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

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Abstract

Inland waters in general and specifically freshwater reservoirs are recognized as a 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 temperature and O_2 and CH_4 concentrations every two weeks for 3.5 years at nine stations in a subtropical monomictic reservoir which was flooded in 2008 (Nam Theun 2 Reservoir, Lao PDR). Based on these results, we quantified CH_4 storage in the water column and diffusive fluxes from June 2009 to December 2012. We compared diffusive emissions with ebullition from Deshmukh et al. (2014) and aerobic methane oxidation and downstream emissions from Deshmukh et al. (2016).

In this monomictic reservoir, the seasonal variations of CH_4 concentration and storage were highly dependent on 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. Concentration and storage 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 CH_4 storage were concomitant with extreme diffusive fluxes (up to 200 mmol m^{-2} d^{-1}). These sporadic emissions occurred mostly in the inflow region in the WW season and during 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 extreme CH_4 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 emission hotspot 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. The hotspot was attributed to the mixing induced by the water intakes (artificial mixing). 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.

1. Introduction

Since the 1990s, hydroelectric reservoirs are known to be a source of methane (CH_4) to the atmosphere. Their contribution to total CH_4 emissions still needs refinement since the

discrepancies among estimates is large, ranging from 1 to 12% of total CH_4 emissions (St Louis et al., 2000;Barros et al., 2011). These two estimates are mostly based on diffusive fluxes at the air-water interface and they overlook emissions from the rivers downstream of the dams (Abril et al., 2005;Guerin et al., 2006;Kemenes et al., 2007;Teodoru et al., 2012;Maeck et al., 2013;Deshmukh et al., 2016), CH_4 ebullition (DelSontro et al., 2010;Deshmukh et al., 2014) and emissions from the drawdown area of reservoirs (Chen et al., 2009;Chen et al., 2011) although these pathways could largely dominate diffusion at the surface of the reservoirs.

Even if CH_4 diffusion at the surface of reservoir is the best-documented emission pathway, little information is available on spatial and temporal variability of CH_4 emissions by diffusive fluxes. In tropical amictic and well-stratified reservoirs with CH_4-rich hypolimnion, the highest diffusive fluxes are usually observed during dry periods and when the stratification weaken at the beginning of the rainy season (Guerin and Abril, 2007). A study of CH_4 emissions from a dimictic reservoir suggests a potential large outgassing of CH_4 during the overturn (Utsumi et al., 1998b) as it is the case in natural monomictic and dimictic lakes (Kankaala et al., 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al., 2012;Fernández et al., 2014). Such hot moments of emissions (McClain et al., 2003) could contribute 45-80% of annual CH_4 emissions by diffusion (Schubert et al., 2012;Fernández et al., 2014). They are rarely taken into account in carbon budgets since they can only be captured by high frequency monitoring. Spatial heterogeneity of CH_4 emissions at the surface of reservoirs is also very high. It mostly depends on the spatial variations of ebullition that is controlled by sedimentation (DelSontro et al., 2011;Sobek et al., 2012;Maeck et al., 2013). The spatial variability of diffusion in reservoirs is less prominent with a few exceptions of higher emissions (1) in areas where dense forest is flooded (Abril et al., 2005), (2) at shallow sites (<10m) (Zheng et al., 2011;Sturm et al., 2014) and (3) at river inflows (Musenze et al., 2014). However, as it was shown for CO_2 emissions from a tropical hydroelectric reservoir, taking into account both spatial and temporal variability of emissions significantly affect positively or negatively carbon budgets and emission factors (Pacheco et al., 2015).

In the framework of a comprehensive project aiming at quantifying greenhouse gas emissions from the Nam Theun 2 Reservoir (NT2R), a recently flooded subtropical reservoir located in Lao People's Democratic Republic (PDR), we studied (1) the spatial and temporal variability of CH_4 ebullitive fluxes (Deshmukh et al., 2014) and (2) the downstream CH_4 emissions (Deshmukh et al., 2016). In the present study, the objective is to quantify the CH_4 diffusive

fluxes at the surface of NT2R and evaluate the significance of the diffusive fluxes in total methane emissions in a subtropical monomictic reservoir with a peculiar water intake that artificially mix the water column. The CH_4 emissions were quantified every two weeks during three and a half year (June 2009 to December 2012) based on a monitoring of CH_4 concentrations in the reservoir water column. This was performed at nine stations flooding different types of ecosystems. On the basis of these results, we discuss the spatial and temporal variations of the CH_4 emissions by diffusive fluxes and the significance of hotspots and hot moments in the total emissions from the surface of the reservoir.

2. Material and methods

2.1. Study area

The NT2 hydroelectric reservoir (17° 59' 49" N, 104° 57' 08" E) was built on the Nam Theun River located in the subtropical region of Lao PDR on the Nakai Plateau. A detailed description of this trans-basin hydroelectric reservoir located on the Nakai Plateau is given in Descloux et al. (2014). Basically, the Nam Theun River is dammed (Nakai Dam, ND in Figure 1) and the water from the Nam Theun River is diverted to the Xebang Fai watershed after passing through water intake (WI in Fig 1) to the powerhouse (PH in Fig 1). The WI is located in a 130 m-width and 9 to 20 m-deep channel on the southwest side on the reservoir and it is located 5 m above the bottom (Figure S1). The powerhouse is located in the valley 200 m below the plateau. The filling of the reservoir began in April 2008 with full water level reached by October 2009 and the power plant commissioned in April 2010.. After that date, turbines were turned on and water was continuously delivered to the turbines and downstream of the reservoir. 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. This low water discharge corresponds to the minimum water discharge of the Nam Theun River before the dam was built.

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 2009 to April 2010 (Figure 2a). After the commissioning (from April 2010 to December 2012) the reservoir surface varied seasonally by a factor of three 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 (Figure 2a). The average water volume is 2.65 km^3 with the lowest volume by the end of the WD season (0.71 in June 2011) and the highest at the end of the rainy seasons (3.97 km^3 in September 2011) (Figure 2b). The seasonal water level variations are about 10 m (Figure 2c), the average depth is 8 m for a maximum depth of 39 m close to the Nakai Dam.

2.2. Sampling strategy

A total of nine stations (RES1-9, Figure 1) located in the reservoir were monitored every two weeks (fortnightly) in order to determine the vertical profiles of temperature and O_2 and CH_4 concentration in the water column. The type of ecosystems flooded, the depth range and the hydrology 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 800 m upstream of the water intake (WI) delivering the water to the powerhouse (Figure 1 and Figure S1). The station RES9 is under the influence of the water column mixing induced by the water withdrawal at the WI, located at the bottom of the reservoir (5m above the bottom) and under 10 to 20 m of water (see discussion). All samples and in situ measurements were taken in the morning or early afternoon from an anchored boat. Most of the time, the boat was attached to a buoy at the sampling station. When no buoy was present, an anchor was used with care in order not to re-suspend surface sediments. As the sampling started from the surface, the bottom water was sampled almost an hour later and should not be influenced by the perturbation generated by the anchor.

2.3. Experimental methods

2.3.1. Vertical profiles of oxygen and temperature

Vertical profiles of O_2 and temperature were measured in situ at all sampling stations with a multi-parameter probe Quanta® (Hydrolab, Austin, Texas) since June 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 every two weeks from May 2009 to December 2012. Surface samples were taken with a surface custom-built water sampler (Abril et al., 2007). Other samples from the water column were taken with an Uwitec water sampler at 3m-depth, at the oxic-anoxic interface 1m above and below the oxic-anoxic interface and every 3 to 5m down to 0.5 m above the bottom. Water samples were stored without air bubble in serum glass vials, capped with butyl stoppers, sealed with aluminium crimps and preserved with $HgCl_2$ (Guerin and Abril, 2007). Samples were analysed within 15 days. Before gas chromatography analysis for CH_4 concentration, a N_2 headspace was created and the vials were vigorously shaken to ensure an equilibration between the liquid and gas phases. The concentration in the water was calculated using the solubility coefficient of Yamamoto et al. (1976).

2.3.3. Gas chromatography

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

2.4. Water column CH_4 storage

Between sampling depths of the vertical CH_4 profiles, concentrations were assumed to change linearly in order to calculate the concentration in each 1-m layer of water. The volume of water in each layer was calculated using the volume-capacity curve (NTPC, 2005). The CH_4

storage was calculated by multiplying the average CH_4 concentrations of each layer by the volume of the layer and summing-up the amount of CH_4 for all depth intervals.

