Dear Editor,

We have now revised our manuscript entitled " Low methane (CH4) emissions downstream of a monomictic subtropical hydroelectric reservoir (Nam Theun 2, Lao PDR)", by Deshmukh and co-workers, submitted for publication to Biogeosciences.

We have carefully considered all the comments by the two reviewers. We accepted most of them and provided clear rebuttal when we believed our assumption were the most adapted to the environmental conditions.

As requested, a copy of the manuscript with Word "track changes" is provided below.

Sincerely,

Frédéric Guérin

REVIEW BY DAMIEN MAHER

The authors thank Damien Maher for his comprehensive and positive review of the manuscript.

RC : A few equations on how various fluxes were calculated would be welcomed – while I am familiar with the floating dome calculations other readers of BG might not be, also some equations on the "degassing" calculations (I am assuming these are a simple mass balance?) would also be good. Also a little bit more on the analysis precision and accuracy would be appreciated (other than just the 5% reproducibility).

ANSWER : Equation for the calculation of diffusive flux from surface concentrations and equation for degassing were added in the sections 2.41 and 2.4.2, respectively.

In the section 2.3.4, the text was modified as follows: "The detection limit is 0.3 ppmv in the headspace and the accuracy is around 4% allowing the determination of nanomolar

concentrations in water samples, depending of the volume of the vials and headspace. Duplicate injection of samples showed reproducibility better than 5%."

RC : My main issue is with the use of a single "averaged" k600 value for the downstream section of the study area, I feel this simplistic approach is not appropriate as the k600 is likely much higher in the immediate downstream area (and also concentrations are likely to be much higher here) therefore there would be an underestimation of the fluxes. Indeed 10 cm/h seems much too low for an area that would have extremely high turbulence, as noted by the authors in the comment on not being able to do floating chamber measurements due to "safety reasons because of strong water currents". Is there any other way of estimating this? Considering the flow is so highly regulated perhaps a similar method can be employed as used for the "degassing" calculations, essentially a CH4 mass balance between an upstream and downstream point. At least some dissussion about the implications of using the single value for gas transfer velocity should be included.

ANSWER: The zone of high turbulence that leads the Nam Theun Power Company to forbid navigation are located immediately downstream of the dam, downstream of the power house and downstream of the aeration weir where there are "artificial waterfall" where degassing occur. Otherwise, the water current velocity in the artificial channel never exceeds 1 m/s and averaged 0.5 m/s. Therefore "safety reasons because of strong water currents" will be rephrased to better describe the sampling conditions. These dangerous areas correspond to the "immediate downstream areas" where degassing was calculated and therefore no k600 was considered and emissions are determined by "mass balance" (see section 2.4.2).

For the other sections where degassing does not occur, a simple mass balance between the upstream and downstream points would lead to an overestimation of the emissions since a fraction of the CH4 would be oxidized (data not show but the specific oxidation rate obtained in the articicial channel is now given in the section 3.4). Modelling is needed in order to take into account both oxidation and diffusion.

As said in the MS and in Guerin et al (2015), the chamber deployment performed in rivers in the watershed gave an average k600 of 10 cm/h. This is very similar to the average k600 value obtained using the formulation k600-wind speed relationships from Guerin et al (2007) obtained downstream of the Petit Saut Reservoir and in small estuaries of the same size with similar water currents like the Scheldt (Borges et al., 2004). We therefore kept 10 cm h-1 as a conservative estimate of the k600 in the artificial channel downstream of the NT2R. k600 was kept constant over the whole period of monitoring since the average of the results obtained by the formulations of Borges et al (2004) and Guerin et al (2007) was 10.06 ± 1.48 cm h-1 according to the limited variation of the monthly average wind speed (1.8 ± 0.46 m s-1).

The section 2.4.1 was modified according to the comments above. We believe that our hypothesis is reliable and its consequences in the methane balance are minor. According to the results presented in table 2, even if we underestimated or overestimated the k600 by 50%, still diffusion would not contribute more than 4% to the total CH4 emissions from the Nam Theun 2 reservoir.

Minor Comments

RC: P 11324 L8-10 Comparisons like this belong in discussion

P11324 L25 – P11325 L2 Discussion

P11325 L9-12 Discussion

ANSWER: The discussion deals with the spatial and temporal variability and the significance of downstream emissions in absolute values (in $Mg(CH_4)$ $month^{-1}$, for instance) and almost no data that could be compared with other studies (like concentrations or diffusive fluxes) are included. Therefore we kept comparison of our dataset with other studies in the result section.

RC : P11325 L13-16 How was the 10km length defined? Is this based on any modelling or just best guess? Some explanation on how this value was calculated should be included

ANSWER : As mentionned line 6-7 (same page) and L20-25 of the previous page, this is based on the measurements at the NTH3 station located immediately downstream of the dam and on the measurements at the NTH4 station located 10 km downstream of the dam. At NTH4, CH4 concentrations and calculated fluxes were always very similar to pristine rivers in the watershed.

The text was slightly modified as follow : "Downstream the station NTH4 located 10 kilometres downstream of the dam, the CH4 emission was similar to what found in pristine river of the watershed and it was 2 orders of magnitude lower than the emissions observed downstream of 10-20 years-old reservoirs (Guérin et al., 2006;Kemenes et al., 2007).

Considering that the CH4 emissions from the Nam Theun River below the dam can be attributed to the reservoir over a maximum length of 10 km and a constant width of 30 m, annual emissions below the Nakai Dam decreased from 20 to 1 Mg-CH4 month-1 between 2009 and 2012, respectively (Figure 3c)."

RC : P11326 L24-25 Maybe a narrow range considering the max of □ 1000 uM but this is still 3 orders of magnitude difference. Perhaps look for different terminology than “narrow range”

ANSWER : The sentence was rewritten as follow : " Whatever the years, in the CD season, surface CH_4 concentrations was lower than 14.5 μmol L^{-1} along the 30 km long watercourse.”

RC: P11328 L5 – 10 Discussion

ANSWER: as mentioned before, these comparisons of our dataset with other studies are kept in the result section.

RC : P11329 L20-23 Give details on how this depth integrated value was calculated in the methods (including equations)

ANSWER : The following text was added in the section 2.3.3

"The kinetics parameters of aerobic methane oxidation obtained from the experiment were combined to the in situ CH4 concentration profiles in order to calculate the integrated aerobic methane oxidation in the oxic water column. As the aerobic methane oxidation rates we obtained were potential, CH4-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 (CH4-ox, mmol.m-2.d-1) is:

$$CH4\text{-}ox = CCH4 \cdot SCH4\text{-}ox \cdot CO2/ (CO2 + Km (O2)) \cdot d \cdot I(z)$$

with CCH4, the CH4 concentration; SCH4-ox, the specific CH4-ox; CO2, the oxygen concentration; Km(O2), the Km of O2 for CH4 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 CH4 oxidation rates were integrated in the oxic water column, from the water surface to the limit of penetration of oxygen."

The sentence pointed out by the reviewer was removed since all explanation is now given in the above-mentioned section.

REVIEW BY REVIEWER#2

The authors want to thank the reviewer for his careful reading of the manuscript and for his positive comments.

RC: The sampling program was designed such that downstream emissions could be partitioned among different features of the system including turbine discharge, aeration pools, and river channels. While it is informative to understand the spatial distribution of downstream emissions, this approach does introduce some complications to the analysis. Specifically, to estimate emissions from the downstream flowing waters the authors needed to make assumptions regarding the air-water gas exchange rate. While this is not a major problem, it does cause the reader to wonder about the accuracy of these estimates, particularly for the section immediately downstream of the turbines where the water was too turbulent to allow for chamber deployments. I suggest the authors also estimate downstream emissions by assuming that all CH4 in excess of atmospheric equilibrium that leaves the reservoir is emitted to the atmosphere.

downstream emissions = [CH4,obs – CH4,eq]Q

where CH4 is the dissolved CH4 concentration that was measured (CH4,obs) and at atmospheric equilibrium (CH4,eq), Q is the rate of water withdrawal from the reservoir. This would provide an upper bound to the downstream emission estimate (i.e. assumes no CH4 oxidation in downstream waters).

ANSWER: In the downstream channel, oxidation occurs and it is significant (specific oxidation rate of 1.5 d-1, now given in the section 3.4). In addition, the CH4 concentration never decreased below 0.11 μmol L^{-1} which is 40 times higher than the concentration at equilibrium. Therefore, the proposed method would lead to a significant overestimation of downstream emissions. The same equation was used for degassing (equation now given in the section 2.4.2) but we used the difference in concentration between upstream and downstream of the "degassing structure". For the assumption about the gas transfer velocity, see the detailed answer to the second comment of Damien Maher.

RC: The authors conclude "The hydrodynamics but also the water residence time significantly impact downstream emissions and must be taken into account for future estimation of total emissions from hydroelectric reservoirs at the global scale". While this is no doubt true, I would like to see a deeper discussion of how we might go about doing this. I very much like the related

discussion on page 11333 (lines 1-15) which suggest that the mixing status of a reservoir is an indicator of potential downstream CH4 emissions. Are there other readily accessible data that can be used in emission inventory guidelines to better estimate downstream emissions? Certainly, downstream emissions scale with discharge, as discussed in page 11331, lines 4-9. Should we recommend that downstream emissions be estimated as a function of reservoir discharge? What about details of the intake structure? I wonder if the discussion of this topic in section 4.3 can be expanded upon. For example, some dams can only withdraw from the hypolimnion, while others can withdraw from multiple depths, including the epilimnion. It seems withdrawal depth is another important factor determining the magnitude of downstream emissions. Overall, I think this paper would be more impactful if it not only said we should estimate downstream emissions in global inventories, but also provided a framework for how we should go about it.

