (Chanudet et al, 2012) show that this is restricted to an area of 3 km2, as stated in the manuscript. Therefore, we are confident with the extension of the area under influence of the water intake.

20-**REVIEWER** : Page 11369, lines 20-24. The authors state that these hot moments only occur a few days in a year. On the same page, lines 26-27, they also say that based on fortnightly measurements, 1 month sampling frequency is sufficient. In my opinion, this conclusion needs more explanation. If this is based on sampling interval of 2 weeks, how the authors can be confident that a significant amount of these hot moments, lasting only few days, were not missed during the study? Especially, since the full CH4 mass balance was not conducted and there are unclear components in CH4 cycle, like possible lateral transport of CH4 (page 11368, lines 6-11).
ANSWER: We obviously cannot be 100% sure that no hot moment was missed, be sure that the peak of emissions was not missed and be sure on the duration of the sporadic events. However, we never observed extreme emissions lasting more than three consecutive samplings, which corresponds to a duration of 1.5-2 months at a single station as it is visible on Figure 7. The text was modified has follow: "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)."

21-**REVIEWER** : Page 11370, lines 8-10. "The high frequency. . .". Seems quite bold to say that one measurement per two weeks is not discrete and that it is high frequency, when it has been shown that e.g. wind speed is a major driving force of gas exchange, and wind speed has ample variation in much shorter time scale than 2 weeks. I suggest to rephrase this sentence since this manuscript actually deals more with the seasonal methane fluxes and discrete sampling and not so much with the actual gas exchange dynamics and high frequency sampling.
ANSWER: Our fortnightly monitoring (over more than 3 years) is "high frequency" as compared to most of the studies on lakes and reservoirs, which are based on "seasonal sampling" (2-4 sampling per year). We removed any mention to high frequency and discrete sampling in the first sentence which was modified as follow:" The fortnightly monitoring of

CH4 diffusive emissions at nine stations revealed complex temporal and spatial variations that could hardly been characterized by seasonal sampling."

22-**REVIEWER** : Figure 2. The panels and axis fonts are way too small. Maybe less measurement sites could be shown and the ones that are shown are larger?
ANSWER: The size of the graphs and fonts was increased but all stations are kept

23- **REVIEWER** : Figure 3. (c) is missing.
ANSWER: Added

24-**REVIEWER** : Figure 7. Check the letters in the panels. (g) is missing and (m) is excess. Also this figure suffers from being very small. The axis labels tick marks are unreadable.
ANSWER: Labelling of the graphs was be corrected and the readability of the figures is improved

ANSWER TO REVIEWER #2

R#2 : Due to the great variability in time, the authors remark in the conclusion that temporal sampling might be at least monthly. This reviewer, however, recommend to the authors to avoid taking data only under the light of nonparametric analysis due to non-normal distribution.
ANSWER: Parametric tests are based on the normal distribution and cannot be used when the dataset follow other distributions

R#2 : Instead, authors should better explore the intrinsic nonlinearities in the underlying CH4 dynamics in hydroelectric reservoirs. Are these distributions power laws, Pareto, log-normal?
ANSWER: As now mentioned in the manuscript (section 3.5) and show in the supplementary material (Figure S3), the dataset (both surface concentrations and calculated diffusive fluxes) follows a loglogistic distribution.

R#2 : If so, what kind of process would lead this sort of distribution outcomes in space and time? Are there literature considering these other kinds of distributions?
ANSWER: Fitting a distribution is only possible with large datasets which are unfortunately rare. Only a few studies consider the statistical distribution of their data and all distributions are heavy-tailed (lognormal or Generalized Pareto Disstribution), indicating that high episodic fluxes are very common for CH4. It confirms that CH4 emissions occur through hotspots and hot moments but it cannot provide any information on the importance of these rare and intense fluxes on the global CH4 budget of the studied ecosystems

R#2 : I do not presume that only intensifying the sampling monitoring would bring novel information, as the distributions maybe the same, nonGaussians. I recommend to the authors to go further on dynamical analysis (complexity) in order to find differential equations or statistical models that come out with those distributions, and might be applicable to any water body. That would be a great advance in CH4 studies and application to hydroelectric reservoirs.
ANSWER: As explained in the section 4.4 and in the answer to the previous comment, defining the type of distribution of a dataset for a given ecosystem requires intense monitoring for at least a year in order to have a dataset with a sufficient number of data encompassing hot moments and the hotspots of emissions to be able to find a statistical distribution. The rare but significant events "shape" the distribution and make them differ from the Gaussian

distribution. Even if we find a general distribution fitting the data of most inland, the parameters of the distribution are unlikely to be constant over all sites and climatic region. Therefore, it will not exclude intense monitoring for adjusting the parameters of the distribution. For reservoir, it is even more complicated since distribution (and their parameters) might change significantly over time with the decrease of emissions with age of the reservoirs since these systems are not at steady state.

ANSWER TO REVIEWER #3

The authors thanks the reviewer for his positive comments on the manuscript

R#3 comment :"I still have some minor concern about the MS in its discussion section. Firstly, authors did not compare their results comprehensively with other studies all over the world. E.g., the diffusive emission from the surface was high or low? Did your results were fallen in the range of emission rates from other studies? The possible reason?"

Answer : At the beginning of the section 4.4, we added a few lines where we compared emissions from the NT2 Reservoir with some other reservoirs in the tropics as follow : "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., 2015 for a detailed discussion)".

R#3 Secondly, for the hotspots, as we know, turbine and water-logged drawdown areas are regarded as the hotspots of hydroelectric reservoirs. Please give some com- parisons with their contribution to the total emission with inflow waters' and highlight how important about this hotspot from the inflow water.