2.5. Aerobic CH_4 oxidation

The depth-integrated CH_4 oxidation rates at each station were calculated on the basis of the specific oxidation rates (d^{-1}) determined at NT2R (Deshmukh et al., 2016) and vertical CH_4 and O_2 profiles in the water column as already described in (Guerin and Abril, 2007). The depth-integrated CH_4 oxidation rates at each station were estimated only from January 2010 since the vertical resolution of the vertical profiles of O_2 and CH_4 was not high enough in 2009.

As the aerobic methane oxidation rates we obtained were potential, $CH_{4\text{-}ox}$ were corrected for two limiting factors, the oxygen availability and the light inhibition as described in Guerin and Abril (2007). The final equation to compute in situ oxidation rates ($CH_{4\text{-}ox}$, mmol m^{-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 $CH_{4\text{-}ox}$ from Deshmukh et al. (2016); C_{O2}, the oxygen concentration; $K_{m(O2)}$, the half-saturation constant (Km) of O_2 for CH_4 oxidation, d, depth of the water layer and I(z), the inhibition of methanotrophic activity by light as defined by Dumestre et al. (1999) at the Petit Saut Reservoir. Finally, the CH_4 oxidation rates were integrated in the oxic water column, from the water surface to the limit of oxygen penetration.

2.6. Diffusive fluxes from surface concentrations

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

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

where F, the diffusive flux at water-air interface; $\Delta C = C_w - C_a$, the concentration gradient between the water (C_w) and the concentration at equilibrium with the overlying atmosphere (C_a) and k_T, the gas transfer velocity at a given temperature (T):

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

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 averaged 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 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 during three deployments at NT2R. 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. The average wind speed (at 10 m height) and rainfall from two meteorological stations located at the Ban Thalang Bridge (close to RES4 station,) and close to the WI (Figure 1) was used for the calculation of fluxes all stations (RES1-8). On average for all stations and all sampling date, the k_{600} was 5.6±5.3 cm h^{-1} ranging between 0.91 to 40.4 cm h^{-1}. The lowest k_{600} were calculated in the CD season (3.43±1.01 cm h^{-1}; 1.65-6.06 cm h^{-1}) while the highest were obtained during the WW season (6.78±6.33 cm h^{-1}; 1.57-40.42 cm h^{-1}) due to high rainfall (up to 113 mm day^{-1}). In the WD season k_{600} averaged 5.58±4.81 cm h^{-1}. This average k_{600} is significantly enhanced by some rainy events in late May-early June in 2010 and 2012 (up to 60 mm d^{-1}).

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 (Figure S1). 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 located downstream of the turbine where we visually observed vortexes similar to those observed at RES9. In the regulating

dam, the k_{600} obtained in May 2009 and March 2010 with a drifting floating chamber as described in Deshmukh et al. (2014) was 19 cm h^{-1} on average for 4 measurements ranging from 9 to 40 cm h^{-1}. 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 the regulating dam that we consider as an area with comparable hydrology/hydrodynamics.

2.7. Total emissions by diffusive fluxes

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

2.8. Statistical and correlation analysis

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

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

concentrations was used to determine the frequency distribution, the skewness of the dataset (third order moment), the auto-correlation of each time series and the correlation between the different stations. All analyses were performed using Matlab.

3. Results

3.1. Temperature and O_2 dynamics in the reservoir

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 3). 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 3) whereas in some occasions, the temperature decreased regularly from the surface to the bottom during sporadic destratification (RES1-3, Figure 3). 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 3) 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 (Figure 4a). 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 4a). During the WW season, the ΔT decreased gradually which means that the overturn occurred over several months.

During the WD season at the stations RES1-8, an oxycline 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 3). 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 3 and 4b). During the CD season (reservoir overturn), the water column was often oxygenated from the top to the bottom of the reservoir (Figure 3). On average over the whole reservoir, the lowest hypolimnic oxygen concentration was observed in 2010 before the reservoir was commissioned (Figure 4b).

After the commissioning of the reservoir and the turbines were powered on in April 2010, the water column located near the intake (RES9) completely mixed as indicated by the

299 homogeneous temperature and oxygen profiles with depth in every season (Figure 3). The
300 water column at RES9 was always well oxygenated (163 ± 62 μmol L^{-1}, Figure 3).

301 **3.2. Seasonal dynamics of the CH_4 concentration in the reservoir**

302 At the station RES1-8, when the water column is thermically stratified with a steep oxicline in
303 the WD and often in the WW seasons, CH_4 concentrations are on average ~150 times higher
304 in the reservoir hypolimnion (246 ± 234 μmol L^{-1}) than in the epilimnion (1.6 ± 7.7 μmol L^{-1})
305 (Figure 3). The gradient of CH_4 concentration at the thermocline/oxicline was steeper during
306 the WD season than during the WW season (Figure 3). During the CD season, the average
307 CH_4 concentration in the reservoir bottom water lowered by a factor of three compare to the
308 WD and the WW seasons. However, the reservoir overturn increased the average CH_4
309 concentrations in the epilimnion by a factor of two (3.4 ± 14.8 μmol L^{-1}) in comparison with
310 the WD and WW seasons. After the commissioning, the CH_4 vertical profiles of concentration
311 before turbine intake (RES9) were homogeneous from the surface to the bottom. The highest
312 average CH_4 concentration from the surface to the bottom peaked up to 215 μmol L^{-1} in July
313 2010 at this station. On a seasonal basis, the CH_4 concentration at RES9 averaged 39.8 ± 48.8,
314 29.9 ± 55.4 and 1.9 ± 4.3 μmol L^{-1} during the WD, WW and CD seasons, respectively (Figure
315 3). The concentrations at RES9 were up to 10 times lower than the maximum bottom
316 concentrations at the other stations for a given season. Since the station RES9 behaved
317 differently from the other stations, results from this station will be treated separately.

318 The overall bottom CH_4 concentration (Figure 4c) and dissolved CH_4 stock in the reservoir
319 (Figure 4d) increased at the beginning of the WD season. The higher bottom CH_4
320 concentration and storage in the reservoir are concomitant with the establishment of anoxia in
321 the hypolimnion and thermal stratification (Figure 4). Hypolimnic CH_4 concentration and
322 storage reached their maxima (up to 508 ± 254 μmol L^{-1} and 4.7 ± 0.5 Gg(CH_4), Figure 4c,d)
323 at the end of the WD-beginning of the WW season when the residence time of water in the
324 reservoir was the lowest (40 days, Figure 4d) and when the reservoir volume was the smallest
325 (Figure 2b). Along the WW season, the thermal stratification weakened (Figure 4a) and the
326 CH_4 concentration and dissolved CH_4 stock decreased (Figure 4c,d) while the residence time
327 of water increased (Figure 4d) and the water volume increased (Figure 2b). In the CD season,
328 the reservoir overturns as evidenced by the low ΔT and the penetration of O_2 to the
329 hypolimnion (Figure 4a,b). During CD season, the bottom CH_4 concentration and the storage
330 reached their minima (down to 1.3 ± 4.5 μmol L^{-1} and 0.01 ± 0.001 Gg(CH_4), Figure 4c,d)

when the residence time of water was the longest (Figure 4d). 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 4).

During the three and a half years of monitoring, the same seasonal pattern as described above is observed although the annual CH_4 bottom concentration and storage was threefold higher in 2009 and 2010 than in the year 2011 (Figure 4c,d). In the dry year 2012, the reservoir bottom CH_4 concentration and storage was almost twice higher than in wet year 2011.

3.3. Aerobic CH_4 oxidation in the reservoir

Between 2010 and 2012, the depth integrated aerobic CH_4 oxidation rates ranged between 0.05 and 380 mmol m^{-2} d^{-1} at the stations RES1-RES8 (Figure 5). 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. In the CD season of the year 2012, the aerobic oxidation rate were exceptionally high compare to the same season in the previous years.