ANSWER: all the parameters that might significantly influence downstream emissions are spread in the last two sections of the discussion as pointed by the reviewer.

As it would be more impactful to group all them in one place, the following paragraph was added at the end of the conclusion : " On the basis of these results, different from those previously published, we recommend that estimates at the global scale of emissions below dams take into account the mixing status of reservoirs, the water residence time and depth of the water intake and its impact on the oxygenation of the water column immediately upstream of the turbines."

Specific comments

RC: Page 11316, line 23: . . .were first reported. . .

ANSWER: changed

RC: Page 11316, line 25: akward to start a sentence with a list of references. Suggest

rephrasing.

ANSWER: The sentence was rephrased as follow: "When all emission pathways from tropical or temperate hydroelectric reservoirs (disregarding the drawdown emissions) are taken into account, downstream emissions could contribute 50 to 90% of total CH_4 emissions (Abril et al., 2005;Kemenes et al., 2007;Maeck et al., 2013)."

RC: Fig. 1: There is a lot going on in this monitoring program. I suggest making this figure as large and clear as possible. Please increase the size of the inset. Consider using colors. I suggest

eliminating the icons used to symbolize the dams, intake structures, etc. They are relatively large and overlap with the sampling locations.

RC: Page 11319, line 26: RES3 and RES7 not included in Fig.1

ANSWER: The map (Figure 1) was modified as suggested by the reviewer and the missing stations were added

RC: Page 11324, line 15: below the dam?

ANSWER: Replaced by "further downstream"

RC: Line 17: Fig 3 cited before Fig 2?

ANSWER: Figure 2 was initially not cited. Now cited in the section 3.1.

RC: Line 20-21: data from NTH4 and NTH5 not shown?

ANSWER: Data from NTH5 are not shown. Data from NTH4 are visible on Fig 3a and 3b as now noted in the text (section 3.2.1 & 3.2.2).

RC: Page 11331, line 2: . . .between X and 1.5. . .

ANSWER: Corrected: …range between 0.2 and 1.5…

Section 4.3: very interesting discussion.

RC: Figs 3 and 4, panel b: y-axis label, "Diffusive emissions (mmol CH4 m-1 d-1)"

ANSWER: Changed by "CH_4 Diffusion mmol m-1 d-1"

RC: Fig. 5. Probably not necessary.

ANSWER: Kept since it is needed for the estimation of the oxidation at the water intake.

Low methane (CH_4) emissions downstream of a monomictic subtropical hydroelectric reservoir (Nam Theun 2, Lao PDR)

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Abstract

Methane (CH_4) emissions from hydroelectric reservoirs could represent a significant fraction of global CH_4 emissions from inland waters and wetlands. Although CH_4 emissions downstream of hydroelectric reservoirs are known to be potentially significant, these emissions are poorly documented in recent studies. We report the first quantification of emissions downstream of a subtropical monomictic reservoir. The Nam Theun 2 Reservoir (NT2R), located in Lao People's

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Democratic Republic, was flooded in 2008 and commissioned in April 2010. This reservoir is a trans-basin diversion reservoir which releases water to two downstream streams: the Nam Theun River below the dam and an artificial channel downstream of the powerhouse and a regulating pond that diverts the water from the Nam Theun watershed to the Xe Bangfai watershed. We quantified downstream emissions during the first four years after impoundment (2009-2012) on the basis of a high temporal (weekly to fortnightly) and spatial (23 stations) resolution of the monitoring of CH_4 concentration.

Before the commissioning of NT2R, downstream emissions were dominated by a very significant degassing at the dam site resulting from the occasional spillway discharge for controlling the water level in the reservoir. After the commissioning, downstream emissions were dominated by degassing which occurred mostly below the powerhouse. Overall, downstream emissions decreased from 10 $GgCH_4\ y^{-1}$ after the commissioning to 2 $GgCH_4\ y^{-1}$ four years after impoundment. The downstream emissions contributed only 10 to 30% of total CH_4 emissions from the reservoir during the study.

Most of the downstream emissions (80%) occurred within 2-4 months during the transition between the warm dry season (WD) and the warm wet season (WW) when the CH_4 concentration in hypolimnic water is maximum (up to 1000 $\mu mol\ L^{-1}$) and downstream emissions are negligible for the rest of the year. Emissions downstream of NT2R are also lower than expected because of the design of the water intake. A significant fraction of the CH_4 that should have been transferred and emitted downstream of the powerhouse is emitted at the reservoir surface because of the artificial turbulence generated around the water intake. The positive counterpart of this artificial mixing is that it allows O_2 diffusion down to the bottom of the water column enhancing aerobic methane oxidation and it subsequently lowering downstream emissions by at least 40%.

1. Introduction

Methane (CH_4) emission from hydroelectric reservoirs at the global scale was recently revised downward and it would represent only 1% of anthropogenic emissions (Barros et al., 2011). This latter estimate is mostly based on CH_4 diffusion at the reservoir surface and in a lesser extent on CH_4 ebullition which are the two best documented pathways to the atmosphere. However, emissions from the drawdown area (Chen et al., 2009;Chen et al., 2011) and emissions

downstream of dams (Galy-Lacaux et al., 1997;Abril et al., 2005;Guérin et al., 2006;Kemenes et al., 2007;Chanudet et al., 2011;Teodoru et al., 2012;Maeck et al., 2013) were poorly studied and are not taken into account in the last global estimate (Barros et al., 2011). Some authors attempted to include these two pathways to the global estimation of greenhouse gas emissions from reservoirs (Lima et al., 2008;Li and Zhang, 2014) and it increased drastically the emission factors of reservoirs.

The downstream emissions include the so-called degassing which occurs just below the turbines. It is attributed to the high turbulence generated by the discharge of the reservoir water into the river below the dam and the large pressure drop that the water undergoes while being transported from the bottom of the reservoir to the surface of the river below the dam. It also includes emissions by diffusion from the river below the dam. Downstream emissions were first reported at the Petit Saut Reservoir (Galy-Lacaux et al., 1997) and this pathway was later confirmed in some Brazilian reservoirs (Guérin et al., 2006;Kemenes et al., 2007). When all emission pathways from tropical or temperate hydroelectric reservoirs (disregarding the drawdown emissions) are taken into account, downstream emissions could contribute 50 to 90% of total CH_4 emissions (Abril et al., 2005;Kemenes et al., 2007;Maeck et al., 2013). At two other sites located in Canada and in Lao People's Democratic Republic (Lao PDR) where this pathway was studied, downstream emissions were found to contribute less than 25% when it exists (Chanudet et al., 2011;Teodoru et al., 2012). According to the differences from one reservoir to the other, it appears that the factors controlling downstream emissions from reservoirs must be identified in order to propose realistic estimations of the global emissions from reservoirs including downstream emissions.

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Supprimé: have shown that downstream emissions would contribute 50 to 90% of total CH_4 emissions from temperate and tropical hydroelectric reservoirs when all emission pathways from reservoirs (disregarding the drawdown emissions) are taken into account.

In the present study, we quantified emissions downstream of the Nam Theun 2 Reservoir (NT2R) located in Lao PDR on the basis of a high temporal (weekly to fortnightly) and spatial (23 stations) resolution monitoring of CH_4 concentration. The significance of the aerobic CH_4 oxidation in the dynamics of CH_4 in the reservoir and the downstream rivers was also evaluated. We characterized the seasonal patterns of downstream emissions and evaluated the contribution of this pathway to CH_4 emissions by ebullition (Deshmukh et al., 2014) and diffusive fluxes at the surface of the reservoir (Guérin et al., 2015). We finally discuss the contribution of downstream emissions according to the reservoir hydrodynamics and the design of the water intake by comparing our results to previously published studies.

2. Material and methods

2.1. Study area

The NT2 hydroelectric Reservoir was built on the Nam Theun River located in the subtropical region of Lao PDR. The NT2 hydroelectric scheme is based on a trans-basin diversion that receives water from the Nam Theun River and releases it into the Xe Bangfai River through a 27 km long artificial downstream channel (Figure 1) (see Descloux et al. (2014) for a detailed description of the study site). Below the powerhouse, the turbinated water reaches first the tailrace channel (TRC1 in Figure 1) and the water is then stored in an 8 Mm^3 regulating pond (RD in Figure 1) located around 3.5 km below the powerhouse. The regulating pond also receives water inputs from the Nam Kathang River (3% of its volume annually). Daily, the water discharge of Nam Kathang River that reaches the regulating pond is returned to the downstream reach of the Nam Kathang River, below the regulating pond. The remaining water from the regulating pond is released to the artificial downstream channel. To prevent potential problem of deoxygenation in the water that passed through the turbines, an aeration weir was built at midway between the turbines and the confluence to the Xe Bangfai River (AW in Figure 1). A continuous flow of 2 $m^3 s^{-1}$ (and occasionally spillway release) is discharged from the Nakai Dam (ND in Figure 1) to the Nam Theun River. 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), leading to a residence time of nearly six months.