Answer : In order to fulfil the reviewer comment we added the table 3 and the section 4.4 was slightly modified as follow : " 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. (2015))."

We also determined emissions from the drawdown area during this study but they are not included in the new table. The results are reported in another manuscript under review. Basically, despite a very large surface area the emissions from the drawdown area are less than 3% of the total emissions from this reservoir.

Effect of sporadic destratification, seasonal overturn and artificial mixing on CH_4 emissions at the surface of a subtropical hydroelectric reservoir (Nam Theun 2 Reservoir, Lao PDR)

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Abstract

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 CH_4 concentrations and physico-chemical 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. (2015).

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.

Frédéric Guérin 11/12/y 17:45
Supprimé: also

Frédéric Guérin 11/12/y 17:46
Supprimé: calculated

Frédéric Guérin 24/9/y 10:02
Supprimé: Based on our fortnightly monitoring, we determined that accurate estimation of the emissions can be determined from measurements made at least at a monthly frequency.

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

463 fluxes at the air-water interface and they overlook emissions from the rivers downstream of
464 the dams (Abril et al., 2005;Guerin et al., 2006;Kemenes et al., 2007;Teodoru et al.,
465 2012;Maeck et al., 2013;Deshmukh et al., 2015), CH_4 ebullition (DelSontro et al.,
466 2010;Deshmukh et al., 2014) and emissions from the drawdown area of reservoirs (Chen et
467 al., 2009;Chen et al., 2011) although these pathways could largely dominate diffusion at the
468 surface of the reservoirs.

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

489 The spatial and temporal variability of CH_4 ebullitive fluxes were intensively studied at the
490 newly flooded subtropical Nam Theun 2 Reservoir (NT2R) (Deshmukh et al., 2014) and
491 downstream as well as total CH_4 emissions are described in Deshmukh et al. (2015). The
492 objective of the present study is to quantify the CH_4 diffusive fluxes at the surface of NT2R.
493 The CH_4 emissions were quantified fortnightly during three and a half year (May 2010 to
494 December 2012) based on a monitoring of CH_4 concentrations that started in June 2009. This
495 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 physico-chemical parameters of the water column and the CH_4 concentrations. 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

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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 poisoned (Guerin and Abril, 2007). Before gas chromatography analysis for CH_4 concentration, a N_2 headspace was created and the vials were vigorously shaken to ensure an equilibration between the liquid and gas phases. The

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Supprimé: Surface and deep-water samples for CH_4 concentration were taken with a surface water sampler

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Supprimé: and a Uwitec water sampler, respectively.

559 concentration in the water was calculated using the solubility coefficient of Yamamoto et al.
560 (1976).

561 **2.3.3. Gas chromatography**

562 Analysis of CH_4 concentrations were performed by gas chromatography (SRI 8610C gas
563 chromatograph, Torrance, CA, USA) equipped with a flame ionization detector. A subsample
564 of 0.5 ml from the headspace of water sample vials was injected. Commercial gas standards
565 (10, 100 and 1010 ppmv, Air Liquid "crystal" standards) were injected after analysis of every
566 10 samples for calibration. Duplicate injection of samples showed reproducibility better than
567 5%.

568 **2.4. Water column CH_4 storage**

569 Between two sampling depth of the vertical profiles of CH_4 concentrations, the CH_4
570 concentrations were assumed to change linearly in order to calculate the concentration in each
571 1-m layer of water. The volume of water in each layer was calculated using the volume-
572 capacity curve (NTPC, 2005). The CH_4 storage was calculated by multiplying the average
573 CH_4 concentrations of each layer by the volume of the layer and summing-up the amount of
574 CH_4 for all depth intervals.

575 **2.5. Aerobic CH_4 oxidation**

576 The depth-integrated CH_4 oxidation rates at each station were calculated on the basis of the
577 specific oxidation rates (d^{-1}) determined at NT2 (Deshmukh et al., 2015) and the vertical
578 profiles of CH_4 and O_2 concentrations in the water column as already described in (Guerin
579 and Abril, 2007). The depth-integrated CH_4 oxidation rates at each station were estimated
580 only from January 2010 since the vertical resolution of the vertical profiles of O_2 and CH_4
581 was not high enough in 2009.

582 **2.6. Estimation of diffusive fluxes from surface concentrations**

583 The diffusive CH_4 fluxes were calculated from the fortnightly monitoring of surface
584 concentrations with the thin boundary layer (TBL) equation at all stations in the reservoir
585 (RES1-9). The CH_4 surface concentrations in water and the average CH_4 concentration in air
586 (1.9 ppmv) obtained during eddy covariance deployments (Deshmukh et al., 2014) were
587 applied in equation (1) to calculate diffusive flux:

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

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 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

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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.

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2.7. Extrapolation of fluxes for the estimation of the NT2 total emissions

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

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(third order moment), the auto-correlation of each time series and the correlation between the different stations. All analyses were performed using Matlab.

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3. Results

3.1. Temperature and O_2 dynamics in the reservoir water column

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.

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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 and storage in the reservoir water column

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).

3.3. Interannual variations of the CH_4 concentrations and storage in the reservoir water column

During the three and a half years of monitoring, the same seasonal pattern 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.

3.4. 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.

3.5. 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^{-}

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782 ^{1}). At the stations RES2 and RES3, the concentrations up to 128 µmol L^{-1} were mostly
783 observed in the CD season when the reservoir overturns. At the station RES4 located at the
784 Nam Xot and Nam Theun confluence and at the stations RES7 and RES8 both located in the
785 inflow region of the Nam Theun River, the high surface concentrations (up to 64.60 µmol L^{-1})
786 were mostly observed during the WW season when the reservoir undergoes sporadic
787 destratification. The auto-correlation function of the time series of the log of the surface CH_4
788 concentrations and diffusive fluxes at the stations RES1-8 indicated that at all stations (except
789 RES1) have a memory effect of 30 to 40 days (Figure S1). This implies that with a sampling
790 frequency of 15 days, we captured most of the changes in the surface CH_4 concentrations. At
791 the station RES1, the changes in CH_4 concentrations are faster than at other stations and
792 would have deserved a monitoring with a frequency higher than 15 days.

