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Annual follow-up of carbon dioxide and methane diffusive emissions from two boreal reservoirs and nearby lakes in Québec, Canada

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Abstract

Surface water pCO$_2$ and pCH$_4$ measurements were made in Québec (Canada) during consecutive seasons from 2006 to 2008 in two boreal reservoirs and nearby lakes. The goal of this follow-up was to evaluate greenhouse gas emissions from the water bodies studied, through flux calculations using the Thin Boundary Layer Model. Our measurements underscored the winter CO$_2$ accumulation due to ice cover and the importance of a reliable estimation of spring diffusive emissions as the ice breaks up. We clearly demonstrated that in our systems, CH$_4$ diffusive fluxes (in terms of CO$_2$ equivalent) were of minor importance in the GHG emissions, with CO$_2$ diffusive fluxes generally representing more than 95% of the annual diffusive fluxes. We also noted the extent of CO$_2$ spring diffusive emissions (16% to 52%) in the annual budget.

1 Introduction

The involvement of freshwater ecosystems in the global carbon budget has long been neglected because of their limited surface coverage on a worldwide scale compared to forest or oceans. However, in a recent review, Cole et al. (2007) demonstrate that lakes, reservoirs and rivers do not behave as mere pipelines transporting organic matter from terrestrial systems to oceans. Actually, half of the carbon annually entering freshwater ecosystems is processed therein and will never reach the ocean.

It has also been demonstrated that the loading of terrestrial dissolved organic carbon can contribute significantly to the energy pathways of lake ecosystems (Tranvik, 1992; Pace et al., 2004; Carpenter et al., 2005), sometimes leading to respiration rates exceeding primary production rates (Del Giorgio et al., 1997). This state, also called ecosystem net heterotrophy, is believed to be largely responsible for the CO$_2$ supersaturation observed in most of the world's lakes (Cole et al., 1994; Del Giorgio et al., 1999; Duarte and Prairie, 2005). Freshwater ecosystems thus clearly appear to be sites for carbon emissions to the atmosphere.

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Direct diffusive flux measurements from natural water bodies and diffusive flux calculations from partial pressure measurements have been carried out for some time in the Northern Hemisphere (Kling et al., 1992, MacIntyre et al., 1995) and in tropical zones (Richey et al., 1988; Keller and Stallard, 1994). Following the statement of Rudd et al. (1993) and St. Louis et al. (2000) about the potential of reservoirs to be net GHG emitters, the techniques developed for GHG measurements in natural systems have been applied to reservoirs. In fact, a comprehensive understanding of the processes involved in the global carbon cycle in reservoirs is becoming more and more crucial to ensure accurate comparisons of energy production methods and determination of national GHG inventories (IPCC, Houghton et al., 2007).

GHGs can be emitted from hydroelectric reservoirs by three major pathways, the first two of which are also observed in natural water bodies: diffusion at the reservoir surface (Huttunen et al., 2003; Rosa et al., 2004), bubbles produced at the sediment-water interface and migrating through the water column and into the atmosphere (Huttunen et al., 2003; Abril et al., 2005), and degassing in the turbulent waters downstream of reservoirs (Soumis et al., 2004; Roehm and Tremblay, 2006). Generally speaking, degassing and bubble emissions are not reported for boreal reservoirs because diffusive emissions were until now considered the major pathway, comprising over 95% of total emissions (Tremblay et al., 2005; Tremblay and Bastien, 2009). This statement may be revised in the future according to preliminary studies on degassing (Bastien, personal communication). In this study, we therefore focused on diffusive fluxes.

In northern temperate and boreal regions, seasonal variations in carbon diffusive emissions (CO_2 and CH_4) from freshwater ecosystems are difficult to measure directly because the ice cover during winter prevents flux measurements with floating chambers, and continuous monitors such as eddy covariance towers are still controversial because of possible interference with the surrounding forest, for example (MacIntyre et al., 1995; Eugster et al., 2003). However, in such climates, temporal variation is crucial information for annual carbon budget estimation since it is commonly recognized that these gases accumulate under ice (Riera et al., 1999; Kortelainen et al., 2000; Striegl

et al., 2001) and are released as diffusive fluxes on spring ice break-up (Michmerhuizen and Striegl, 1996; Huttunen et al., 2004; Duchemin et al., 2006).