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). During the CD season, the reservoir water column overturns and during the WW season, sporadic destratification occurs allowing oxygen to diffuse down to the bottom of the water column (Chanudet et al., 2012;Guérin et al., 2015). Daily average air temperature varies between 12°C (CD season) to 30°C (WD season). The mean annual rainfall is about 2400 mm and occurs mainly (80%) in the WW season (NTPC, 2005).

The filling of the reservoir began in April 2008, the full water level was first reached in October 2009 and stayed nearly constant until the power plant was commissioned in March 2010. After the commissioning, during the studied period the reservoir surface varied seasonally and reached

its maxima (489 km^2) and minima (between 168 and 221 km^2 depending on the year) during the WW and WD seasons, respectively.

2.2. Sampling strategy

A total of 23 stations were monitored weekly to fortnightly in order to determine physico-chemical parameters and the CH_4 concentrations and emissions in pristine rivers, the reservoir, and all rivers and channels located downstream of the reservoir. In the reservoir, two stations were monitored (RES1 and RES9, Figure 1). The station RES1 is located 100 m upstream of the Nakai Dam and RES9 is located ~1 km upstream of the water intake which transports water to the turbines.

Below the powerhouse, the water was monitored at nine stations: in the tailrace channel (TRC1), regulating pond (REG1), artificial downstream channel (DCH1, DCH2, DCH3 and DCH4), and the Xe Bangfai River (XBF2, XBF3 and XBF4). Owing to existence of the above-listed civil structures downstream of the powerhouse, three sections were defined in order to calculate emissions and degassing downstream of the powerhouse, the regulating pond and the aeration weir (Figure 1). The influence of the water released from the regulating pond on the Nam Kathang River was evaluated by the monitoring of two pristine stations (NKT1 and NKT2) upstream of the regulating pond and three stations (NKT3-NKT5) below the regulating pond (Figure 1).

Below the Nakai Dam, 4 sampling stations (NTH3-NTH5 and NTH7) were used for the monitoring of the Nam Theun River. The section 4 refers to the Nam Theun River section located between the stations NTH3 and NTH4 (Figure 1).

Additionally, we monitored the pristine Xe Bangfai River (XBF1) upstream of the confluence with the artificial channel and one of its pristine tributaries (Nam Gnom River: NGM1) and a pristine tributary of the Nam Theun River (Nam Phao River: NPH1) downstream of the Nakai Dam.

During various field campaigns (March 2010, June 2010, March 2011, June 2011 and June 2013), aerobic methane oxidation rates (AMO) were determined at three stations in the reservoir (RES1, RES3 and RES7, Figure 1). Additionally, AMO was also determined in the reservoir at the water intake (RES9) in June 2013,

2.3. Experimental methods

2.3.1. In situ water quality parameter

Oxygen 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 of the vertical profiles was 0.5 m above the oxic–anoxic limit and 1-5 m in the hypolimnion, whereas it was only measured in surface waters (0.2 m) in the tailrace channel, downstream channel and rivers.

2.3.2. Methane concentration in water

The CH_4 concentrations at all stations have been monitored between May 2009 and December 2012 on a fortnightly basis. Surface and deep-water samples for CH_4 concentration were taken with a surface water sampler (Abril et al., 2007) and a Uwitec water sampler, respectively. Water samples were stored in serum glass vials, capped with butyl stoppers, sealed with aluminium crimps and poisoned (Guérin and Abril, 2007). A N_2 headspace was created and the vials were vigorously shaken to ensure an equilibration between the liquid and gas phases prior to CH_4 concentration gas chromatography (GC) analysis. The concentration in the water was calculated using the solubility coefficient of Yamamoto et al. (1976).

2.3.3. Aerobic methane oxidation

In the reservoir, water samples for AMO rate measurements were collected in the epilimnion and in the metalimnion (at the oxicline). At RES9, the samples were taken in the middle of the water column since the water column was well mixed. AMO was also performed at TRC1 (immediately downstream of the powerhouse). The water was collected in 1L HDPE bottles, homogenized, oxygenated and redistributed to twelve serum vials (160 mL). Each vial contained 60 mL of water and 100 mL of air. Vials were covered with aluminium foil to avoid effect of light on any bacterial activity and incubated in the dark (Dumestre et al., 1999;Murase and Sugimoto, 2005) at 20°C to 30°C, depending on in situ temperatures. According to in situ concentration of CH_4 in the water, different amounts of CH_4 were added by syringe while withdrawing an equal volume of air from the headspace with a second syringe in order to obtain concentrations of dissolved CH_4 in the incubated water ranging from in situ to four times in situ.

Incubations were performed with agitation to ensure continuous equilibrium between gas and water phases. Total CH_4 concentrations in the vials were measured 5-times in a row at a 12 h interval, and oxidation rates were calculated as the total loss of CH_4 in the vial (Guérin and Abril, 2007). The oxidation rate for each concentration was the average value of the triplicates with standard deviation (±SD).

The kinetics parameters of aerobic methane oxidation obtained from the experiment were combined to the in situ CH_4 concentration profiles in order to calculate the integrated aerobic methane oxidation in the oxic water column. 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^{-2} d^{-1}) is:

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

with C_{CH4}, the CH_4 concentration; $S_{CH4\text{-}ox}$, the specific $CH_{4\text{-}ox}$; C_{O2}, the oxygen concentration; $K_{m(O2)}$, the K_m 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.

2.3.4. 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 and mixture of N_2 with 100% CH_4) were injected after analysis of every 10 samples for calibration. The detection limit is 0.3 ppmv in the headspace and the accuracy is around 4% allowing the determination of nanomolar concentrations in water samples, depending of the volume of the vials and headspace. Duplicate injection of samples showed reproducibility better than 5%.

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2.4. Calculations

2.4.1. Estimation of diffusive fluxes from surface concentrations

The diffusive CH_4 fluxes downstream of the powerhouse (section 1-3 in Figure 1), and downstream of the Nakai Dam (NTH3-7, Figure 1) were calculated with the thin boundary layer (TBL) equation (Liss and Slater, 1974) from the difference between the water surface CH_4 concentrations and the average CH_4 concentration in air (1.9 ppmv) obtained during eddy covariance deployments (1.9 ppmv) (Deshmukh et al., 2014) combined with a gas transfer velocity (k_{600}) as follow:

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

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) and n = 1/2 for turbulent water (Borges et al., 2004;Guerin et al., 2007).

The artificial channel and the Nam Theun River downstream of the dam are closed for navigation because of the potential high water level changes due to reservoir management and because of the presence of zone of very high turbulence immediately downstream of the powerhouse and downstream of the regulation pond. In the artificial channel, water current velocity never exceeds 1 m s^{-1} and averaged 0.5 m s^{-1}. Floating chamber measurement was not possible for the accurate determination of the k_{600}. In a handful occasions, k_{600} was calculated from floating chamber measurements (Deshmukh et al., 2014) and concomitant CH_4 water surface concentrations in the turbulent waters downstream of the powerhouse (section 1 at stations TRC1 and REG1), in the Xe Bangfai River downstream of its confluence with the artificial channel (XBF2) and in pristine rivers (XBF1, Nam On River and Nam Noy River). The gas transfer velocity reached up to 45 cm h^{-1} and averaged 10.5±12.1 cm h^{-1} (data not showed).

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Mis en forme: Exposant

Frédéric Guérin 31/1/y 10:33
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This is very similar to the average k_{600} value obtained using the formulation k_{600}-wind speed relationships from Guerin et al. (2007) obtained downstream of the Petit Saut Reservoir and in small estuaries of the same size with similar water currents like the Scheldt by Borges et al. (2004). We therefore kept 10 cm h^{-1} as a conservative estimate of the k_{600} in the artificial channel downstream of the NT2R. The gas transfer velocity for the artificial channel, the Xe Bangfai River and downstream of the Nakai Dam (NTH3-7) was kept constant over the whole period of monitoring since the average of the results obtained by the formulations of Borges et al (2004) and Guerin et al (2007) was 10.06 ± 1.48 cm h^{-1} according to the limited variation of the monthly average wind speed (1.8 ± 0.46 m s^{-1}).

2.4.2. Degassing

Although the so-called "degassing" usually occurs only below dams (Galy-Lacaux et al., 1997;Abril et al., 2005;Kemenes et al., 2007;Maeck et al., 2013), degassing occur at 4 sites at NT2R: (1) the Nakai Dam, (2) the turbine release in the tailrace channel, (3) the regulating dam and (4) the aeration weir using the following equation:

$$Degassing = (C_{upstream} - C_{downstream}) \times water\ discharge$$

where $C_{upstream}$ is the CH_4 upstream of the site where degassing might occur and $C_{downstream}$ is the CH_4 concentration in the water downstream of the degassing site. On each of these structures, the degassing was calculated using the water discharges and the difference of CH_4 concentration between the stations: (1) NTH3 located below the Nakai Dam and RES1, (2) TRC1 located below the turbines and RES9, (3) NKT3 below the Regulating Dam and REG1, and (4) DCH3 below the Aeration Weir and DCH2 (Figure 1). In addition, degassing was calculated for the occasional spillway releases from the Nakai Dam.

The estimation of the concentration upstream of the degassing sites was different for the four sites. For the degassing below the turbines and below the regulating dam, the average of the vertical profile of CH_4 concentrations at RES9 and REG1 were considered as concentrations before degassing, respectively. Surface concentration at DCH2 was considered for the degassing at the aeration weir. For the degassing below the Nakai Dam, since the continuous flow of 2 m^3 s^{-1} was released from the surface water layer, we considered the average CH_4 concentration in the

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Supprimé: We therefore used a k_{600} of 10 cm h^{-1} for the artificial channel, the Xe Bangfai River and downstream of the Nakai Dam (NTH3-7).