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

The surface concentrations at the stations RES1-8 ranged from 0.02 to 150 μmol L^{-1} and were 2.0±10.5 μmol L^{-1} (median = 0.9), 1.5±5.5 μmol L^{-1} (median = 0.4) and 3.4±14.7 μmol L^{-1} (median = 0.2) on average for the CD, WD and WW season, respectively. The surface concentration followed a loglogistic distribution, which indicates the existence of extremely high values. This is confirmed by the fact that the skewness of the time series of the log of the CH_4 concentrations for all stations is positive (Figure S2), especially at the stations RES1, RES3 and RES7 for which the skewness is >1 (Figure S2). 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 in all season (Figure S3a). The normalized distribution of concentrations (in log) according to seasons (Figure 6) indicates that these high concentrations were observed without any clear seasonal trend at the station RES1, RES5 and RES6 (<1 up to 150 μmol L^{-1}). At the stations RES2 and RES3, the concentrations up to 128

μmol L^{-1} were mostly observed in the CD season when the reservoir overturns. At the station RES4 located at the Nam Xot and Nam Theun confluence and at the stations RES7 and RES8 both located in the inflow region of the Nam Theun River, the high surface concentrations (up to 64.60 μmol L^{-1}) were mostly observed during the WW season when the reservoir undergoes sporadic destratification. The auto-correlation function of the time series of the log of the surface CH_4 concentrations and diffusive fluxes at the stations RES1-8 indicated that all stations (except RES1) have a memory effect of 30 to 40 days (Figure S4). This implies that with a sampling frequency of 15 days, we captured most of the changes in the surface CH_4 concentrations. At station RES1, the changes in CH_4 concentrations are faster than at other stations and would have deserved a monitoring with a frequency higher than 15 days.

During the monitoring at RES1-RES8 stations, the average diffusive flux was 2.8 ± 12.2 mmol m^{-2} d^{-1} ranging from 0.01 to 201.86 mmol m^{-2} d^{-1} without any clear interannual and seasonal trends (Figure S3b). As for the concentrations, flux data followed a loglogistic distribution. The median flux in the WD season is 40 to 80% higher than the median in the WW and CD season, respectively. During the WW and the CD seasons, more than 60% of the calculated fluxes were lower than 1 mmol m^{-2} d^{-1}, which corresponds to classical flux in pristine rivers. However, the average fluxes in the WW and CD season are 30% higher than in the WD season (Table 2). This confirms the presence of extremely high values during WD and CD seasons, as expected from the surface concentrations. All seasons together, around 7% of the diffusive fluxes that we observed were higher than 5 mmol m^{-2} d^{-1} which corresponds to extremely high diffusive fluxes in comparison with data from the literature for reservoirs and lakes (Bastviken et al., 2008;Barros et al., 2011). The median and average of these extreme fluxes higher than 5 mmol m^{-2} d^{-1} were 2 times higher in the WW and CD seasons than in the WD season (Table 2).

At NT2R, diffusive CH_4 fluxes covered the whole range of fluxes reported for tropical reservoirs, depending on the season. Most of the fluxes at the NT2R Reservoir were around one order of magnitude lower than those at Petit Saut Reservoir (French Guiana) just after the impoundment (Galy-Lacaux et al., 1997), and in the same order of magnitude as reported for reservoirs 10 to 18 years older (Abril et al., 2005;Guerin et al., 2006;Kemenes et al., 2007;Chanudet et al., 2011). However, some diffusive fluxes at the stations RES1-8 in the WW and the CD seasons (up to 202 mmol m^{-2} d^{-1}) are among the highest ever reported at the

surface of a hydroelectric reservoir or a lake (Bastviken et al., 2011;Barros et al., 2011) and rivers downstream of dams (Abril et al., 2005;Guerin et al., 2006;Deshmukh et al., 2016).

3.5. 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 7a) located at the water intake were up to 30 times higher than at any other stations Following the commissioning of the reservoir and powering of the turbines, CH_4 concentrations at station RES9, which was located at the water intake, were up to 30 times higher than at the other stations (Figure 7a). On average CH_4 concentrations were 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 CD season ($p = 0.0002$ and Figure 7a). 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 7b 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.

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 3) 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

421 the oxic-anoxic interface (Bedard and Knowles, 1997;Bastviken et al., 2002;Guerin and Abril,
422 2007;Deshmukh et al., 2016).

423 CH_4 concentrations and storage increase concomitantly with the surface water temperature
424 and the establishment of the thermal stratification during the WD season and peak at the end
425 of the WD season-beginning of the WW season when the surface and the volume of the
426 reservoir was minimum (Figure 2, 3 and 4). The fact that the storage reached its maximum
427 when the reservoir volume is at its minimum shows that the increase of concentration at the
428 bottom of the reservoir is highly significant. During the WW season, CH_4 concentrations and
429 storage decrease slowly (Figure 4) while aerobic methane oxidation reaches its maximum
430 (Figure 5). When the reservoir overturns at the beginning of the CD season, the CH_4
431 hypolimnic concentrations and storage reach their minima (Figure 4). The overturn favours
432 the penetration of oxygen down to the bottom (Figure 3 and 4b). The sharp decrease of the
433 CH_4 concentrations and CH_4 storage during this period is expected to result from sudden
434 outgassing (Section 4.2) together with an enhancement of the aerobic CH_4 oxidation as
435 observed in lakes that overturn (Utsumi et al., 1998b;Utsumi et al., 1998a;Kankaala et al.,
436 2007;López Bellido et al., 2009;Schubert et al., 2010;Schubert et al., 2012;Fernández et al.,
437 2014). Significant increase in methane oxidation during overturn in the CD season was not
438 observed except for the year 2012 (Figure 5) when hypolimnetic CH_4 concentrations were still
439 quite high (Figure 4c,d). The absence of a clear enhancement of the CH_4 oxidation in the
440 water column of NT2R can be attributed to the slow erosion of the thermal stratification
441 before the reservoir really overturns.

442 As the reservoir overturns during the period over which the water residence time is the
443 longest, the temporal evolution of the concentrations is anti-correlated with the residence time
444 (Figure 4c,d). The seasonal dynamics of the CH_4 in the monomictic NT2R differs from
445 permanently stratified reservoirs like Petit Saut Reservoir where CH_4 concentration increased
446 with retention time (Abril et al., 2005). However, at the annual scale the water residence time
447 has a strong influence on CH_4 concentration and storage in the reservoir. Before the reservoir
448 was commissioned (April 2010), the water residence time was up to 4 years and the CH_4
449 storage was up to four times higher than in 2011 and 2012 (Figure 4d). Although a decrease
450 of concentration and storage with the age of the reservoir was expected (Abril et al., 2005),
451 storage in the dry year of 2012 was twice that of the wet year of 2011, likely due to a 25%
452 increase in residence time between 2011 and 2012 due to a decrease in rainfall and water
453 inputs. In wet years like 2011, the thermal stratification is weaker than in dry years since the

warming of the epilimnion is less efficient due to (1) lower insulation and cold water inputs from above and (2) the high riverine inputs of water alters the stability of the reservoir thermal stratification as shown by the sharper decrease of the thermal stratification illustrated by the decrease of the stratification index (ΔT) in 2011 than in 2012 (Figure 4a). As a consequence, the oxygen diffusion to the hypolimnion was higher in 2011 than in 2012 (Figure 4b) and it enhanced aerobic methane oxidation by 20% in the water column in the WW season in 2011 as compared to 2012 (Figure 5). Therefore our results suggest that the hydrology affects both the thermal stratification and therefore the diffusion of O_2 in the water column. The enhancement of O_2 penetration in rainy years favours the CH_4 oxidation and therefore contributes to the CH_4 storage reduction. With less CH_4 in the water column, the potentiality for downstream emissions (Deshmukh et al., 2016) and emissions through hotspots and hot moments (see below) is highly reduced.

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

Figure 8 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 S2) 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 in both the CD and WW seasons only when ΔT decreased sharply (>4°C, Figure 8a,d,g,j) and were usually followed by a sharp decrease of the CH_4 storage in the water column (Figure 8b,e,h,k).

Hot moments of emissions occurred during overturn in the CD at the stations RES1 and RES3 as illustrated in Figure 7. 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 8b,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 (see stations RES7 and 8 in Figure 8). This is contrasting with the observations in reservoir older 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 matter from the watershed (Musenze et al., 2014).