The objectives of our study were (1) to present a follow-up from 2006 to 2008 of GHG concentrations and fluxes measured at different seasons in two regions, Eastmain River and La Grande River; (2) to address the question of gas accumulation under the ice cover in both lakes and reservoirs by comparing the results of winter and summer field campaigns; and (3) to propose a way to evaluate spring diffusive fluxes in order to provide estimates of annual greenhouse gas diffusive fluxes. We present a comparison of emissions from reservoirs and nearby lakes in order to document the effect of anthropogenic reservoir creation on aquatic GHG emissions (as performed by Huttunen et al., 2003). The comparison of reservoirs with nearby lakes is commonly accepted (Åberg et al., 2004; Harrison et al., 2009), given that reservoirs more than 10 years old can generally present limnological features comparable to lakes in the same region (Schetagne, 1994).

2   Methodology

2.1   Study sites

Sampling stations were located in the boreal zone, in northwestern Québec, Canada (Figs. 1 and 2). Within this region, mean monthly temperatures vary between $-23\,°C$ and $14\,°C$, and total rainfall and total snow precipitation are up to 430 mm and 260 mm, respectively. Two reservoirs and four lakes were sampled in the Eastmain River watershed and the La Grande River watershed (Table 1). The catchments of Robert-Bourassa and Eastmain 1 reservoirs are dominated by coniferous forest, shallow podzolic and peat soils, and igneous bedrock. They are described as oligotrophic systems with an overall low primary production (Planas et al., 2005). The studied reservoirs and lakes are partially to totally covered by ice from 15 December to 15 May, approximately (source: Hydro-Québec). Sampling dates between 2006 and 2008, and the number of

sampling stations (visited once per sampling period) for each system are described in Table 2. The sampling stations were distributed over the reservoirs and showed various water column depths (Figs. 1 and 2, all campaigns combined); their locations remained the same for all the field campaigns, but the number of stations sampled depended on weather conditions.

2.2 Temperature and dissolved oxygen

Temperature and dissolved oxygen saturation profiles were performed at each sampling station for the reservoirs and Mistumis Lake with a YSI sensor (600QS). First, water column depth at each station was measured with a depth sounder (Digital Hondex®). Samples were then taken every metre for stations <10 m deep, and every two metres for stations >10 m deep, up to 1 m from the bottom to avoid sensor damage. Because of the sensor cable length, the maximum sampling depth was 28 m. In order to obtain a broad description of the water bodies studied in all the field campaigns, we calculated the mean temperature and dissolved oxygen measured at each sampling depth and thus obtained mean profiles.

2.3 CO_2 and CH_4 partial pressures

CO_2 and CH_4 water partial pressures were measured at the water surface (0.1 m), and for some stations, profiles were performed (three depths chosen according to the station's water depth), using a peristaltic pump to sample the water. At Eastmain 1 reservoir, pCO_2 and pCH_4 profiles were made for 11 sampling stations in March 2007, for 17 stations in January 2008, for 2 stations in March 2008, for 13 stations in July 2008 and for 13 stations in September 2008. At Mistumis Lake, pCO_2 and pCH_4 profiles were made at one station for each field campaign. Water pCO_2 was measured in situ with a non-dispersive infrared (NDIR) sensor (EGM-4 from PP-Systems) coupled with a gas exchanger (Celgard from Membrana). Ten consecutive measurements (one per

minute) were averaged to obtain the pCO_2 at each station (mean variation coefficient of 1.3%).

For pCH_4 determination, three 30-mL samples were collected in 60-mL polypropylene syringes from each depth and kept chilled in a dark cooler for transport to the laboratory. There, 30 mL of nitrogen gas (N_2) was added. Water and N_2 were equilibrated by shaking the syringe vigorously for two minutes. Headspace CH_4 partial pressure (pCH_4 HS) was quantified on a gas chromatograph (with a flame ionization detector) within the next 24 h. Partial pressure before equilibration ($pCH_4 f$) was determined from Eqs. (1) and (2):

$$pCH_4 f = \frac{(pCH_4 HS \times K_{H\,Eq.}) + \left(HSR \times \frac{(pCH_4 HS - pCH_4 i)}{V_m}\right)}{K_{H\,Sample}} \tag{1}$$