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516 upper 3 m water layer at RES1 located ~100 m upstream of dam. For the spillway release of the
517 Nakai Dam, as the spillway gate is located at 12 m below the maximum reservoir water level, the
518 degassing due to spillway release was calculated using the average CH_4 concentration in the
519 upper 15 m water layer at RES1.

520 ## 3. Results

521 ### 3.1. Temperature, O_2 and CH_4 concentrations in the reservoir (RES1 and RES9)

522 Before the commissioning of the power plant, the vertical profiles of temperature and oxygen
523 and CH_4 concentrations at the stations RES1 located at the Nakai Dam and RES9 located at the
524 water intake were similar (Figure 2). As already shown in Chanudet et al. (2015) and Guérin et
525 al. (2015), the reservoir was thermally stratified with higher temperature at the surface than at the
526 bottom during the WD (surface: 26.8±2.7°C and bottom: 18.9±1.6°C) and WW (surface:
527 28.0±1.6°C and bottom: 21.5±1.7°C) seasons and it overturns during the CD season (Average =
528 23.2±3.9°C) (Figure 2). During the WD and WW season, the epilimnion was always oxygenated
529 with surface O_2 concentrations ranging from 14 to 354 μmol L^{-1} (5 to 137% saturation) and the
530 hypolimnion was anoxic. In the CD season, the reservoir water column was poorly but entirely
531 oxygenated during a few weeks/month (127±93 μmol L^{-1}). In the WD and WW seasons, the CH_4
532 concentrations ranged between 0.02 and 201.7 μmol L^{-1} in the epilimnion and 0.02 to 1000 μmol
533 L^{-1} in the hypolimnion. In the CD season, the CH_4 concentrations are only slightly higher in the
534 hypolimnion than in the epilimnion. After the starting of turbines, the hydrodynamics of the
535 water column at RES1 followed the same seasonal pattern as described before whereas the CH_4
536 vertical profiles of concentration at RES9 located upstream of the water intake were
537 homogeneous from the surface to the bottom. At RES9 during the years 2010 to 2012, the
538 temperature was constant from the bottom to the surface whatever the season and the water
539 column was always oxygenated (O_2 = 166 μmol L^{-1}) (Figure 2). During this period, CH_4
540 concentration peaked up to 215 μmol L^{-1} with averages of 39.8 ± 48.8, 29.9 ± 55.4 and 1.9 ± 4.3
541 μmol L^{-1} during the WD, WW and CD seasons, respectively. For the two stations, the average
542 CH_4 concentrations over the water column were always the highest in the WD season,
543 intermediate in the WW season and the lowest in the CD season. At the two stations, the average
544 concentrations were significantly higher in 2009 and 2010 than they were in 2011 and 2012. The

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546 average CH_4 concentrations at NT2R were in the range reported for tropical reservoir flooded
547 10-20 years ago (Abril et al., 2005;Guérin et al., 2006;Kemenes et al., 2007).

548 ### 3.2. Emissions downstream of the Nakai Dam

549 #### 3.2.1. CH_4 and O_2 concentrations below the Nakai Dam

550 Downstream of the Nakai Dam (NTH3) after the commissioning, the average O_2 concentration
551 was 224 μmol L^{-1} that is 87% saturation and the concentration increased further downstream.
552 When excluding the periods of spillway releases, the CH_4 concentration at NTH3 ranged from
553 0.03 to 6 μmol L^{-1} (average: 0.94 ± 1.2 μmol L^{-1}) with the highest CH_4 concentrations in the WW
554 season and the lowest in the CD season (Figure 3a). High CH_4 concentrations (up to 69 μmol L^{-1})
555 were occasionally observed when CH_4-rich water was released from the spillway, especially in
556 2009. Ten kilometers downstream of the Nakai Dam, CH_4 concentration decreased down to
557 0.41±0.32 μmol L^{-1} at NTH4 and NTH5 without any clear seasonal pattern (Fig. 3 a).

558 The concentrations observed below the Nakai Dam at the stations NTH4 and NTH5 were similar
559 to the CH_4 concentrations found in the pristine Nam Phao River (NPH1) in the watershed and
560 40% lower than the CH_4 concentrations at the station NTH7 located 50 km downstream of the
561 dam. They were 2 orders of magnitude lower than the concentrations observed downstream of
562 10-20 years old-reservoirs in Brazil and in French Guiana (Guérin et al., 2006;Kemenes et al.,
563 2007).

564 #### 3.2.2. Diffusive fluxes below the Nakai Dam

565 The average diffusive flux downstream of the Nakai Dam was 3.3 ± 3.9 mmol m^{-2} d^{-1} for the year
566 2010 and fluxes decreased down to 1.9±2.5 and 1.4 ± 0.9 mmol m^{-2} d^{-1} for the years 2011 and
567 2012, respectively (Figure 3b). Ten kilometres downstream from the Nakai Dam at NTH4 and at
568 NTH5 downstream of the confluence of the Nam Phao River, the CH_4 fluxes decreased down to
569 1.14±0.92 mmol m^{-2} d^{-1} on average (Fig 3b). As for the concentrations, no seasonal or
570 interannual trends were found. Downstream the station NTH4 located 10 kilometres downstream
571 of the dam, the CH_4 emission was similar to what found in pristine river of the watershed and it
572 was 2 orders of magnitude lower than the emissions observed downstream of 10-20 years-old
573 reservoirs (Guérin et al., 2006;Kemenes et al., 2007).

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Considering that the CH_4 emissions from the Nam Theun River below the dam can be attributed to the reservoir over a maximum length of 10 km and a constant width of 30 m, annual emissions below the Nakai Dam decreased from 20 to 1 Mg-CH_4 $month^{-1}$ between 2009 and 2012, respectively (Figure 3c). The very high emissions in 2009 were due to spillway releases (see below).

3.2.3. Degassing below the Nakai Dam

Due to the low water discharge at the Nakai Dam (2 m^3 s^{-1}), CH_4 emissions by degassing reached a maximum of 0.1 MgC-CH_4 d^{-1} at NTH3 (Figure 3e). The occasional spillway releases occurred mostly in 2009 before the commissioning of the power plant and in the CD after the commissioning. They led to very intense degassing (up to 72 Mg-CH_4 d^{-1}, August 2009, Figure 3d). In total, 99% of the degassing below the Nakai Dam is due to the spillway releases in 2009 which represent 32% of total emissions downstream of the Nakai Dam during the study (2009-2012). Total degassing below the Nakai Dam was very significant in 2009 due to the spillway releases and it dropped below 3 Mg-CH_4 $month^{-1}$ when only 2 m^3 s^{-1} were released for the years 2010 to 2012.

3.3. Emissions downstream of the powerhouse

3.3.1. CH_4 and O_2 concentrations below the powerhouse

Downstream of the turbines at the station TRC1 after the commissioning, the average O_2 concentration was 174 ± 58 μmol L^{-1} that is 67 ± 20% saturation. After the commissioning of the power plant, the O_2 saturation downstream of the station DCH4 located 30 km below the turbines was always around 100% saturation in the artificial downstream channel. Just below the regulating dam, in the Nam Kathang River (NKT3), the average O_2 concentration was 237 μmol L^{-1} that is 93% saturation. There was no marked inter-annual change in the O_2 concentration.

Surface CH_4 concentration at the station TRC1, which is located below the turbines and receives water from the homogenized water column in the reservoir (RES9), varied by four orders of magnitude; from 0.01 μmol L^{-1} (August-February, WW and CD seasons) to 221 μmol L^{-1} (June, end of the WD and beginning of the WW season) (Figure 4a). The seasonal pattern of the CH_4 concentrations at TRC1 mimicked the concentrations at RES9. In 2010, the surface CH_4

concentration decreased from 117 ± 71 μmol L^{-1} at TRC1 to 1.55 ± 1.15 μmol L^{-1} at DCH4 in the WD season and from 88 ± 84 to 1.26 ± 1.59 μmol L^{-1} in the WW season. In 2011 and 2012, the average CH_4 concentrations just below the turbines at TRC1 were fourfold (33.4 ± 32.0 μmol L^{-1}) and ninefold (9.8 ± 29.6 μmol L^{-1}) lower than in 2010 for the WD and WW seasons, respectively. At DCH4, the surface CH_4 concentration drops to 1.1 ± 2.4 μmol L^{-1} (WD) and 0.3 ± 0.5 μmol L^{-1} (WW) in the years 2011 and 2012 that is similar to what was observed in 2010. Whatever the years, in the CD season, surface CH_4 concentrations was lower than 14.5 μmol L^{-1} along the 30 km long watercourse (0.02 – 14.5 μmol L^{-1}).

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On average, at the station DCH4 (30 km below the turbines) and at the station XBF4 located 90 km below the confluence of the downstream channel and the Xe Bangfai River, the CH_4 concentrations were 0.54 ± 0.95 and 0.3 ± 0.4 μmol L^{-1}, respectively. These concentrations are the same as those found in the pristine Xe Bangfai River (0.78 ± 0.86 μmol L^{-1} at XBF1 station).

