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

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## Abstract

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

We measured fortnightly $CH_4$ concentrations and physico-chemical parameters at nine stations in a subtropical monomictic reservoir which was flooded in 2008 (Nam Theun 2 Reservoir, Lao PDR). Based on these results, we quantified $CH_4$ storage in the water column and diffusive fluxes from June 2009 to December 2012. We also compared emissions with aerobic methane oxidation calculated from Deshmukh et al. (2015).

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

During the transition between the WD and WW seasons, a new hotspot of emissions was identified upstream of the water intake where diffusive fluxes peaked at 600 $mmol\,m^{-2}\,d^{-1}$ in 2010 down to 200 $mmol\,m^{-2}\,d^{-1}$ in 2012. In the CD season, diffusive fluxes from this area were the lowest observed at the reservoir surface. Emissions from this area contributed 15–25 % to total annual emissions although they occur on






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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 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; Guérin et al., 2006; Kemenes et al., 2007; Teodoru et al., 2012; Maeck et al., 2013; Deshmukh et al., 2015), $CH_4$ ebullition (Del-Sontro et al., 2010; Deshmukh et al., 2014) and emissions from the drawdown area of reservoirs (Chen et al., 2009, 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 variabilities of $CH_4$ emissions by diffusive fluxes. In tropical amictic reservoirs, the highest diffusive fluxes are usually observed during dry periods and when the stratification weaken at the beginning of the rainy season (Guérin and Abril, 2007). A study of $CH_4$ emissions from a dimictic reservoir suggests a potential large outgassing of $CH_4$ during the reservoir overturns (Utsumi et al., 1998b) as it is the case in natural monomictic and dimictic lakes (Kankaala et al., 2007; López Bellido et al., 2009; Schubert et al., 2010, 2012; Fernández et al., 2014). Such hot moments of emissions (McClain et al., 2003) could contribute 45–80 % of $CH_4$ annual emissions by diffusion (Schubert et al., 2012; Fernández et al., 2014). They are rarely taken into account in carbon budgets since they can only be captured by high frequency monitoring. Spatial heterogeneity of $CH_4$ emissions at the surface of reservoirs is also very high. It mostly depends on the spatial vari-





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ations of ebullition that is controlled by sedimentation (DelSontro et al., 2011; Sobek et al., 2012; Maeck et al., 2013). The spatial variation of diffusion appears to be low with emissions being slightly higher (1) in area where dense forest is flooded as compare to the former riverbed (Abril et al., 2005), (2) at shallow sites than at deeper ones (Zheng et al., 2011; Sturm et al., 2014) and (3) in inflow zones of reservoirs compare to the main body (Musenze et al., 2014). However, as it was shown for $CO_2$ emissions from a tropical hydroelectric reservoir, taking into account both spatial and temporal variability of emissions significantly affect carbon budgets and emission factors (Pacheco et al., 2015).

The objective of the study is to quantify the $CH_4$ diffusive fluxes at the surface of a newly flooded subtropical reservoir (Nam Theun 2 Reservoir, NT2R). The $CH_4$ emissions were quantified fortnightly during three and a half year (May 2010 to December 2012) based on a monitoring of $CH_4$ concentrations that started in June 2009. This was performed at nine stations flooding different types of ecosystems. On the basis of these results, we discuss the spatial and temporal variations of the $CH_4$ emissions by diffusive fluxes and the significance of hotspots and hot moments in the total emissions from the surface of the reservoir.

## 2 Material and methods

### 2.1 Study area

The NT2 hydroelectric reservoir was built on the Nam Theun River located in the subtropical region of Lao People's Democratic Republic (Lao PDR) on the Nakai Plateau. A detailed description of the study site is given in Descloux et al. (2014). The filling of the reservoir began in April 2008, the full water level was first reached in October 2009 and the power plant was commissioned in April 2010. Annually, the NT2 Reservoir receives around 7527 $Mm^3$ of water from the Nam Theun watershed, which is more than twice the volume of the reservoir (3908 $Mm^3$). A continuous flow of 2 $m^3 s^{-1}$ (and occa-





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sionally spillway release) is discharged from the Nakai Dam (ND in Fig. 1) to the Nam Theun River. The water used for electricity production is delivered from water intake (WI in Fig. 1) to the powerhouse (PH in Fig. 1). The powerhouse is located in the valley 200 m below the plateau.

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

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

### 2.2 Sampling strategy

A total of nine stations (RES1–9, Fig. 1) located in the reservoir were monitored fortnightly in order to determine the vertical profiles of physico-chemical parameters of the water column and the $CH_4$ concentrations. The characteristics of the stations are given in the Table 1. Basically, three stations are located on the thalweg of the former Nam Theun River (RES2, RES4, RES6) whereas four other stations are located in a small embayment in the flooded dense forest (RES3), flooded degraded forest (RES5), flooded swamp area (RES7) and flooded agricultural land (RES8). The RES1 station is located 100 m upstream of the Nakai Dam, and RES9 station is located ~ 1 km upstream of the water intake delivering the water to the powerhouse.





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## 2.3 Experimental methods

### 2.3.1 In situ physico-chemical parameter

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

### 2.3.2 Methane concentration in water

The evolution of $CH_4$ concentrations has been monitored from May 2009 to December 2012 on a fortnightly basis. Surface 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). 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 %.





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## 2.4 Water column $CH_4$ storage

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

## 2.5 Aerobic $CH_4$ oxidation

The depth-integrated $CH_4$ oxidation rates at each station were calculated on the basis of the specific oxidation rates ($d^{-1}$) determined at NT2 (Deshmukh et al., 2015) and the vertical profiles of $CH_4$ and $O_2$ concentrations in the water column as already described in Guérin 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.