793 During the monitoring at RES1-RES8 stations, the average diffusive flux was 2.8 ± 12.2
794 mmol m^{-2} d^{-1} ranging from 0.01 to 201.86 mmol m^{-2} d^{-1} without any clear interannual and
795 seasonal trends (Figure S2b). As for the concentrations, flux data followed a loglogistic
796 distribution. The median flux in the WD season is 40 to 80% higher than the median in the
797 WW and CD season, respectively. However, the average fluxes in the WW and CD season are
798 30% higher than in the WD season (Table 2). This confirms the presence of extremely high
799 values during WD and CD seasons, as expected from the surface concentrations. All seasons
800 together, around 7% of the diffusive fluxes that we observed were higher than 5 mmol m^{-2} d^{-1}
801 which corresponds to extremely high diffusive fluxes in comparison with data from the
802 literature for reservoirs and lakes (Bastviken et al., 2008;Barros et al., 2011). The median and
803 average of these extreme fluxes higher than 5 mmol m^{-2} d^{-1} were 2 times higher in the WW
804 and CD seasons than in the WD season (Table 2).

805 At NT2, diffusive CH_4 fluxes covered the whole range of fluxes reported for tropical
806 reservoirs, depending on the season. Most of the fluxes at the NT2R Reservoir were around
807 one order of magnitude lower than the ones at Petit Saut Reservoir (French Guiana) just after
808 the impoundment (Galy-Lacaux et al., 1997), and in the same order of magnitude as reported
809 for reservoirs older by 10 to 18 years (Abril et al., 2005;Guerin et al., 2006;Kemenes et al.,
810 2007;Chanudet et al., 2011). However, some diffusive fluxes at the stations RES1-8 in the
811 WW and the CD seasons (up to 202 mmol m^{-2} d^{-1}) are among the highest ever reported at the
812 surface of a hydroelectric reservoir or a lake (Bastviken et al., 2011;Barros et al., 2011) and
813 rivers downstream of dams (Abril et al., 2005;Guerin et al., 2006;Deshmukh et al., 2015).

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3.6. Surface methane concentrations and diffusive fluxes at the water intake (RES9)

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

The diffusive fluxes ranged between 0.03 and 605.38 mmol m^{-2} d^{-1} (Figure 6b and Table 2). On average, the CH_4 diffusive fluxes at RES9 were two to forty times higher than at the other stations in the CD, WD and WW season. Diffusive fluxes at this station are usually higher than 10 mmol m^{-2} d^{-1} from April to July that corresponds to the WD season and the very beginning of the WW season. In 2010, diffusive fluxes were on average 241 ± 219 and 239 ± 228 mmol m^{-2} d^{-1} respectively for the WD and WW seasons. In 2011 and 2012, the fluxes 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., 2015).

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., 2015).

849 CH_4 concentrations and storage increase concomitantly with the surface water temperature
850 and the establishment of the thermal stratification during the WD season and peak at the end
851 of the WD season-beginning of the WW season (Figure 2 and 3). During the WW season,
852 CH_4 concentrations and storage decrease slowly (Figure 3) while aerobic methane oxidation
853 reaches its maximum (Figure 4). When the reservoir overturns at the beginning of the CD
854 season, the CH_4 hypolimnic concentrations and storage reach their minima (Figure 3). The
855 overturn favours the penetration of oxygen down to the bottom (Figure 2 and 3b). The sharp
856 decrease of the CH_4 concentrations and CH_4 storage during this period is expected to result
857 from sudden outgassing (Section 4.2) together with an enhancement of the aerobic CH_4
858 oxidation as already observed in lakes that overturn (Utsumi et al., 1998b;Utsumi et al.,
859 1998a;Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al.,
860 2012;Fernández et al., 2014). A large increase of the aerobic methane oxidation was only
861 observed in the CD season in the dry year 2012 (Figure 4) because the amount of hypolimnic
862 CH_4 to be oxidized at the beginning of the CD season was still high in the water column
863 (Figure 3c,d).

864 As the reservoir overturns during the period over which the water residence time is the longest
865 in the reservoir, the temporal evolution of the concentrations is anti-correlated with the
866 residence time (Figure 3c,d). The seasonal dynamics of the CH_4 in the monomictic NT2R
867 differs from permanently stratified reservoirs like Petit Saut Reservoir where CH_4
868 concentration increased with retention time (Abril et al., 2005). However, at the annual scale
869 the water residence time has a strong influence on CH_4 concentration and storage in the
870 reservoir. Before the reservoir was commissioned (April 2010), the water residence time was
871 up to 4 years and the CH_4 storage was up to four times higher than in 2011 and 2012 (Figure
872 3d). Although a decrease of concentration and storage with the age of the reservoir was
873 expected (Abril et al., 2005), the storage in the dry year 2012 was twice higher than in the wet
874 year 2011 due to an increase of the water residence time by 25% between 2011 and 2012. In
875 wet years like 2011, the thermal stratification is weaker than in dry years since the warming of
876 surface water is less efficient and the high water inputs alters the stability of the reservoir
877 thermal stratification as shown by the sharper decrease and the larger range of ΔT in 2011
878 than in 2012 (Figure 3a). As a consequence, the oxygen diffusion to the hypolimnion was
879 higher in 2011 than in 2012 (Figure 3b) and it enhanced aerobic methane oxidation by 20% in
880 the water column in the WW season in 2011 as compared to 2012 (Figure 4). It therefore
881 suggests that the hydrology affects both the thermal stratification and the hypolimnic storage

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of CH_4 in reservoirs, indirectly controls aerobic methane oxidation, and ultimately influences emissions.