At the station NKT3 located in the Nam Kathang River just below the regulating dam, the average surface CH_4 concentration was 0.87 ± 0.77 μmol L^{-1}. At the station NKT5 located 15 km downstream of the regulating dam, the average CH_4 concentration was 1.34 ± 2.09 μmol L^{-1}. These concentrations are not statistically different from the concentrations found in the pristine Nam Kathang Noy River (0.42 ± 0.49 μmol L^{-1} at NKT1 station), the pristine Nam Kathang Gnai River (1.01 ± 1.73 μmol L^{-1} at NKT2 station) and the pristine Nam Gnom River (1.08 ± 1.45 μmol L^{-1} at NGM1) all located in the same watershed.

3.3.2. Diffusive fluxes below the Powerhouse

In 2010, in the section 1, the flux was 198 ± 230 mmol m^{-2} d^{-1}, which was two times higher than in section 2 (94 ± 102 mmol m^{-2} d^{-1}) (Figure 4c). In the section 3 (below the aeration weir), fluxes were fifteen times lower than the fluxes in section 1 (12.7 ± 18.6 mmol m^{-2} d^{-1}). After the confluence with the Xe Bangfai River, CH_4 fluxes dropped down to 0.95 ± 0.76 mmol m^{-2} d^{-1} for the next 30 km. For the years 2011 and 2012, the average diffusive fluxes below the powerhouse decreased by a factor of four as compare to 2010. In 2010, most of the diffusive fluxes occurred from the middle of the WD season until the late WW season (155 ± 127 mmol m^{-2} d^{-1}) whereas diffusive fluxes in the CD season were 100 times lower (1.4 ± 1.1 mmol m^{-2} d^{-1}). In 2011 and 2012, most of the emissions occurred during the WD season (61.9 ± 50 mmol m^{-2} d^{-1}) whereas

635 emissions were twentyfold lower during both the WW and the CD seasons (3.7 ± 3.9 mmol m^{-2}
636 d^{-1}).

637 As observed for the concentrations, emissions downstream of DCH4 in the downstream channel
638 (1.5 ± 2.7 mmol m^{-2} d^{-1}) and at NKT3 downstream of the regulating dam in the Nam Kathang
639 River (2.03 ± 2.23 mmol m^{-2} d^{-1}) (Figure 4b) were not significantly different from those
640 calculated for the pristine Xe Bangfai River (2.2 ± 2.6 mmol m^{-2} d^{-1} at XBF1 station), Nam
641 Kathang Noy River (NKT1 station) and Nam Kathang Gnai River (NKT2 station) (1.98 ± 4.01
642 mmol m^{-2} d^{-1}).

643 The average diffusive flux for the sections 1 to 3 during the monitoring was 12 ± 22 mmol m^{-2} d^{-1},
644 which is 7 times lower than the diffusive flux along the 40 km reach below the Petit Saut Dam
645 (90 mmol m^{-2} d^{-1}) (Guérin and Abril, 2007) 10 years after impoundment and twelve times lower
646 than the diffusive flux along the 30 km reach downstream of the Balbina Dam (140 mmol m^{-2} d^{-1})
647 (Kemenes et al., 2007) 18 years after impoundment.

648 The sum of the CH_4 emissions by diffusion from the sections 1, 2 and 3 (Figure 1) peaked at 333
649 Mg-CH_4 $month^{-1}$, 156 Mg-CH_4 $month^{-1}$ and 104 Mg-CH_4 $month^{-1}$ at the end of the WD-beginning
650 of the WW season in 2010, 2011 and 2012, respectively (Figure 4c). Diffusion was negligible for
651 more than half of the year. The results clearly show that emissions decrease with time within the
652 first four years after flooding.

653 ### 3.3.3. Degassing below the Powerhouse

654 The degassing mainly occurred within 3 to 5 months around the transition between the WD and
655 the WW seasons (Figure 4d). Below the powerhouse (TRC1), the degassing reached up to 385
656 Mg-CH_4 $month^{-1}$ at the end of the WD season and beginning of the WW season in 2010, just
657 after the turbines were operated (Figure 4d). Below the regulating dam, the degassing was almost
658 three times higher (1240 Mg-CH_4 $month^{-1}$) than below the turbines, and the degassing from the
659 release to the Nam Kathang River was 55 Mg-CH_4 $month^{-1}$ in the WD season. Even if CH_4
660 concentrations at DCH2 were 50% lower than at TRC1, still up to 756 Mg-CH_4 $month^{-1}$ were
661 emitted at the aerating weir. This shows the very high degassing efficiency of the aeration weir
662 (up to 99%), especially in the WD season (Descloux et al., 2015). Therefore, most of the
663 degassing emissions occurred below the regulating dam and at the aerating weir.

664 In 2010, most of the degassing occurred from April to August whereas it occurred only from
665 March to June in 2011 and 2012. The annual degassing emissions almost deceased by a factor of
666 four in 2011 and 2012 compare to 2010 (Figure 4e).

667 ### 3.4. Aerobic CH_4 oxidation in the reservoir and downstream of the powerhouse
668 ### and the Nakai Dam

669 In the reservoir, the potential AMO rates increased linearly with the CH_4 concentration (Figure
670 5a,b,c) in both epilimnetic and metalimnic waters at the stations RES1, RES3 and RES7. The
671 AMO rates in the middle of the well-mixed water column at the station RES9 were not
672 statistically different from the AMO rates in the metalimnion at the other stations of the
673 reservoirs. Therefore, the AMO rates from RES9 were plotted versus the initial CH_4
674 concentration together with AMO rates from the metalimnion. The slope of the linear
675 correlation, or the so-called specific oxidation rate (SOR, d^{-1}) in the metalimnion was similar for
676 the CD and WD seasons (SOR = 0.88 ± 0.03 d^{-1}) (Figure 5a). In the epilimnion the SOR was
677 twice higher in the WD season (5.28 ± 0.43 d^{-1}) than in the CD season (2.24±0.41 d^{-1}) (Figure
678 5b,c). Overall, the SOR in the epilimnion was two to fourfold higher than the SOR in the
679 metalimnion. Downstream of the powerhouse, the SOR was 1.47 ± 0.07 d^{-1}, that is intermediate
680 between the observation in the epilimnion and the metalimnion (data not show). The values of
681 SOR observed at the NT2R are in same range as those reported at the Petit Saut Reservoir (2.64-
682 4.13 d^{-1}) (Dumestre et al., 1999;Guérin and Abril, 2007) and boreal experimental reservoirs
683 during the summer period (0.36 - 2.4 d^{-1}) (Venkiteswaran and Schiff, 2005).

684 The depth-integrated oxidation rates ranged from 0.16 to 931 mmol m^{-2} d^{-1} at RES9 and from
685 0.13 to 310 mmol m^{-2} d^{-1} at RES1 upstream of the Nakai Dam. Overall, for the years 2010, 2011
686 and 2012, the average integrated oxidation rate at RES9 is 122 mmol m^{-2} d^{-1} that is more than
687 three times higher than the average integrated oxidation rate at RES1 (35 mmol m^{-2} d^{-1}). Since
688 oxidation occurs from the surface to the bottom of the water column at RES9 and mostly around
689 the oxicline at RES1, the depth-integrated oxidation rates were 5-20 times higher at RES9 than at
690 RES1 during the WD season and no clear tendency can be drawn for the WW and CD seasons
691 (Table 1). At RES9, the total amount of oxidized CH_4 decreased from 5 to 1 $Gg(CH_4)$ y^{-1}

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between 2010 and 2012 whereas it ranged between 0.4 and 0.7 Gg(CH_4) y^{-1} without clear trend at RES1 (Table 1).

4. Discussion

4.1. Spatial and temporal variations of downstream emissions

Before the power plant was commissioned in March 2010, only a few m^3 of water was discharged at the powerhouse for testing the turbines and most of the water was discharged at the Nakai Dam. The continuous water discharge at the Nakai Dam was about 2 m^3 s^{-1} and occasionally, water was spilled in order to prevent dam overflow. The continuous discharge at the Nakai Dam mimics the lowest annual water flow in the Nam Theun River before it was dammed. Since it expels CH_4-poor water (0.95 μmol L^{-1}) from the surface associated with a very low discharge, subsequent degassing and diffusive emissions below the Nakai Dam were lower than 4 Mg-CH_4 $month^{-1}$ in 2010 just after the commissioning and lower than 1 Mg-CH_4 $month^{-1}$ in 2012 (Figure 3e). Degassing was four fold higher in 2010 than in 2012 because of the very high CH_4 concentrations in the water column resulting from the long residence time of water in the reservoir before the first water releases. In 2011, the concentrations were lower than in 2012 due to the high water discharges from the inflows that decreased the CH_4 concentrations by dilution (Guérin et al., 2015). The spillway releases reached up to 5309 m^3 s^{-1} and water from the top 15 m of the water column having an average concentration around 100 μmol L^{-1} at RES1 were released at these occasions. During these short releases, up to 3000 Mg-CH_4 $month^{-1}$ were released in 2009 (Figure 3d). After the commissioning, the spillways were used only twice in October 2010 and September 2011. The diffusive fluxes in the Nam Theun River below the Nakai Dam were only highly significant during the spillway releases when it reached up to 20 Mg $month^{-1}$ in 2009. After the commissioning, the diffusion ranged between 0.2 and 1.5 Mg-CH_4 $month^{-1}$ (Figure 3c) and contributed to only a few percent of total downstream emissions below the Nakai Dam (Figure 3f). Emissions below the Nakai Dam are low compare to emissions below the powerhouse because, except during spillway releases, only a small amount of water is discharged downstream and this water has a low CH_4 concentration since surface water is released. However, we show here that short spillway releases with high water discharge and moderate CH_4 concentrations could contribute up to 30% of downstream emissions in 4 years.