where HSR is the headspace ratio (here equal to 1); $pCH_4 i$ is equal to zero, since the only gas present in the air inside the syringe before equilibration was N_2; V_m is the molar volume (according to Avogadro's law); $K_{H\,Eq.}$ and $K_{H\,Sample}$ are the gas partition constants at equilibrium (20 °C) and at sampling temperature, respectively, calculated according to Lide, 2007:

$$\ln K_H(CH_4) = -115.6477 + \frac{155.5756}{(T_K/100)} + 65.2553 \times \ln\left(\frac{T_K}{100}\right) - 6.1698 \times \left(\frac{T_K}{100}\right) \tag{2}$$

where T_K is the temperature in degrees Kelvin. $K_H(CH_4)$ is in molar fraction atm^{-1} (Lide, 2007) but converted in mole L^{-1} atm^{-1} using the following factor,

$$\left(\frac{1000 \text{ g/L [water density]}}{18.0153 \text{ g/mole [water molecular weight]}}\right).$$

The field pCH_4 was obtained by averaging the results from three sampling syringes.

2.4 CO_2 and CH_4 fluxes

We calculated mean pCO_2 and pCH_4 for ice-free periods and in spring, and thereby estimated mean fluxes, using corresponding mean water temperatures (data from automated systems at Eastmain-1 and Robert-Bourassa generating stations) and wind speeds (data from Nemiscau Airport meteorological station for the Eastmain River system [69 km away] and La Grande River Airport meteorological station for the La Grande River system [33 km away]).

Flux calculations require conversion of the partial pressures in concentration. CO_2 water concentration and CH_4 water concentration (CO_2wc and CH_4wc, respectively) were calculated from pCO_2 (CO_2wp) and pCH_4 (CH_4wp), according to Eqs. (3) and (4) (Morel, 1982; Anderson, 2002):

$$CO_2wc = K_H \times CO_2wp \qquad (3)$$

$$CH_4wc = K_H \times CH_4wp \qquad (4)$$

with K_H (CO_2) and K_H (CH_4) determined according to Eqs. (5) and (2), respectively:

$$\ln K_H(CO_2) = -58.0931 + 90.5069 \times \left(\frac{100}{T_K}\right) + 22.294 \times \ln\left(\frac{T_K}{100}\right) \qquad (5)$$

where K_0 (CO_2) is the gas partition constant of CO_2 in water at sampling temperature, expressed in mole L^{-1} atm^{-1}, and T_K is the temperature in degrees Kelvin (Weiss, 1974).

Then, CO_2 and CH_4 fluxes were calculated by Eq. (6) (MacIntyre et al., 1995),

$$Flux = k_x(C_w - C_a) \qquad (6)$$

with C_a being the gas concentration in water exposed to the atmosphere (385 ppm for CO_2, NOAA October 2008; and 1.745 ppm for CH_4, Houghton et al., 2001), k_x being the Mass Transfer Coefficient (cm h^{-1}):

$$k_x = k_{600}\left(\frac{Sc}{600}\right)^{-x} \qquad (7)$$

where x is equal to 0.66 for wind speed $\leq 3\,\mathrm{m\,s^{-1}}$ and is equal to 0.5 for wind speed $> 3\,\mathrm{m\,s^{-1}}$; Sc is the Schmidt number for CO_2 or CH_4, which is dependent on temperature (t) according to Eqs. (8) and (9) (Wanninkhof, 1992):

$$Sc(CO_2) = 1911.1 - 118.11t + 3.4527t^2 - 0.04132t^3 \tag{8}$$

$$Sc(CH_4) = 1897.8 - 114.28t + 3.2902t^2 - 0.039061t^3 \tag{9}$$

and k_{600} is estimated from the wind speed, according to Cole and Caraco (1998):

$$k_{600} = 2.07 + (0.215 \times U_{10}^{1.7}) \tag{10}$$

Fluxes obtained from Eq. (6) are converted from molar to gram basis using the respective molecular weights of CO_2 and CH_4 (44.0098 and 16.04276 $\mathrm{g\,mol^{-1}}$). The GHG flux can thus be calculated by adding the CO_2 and CH_4 fluxes in CO_2eq (Eq. (11), using the Global Warming Potential of 25 for CH_4, Forster et al., 2007):

$$\mathrm{GHGFlux(CO_2eq)} = CO_2\mathrm{flux} + 25 \times (CH_4\mathrm{flux}) \tag{11}$$

3 Results

3.1 Comparison between reservoirs and nearby lakes

Under the assumption of an atmospheric CO_2 concentration of 385 ppm (NOAA, 2008), we observed that the six water bodies studied are supersaturated in CO_2, whatever the season, with pCO_2 being from one (Clarkie Lake, September 2008) to seven (Eastmain 1 reservoir, March 2007) times higher than the atmospheric equilibration concentration (Table 3).