In the WD season, some sporadic emissions occurred but they were always lower than 20 mmol m^{-2} d^{-1} that is up to ten times lower than extreme fluxes in the WW and CD season. Those high fluxes occurred at RES3, RES7 and RES8 (Figure 8d,g,j) and were associated with ΔT variations lower than 2°C. The CH_4 storage decreases associated with these fluxes, however, were not as sharp as those observed during other seasons (Figure 8e,h,k). These high emissions were actually 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 (1) in the inflow region during the wet season and (2) in area away from inflow zone 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 8). 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 finally overturns as was 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. Therefore, during overturn in the CD season, a significant amount of CH_4 is oxidized, but 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 Pääjärvi 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 7b) whereas fluxes are 3 orders of magnitude lower in the CD season. These high fluxes are the highest reported at the surface of an aquatic ecosystem (Abril et al., 2005;Guerin et al., 2006;Bastviken et al., 2011;Barros et al., 2011;Deshmukh et al., 2016). To the best of our knowledge, this is the first time it is reported that the artificial mixing induced by the water intakes upstream of a dam or a power station enhance significantly emissions. At NT2R, the intake is located at the bottom of a narrow (130 m) and shallow channel (depth =9-20 m) on the side of the reservoir (Figure S1). This design enhances horizontal water current velocities,

550 the vertical mixing and therefore the emissions. The existence of such a hotspot at other
551 reservoirs might be highly dependant on the design of the water intake (depth among other
552 parameters) and its effect on the hydrodynamics of the reservoir water column.

553 **4.4. Estimation of total diffusive fluxes from the reservoir**

554 Yearly emissions by diffusive fluxes peaked at more than 9 Gg(CH_4) in 2010 when the
555 reservoir was commissioned and they decreased down to ≈ 5 Gg(CH_4) in 2011 and 2012
556 (Figure 9a and Table 3). Yearly integrated at the whole reservoir surface, these emissions
557 correspond to diffusive fluxes of 1.5 to 4 mmol m^{-2} d^{-1}. These emissions are significantly
558 lower than diffusive fluxes measured at the Petit Saut Reservoir during the first two years
559 after flooding but similar to those determined in the following years (Abril et al., 2005) and
560 values reported for diffusive fluxes from tropical reservoirs in Barros et al. (2011). In absence
561 of the extreme emissions (both hotspots and hot moments), diffusive emissions from NT2R
562 would have been one order of magnitude lower than emissions from tropical reservoirs as
563 expected from the lower flooded biomass compare to Amazonian reservoirs (Descloux et al.,
564 2011). Due to the specific dynamic of diffusive fluxes at NT2R with hotspots and hot
565 moments, diffusion at the reservoir surface contribute 18 to 27% of total emissions (Table 3)
566 that is significantly higher than at other reservoirs tropical reservoirs where it was measured
567 (See also Deshmukh et al., 2016).

568 Most of the increase of CH_4 emissions by diffusive fluxes from 4 to 9 Gg(CH_4) between 2009
569 and 2010 is due to very significant emissions of 2-3 Gg(CH_4) at the water intake after the
570 commissioning of the reservoir and resulting artificial mixing (Figure 9a). This increase might
571 be overestimated because we have no measurements between Jan. and April but this
572 overestimation might be reasonable since those months are usually associated with the lowest
573 emissions of the year (Figure 9b). After the commissioning, the outgassing of CH_4 was
574 triggered by the artificial mixing generated by the withdrawal of water from the reservoir to
575 the turbines. Although the area under the influence of the water intake is less than 1% of the
576 total area of the reservoir, emissions at the water intake contributed between 13 and 25% of
577 total diffusive emissions and 4 to 10 % if considering both ebullition and diffusion,
578 disregarding the year 2009 (Table 3). It is worth to note that emissions at this site are only
579 significant within 3-5 month per year at the end of the WD season-beginning of the WW
580 season when the storage of CH_4 reach its maximum in the reservoir (Figure 9b). This new
581 hotspot equals 20 to 40% of downstream emissions and contributes between 3 and 7% of total

582 emissions from NT2R surface when including ebullition and downstream emissions (Table 3
583 and Deshmukh et al. (2016)). Localized perturbation of the hydrodynamics, especially in
584 lakes or reservoirs with CH_4-rich hypolimnion, can generate hotspots of emissions
585 contributing significantly to the total emissions from a given ecosystem. These hotspots could
586 be found upstream of dams and water intake in reservoirs but also around aeration stations
587 based on air injection or artificial mixing that could be used for improving water quality in
588 water bodies (Wüest et al., 1992).

589 The contribution of extreme diffusive fluxes (with daily values being > 5 up to 200 mmol m^{-2}
590 d^{-1}) to total emission by diffusion range from 30 to 50% on a yearly basis (Figure 9a) and
591 from 40 up to 70% on a monthly basis (Figure 9b) although these hot moments represent less
592 than 10% of the observations during the monitoring. In the literature, the statistical
593 distribution of CH_4 emissions dataset always follows heavy-tailed and right skewed
594 distribution like the log-normal, the Generalized Pareto Distribution (Windsor et al.,
595 1992;Czepiel et al., 1993;Ramos et al., 2006;DelSontro et al., 2011) or loglogistic (this study)
596 which indicates that CH_4 emissions are always characterized by high episodic fluxes. The
597 quantification of emissions thus requires the highest spatial and temporal resolutions in order
598 to capture as many hot moments as possible. At a single station, there is a possibility that we
599 did not catch the peak of emissions but extreme emission events never lasted more than 2
600 months (3 consecutive sampling dates) and probably lasted less than 15 days most of the time
601 (Figure 8). The auto-correlation function of the concentration time series indicates that a
602 minimum sampling frequency of 1 month is required in this monomictic reservoirs for an
603 accurate description of the change in the surface concentrations and estimation of the
604 emissions (Figure S4). Although a better temporal resolution than 15 days or monthly would
605 probably improve the estimation of CH_4 emissions from this reservoir, a lower temporal
606 resolution could significantly affect (positively or negatively) the emission factor of this
607 reservoir that overturn gradually over several month. Therefore, the monthly frequency
608 defined for this specific reservoir is probably not applicable to every aquatic ecosystem,
609 especially in lakes or reservoirs that overturn within a week or less (Kankaala et al.,
610 2007;López Bellido et al., 2009;Schubert et al., 2012). However, it suggests that
611 quantification of emissions based on 2-4 campaigns in a year might significantly affect
612 emissions factors and carbon budgets of ecosystems under study.

5. *Conclusion*

The monitoring of CH_4 diffusive emissions every two weeks 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 far from 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 suggest that sporadic emissions cannot be integrated properly in the quantification of emissions and establishments of carbon budgets based only on seasonal sampling (2-4 campaigns).

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.

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802
803 Table 1: Characteristics of the nine monitoring stations in the Nam Theun 2 Reservoir
804

Station	Depth (min-max) (m)	Flooded ecosystem[1]	Hydrology	Water residence time index[2]
RES1	25.4-35	Dense forest	100 m upstream of the Nakai Dam	**
RES2	18.4-28	Dense forest	Thalweg of the Nam Theun River	**
RES3	6.4-16	Dense forest	Embayment	***
RES4	17.4-27	Degraded forest	Confluence Nam Theun-Nam Xot Rivers	**
RES5	8.4-18	Degraded forest	Aside from the main stream	**
RES6	15.4-25	Degraded forest	Thalweg of the Nam Theun River	*
RES7	5.4-15	Swamp	Between inflows and water intake	*
RES8	11.4-21	Agricultural soils	Between inflows and water intake	*
RES9	9.4-19	Civil construction	Water intake	*

805 [1]Descloux et al. (2011)

806 [2]Water renewal index in arbitrary units, (***) stands for longer residence time, (**) for
807 average residence times and (*) for shorter residence times than average for the whole
808 reservoir
809

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

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

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 (Ebullition + diffusion at all stations)	15.66±1.02	23.73±2.32	18.39±0.82	17.25±1.09
Contribution of RES9 (%) to total emissions from reservoir	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. (2016)

Figure captions

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

Figure 2: Variations of (a) surface of the reservoir (km^2), volume of water (km^3) and water level (masl) at the Nam Theun 2 Reservoir between January 2009 and December 2012.

Figure 3: 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. Note that for the stations RES3 and RES9, the scale is different for the vertical profiles of CH_4.