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728 Downstream of the powerhouse, maximum yearly emissions were dominated by degassing
729 (Figure 4e). They ranged between 1 and 3 Gg month^{-1} and had a clear seasonal pattern. Emissions
730 below the powerhouse peaked during the WD season until the beginning of the WW season
731 when the CH_4 concentration in the hypolimnion of the reservoir is up to 1000 μmol L^{-1} (Guérin et
732 al., 2015) and concentration at RES9 higher than 100 μmol L^{-1}. Emissions were negligible in the
733 late WW and during the CD seasons when hypolimnic concentration in the reservoir and
734 concentration at RES9 decreased down to 5 μmol L^{-1} (Guérin et al., 2015). Due to the
735 accumulation of CH_4 in the reservoir in absence of turbining until commissioning, emissions
736 downstream of the powerhouse in 2010 were higher than in 2011 and 2012 and lasted from the
737 commissioning to the beginning of the next CD season in 2010. After the commissioning, the
738 high emissions downstream of the powerhouse occurred within 3-5 month in the WD season and
739 the very beginning of the WW season. During the wet 2011-year, emissions became negligible
740 after the first rainfalls. For all years, downstream emissions were negligible in the CD season.
741 These results show the very high seasonal variations over 3-4 orders of magnitude for
742 downstream emissions as already observed in tropical reservoirs flooding primary forest (Abril et
743 al., 2005;Kemenes et al., 2007). However, we show in this monomictic reservoir that
744 downstream emissions are negligible most of the year and this is mostly due to the seasonal
745 overturn in the CD and some sporadic destratification events and dilution of the hypolimnoion in
746 the WW season. Overall, these results highlight the fact that the precise determination of
747 downstream emissions cannot be done on the basis of discrete sampling one to four times in a
748 year. It requires weekly to monthly monitoring in order to (1) capture the hot moment(s) of
749 emissions and (2) determine their duration. For instance, downstream emissions reported for the
750 Nam Ngum and Nam Leuk Reservoirs located in the same region were obtained at the beginning
751 of the WD season when downstream emissions are moderate and during the CD and WW season
752 when no emission occur (Chanudet et al., 2011). Therefore, emissions were probably
753 underestimated since the peak of downstream emissions at the end of the WD season-beginning
754 of the WW season was missed.

755 ### 4.2. Contribution of downstream emissions to CH_4 gross emissions

756 Table 2 reports CH_4 emissions by ebullition and diffusion at the surface of the reservoir from the
757 Deshmukh et al. (2014) and Guérin et al. (2015), respectively. These estimates take into account

758 the seasonal variations of the reservoir water surface and the variations of depth. Between June
759 and December 2009, the spillway releases contributed to 30% of total gross emissions from the
760 NT2R. In 2010, downstream emissions (degassing + diffusive fluxes) contributed to more than
761 30% of total gross emissions (disregarding drawdown emissions). In 2011 and 2012, downstream
762 emissions contributed to about 10% of total gross emissions. This contribution of downstream
763 emissions to total emissions is low compare to tropical reservoirs located in South America
764 (Abril et al., 2005;Kemenes et al., 2007). Disregarding the first two years of monitoring (2009
765 and 2010) during which the quantification highly depends on the management of the reservoir,
766 the contribution of downstream emissions to total emissions is even lower than in boreal
767 reservoirs (Teodoru et al., 2012). The low downstream emissions arise from the fact that the
768 reservoir is monomictic. Each time the reservoir overturns in the CD season, 1-3 Gg of CH_4 are
769 emitted to the atmosphere within a few days and up to a month which purge the reservoir water
770 column (Guérin et al., 2015). As a consequence, bottom concentrations decrease from 500 to less
771 than 5 μmol L^{-1} during these events and the amount of CH_4 transferred from the reservoir to the
772 downstream reaches decrease by two orders of magnitude and stays low during 8 to 9 months,
773 before the CH_4 concentration in the reservoir increases again. Monomictic reservoirs like Nam
774 Theun 2, Nam Leuk, Nam Ngum in Lao PDR (Chanudet et al., 2011), the Three Gorges Dam in
775 China (Li et al., 2014) and the Cointzio Reservoir in Mexico (N. Gratiot, Pers. Com.) are
776 common in the subtropics and especially in Asia where 60% of the worldwide hydroelectric
777 reservoirs are. Although CH_4 emissions below amictic reservoirs like Petit Saut and Balbina are
778 high and very significant in the total emissions (Abril et al., 2005;Kemenes et al., 2007), low
779 emission downstream of monomictic/dimictic/polymictic reservoirs is likely to be a general
780 feature. The thermal stratification of hydroelectric reservoirs has to be taken into account for the
781 estimation of global downstream emissions from hydroelectric reservoirs. Therefore, global
782 estimates of CH_4 emissions from hydroelectric reservoirs that include downstream emissions
783 (Lima et al., 2008;Li and Zhang, 2014) calculated on the basis of the results from Amazonian
784 reservoirs (Abril et al., 2005;Guérin et al., 2006;Kemenes et al., 2007) must be considered with
785 caution.

4.3. Consequence of outgassing and aerobic CH_4 oxidation at the water intake for the emissions below the powerhouse

In addition to the dynamic of the thermal stratification of the NT2R, the design of the water intake contributes to lower the emissions downstream of the powerhouse. After the power plant was commissioned, the water column at the station RES9 was always completely mixed from the top to the bottom as revealed by the vertical profiles of temperature. Consequently, O_2 penetrated down to the bottom of the water column and CH_4 concentration were higher than 100 μmol L^{-1} from the top to the bottom of the water column in the WD season and at the beginning of the WW season. The overturn of the water column at RES9 results from the artificial mixing due to the advection of water caused by the water current generated by the water intake localized around 11-20 m under the water surface depending on the water level. The water intake is responsible for the mixing of the whole water column over an area of 3 km^2 according to the hydrodynamic model of Chanudet et al. (2012). This mixing has a strong effect on both the outgassing (Guérin et al., 2015) and the aerobic oxidation of CH_4 around the water intake and on the oxidation of CH_4 below the powerhouse.

In the area of influence of the water intake where RES9 is, large amount of CH_4 (up to 600 mmol m^{-2} d^{-1}) are emitted by diffusive fluxes at the end of the WD season-beginning of the WW (Guérin et al., 2015). The artificial mixing at RES9 generated a hotspot of CH_4 emissions where diffusive fluxes are 15 to 150 times higher than at other stations in the reservoir for the years 2010 to 2012 (Guérin et al., 2015). The emissions at RES9 correspond to 20 to 40% of the total downstream emissions (Table 2). Therefore, a very significant amount of CH_4 that could be emitted downstream is emitted at the reservoir surface and this contributes to lower downstream emissions.

However, the mixing at the water intake has a strong impact on aerobic CH_4 oxidation. The vertical mixing allows O_2 to penetrate down to the bottom in the vicinity the water intake and enhances both oxidation at the water intake and downstream of the powerhouse. On average, depth-integrated CH_4 oxidation at RES9 upstream of the water intake is one order of magnitude higher than at the station RES1 upstream of the Nakai Dam where the water column is thermally stratified. Over the 3-km^2-area representative for RES9 between 2010 and 2012, aerobic CH_4 oxidation consumed an amount of CH_4 that is equivalent to 50% of total CH_4 downstream

emissions (Table 1 and 2). In absence of artificial mixing, aerobic CH_4 oxidation would only remove an amount of CH_4 that is equivalent to the amount of CH_4 removed by oxidation at RES1 that is on average, that is 11% of total downstream emissions over the three years of monitoring (Table 1 and 2). Total downstream emissions were therefore lowered by 20% due to the enhancement of aerobic CH_4 oxidation at RES9 if we compare total downstream emissions to total downstream emissions plus the amount of CH_4 that would not be oxidized in absence of mixing (oxidation at RES9 minus oxidation at RES1). In addition, aerobic methane oxidation in the downstream channel might be enhanced too since water from RES9 being transferred to the artificial downstream channel is better oxygenated that it would be in absence of artificial mixing.

Overall, the design of the water intake that mixes the whole water column decreases virtually downstream emissions since part of the CH_4 is outgassed at the reservoir surface instead of being transported and emitted downstream. The very positive counterpart of this artificial mixing at the water intake is that the mixing allows O_2 to penetrate down to the bottom of the water column enhancing aerobic methane oxidation both at the water intake and in the river/channel downstream of the powerhouse. Roughly, CH_4 emissions from NT2 Reservoir are lowered by 40% or more due to the artificial mixing of the water column at the water intake.

5. Conclusion

This first quantification of CH_4 emissions downstream of a subtropical monomictic hydroelectric reservoir shows that emissions are negligible most of the year due to low CH_4 concentration in the hypolimnion. They occurred only during 2-4 month per the year at the end of the warm season-beginning of the wet season and globally contribute to 10% of total emissions as observed during normal reservoir operation years (2011 and 2012). The monitoring of downstream emissions before and just after the commissioning (2009 and 2010) after a period with long water residence time in the reservoir (up to 5 years) with occasional use of spillways stresses that reservoir management can have very significant impact on emissions by enhancing diffusive emissions and downstream emissions resulting from the use of spillways.