Surface pCO_2 measured at Eastmain 1 reservoir is higher than pCO_2 measured at the two lakes nearby. This observation is not surprising: Eastmain 1 reservoir is four years old, and it has been observed that it can take around 10 years after reservoir

creation for the labile flooded organic matter to be decomposed and for diffusive GHG fluxes to return to levels observed for natural lakes before reservoir creation (Chartrand et al., 1994; Tremblay et al., 2005). Indeed, there was no difference in surface pCO_2 measured at Robert-Bourassa reservoir, an old reservoir created in 1979, and nearby lakes.

For the different sampling periods, there were no significant differences between pCH_4 measured at the reservoirs and nearby lake surfaces, because of the high variability observed in the reservoirs (ANOVA, $p > 0.05$).

3.2 Temperature and dissolved gas profiles in Eastmain River watershed area

Temperature and dissolved gas profiles performed at Eastmain 1 reservoir and Mistumis Lake are quite similar (no temperature stratification for both), whatever the season (Fig. 3a and b). Consequently, because of the absence of hypolimnetic waters in these water bodies, it was not surprising to observe no oxycline even though dissolved oxygen saturation decreased slightly with depth at all seasons (Fig. 3c and d).

There was a general increase in pCO_2 with depth at Eastmain 1 reservoir, whatever the season, with the greatest difference between surface and bottom observed in March 2007 and 2008 (Fig. 3e), reflecting CO_2 accumulation under the ice. At Mistumis Lake, three stations were sampled and a GHG profile was made at a single station (explaining the absence of an error bar in Fig. 3g and h). As at Eastmain 1 reservoir, an increase of pCO_2 with depth was observed for under-ice measurements, also reflecting winter CO_2 accumulation (Fig. 3f).

At Mistumis Lake, pCH_4 presented no trend with depth, except in September 2008 (Fig. 3h). In fact, pCH_4 increased from 20 ppm at 7.5 m deep to 152 ppm at 9 m deep. Unfortunately, no other summer or fall data are available at a depth of 9 m, preventing us from reaching any conclusion about the repetition of such an event. Since the water column temperature was higher in July 2006, 2007 and 2008, and because methanogenesis is related to temperature (Wang et al., 1996), we hypothesize that the deep-water pCH_4 at these periods would have been at least equal to that measured in

September 2008. We note that high pCH$_4$ values found at depth are not reflected by the surface pCH$_4$, indicating that methane may have been degraded by methanotrophic bacteria along the water column and the gradient of dissolved oxygen (Fig. 3d; Wang et al., 1996).

⁵ At Eastmain 1 reservoir, pCH$_4$ presented no trend with depth for all campaigns, except in March 2007 below a depth of 10 m (Fig. 3g). During this campaign, we measured pCH$_4$ at depths of 10 m (stations PK 220-7 and PK245-3), 11 m (stations Fed-6, PK235-2 and PK260-4), 12 m (station PK255-6), 13 m (stations PK245-3 and Tour-3), 18 m (PK220-7) and 21 m (stations Fed-6, PK260-4 and Tour-3); it should be
¹⁰ recalled that sampling depths at each station were chosen according to the maximum depth at the station. Most of the pCH$_4$ measurements were in the same range as those above a depth of 10 m. However, for stations PK 245-3 (at depths of 10 m and 13 m), PK 260-4 (at 21 m only) and Tour-3 (at 21 m only), high pCH$_4$ was measured (Fig. 3g), corresponding to low to very low dissolved oxygen saturation (50%, 15.5%,
¹⁵ 16.7% and 21.6%, respectively), thus suggesting CH$_4$ accumulation in deep zones. We cannot confirm a possible under-ice CH$_4$ accumulation, since January 2008 profiles at the same stations showed no increase with depth and these same profiles are not available for the March 2008 campaign. However, low dissolved oxygen saturation experienced in March 2008 below a depth of 20 m (Fig. 3c) may have led to similar
²⁰ favourable conditions for CH$_4$ formation in deep zones. For the reservoir as a whole, our results showed that, as at Mistumis Lake, high values measured at depth are not reflected by the overall surface pCH$_4$ (surface pCH$_4$ measured at 39 stations in March 2007), also suggesting a CH$_4$ degradation along the water column. In March 2008, three stations (among the 34 where pCH$_4$ measurements were done) showed very
²⁵ high surface pCH$_4$ (1442 ppm, 2221 ppm and 5439 ppm), leading to the high mean surface concentration observed in Fig. 3g (303 ppm, SD±1009); without these data the mean pCH$_4$ would be 41 ppm, which is not significantly different from the surface pCH$_4$ observed during the other campaign.