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

We therefore confirm the occurrence of hot moments of emissions during the reservoir overturn in the CD season as already observed in lakes that overturn in temperate regions (Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al., 2012;Fernández et al., 2014). The highest emissions determined at NT2R are one order of magnitude higher than previously reported outgassing during overturn and they occur mostly in the section of the reservoir that has the longest water residence time (RES1-3, Table 1) and the largest CH_4 storage (Figure 7b,e,h,k). This suggests that the impact of reservoir overturn can be very critical for the whole-reservoir CH_4 budget in tropical hydroelectric reservoirs and especially in young ones where hypolimnic concentration could reach up to 1000 µmol L^{-1}. Hot moments of emissions also occur during sporadic destratifications in the WW season in the inflow region (RES4 and RES6-8) where the inflow of cool water from the watershed might disrupt the thermal stratification in reservoirs. This is contrasting with the observations in older reservoir than NT2R where high emissions from the inflow region were recently attributed to an enhancement of CH_4 production fuelled by the sedimentation of organic

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matter from the watershed (Musenze et al., 2014). The high emissions in the WD seasons were associated with early rains and associated high winds that occur sometimes in the last fifteen days of May. This shows that a moderate erosion of the stratification when hypolimnic CH_4 concentrations are high could enhance vertical transport of CH_4 toward the surface and emissions to the atmosphere. Basically, this intense monitoring shows that spatial and temporal variations of CH_4 emissions are largely controlled by the hydrodynamics of the reservoir with extreme emissions occurring mostly in the inflow region during the wet season and mostly in area remotely located from the inflow zone and the riverbed during reservoir overturns in the CD season. Even if less frequent, moderate erosion of the stable and steep thermal stratification during warm seasons, could also lead to high emissions.

The evolution of depth-integrated aerobic CH_4 oxidation is not clearly related with the reservoir overturns and the CH_4 burst (Figure 7). Significant increases in the aerobic CH_4 oxidation occurred mostly during the first half of the WD season when the stratification was unstable and at the very beginning of the destratification in the WW, when ΔT started to decrease. The oxidation could reach high values (up to 380 mmol m^{-2} d^{-1}) during these two periods since the yield of CH_4 in the water column to sustain the activity of methanotrophs is higher than in the CD season when the reservoir overturns. It shows that in reservoirs or lakes like NT2R that destratify progressively before the overturn, there is no substantial increase of the CH_4 oxidation when the water body overturns as it could be observed in lakes that overturn within a few days (Kankaala et al., 2007). In addition, the contribution of CH_4 oxidation to the total loss of CH_4 (sum of diffusion and oxidation) in the WD and WW seasons was 90-95% during the entire monitoring whereas it was 85% in the CD season. During overturns, a significant amount of CH_4 is oxidized (Utsumi et al., 1998a;Utsumi et al., 1998b;Kankaala et al., 2007;Schubert et al., 2012) but it also indicates that the removal of CH_4 during overturn is not as efficient as during seasons with a well established thermal stratification.

During the periods with major loss in the CH_4 storage with concomitant CH_4 burst, we compared the change in the yield of CH_4 with the sum of emissions and oxidation. Most of the time, the emissions alone and/or the sum of emissions and oxidation were significantly higher than the amount of CH_4 that was lost from the water column. At the Pääjäri Lake in Finland (López Bellido et al., 2009), the fact that measured or calculated emissions exceed the loss of CH_4 in the water column was attributed to a probable underestimation of the CH_4

storage in the lake by under-sampling the shallow area of the lake. In this study, emissions, storage and oxidation were estimated at the same stations, avoiding such sampling artefacts. Therefore, it suggests that CH_4 is provided by lateral transport or by production in the flooded soil and biomass (Guerin et al., 2008) at a higher rate than the total loss of CH_4 from the water column by emissions and oxidation. This hypothesis could only be verified by a full CH_4 mass balance including production and total emissions from the reservoir, which is beyond the scope of this article.

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

After the commissioning of the reservoir, the temperature and the oxygen and CH_4 concentrations were constant from the surface to the bottom of the reservoir at the vicinity of the water intake. On the basis of physical modelling and measurements of water current velocities (Chanudet et al., 2012), the vertical mixing at this station was attributed to the water withdrawal at the intake generating turbulence and water currents over a surface area of 3 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.

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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., 2015 for a detailed discussion).

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. (2015)). 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

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

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

Acknowledgements

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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[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 residence time in arbitrary units, (***) stands for long residence time, (**) for intermediate residence times and (*) for short residence times

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

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Supprimé: mmol m^{-2} d^{-1}

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

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

[1]Deshmukh et al. (2014)
[2]Deshmukh et al. (2015)

Figure captions

Figure 1: Map of the sampling stations and civil structures at the Nam Theun 2 Reservoir (Lao PDR).

Figure 2: Vertical profiles of temperature (°C), oxygen (µmol L^{-1}) and methane (µmol L^{-1}) at the stations RES1, RES3, RES7, RES8 and RES9 in the Nam Theun 2 Reservoir. Representative profile of the years 2010 (circle), 2011 (square) and 2012 (triangle) are given for each seasons: cool dry in blue, warm dry in red, and warm wet in grey.