## 2.6 Estimation of diffusive fluxes from surface concentrations

The diffusive $CH_4$ fluxes were calculated from the fortnightly monitoring of surface concentrations with the thin boundary layer (TBL) equation at all stations in the reservoir (RES1–9). The concomitant water and air $CH_4$ concentrations were applied in Eq. (1) to calculate diffusive flux:

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

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





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was computed from $k_{600}$ with the following equation:

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

with $\mathrm{Sc}_T$, 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 ($< 3.7\,\mathrm{m\,s^{-1}}$) or 1/2 for higher wind speed and turbulent water (Jahne et al., 1987).

For the determination of $k_{600}$, we used the formulations of from Guérin et al. (2007) and MacIntyre et al. (2010). As shown by Deshmukh et al. (2014), the average of the fluxes obtained from these two relationships compared well with fluxes measured at the reservoir surface. For calculation purpose, wind speed (at 10 m height) and rainfall from two adjacent meteorological stations located at Nakai Village (close to RES9 station) and at the Ban Thalang Bridge (close to RES4 station, Fig. 1) were used. In these stations, the average $k_{600}$ was $6.5\,\mathrm{cm\,h^{-1}}$ over the course of the year. At the water intake (RES9), it was impossible to quantify the $k_{600}$ since the boat drifted quickly to the shoreline because of water currents in the narrow channel. According to Chanudet et al. (2012), water current velocity in this area of the reservoir is about $0.2\,\mathrm{m\,s^{-1}}$. After Borges et al. (2004), the contribution of such water currents in a water body with depth ranging from 9 to 20 m is $2 \pm 0.5\,\mathrm{cm\,h^{-1}}$ which should be summed up with the contribution of wind, rainfall and buoyancy flux from Guerin et al. (2007) and MacIntyre et al. (2010). It gives an average of $9\,\mathrm{cm\,h^{-1}}$. The $k_{600}$ was determined in the regulating dam (Deshmukh et al., 2015) located downstream of the turbine where we observed vortexes similar to those observed at RES9. In the regulating dam where we observed the same vortexes as in RES9, the $k_{600}$ was $19\,\mathrm{cm\,h^{-1}}$ on average for 4 measurements. In order to be conservative for the estimation of emissions from the water intake, we considered a constant value of $k_{600}$ ($10\,\mathrm{cm\,h^{-1}}$) 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) $k_{600}$ determined in area with comparable hydrology/hydrodynamics.





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

Based on physical modelling (Chanudet et al., 2012), it has been showed that the station RES9 located at the water intake is representative of an area of ~ 3 $km^2$ (i.e. 0.6 % of reservoir water surface), whatever the season. This 3 $km^2$ 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. Since the Kolmogorv–Sminrov test indicated the non-normal distribution of the data, Kruskal–Wallis and Mann–Whitney tests were performed with GraphPad Prism (GraphPad Software, Inc., v5.04).

For each station, the time series of the log of the $CH_4$ surface concentrations were linearly interpolated and re-sampled every 15 days. The log of the concentrations were 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 log-time series from different stations. All analysis were performed using Matlab.





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## 3 Results

### 3.1 Temperature and $O_2$ dynamics in the reservoir water column

During the three and half year of monitoring at the stations RES1–8, the NT2R was thermally stratified with a thermocline at 4.5 ± 2.6 m depth in the WD (February–June) season as revealed by the vertical profiles of temperature (Fig. 2). In the WW season, the temperature vertical profiles at the stations RES1–8 either showed a thermocline (RES7 and RES8 in 2010 and 2011, Fig. 2) whereas in some occasions, the temperature decreased regularly from the surface to the bottom during sporadic destratification (RES1–3, Fig. 2). On average during the WW season, a thermocline was located at 5.8 ± 4.8 m depth. During the CD season, the reservoir overturned as already mentioned by Chanudet et al. (2012) and the temperature was constant from the surface to the bottom (Fig. 2) in the different years. In order to illustrate the destratification, a stratification index ($\Delta T$) which corresponds to the difference between the surface and bottom water temperature was defined. During the periods of stratification in the WD seasons, $\Delta T$ was up to 10 °C higher than during reservoir overturn in the CD season with $\Delta T$ close to zero (Fig. 3a). During the WW season, the $\Delta T$ decreased gradually.

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

After the commissioning of the reservoir (April 2010), the water column located near the water intake (RES9) got totally mixed as revealed by the homogeneous temperature





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and oxygen profiles from the surface to the bottom whatever the season (Fig. 2). The water column at RES9 was always well oxygenated ($163 \pm 62\,\mu mol\,L^{-1}$, Fig. 2).

### 3.2 Seasonal dynamics of the $CH_4$ concentration and storage in the reservoir water column

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

The overall bottom $CH_4$ concentration (Fig. 3c) and dissolved $CH_4$ stock in the reservoir (Fig. 3d) increased at the beginning of the WD season. The higher bottom $CH_4$ concentration and storage in the reservoir are concomitant with both the establishment of anoxia in the hypolimnion and the reservoir thermal stratification (Fig. 3). Hypolimnic $CH_4$ concentration and storage reached their maxima (up to $508 \pm 254\,\mu mol\,L^{-1}$ and $4.7 \pm 0.5$ Gg ($CH_4$), Fig. 3c and d) at the end of the WD-beginning of the WW season when the residence time of water in the reservoir was the lowest (40 days, Fig. 3d). Along the WW season, the thermal stratification weakened (Fig. 3a) and the $CH_4$ concentration and dissolved $CH_4$ stock decreased (Fig. 3c and d) while the residence time





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of water increased (Fig. 3d). In the CD season, the reservoir overturns as evidenced by the low $\Delta T$ and the penetration of $O_2$ to the hypolimnion (Fig. 3a and b). During CD season, the bottom $CH_4$ concentration and the storage reached their minima (down to $1.3 \pm 4.5\,\mu mol\,L^{-1}$ and $0.01 \pm 0.001\,Gg(CH_4)$, Fig. 3c and d) when the residence time of water was the longest (Fig. 3d). The sharp decrease of $CH_4$ storage and concentration in the transition from the WW to the CD seasons is concomitant with a sharp increase of $O_2$ concentration at the bottom (up to $160 \pm 89\,\mu mol\,L^{-1}$, Fig. 3).