Figure 4: (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 5: 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., 2016). 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 6: 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 7: (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 8: (a, d, g, j) stratification index (ΔT, red line, see text) and diffusive fluxes, (b,e,h,k) CH_4 storage and (c,f,i,l) depth-integrated aerobic CH_4 oxidation (mmol m^{-2} d^{-1}, black line) calculated from the aerobic oxidation rates determined by (Deshmukh et al., 2016) and ΔT (red line) between June 2009 and December 2012 at the stations RES1, RES3, RES7 and RES8 at the Nam Theun 2 Reservoir. Julian day 0 is 1st of January, 2009. The red, grey and blue colour dots indicate the warm dry (WD), warm wet (WW) and cold dry (CD) seasons, respectively.

Figure 9: (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.

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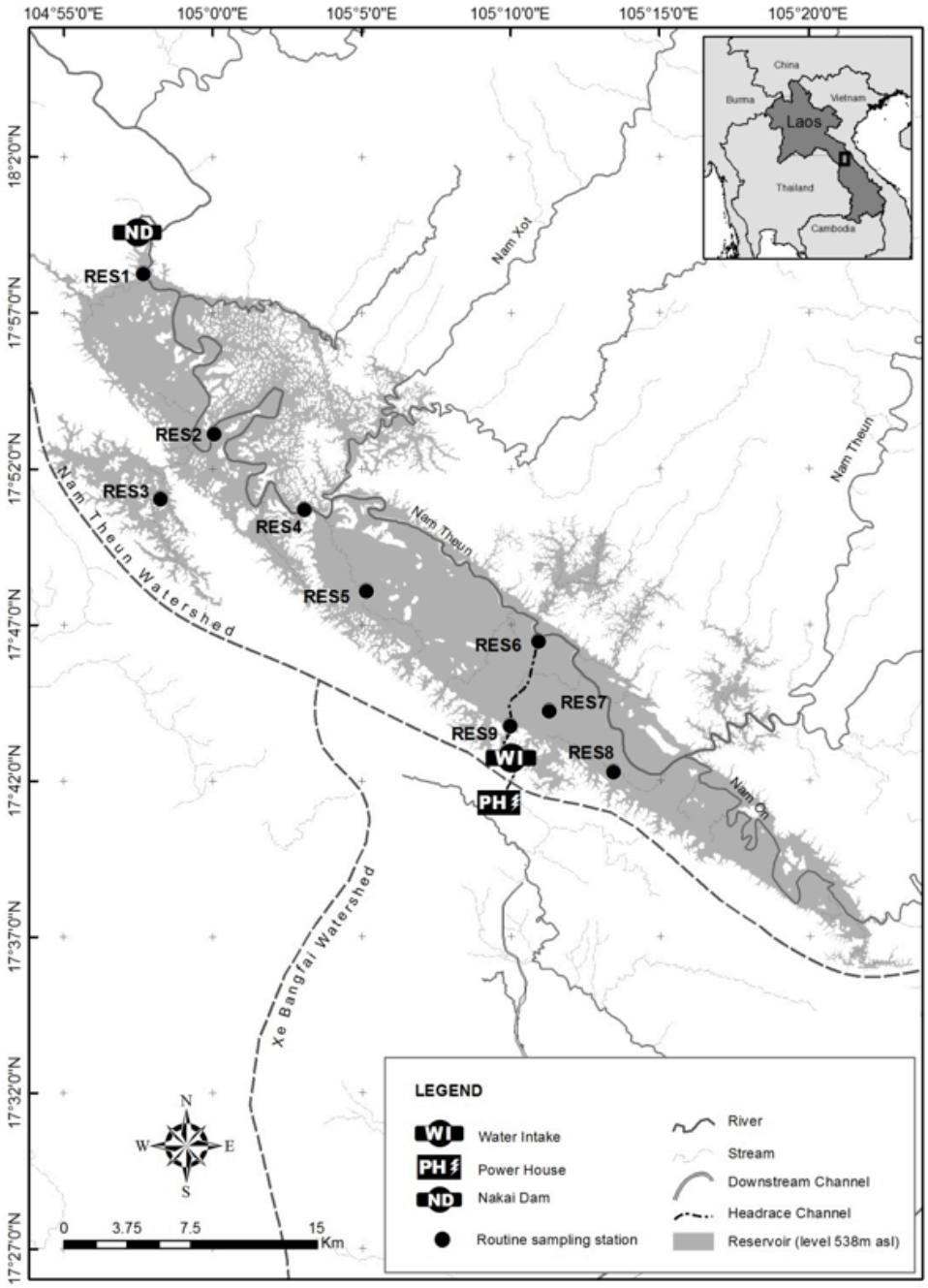

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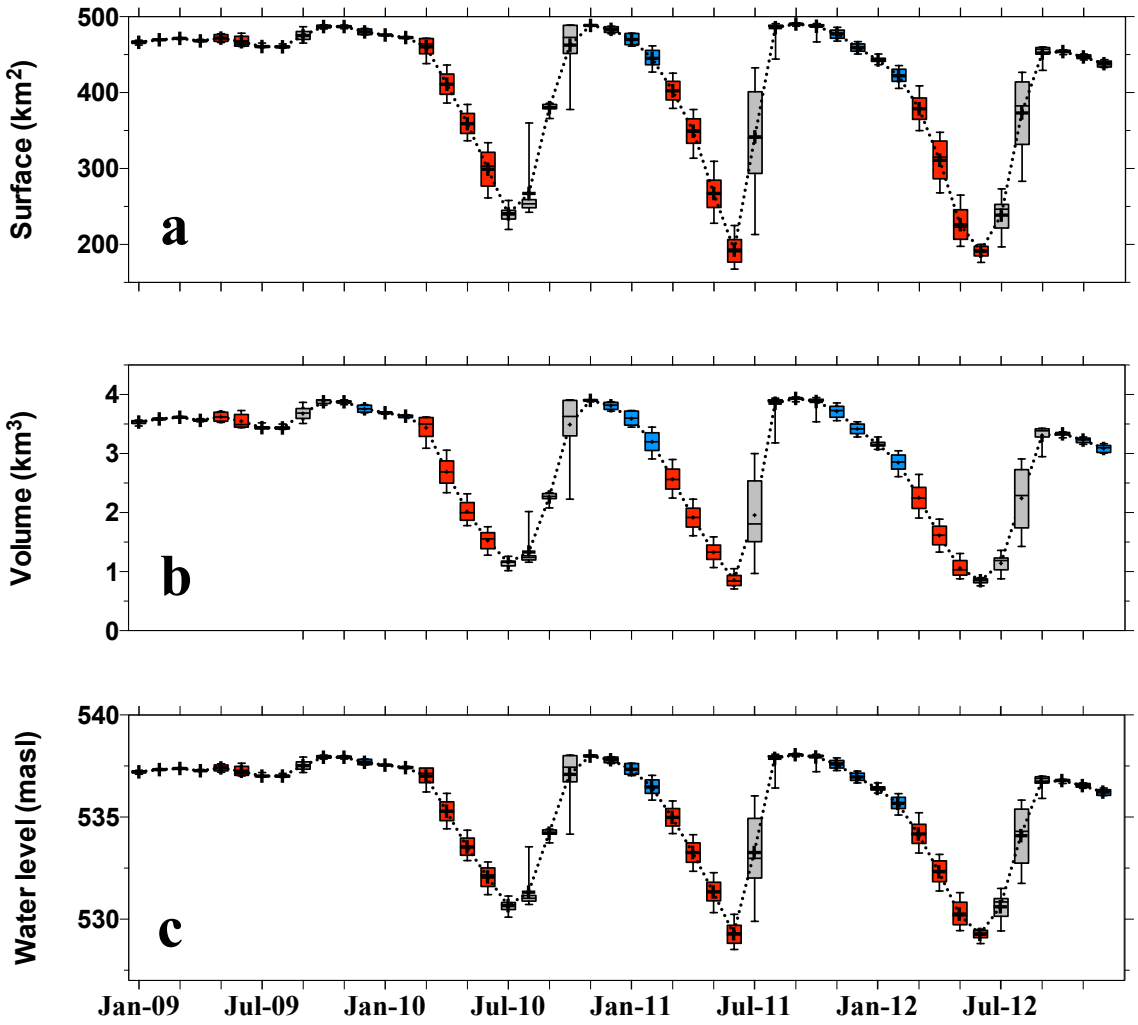