Figure 3: (a) Stratification index (ΔT, see text), (b) O_2 concentration in the hypolimnion (µmol L^{-1}), (c) CH_4 concentration in the hypolimnion (µmol L^{-1}) and (d) CH_4 storage in the water column ($Gg(CH_4)$ $month^{-1}$, bars) and water residence time (days, black line with circles) in the Nam Theun 2 Reservoir (Lao PDR) between 2009 and 2012. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool dry (CD) seasons, respectively. For the panels (a), (b) and (c), the boxes show the median and the interquartile range, the whiskers denote the full range of values and the plus sign (+) denotes the mean.

Figure 4: Seasonal variations between 2010 and 2012 of the depth-integrated aerobic CH_4 oxidation (mmol m^{-2} d^{-1}) at the stations RES1-RES8 calculated from the aerobic oxidation rates determined by Deshmukh et al. (2015). WD stands for warm dry (in red), WW for warm wet (in grey) and CD for cool dry (in blue). The boxes show the median and the interquartile range, the whiskers denote the full range of values and the plus sign (+) denotes the mean.

Figure 5: Frequency distribution of the log of CH_4 concentrations (µmol L^{-1}) at the nine monitoring stations of the Nam Theun 2 Reservoir. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool dry (CD) seasons, respectively.

Figure 6: (a) Surface concentrations and (b) diffusive fluxes between June 2009 and December 2012 at the station RES9 located at the water intake. Julian day 0 is 1st of January, 2009. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool 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. (2015) 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.

Figure 1

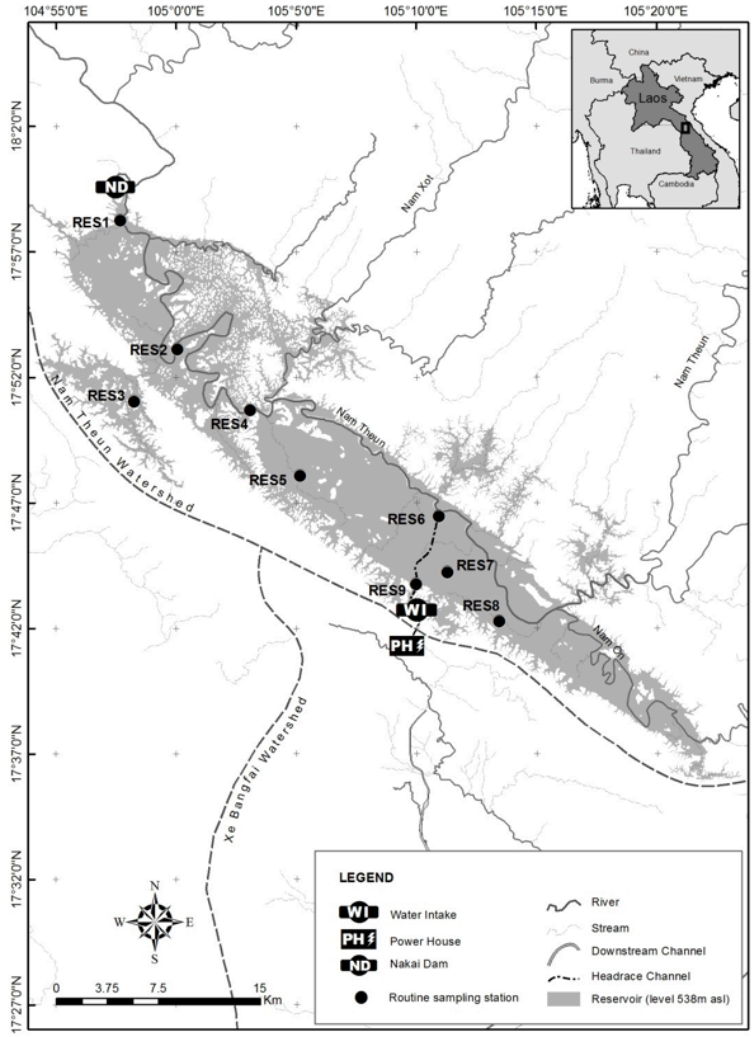

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Figure 2

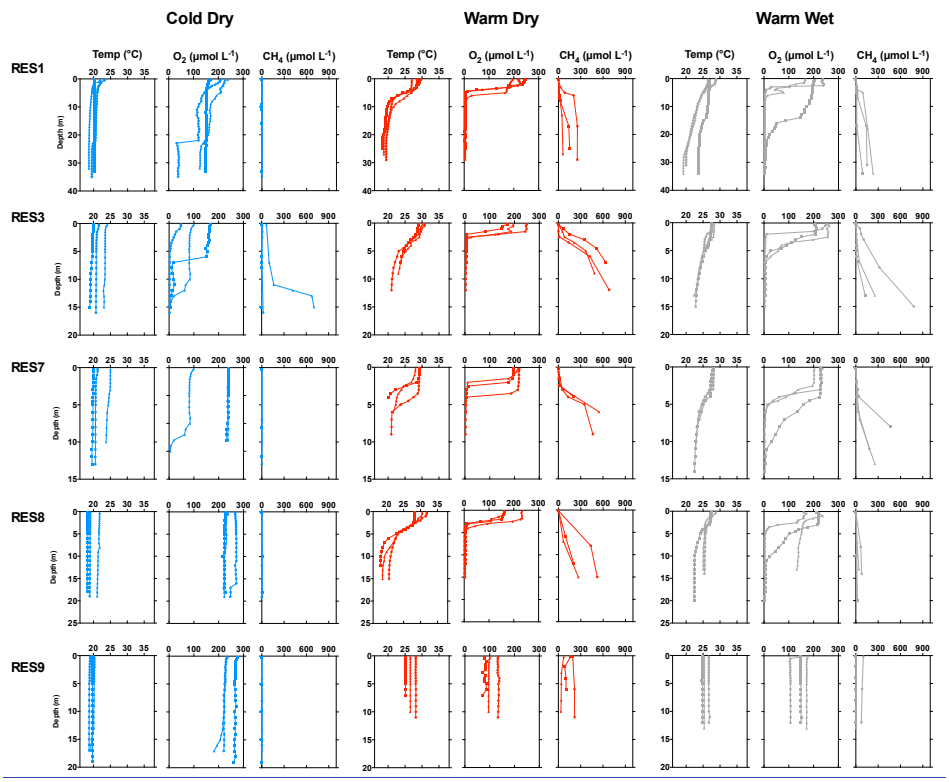

Cold Dry
Warm Dry
Warm Wet
Temp (°C)
O2 (µmol L-1)
CH4 (µmol L-1)
RES1
RES3
RES7
RES8
RES9
Depth (m)