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

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

### 3.4 Aerobic $CH_4$ oxidation in the reservoir

Between 2010 and 2012, the depth integrated aerobic $CH_4$ oxidation rates ranged between 0.05 and $380\,mmol\,m^{-2}\,d^{-1}$ at the stations RES1–8 (Fig. 4). On average, aerobic oxidation was higher in the WW season ($55 \pm 63\,mmol\,m^{-2}\,d^{-1}$) than in the CD ($30 \pm 46\,mmol\,m^{-2}\,d^{-1}$) and WD ($36 \pm 32\,mmol\,m^{-2}\,d^{-1}$) seasons and it was not statistically different for the three years. In the WD season, aerobic $CH_4$ oxidation was on average twice higher in 2010 than for the two following years whereas in the CD season, the highest aerobic oxidation rate was observed in 2012.





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### 3.5 Spatial and seasonal variability of surface $CH_4$ concentration and diffusive fluxes at the reservoir surface (RES1–8)

The statistical analysis of the time series of the log of the surface $CH_4$ concentrations at the stations RES1–8 with the auto-correlation function indicated that at all stations (except RES1) have a memory effect of 30 to 40 days (Supplement Fig. S1) which implies that with a sampling frequency of 15 days we capture most of the changes in the surface $CH_4$ concentrations. At the station RES1, the changes in $CH_4$ concentrations are faster than at other stations and would have deserved a monitoring with a frequency higher than 15 days.

The surface concentrations at the stations RES1–8 ranged from 0.02 to 150 $\mu mol\,L^{-1}$ and were $2.0 \pm 10.5\,\mu mol\,L^{-1}$ (median = 0.9), $1.5 \pm 5.5\,\mu mol\,L^{-1}$ (median = 0.4) and $3.4 \pm 14.7\,\mu mol\,L^{-1}$ (median = 0.2) on average for the CD, WD and WW season, respectively. Over the course of the three and a half year of survey, no clear temporal and spatial trends were observed (Fig. S2a). The surface concentrations were not statistically different between all stations and no statistically significant seasonal variations were observed The skewness of the time series of the log of the $CH_4$ concentrations for all stations is positive (Fig. S3) indicating the existence of extremely high values, especially at the stations RES1, RES3 and RES7 for which the skewness is $> 1$. The normalized distribution of concentrations (in log) according to seasons (Fig. 5) indicates that these high concentrations were observed without any clear seasonal trend at the station RES1, RES5 and RES6 ($< 1$ up to 150 $\mu mol\,L^{-1}$). At the stations RES2 and RES3, the concentrations up to 128 $\mu mol\,L^{-1}$ 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 $\mu mol\,L^{-1}$) were mostly observed during the WW season when the reservoir undergoes sporadic destratification.





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During the monitoring at RES1–8 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 (Fig. S2b). The median flux in the WD season is 40 to 80 % higher than the median in the WW and CD season, respectively. However, the average fluxes in the WW and CD season are 30 % higher than in the WD season (Table 2). This confirms the presence of extremely high values during WD and CD seasons, as expected from the surface concentrations. All seasons 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 NT2, 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 the ones 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 older by 10 to 18 years (Abril et al., 2005; Guérin 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; Guérin et al., 2006; Deshmukh et al., 2015).

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

After the commissioning of the reservoir (Julian day 450), the concentrations at the stations RES9 (Fig. 6a) located at the water intake were up to 30 times higher than at any other stations that is 36.6 ± 35.8 µmol $L^{-1}$ (median = 24.3), 37.6 ± 67.0 µmol $L^{-1}$ (median = 0.9) and 1.0 ± 1.7 µmol $L^{-1}$ (median = 0.3) in the WD, WW and CD season, re-





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spectively. The surface concentrations at RES9 were significantly higher in the WD and WW seasons than in the WW and CD seasons ($p$ = 0.0002 and Fig. 6a). The highest concentration was observed each year at the end of the WD season-beginning of the WW season in between June and August. These maxima decreased from 215 µmol $L^{-1}$ in August 2010 to 87 µmol $L^{-1}$ in June 2012.

The diffusive fluxes ranged between 0.03 and 605.38 mmol $m^{-2}$ $d^{-1}$ (Fig. 6b and Table 2). On average, the $CH_4$ diffusive fluxes at RES9 were two to forty times higher than at the other stations in the CD, WD and WW season. Diffusive fluxes at this station are usually higher than 10 mmol $m^{-2}$ $d^{-1}$ from April to July that corresponds to the WD season and the very beginning of the WW season. In 2010, diffusive fluxes were on average 241 ± 219 and 239 ± 228 mmol $m^{-2}$ $d^{-1}$ respectively for the WD and WW seasons. In 2011 and 2012, the fluxes dropped down by a factor of two in the WD season (112 ± 110 mmol $m^{-2}$ $d^{-1}$) and almost by a factor of forty in the WW season (6.8 ± 14.4 mmol $m^{-2}$ $d^{-1}$). Overall, emissions at RES9 decreased by a factor of two between 2010 and 2012.