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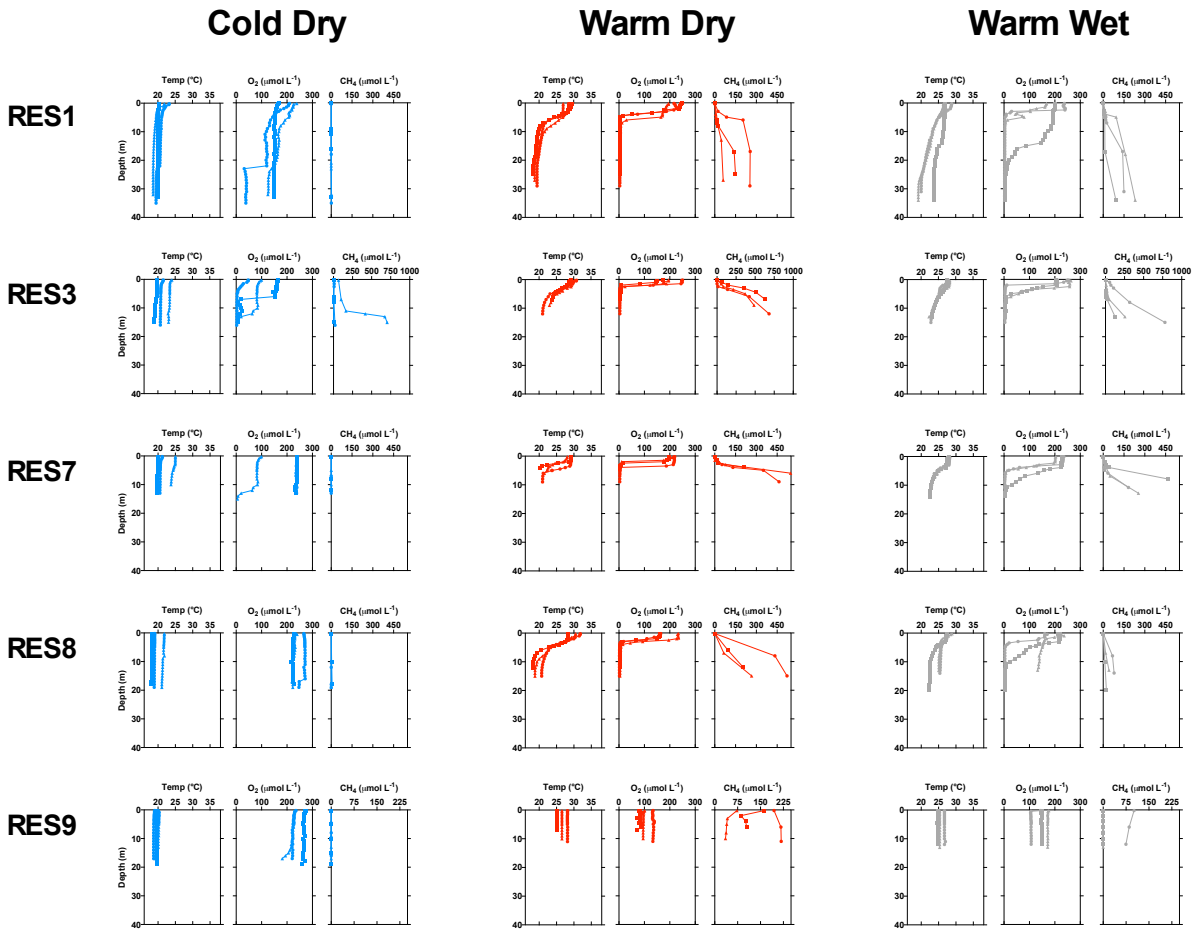

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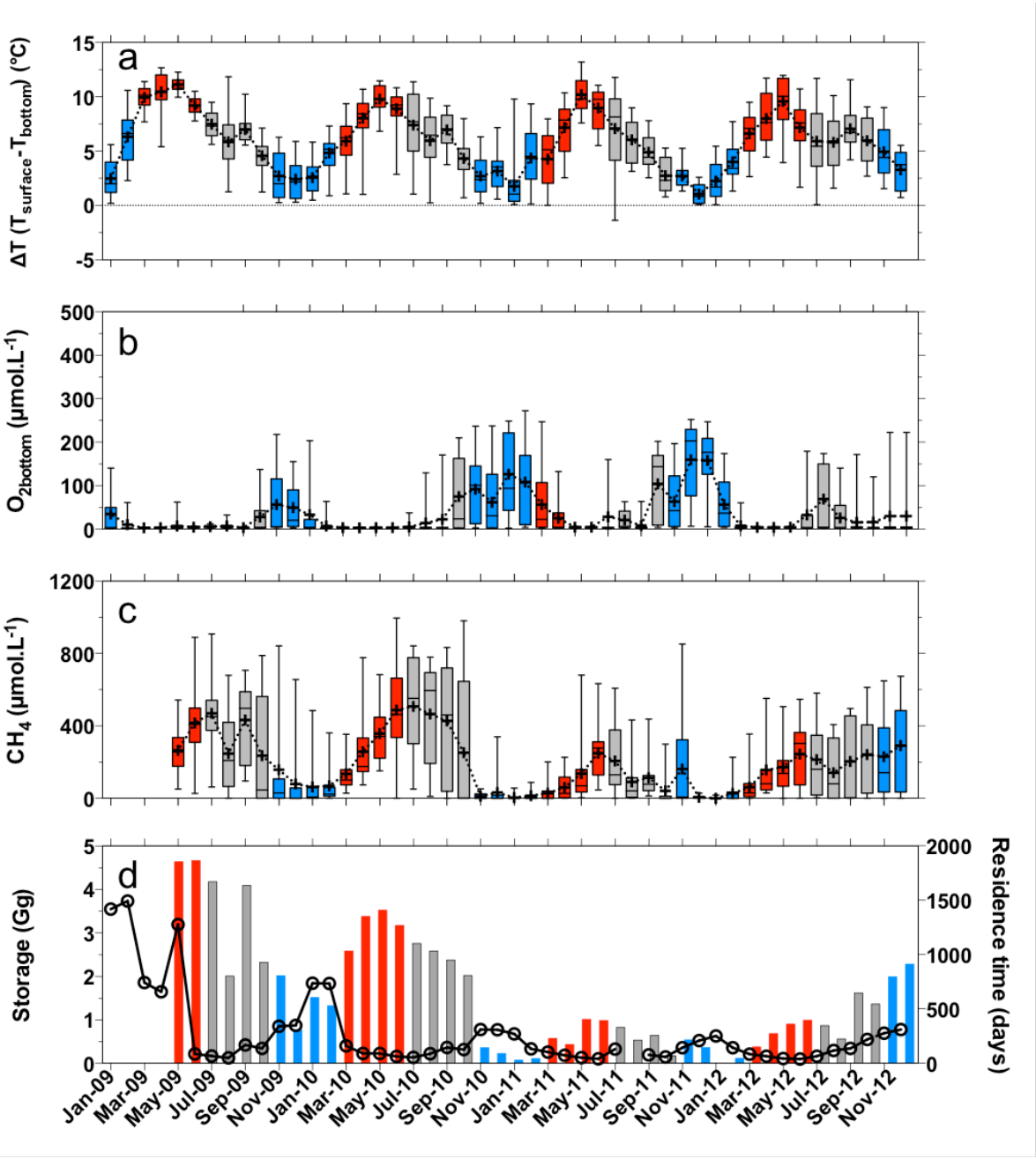