Emissions downstream of the Nam Theun 2 Reservoir have a low contribution to total emissions also because a very significant amount of CH_4 that could be emitted downstream of the reservoir

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is (1) emitted upstream of the water intake and (2) is oxidized in the vicinity of the water intake because of the artificial mixing it generates. This artificial mixing contributes to improve the water quality downstream of the turbines since the water that passes through is well oxygenated (70% saturation). The other positive counterpart is that it generates a hotspot of aerobic methane oxidation that contributes to the oxidation of 20% of the CH_4 that would potentially be emitted at the water intake or downstream of the turbines. This study shows that downstream emissions from future or existing reservoirs could be significantly mitigated by the adoption of water intake-design or the installation of devices enhancing artificial water column destratification and oxygenation upstream of the turbines.

On the basis of these results, different from those previously published, we recommend that estimates at the global scale of emissions below dams take into account the mixing status of reservoirs, the water residence time and depth of the water intake and its impact on the oxygenation of the water column immediately upstream of the turbines.

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Acknowledgements

The authors thank everyone who contributed to the NT2 monitoring programme, especially the Nam Theun 2 Power Company (NTPC) and Electricité de France (EDF) for providing financial, technical and logistic support. We are also grateful to the Aquatic Environment Laboratory of the Nam Theun 2 Power Company whose Shareholders are EDF, Lao Holding State Enterprise and Electricity Generating Public Company Limited of Thailand. CD benefited from a PhD grant by EDF.

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Table 1: Depth-integrated methane oxidation rates (mmol m^{-2} d_{-1}) and annual amount of oxidized CH_4 ($Gg(CH_4)$ y^{-1}) at the stations RES9 and RES1 of the Nam Theun 2 Reservoir. The depth-integrated CH_4 oxidation rates are given for each season: cold dry (CD), warm dry (WD) and warm wet (WW) for each year.

		RES9		RES1	
Year	Season	mmol m^{-2} d^{-1}	$Gg(CH_4).y^{-1}$	mmol m^{-2} d^{-1}	$Gg(CH_4).y^{-1}$
2010	CD	11.6±5.5		2.8±1.0	
	WD	444.1±106.1	5.2±1.2	18.2±6.5	0.7±0.2
	WW	442.3±93.6		96.3±29.8	
2011	CD	1.0±0.2		7.5±2.7	
	WD	128.2±46.2	1.0±0.5	5.3±2.4	0.4±0.2
	WW	46.9±31.8		50.2±26.3	
2012	CD	33.9±9.6		34.7±11.3	
	WD	94.1±19.4	1.2±0.3	41.9±21.8	0.6±0.2
	WW	80.7±24.2		26.13±5.3	

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

$Gg(CH_4)\ year^{-1}$	2009	2010	2011	2012
Emission from reservoir				
Ebullition[1]	11.21±0.16	14.39±0.11	14.68±0.10	12.29±0.09
Diffusion at RES9 only[2]	0.02±0.01	2.33±0.21	0.86±0.12	0.66±0.11
Diffusion at RES1 only[2]	0.06±0.03	0.09±0.07	0.01±0.00	0.01±0.00
Total diffusion[2]	4.45±1.01	9.34±2.32	3.71±0.81	4.95±1.09
Total emissions from reservoir	15.66±1.02	23.73±2.32	18.39±0.82	17.25±1.09
Emissions from downstream				
Degassing (continuous release)	0.49±0.03	8.48±0.74	1.83±0.41	1.67±0.31
Degassing (Spillway release)	7.20±0.90	0.92±0.39	0.14±0.00	0.00±0.00
Diffusion	0.10±0.02	1.33±0.03	0.32±0.02	0.33±0.03
Total downstream emissions	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
Downstream emissions (%)	33	31	11	10

[1]Deshmukh et al. (2014)

[2]Guérin et al. (2015)

983 **Figure captions**

984 Figure 1. Map of the Nam Theun 2 Hydroelectric Reservoir (Lao People's Democratic Republic)

985 Figure 2: Vertical profiles of temperature, oxygen and methane concentrations at the stations
986 RES1 and RES9 in the Nam Theun 2 Reservoir during the three seasons in 2010, 2011 and 2012

987 Figure 3: Methane concentrations and emissions downstream of the Nakai Dam at the Nam
988 Theun 2 Reservoir between 2009 and 2012. (a) Time series of CH_4 concentrations at the stations
989 NTH3 and NTH4, (b) diffusive fluxes at the stations NTH3 and NTH4, (c) emissions by
990 diffusive fluxes in the section 4 (between NTH3 and NTH4), (d) degassing due to spillway
991 release below the Nakai Dam, (e) degassing below the Nakai Dam due to the continuous water
992 discharge of 2 m3 s-1 and (f) Total emissions by degassing and diffusion downstream of the
993 Nakai Dam.

994 Figure 4: Methane concentrations and emissions downstream of the powerhouse of the Nam
995 Theun 2 Reservoir between 2009 and 2012. (a) Time series of CH_4 concentrations at the stations
996 TRC1, DCH1, DCH3 and DCH4, (b) diffusive fluxes at the stations TRC1, DCH1, DCH3 and
997 DCH4, (c) emissions by diffusive fluxes in the section 1, 2 and 3 (see Figure 1), (d) degassing
998 downstream of the powerhouse, the regulating dam and the aeration weir, (e) Total emissions by
999 degassing and diffusion downstream of the Nakai Dam.

1000 Figure 5: Linear relationships between methane (CH_4) concentrations and aerobic methane
1001 oxidation in the (a) metalimnion, (b) the epilimnion in the cool dry season and (c) the epilimnion
1002 in the warm dry season at the Nam Theun 2 Reservoir.

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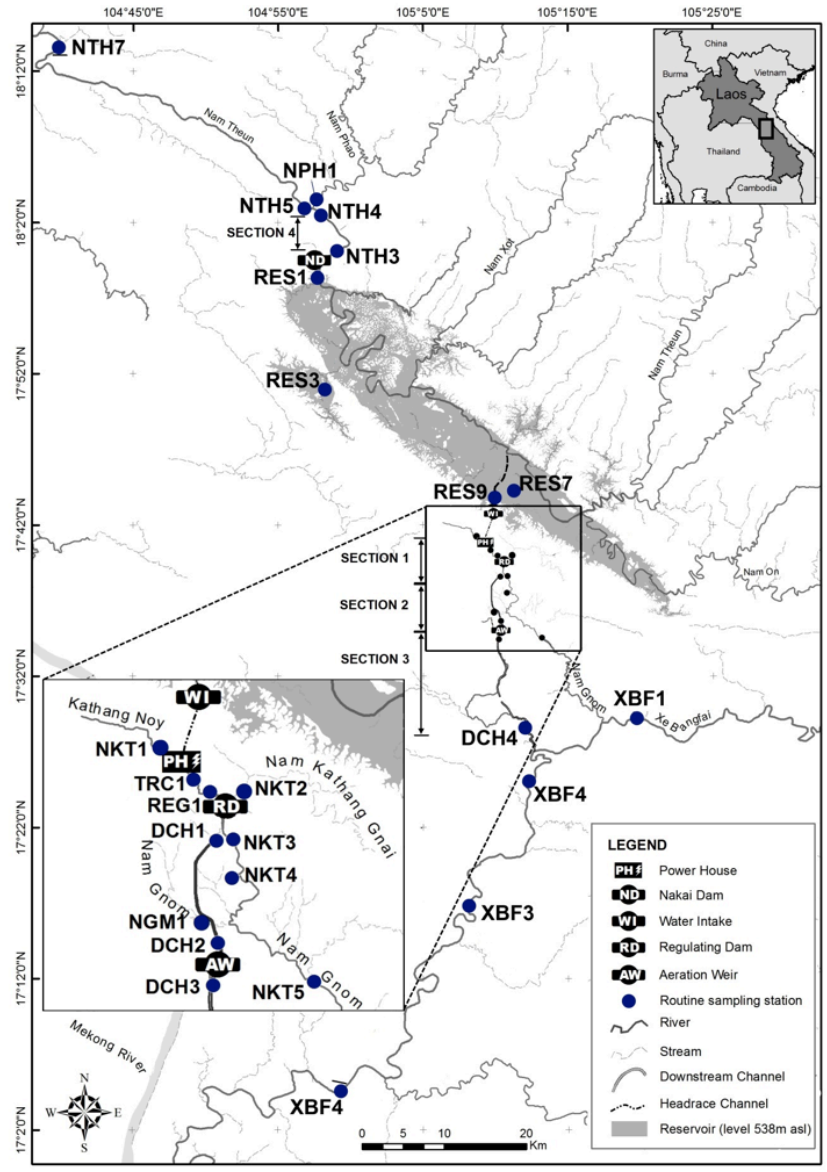
1004 Figure 1



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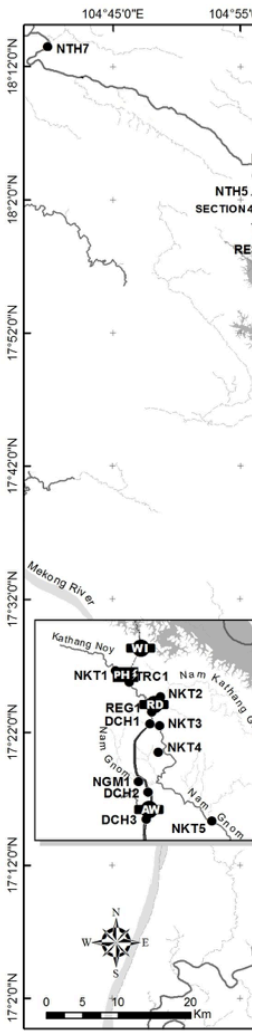