At the water intake, $CH_4$ diffusive fluxes during the transition between the WD and WW seasons (up to 600 mmol $m^{-2}$ $d^{-1}$) are the highest reported at the surface of an aquatic ecosystem (Abril et al., 2005; Guérin et al., 2006; Bastviken et al., 2011; Barros et al., 2011; Deshmukh et al., 2015).

## 4 Discussion

### 4.1 $CH_4$ dynamic in the reservoir water column

The gradual decrease of the $CH_4$ concentration from the anoxic bottom water column to the metalimnion and the sharp decrease around the oxicline in the metalimnion (Fig. 2) is typical in reservoirs and lakes where $CH_4$ is produced in anoxic sediments and flooded soils (Guérin et al., 2008; Sobek et al., 2012; Maeck et al., 2013), and





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where most of it is oxidized at the oxic–anoxic interface (Bedard and Knowles, 1997; Bastviken et al., 2002; Guérin and Abril, 2007; Deshmukh et al., 2015).

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

As the reservoir overturns during the period over which the water residence time is the longest in the reservoir, the temporal evolution of the concentrations is anti-correlated with the residence time (Fig. 3c and d). The seasonal dynamics of the $CH_4$ in the monomictic NT2R differs from permanently stratified reservoirs like Petit Saut Reservoir where $CH_4$ concentration increased with retention time (Abril et al., 2005). However, at the annual scale the water residence time has a strong influence on $CH_4$ concentration and storage in the reservoir. Before the reservoir was commissioned (April 2010), the water residence time was up to 4 years and the $CH_4$ storage was up to four times higher than in 2011 and 2012 (Fig. 3d). Although a decrease of concentration and storage with the age of the reservoir was expected (Abril et al., 2005), the storage in the dry year 2012 was twice higher than in the wet year 2011 due to an increase of the water residence time by 25 % between 2011 and 2012. The short





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residence time (3 month) and the high water inputs in 2011 weakened the reservoir thermal stratification or altered its stability as shown by the sharper decrease and the larger range of $\Delta T$ in 2011 than in 2012 (Fig. 3a). As a consequence, the oxygen diffusion to the hypolimnion was higher in 2011 than in 2012 (Fig. 3b) and it enhanced aerobic methane oxidation by 20 % in the water column in the WW season in 2011 as compared to 2012 (Fig. 4). It therefore suggests that the residence time of water controls aerobic methane oxidation in the reservoirs and ultimately emissions.

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

The Fig. 7 illustrates the evolution of the diffusive fluxes, the stratification index ($\Delta T$), the $CH_4$ storage and the aerobic $CH_4$ oxidation at the stations RES1, RES3, RES7 and RES8. It shows that the large bursts of $CH_4$ (from 5 up to 200 mmol $m^{-2}$ $d^{-1}$) always occurred when $\Delta T$ decreased sharply (> 4 °C, Fig. 7a, d, g and j) and are usually followed by a sharp decrease of the $CH_4$ storage in the water column (Fig. 7b, e, h and k). These hot moments of emissions occurred mostly in the CD at the stations RES1 and RES3 whereas it was in the WW season at the stations RES7 and RES8 (Fig. 7). In the WD season, diffusive fluxes gradually increased together with the $CH_4$ storage in the water column (Fig. 7a, d, g and j) and they remained always lower than 20 mmol $m^{-2}$ $d^{-1}$. These sporadic high fluxes occurred in the WD season at RES3, RES7 and RES8 (Fig. 7d, g and j). They are usually associated with $\Delta T$ variations lower than 2 °C and the $CH_4$ storage decrease that is associated with these fluxes is not as sharp as the one observed in the CD and WW season (Fig. 7e, h and k).

We therefore confirm the occurrence of hot moments of emissions during the reservoir overturn in the CD season as already observed in lakes that overturn (Kankaala et al., 2007; López Bellido et al., 2009; Schubert et al., 2010, 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 sec-





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tion of the reservoir that has the longest water residence time (RES1–3, Table 1) and the largest $CH_4$ storage (Fig. 7b, e, h and 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. In older reservoir than NT2R, 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).

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

During the periods with major loss in the $CH_4$ storage with concomitant $CH_4$ burst, we compared the change in the yield of $CH_4$ with the sum of emissions and oxidation. Most of the time, the emissions alone and/or the sum of emissions and oxidation were





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





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### 4.4 Estimation of total diffusive fluxes from the reservoir

Yearly emissions by diffusive fluxes peaked at more than 9 Gg ($CH_4$) in 2010 when the reservoir was commissioned and they decreased down to ≈ 5 Gg ($CH_4$) in 2011 and 2012 (Fig. 8a).

Most of the increase of $CH_4$ emissions by diffusive fluxes from 4 to 9 Gg($CH_4$) between 2009 and 2010 is due to very significant emissions of 2–3 Gg ($CH_4$) at the water intake (Fig. 8a). This outgassing of $CH_4$ was triggered by the vertical mixing generated by the withdrawal of water from the reservoir to the turbines. Although the area under the influence of the water intake is less than 1 % of the total area of the reservoir, emissions at the water intake contributed between 15 and 25 % of total diffusive emissions after the commissioning of the reservoir. It is worth to note that emissions at this site are only significant within 3–5 month per year at the end of the WD season-beginning of the WW season when the storage of $CH_4$ reach its maximum in the reservoir (Fig. 8b). Very localized perturbation of the hydrodynamics, especially in lakes or reservoirs with $CH_4$-rich hypolimnion, can generate hotspots of emissions contributing very significantly to the total emissions from a given ecosystem. These hotspots can be found upstream of dams and water intake in reservoirs but also around aeration stations based on air injection or artificial mixing that could be used for improving water quality in water bodies (Wüest et al., 1992).