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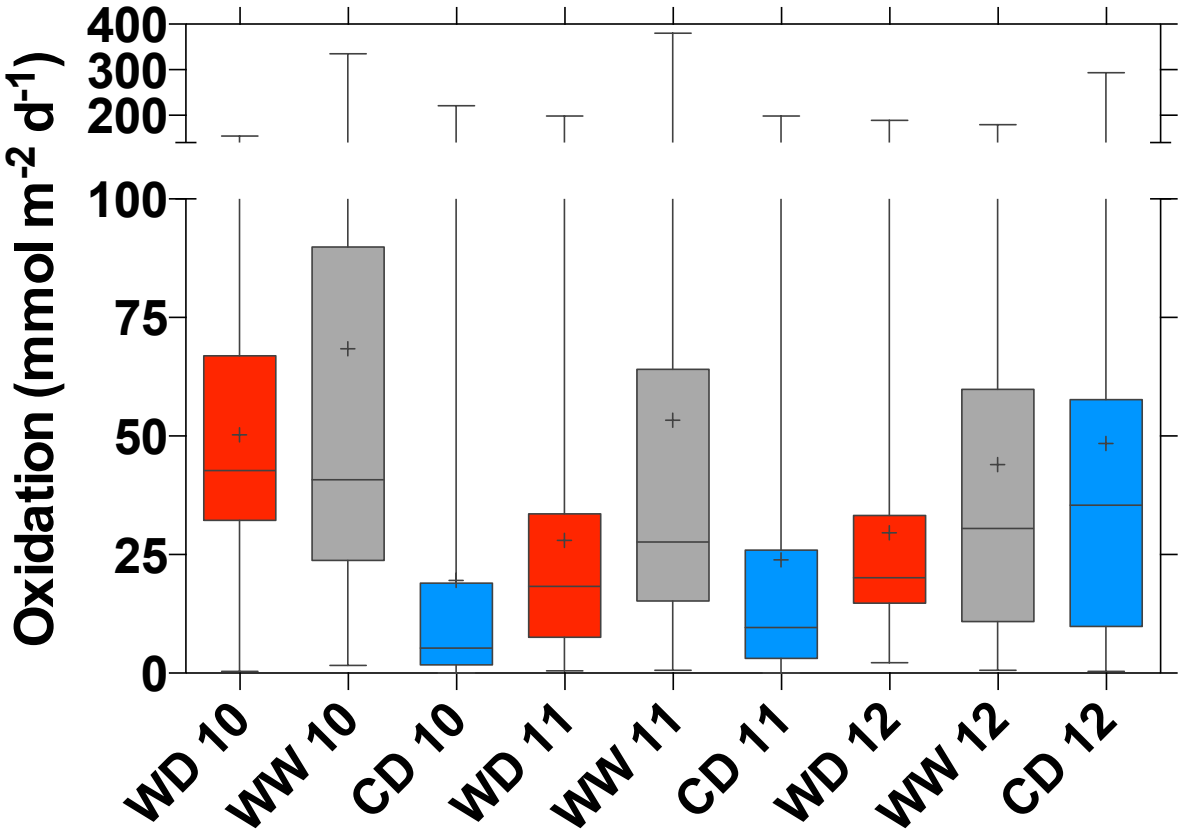
Figure 5


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

903
904 Figure 6
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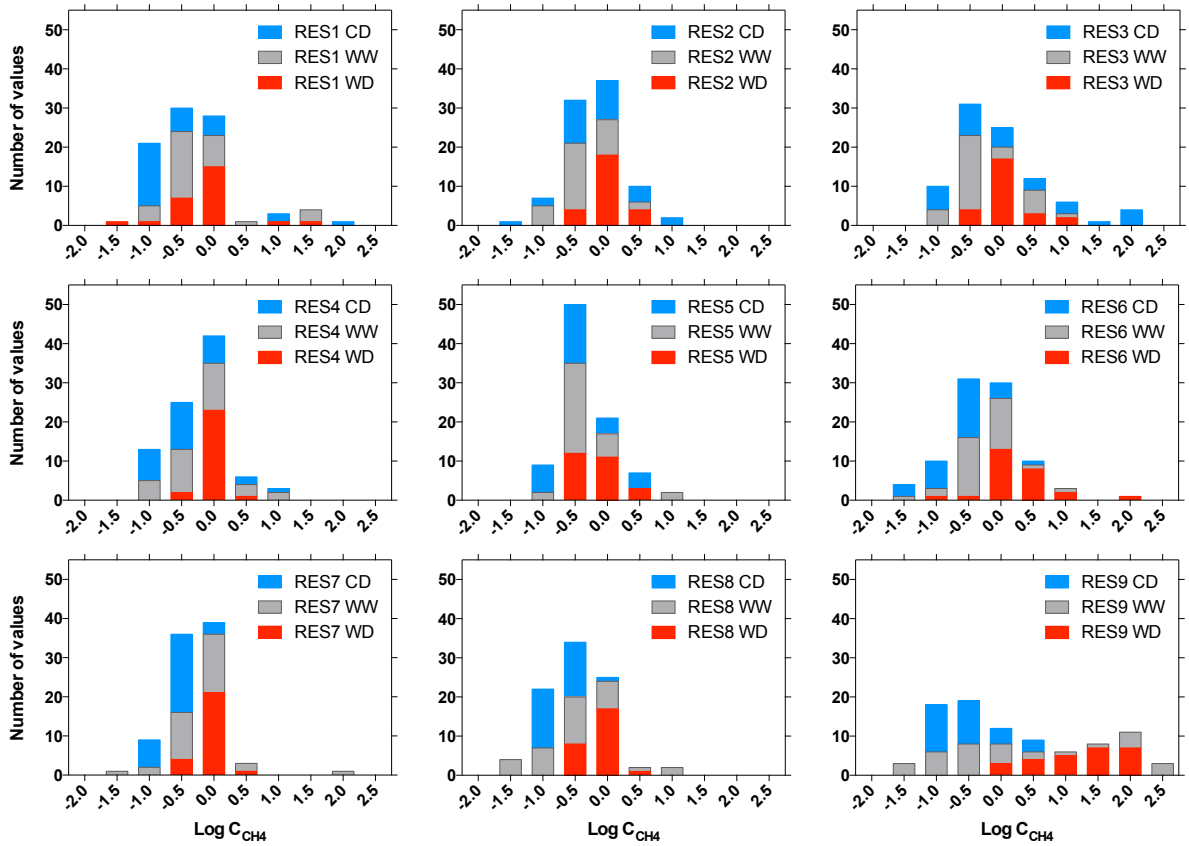

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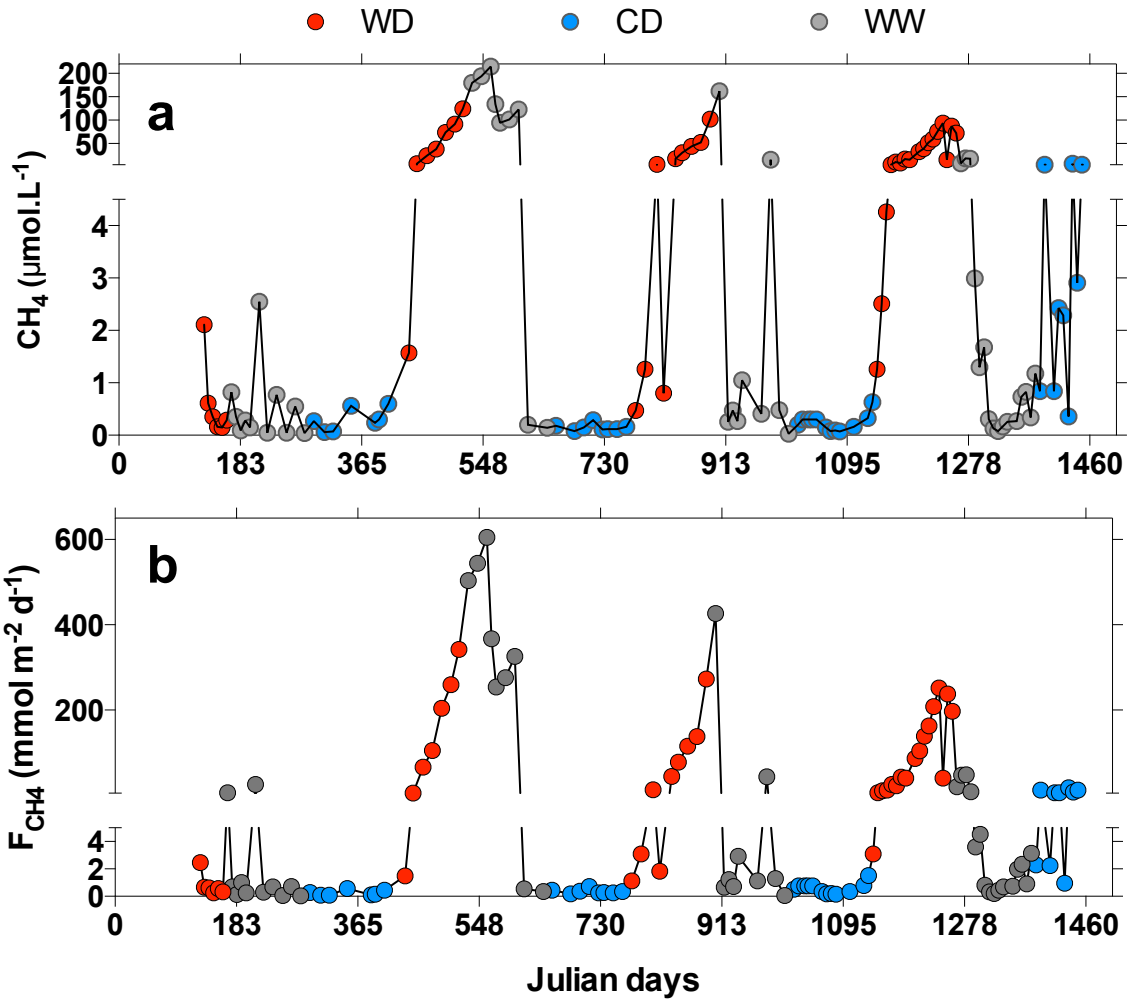