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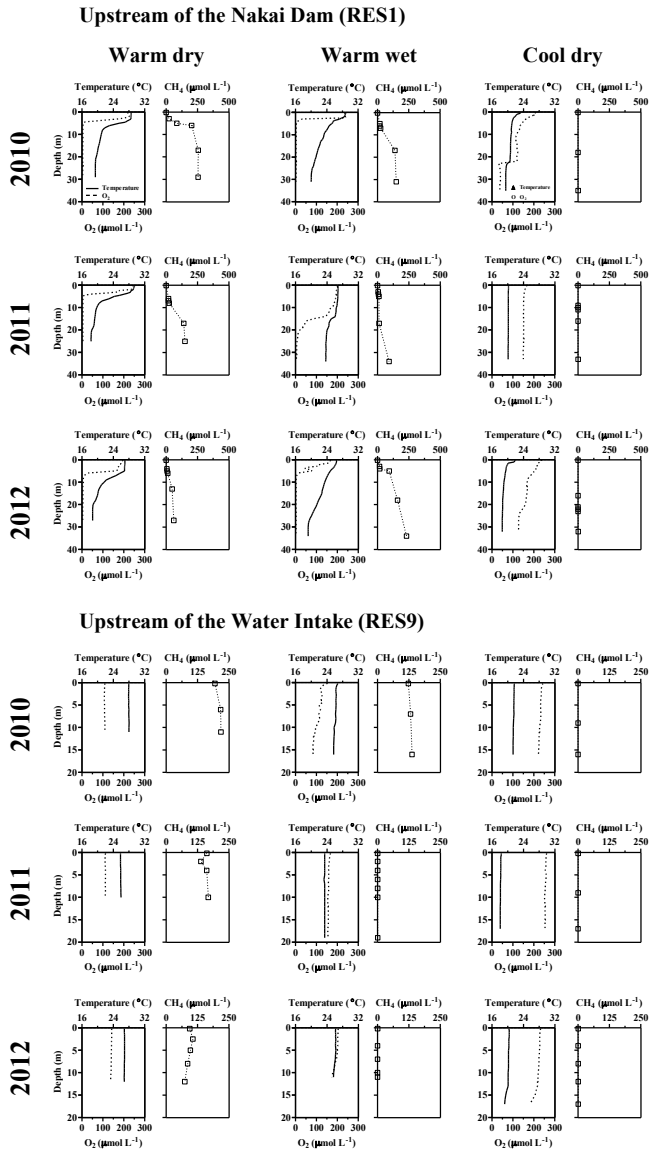

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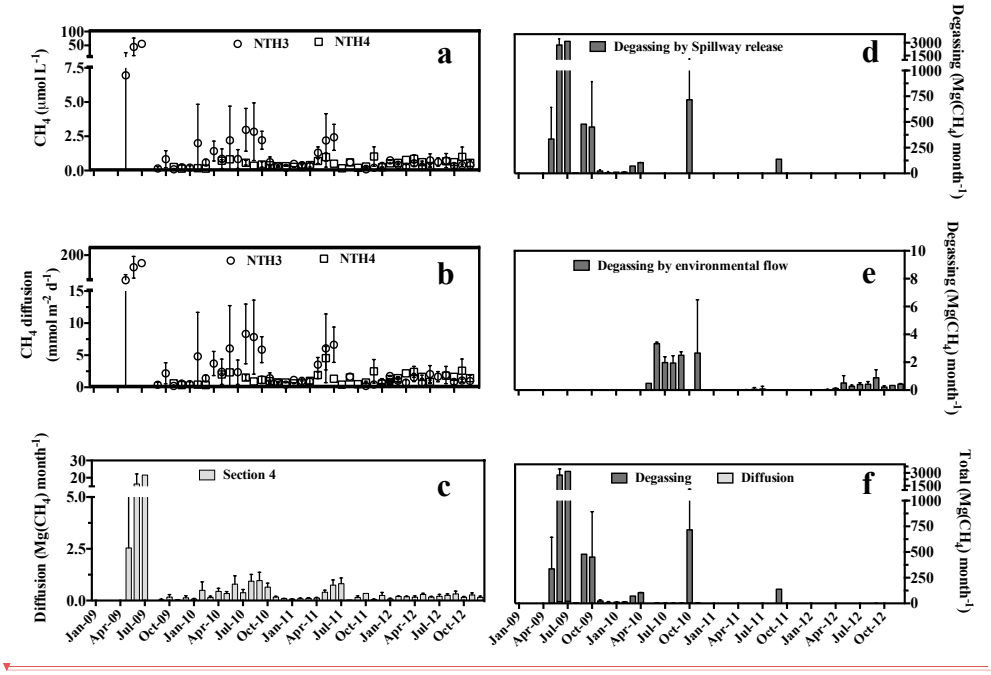

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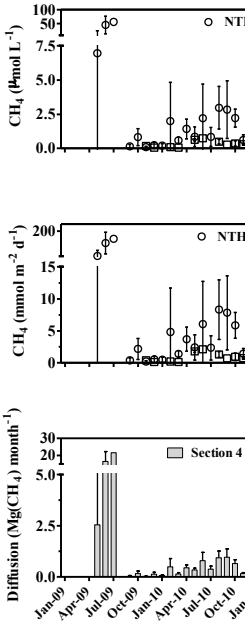

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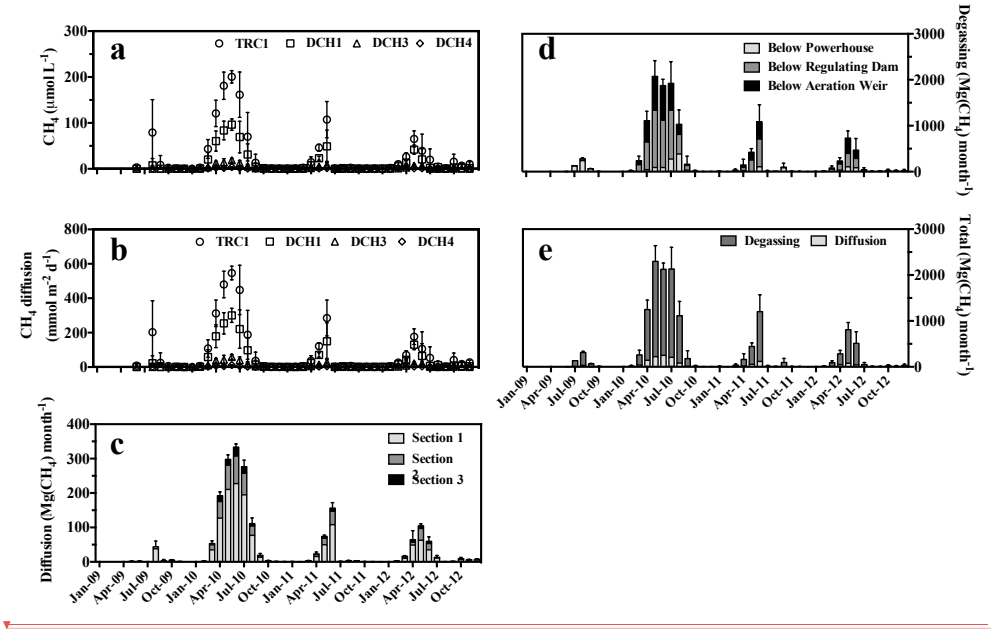

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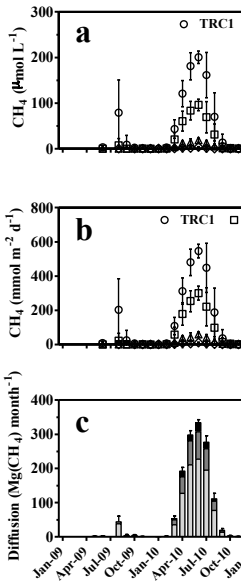

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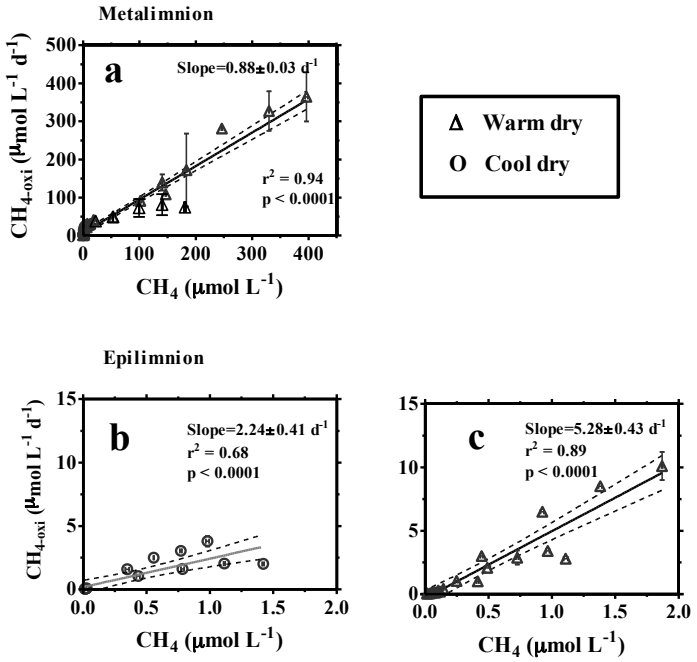

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