The contribution of extreme diffusive fluxes (> 5 up to 200 mmol $m^{-2}$ $d^{-1}$) to total emission by diffusion range from 30 to 50 % on a yearly basis (Fig. 8a) and from 40 up to 70 % on a monthly basis (Fig. 8b) although these hot moments represent less than 10 % of the observations during the monitoring and they only occur a few days in a year during sporadic destratification and reservoir overturn. This study shows that diffusive fluxes can be as sporadic as ebullition. Their precise quantification thus requires high spatial and temporal resolution. Based on our fortnightly monitoring we defined that a minimum sampling frequency of 1 month is required in monomictic reservoirs for an accurate estimation of the emissions (Fig. S1). A lower temporal resolution can





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significantly affect (positively or negatively) the emissions factors of non-permanently stratified freshwater reservoirs. This is particularly critical in the inflow regions when water inputs from the watershed increase in the rainy season in all reservoirs and at the beginning of the overturn in regions of the world where reservoirs are not permanently stratified like in Asia (Chanudet et al., 2011) which concentrate 60 % of the worldwide hydroelectric reservoirs (Kumar et al., 2011).

## 5 Conclusion

The high frequency monitoring of $CH_4$ diffusive emissions associated at nine stations revealed complex seasonal variations that could hardly been characterized by discrete sampling. The highest emissions occur sporadically during hot moments in the rainy season and when the reservoir overturns. In the rainy season, they mostly occur in the inflow region because the increase of the discharge of cool water from the reservoir tributaries contributes to sporadic thermal destratification. During the reservoir overturn, extreme emissions occur mostly in area remotely located from the inflows and outflows that are supposed to have the highest water residence time. It shows that diffusive emissions can be sporadically as high as ebullition and that these hot moments could contribute very significantly to the total emissions from natural aquatic ecosystems and reservoirs. Our results showing that a monthly monitoring is the minimum required frequency to capture all emissions is probably not applicable to every aquatic ecosystems. However, it suggests that quantification of emissions based on 2–4 campaigns in a year might significantly affect emissions factors and carbon budgets of ecosystems under study.

We also identified a new hotspot of emissions upstream of the water intake resulting from the artificial destratification of the water column due to vertical water current 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





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(immediately upstream of dams, typically) in order to evaluate if such results can be generalized.

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

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

[1] Descloux et al. (2011).
[2] Water residence time in arbitrary units, (***) stands for long residence time, (**) for intermediate and (*) for short residence times.





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**Table 2.** Median, average, ranges and proportion of diffusive fluxes ($F_{CH_4}$) < 1 and > 5 mmol $m^{-2}$ $d^{-1}$ for three seasons

| Station | | Warm Dry (WD) | Warm Wet (WW) | Cool Dry (CD) |
|---|---|---|---|---|
| RES1–8 | *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 |
| | % $F_{CH_4}$ < 1 | 48 % | 63 % | 86 % |
| | % $F_{CH_4}$ > 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 |





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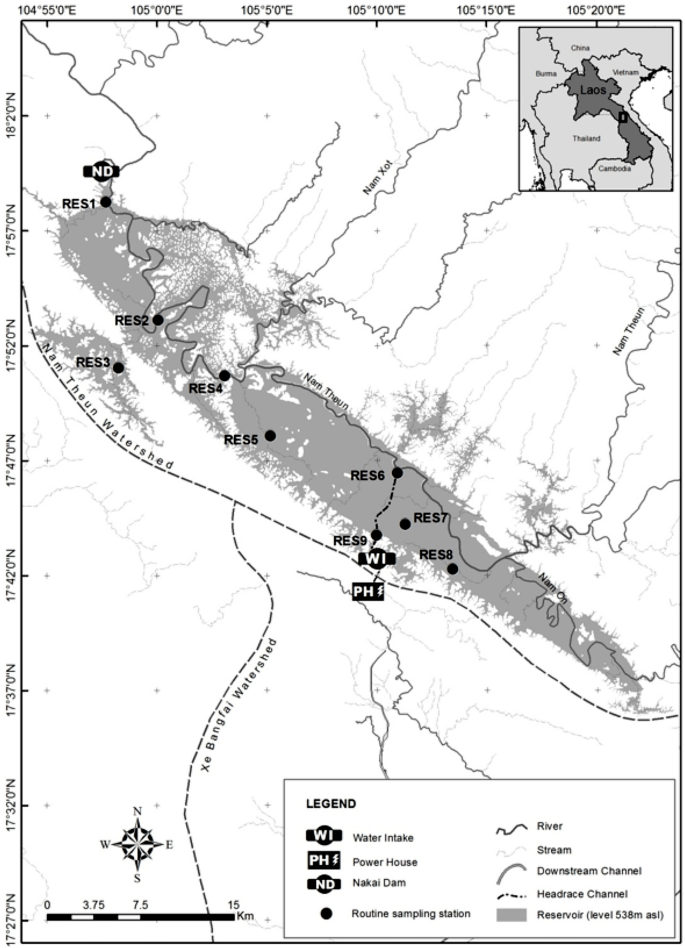