Figure 3

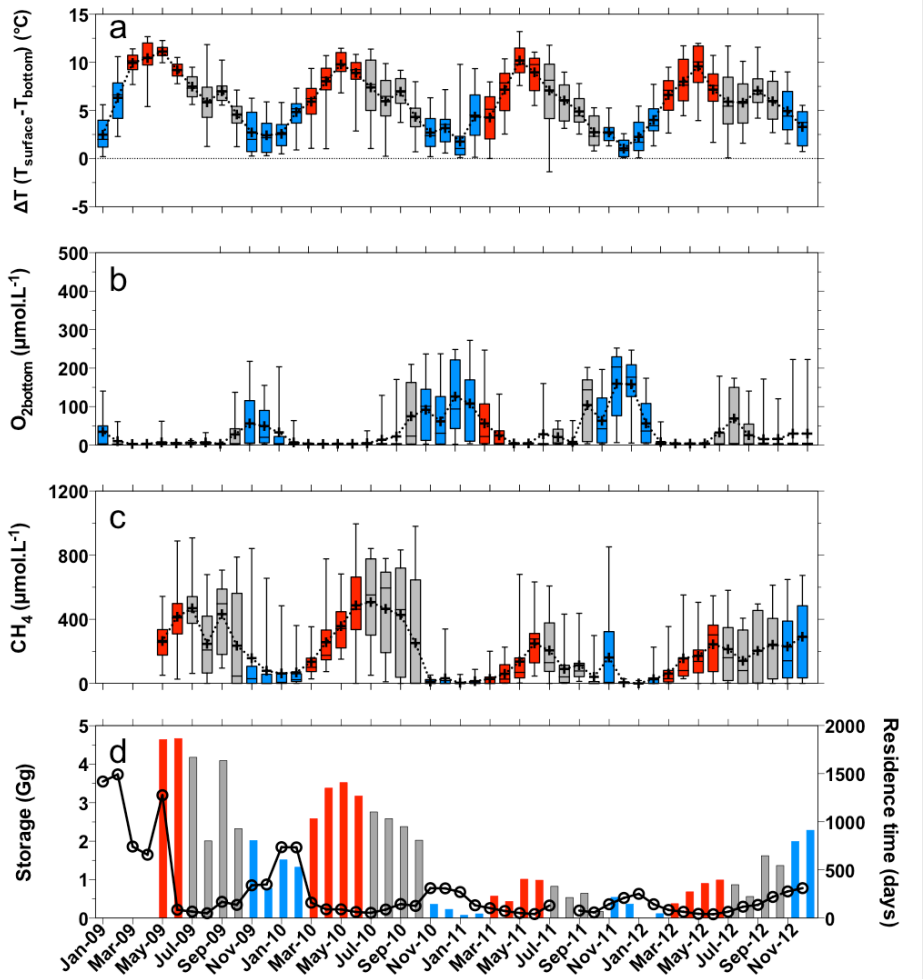

a
b
c
d
ΔT (T surface -T bottom) (°C)
O2bottom (µmol.L-1)
CH4 (µmol.L-1)
Storage (Gg)
Residence time (days)
Jan-09
Mar-09
May-09
Jul-09
Sep-09
Nov-09
Jan-10
Mar-10
May-10
Jul-10
Sep-10
Nov-10
Jan-11
Mar-11
May-11
Jul-11
Sep-11
Nov-11
Jan-12
Mar-12
May-12
Jul-12
Sep-12
Nov-12