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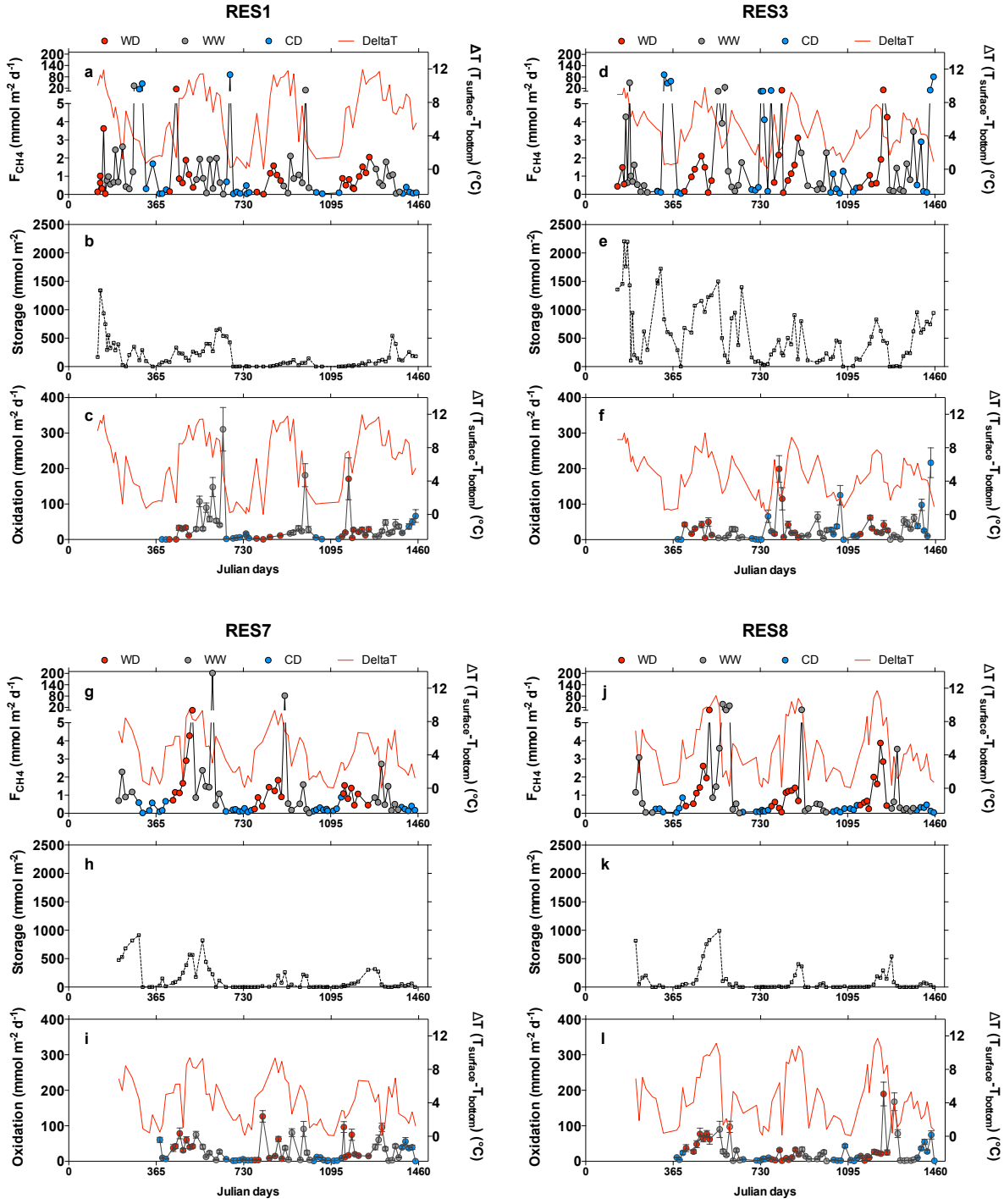

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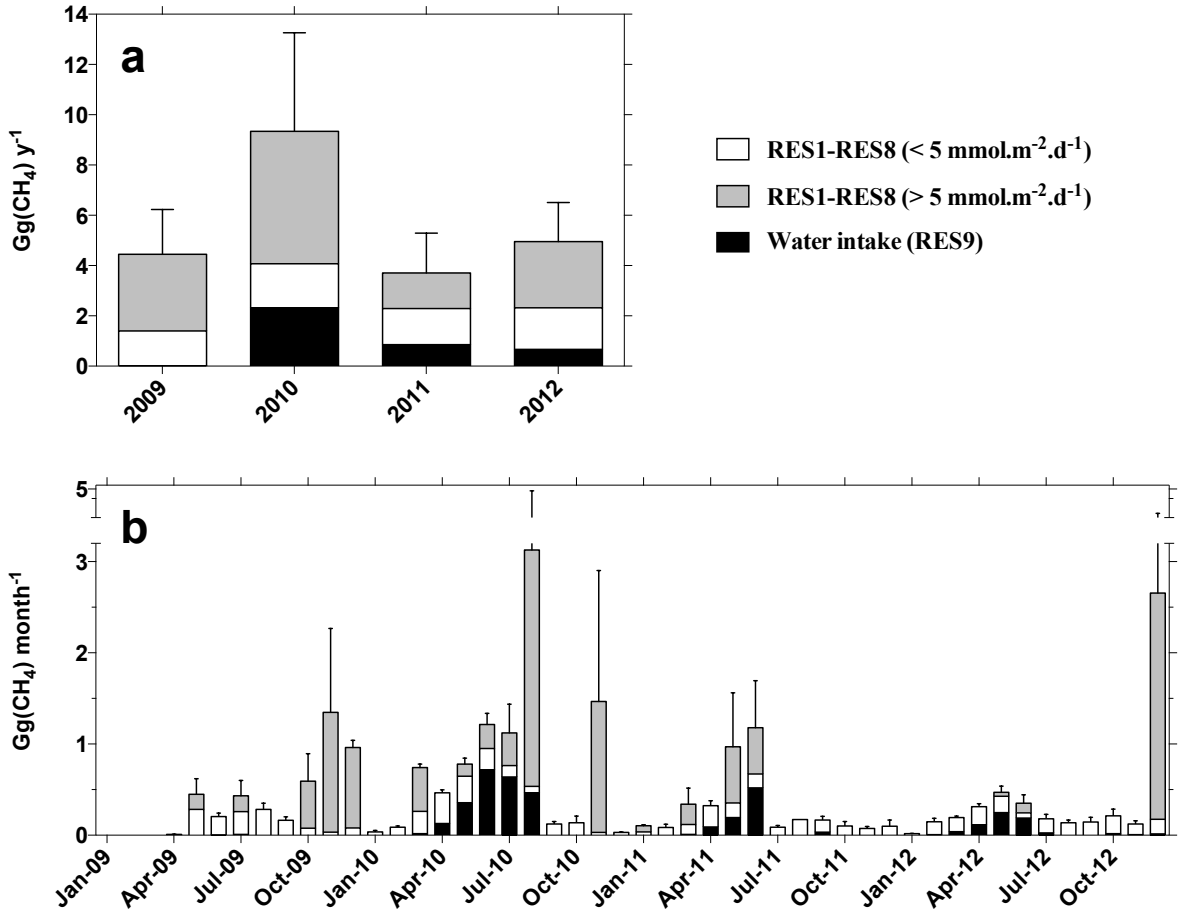

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