**Figure 1.** Map of the sampling stations and civil structures at the Nam Theun 2 Reservoir (Lao PDR).





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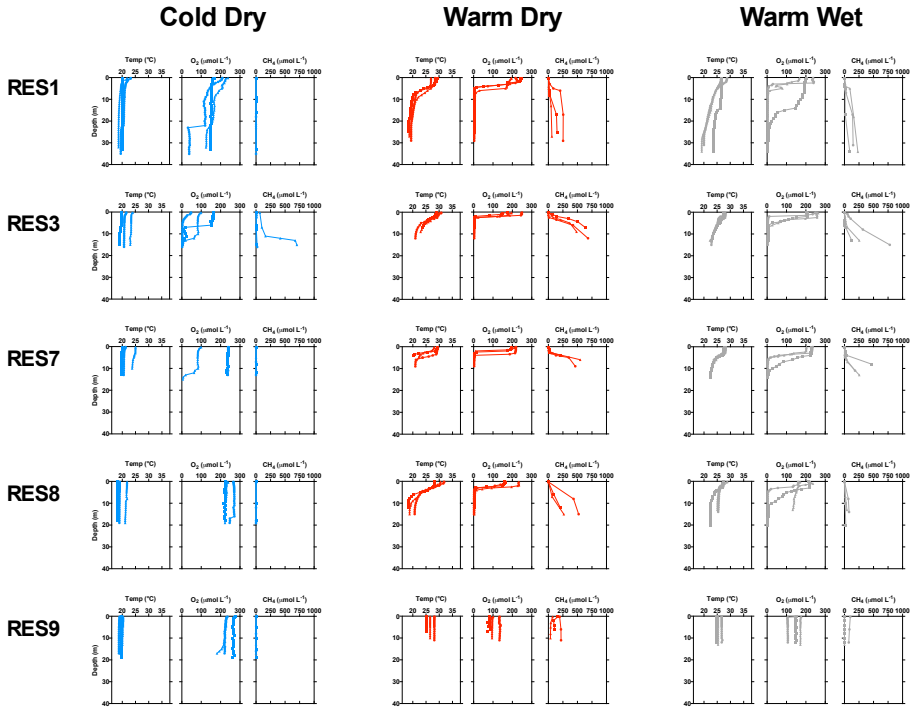


**Figure 2.** Vertical profiles of temperature (°C), oxygen (µmol $L^{-1}$) and methane (µmol $L^{-1}$) at the stations RES1, RES3, RES7, RES8 and RES9 in the Nam Theun 2 Reservoir. Representative profile of the years 2010 (circle), 2011 (square) and 2012 (triangle) are given for each seasons: cool dry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





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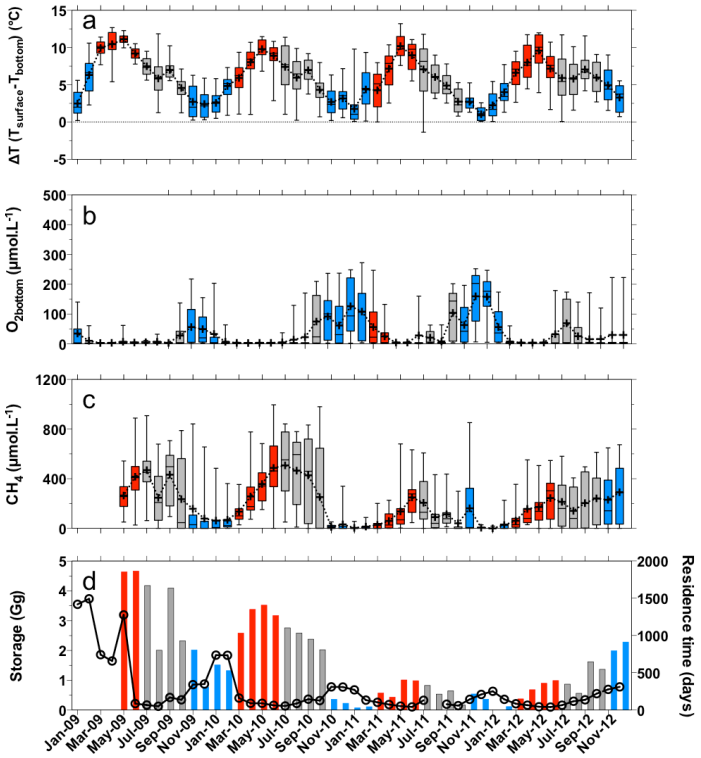


**Figure 3. (a)** Stratification index ($\Delta T$, see text), **(b)** $O_2$ concentration in the hypolimnion ($\mu mol\,L^{-1}$), $CH_4$ concentration in the hypolimnion ($\mu mol\,L^{-1}$) and **(d)** $CH_4$ storage in the water column (Gg $(CH_4)$ $month^{-1}$, bars) and water residence time (days, 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 (**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.