Figure 4

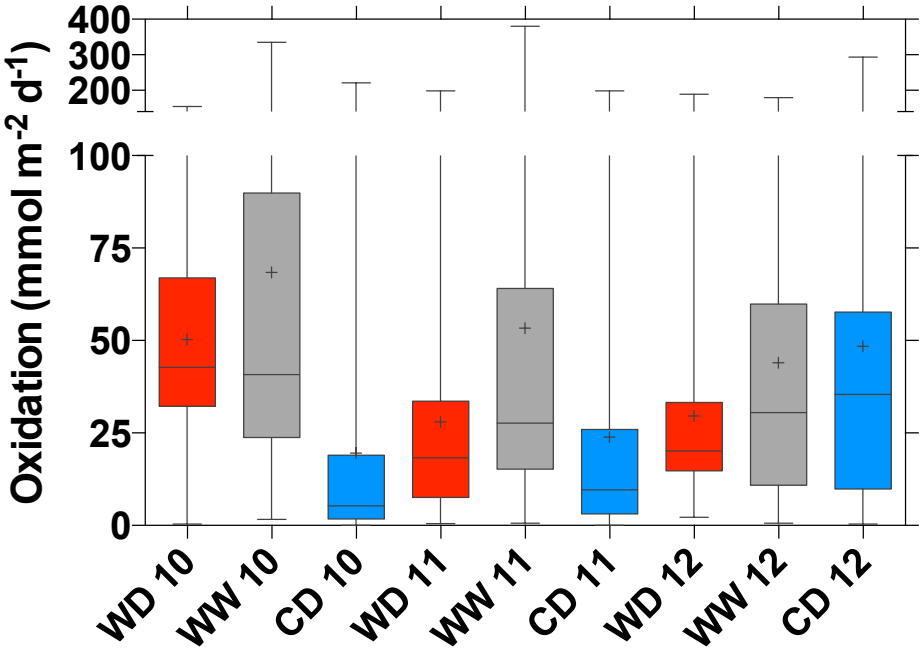
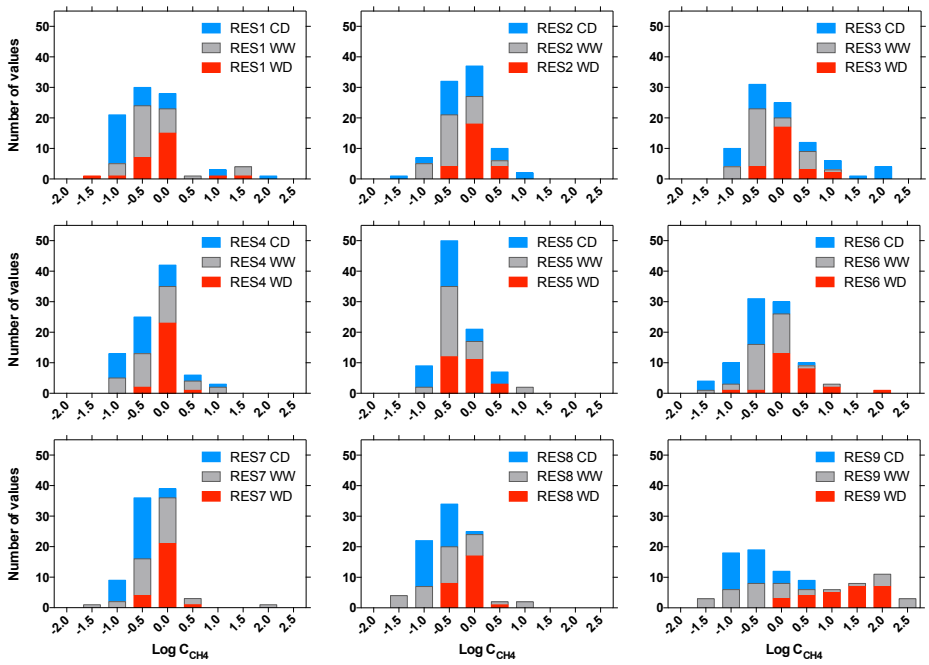
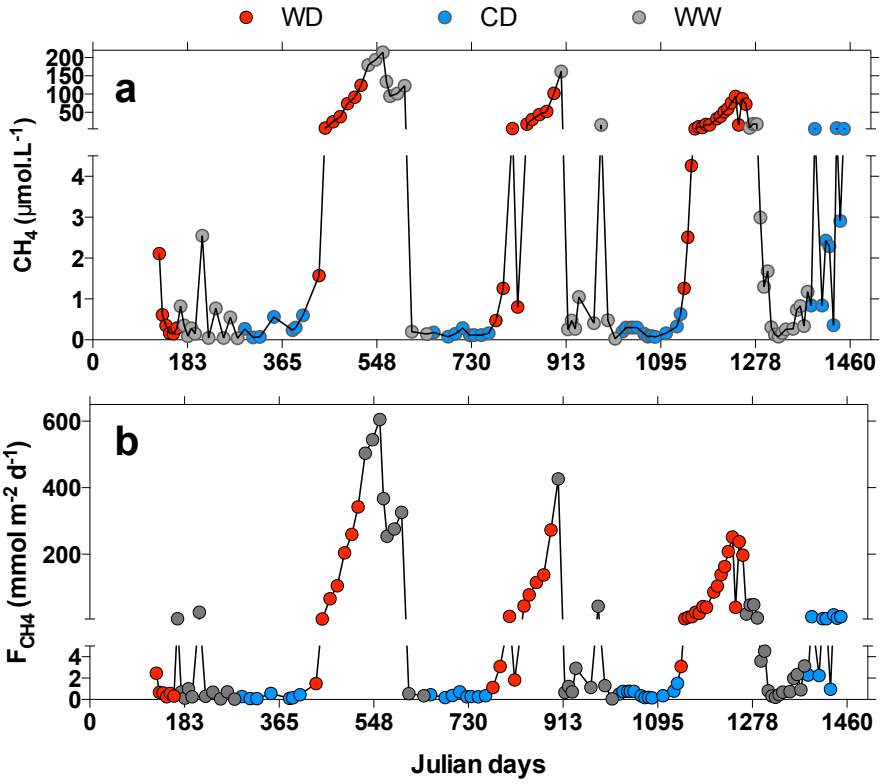

Oxidation (mmol m-2 d-1)
400
300
200
100
75
50
25
0
WD 10
WW 10
CD 10
WD 11
WW 11
CD 11
WD 12
WW 12
CD 12