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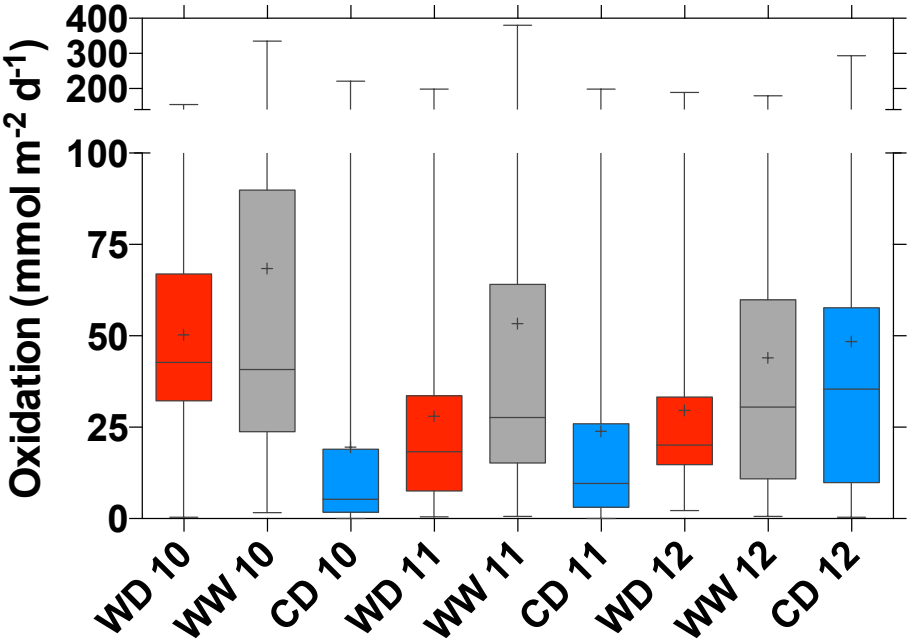


**Figure 4.** Seasonal variations between 2010 and 2012 of the depth-integrated aerobic $CH_4$ oxidation (mmol $m^{-2}$ $d^{-1}$) at the stations RES1–8 calculated from the aerobic oxidation rates determined by Deshmukh et al. (2015). WD stands for warm dry (in red), WW for warm wet (in grey) and CD for cool dry (in blue). The boxes show the median and the interquartile range, the whiskers denote the full range of values and the plus sign (+) denotes the mean.





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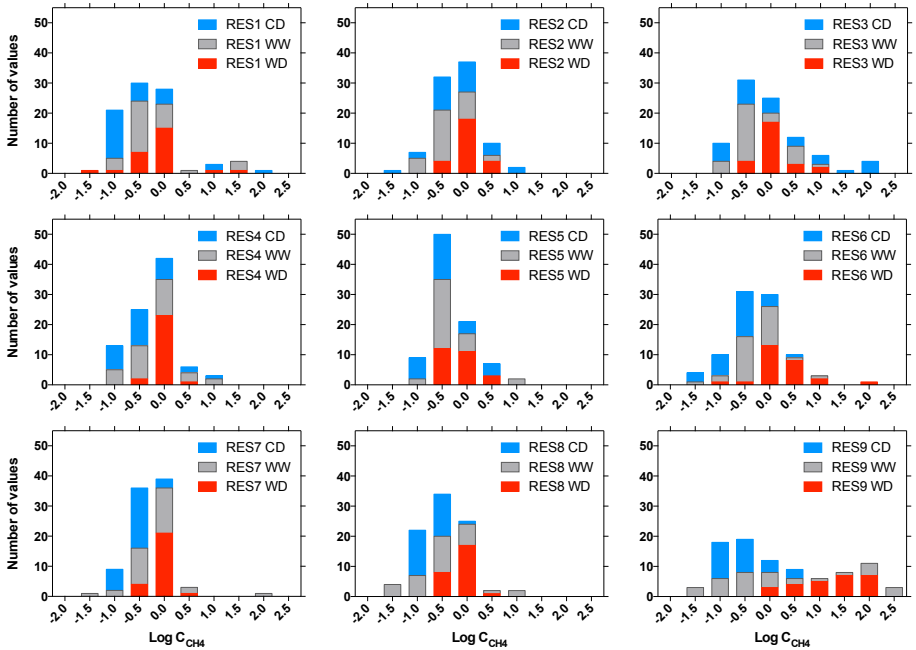


**Figure 5.** Frequency distribution of the log of $CH_4$ concentrations ($\mu mol\,L^{-1}$) at the nine monitoring stations of the Nam Theun 2 Reservoir. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool dry (CD) seasons, respectively.





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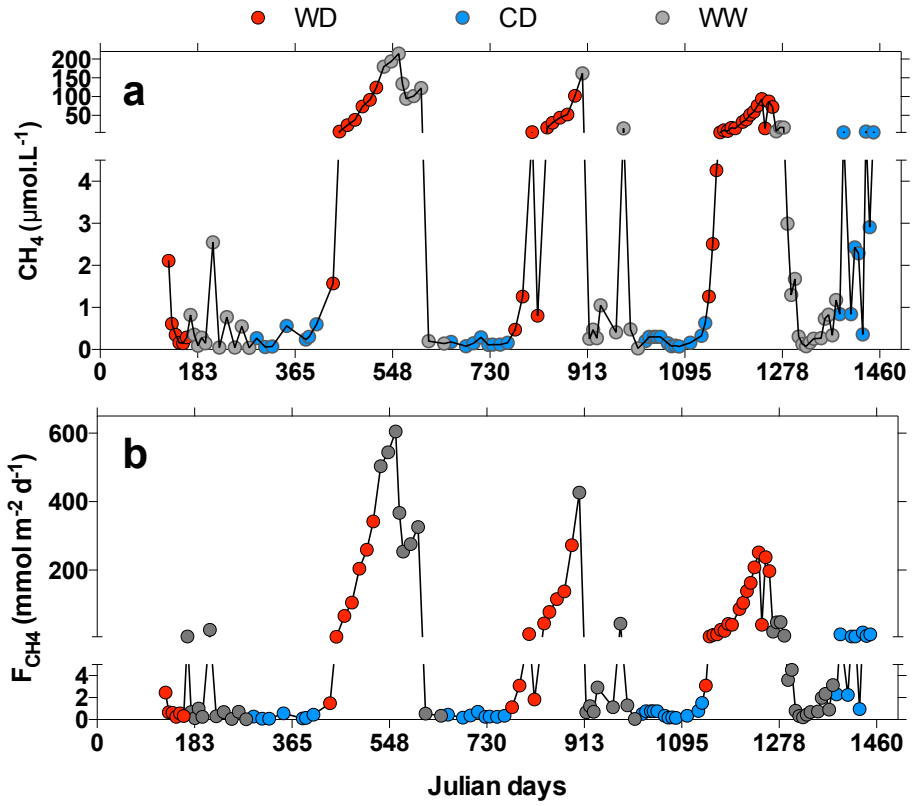