Figure 5

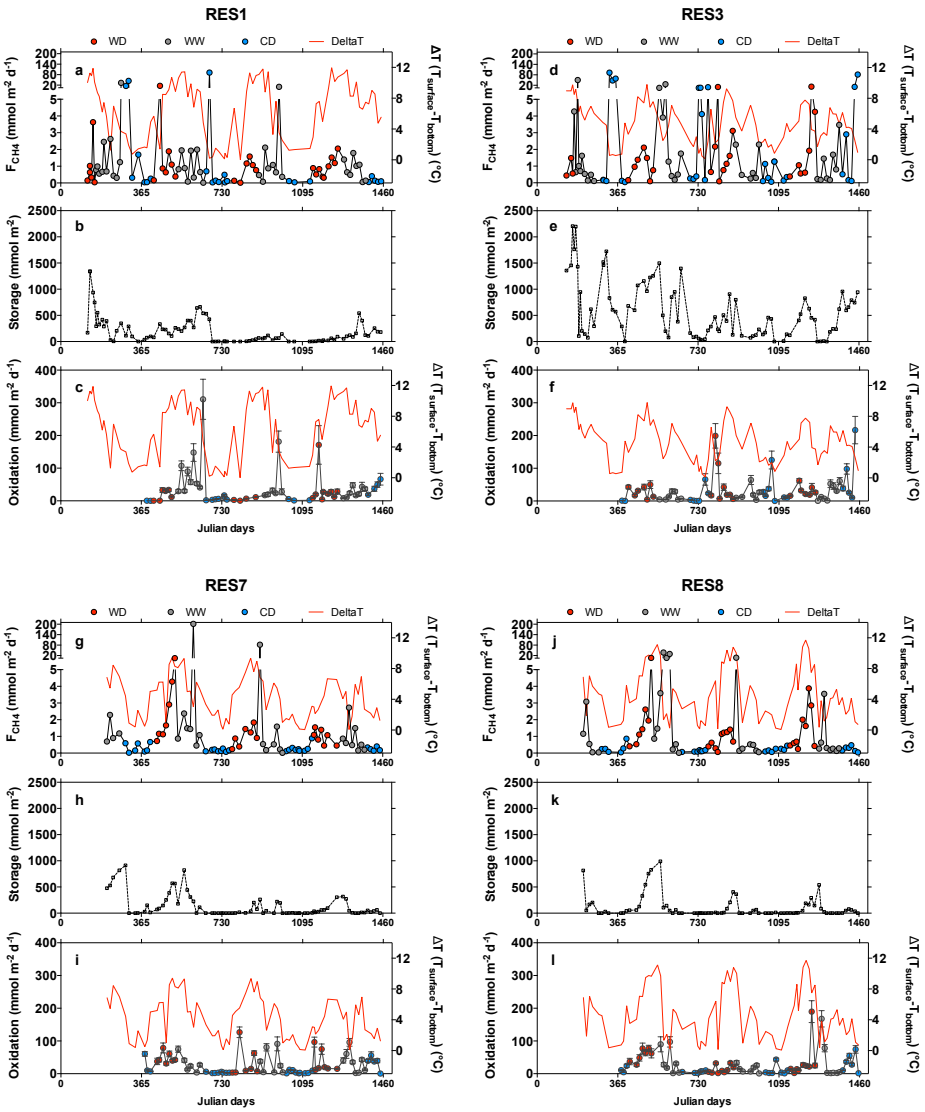

RES1 CD
RES1 WW
RES1 WD
RES2 CD
RES2 WW
RES2 WD
RES3 CD
RES3 WW
RES3 WD
RES4 CD
RES4 WW
RES4 WD
RES5 CD
RES5 WW
RES5 WD
RES6 CD
RES6 WW
RES6 WD
RES7 CD
RES7 WW
RES7 WD
RES8 CD
RES8 WW
RES8 WD
RES9 CD
RES9 WW
RES9 WD
Number of values
Log C_{CH4}

1361
1362 Figure 6
1363

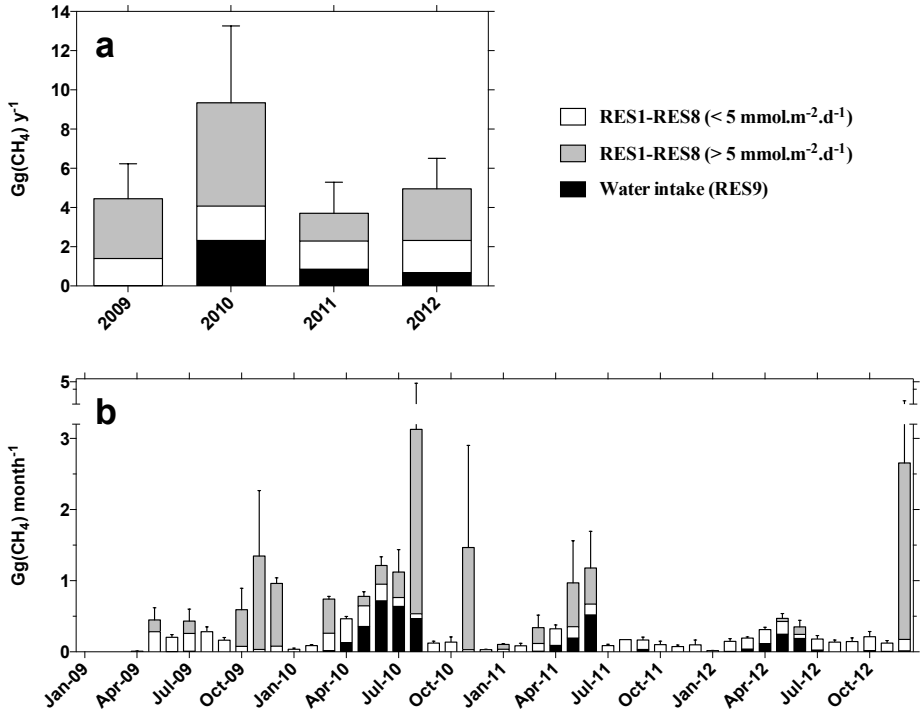


1364
1365
1366

Figure 7


RES1
RES3
RES7
RES8
WD
WW
CD
DeltaT
a
b
c
d
e
f
g
h
i
j
k
l
Julian days
Storage (mmol m-2)
ΔT (Tsurface-Tbottom) (°C)

1373
1374 Figure 8
1375



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