**Figure 6. (a)** Surface concentrations and **(b)** diffusive fluxes between June 2009 and December 2012 at the station RES9 located at the water intake. Julian day 0 is 1 January 2009. The red, grey and blue colours indicate the warm dry (WD), warm wet (WW) and cool dry (CD) seasons, respectively.





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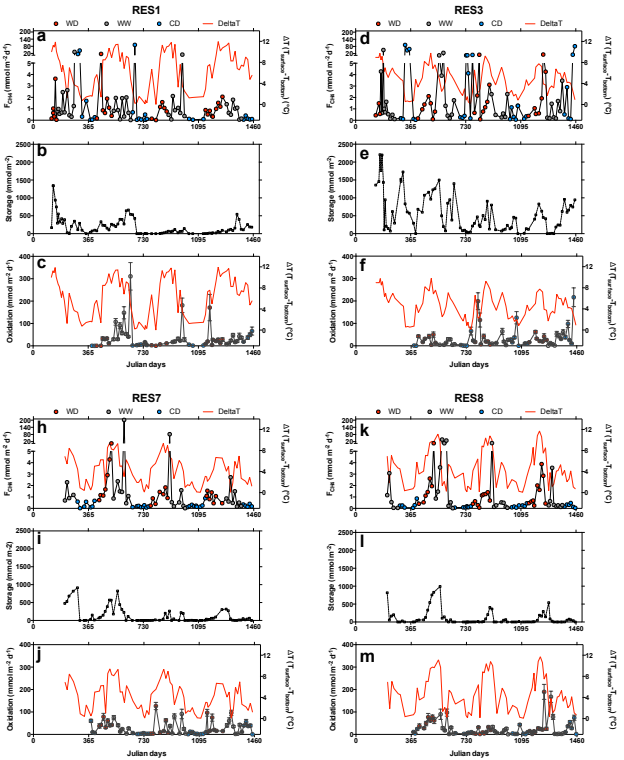


**Figure 7. (a, d, g, j)** stratification index ($\Delta T$, red line, see text) and diffusive fluxes, **(b, e, h, k)** $CH_4$ storage and **(c, f, i, l)** depth-integrated aerobic $CH_4$ oxidation (mmol $m^{-2}$ $d^{-1}$, black line) calculated from the aerobic oxidation rates determined by Deshmukh et al. (2015) and $\Delta T$ (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 1 January 2009. The red, grey and blue colour dots indicate the warm dry (WD), warm wet (WW) and cold dry (CD) seasons, respectively.





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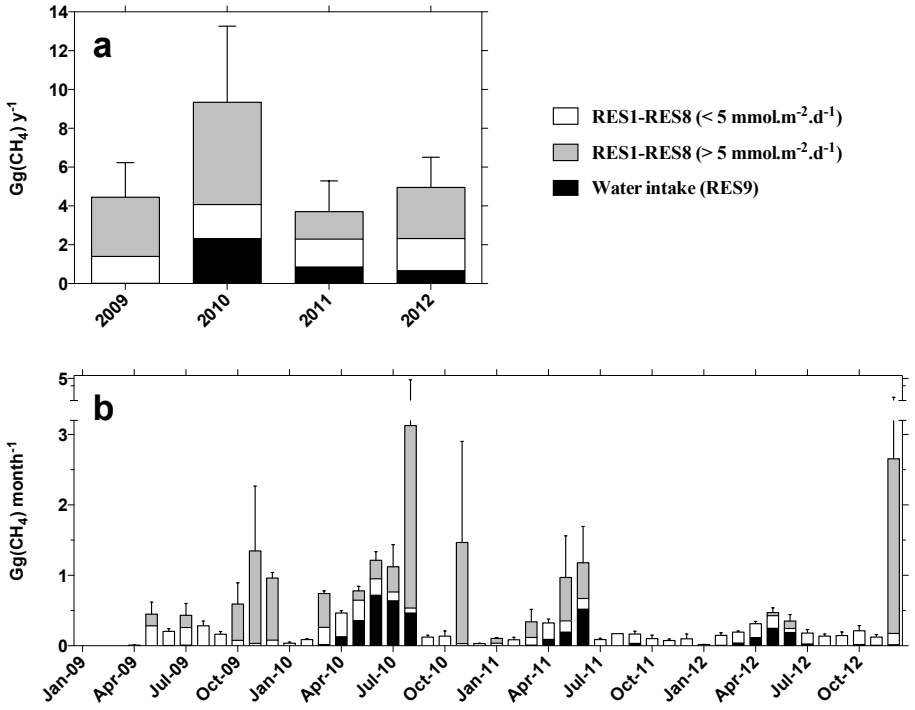


**Figure 8. (a)** Total emissions by diffusive fluxes in 2009, 2010, 2011 and 2012, and **(b)** monthly emissions by diffusive fluxes between May 2009 and December 2012. Emissions from RES9 (water intake) are shown in black, emissions resulting from diffusive fluxes lower than 5 mmol $m^{-2}$ $d^{-1}$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





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