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In a parallel study using automated systems for GHG measurements for the reservoir as a whole, we showed that there was no CH_4 accumulation under ice at Eastmain 1 reservoir (unlike CO_2, Demarty et al., 2009). Consequently, in view of this information and the preceding analysis concerning the variations in pCH_4 observed, we will assume, for subsequent analysis, that CH_4 did not accumulate under the ice at either Eastmain 1 reservoir or Mistumis Lake.

3.3 Seasonal trends in mean surface water pCO_2 and pCH_4

For Eastmain 1 reservoir, Mistumis Lake and Clarkie Lake, mean surface pCO_2 measured during the 2007 and 2008 spring campaigns (i.e., in March–April) is clearly higher than that measured during the other sampling period (ANOVA, $p<0.05$; Table 4 and Fig. 4). The results thus demonstrate CO_2 accumulation under the ice for these water bodies. For statistical analysis, March and April data (hereafter called "spring") have been pooled for Robert-Bourassa reservoir, Yasinski Lake and Duncan Lake, since they did not present a significant difference (Student Test, $p>0.05$; Table 3). Accordingly, surface pCO_2 at Robert-Bourassa reservoir and Yasinski Lake is higher in spring than in summer, thereby demonstrating CO_2 accumulation under the ice (Student Test, $p<0.05$; Fig. 4). This was not the case for Duncan Lake (Student Test, $p>0.05$), possibly reflecting a slower winter metabolism than in the other systems studied, without measurable CO_2 production under ice.

As explained above, we were not able to conclude that there was an accumulation of CH_4 under the ice for either Eastmain 1 reservoir or Mistumis Lake. For the ice-free periods, we observed no significant difference in pCH_4 among the campaigns (ANOVA and Tukey test, $p>0.05$). As well, no significant differences in surface pCH_4 among the field campaigns were observed for Clarkie Lake (ANOVA and Tukey test, $p>0.05$). Because of field work constraints, we only have one period of measurement for pCH_4 at Robert-Bourassa reservoir, Yasinski Lake and Duncan Lake (April 2006 field campaign); we will therefore base subsequent calculations on a constant pCH_4 throughout the year.

3.4 Estimation of maximum dissolved pCO$_2$ reached before ice break-up

The preceding results showed a CO$_2$ accumulation under the ice cover in all sampled water bodies (Eastmain 1 and Robert-Bourassa reservoirs; Mitsumis, Clarkie and Yasinski lakes). No difference in summer, fall and early winter (before January) mean dissolved pCO$_2$ was observed (ANOVA, $p > 0.05$). Taken together, these data thus represent the baseline pCO$_2$ of the year (Fig. 4). Using the winter data, we observed higher dissolved pCO$_2$ in March than in January, and a linear increase of dissolved pCO$_2$ is expected under the ice cover from January to May, when the ice break-up occurs. Linear regressions between baseline pCO$_2$ values and March pCO$_2$ values were then made to calculate the under-ice daily rate of pCO$_2$ increase (75 days between 15 January and 30 March; Table 4). Taking the date of 15 May as the beginning of the ice degassing period (based on observations from automated systems, Demarty et al., 2009), we thus estimated potential pCO$_2$ before the ice break-up. Since our goal was to obtain a broad estimation of the potential spring CO$_2$ flux, the 2005 baseline pCO$_2$ (which was not measured but is necessary for the calculation) was assumed to be equal to the 2006 baseline pCO$_2$ for Robert-Bourassa reservoir and Yasinski Lake (for which we only have one summer field campaign and two winter field campaigns in 2006). From these calculations, it is clear that Eastmain 1 reservoir tends to present the highest pCO$_2$ before the spring emission period (Table 4).

3.5 Estimation of potential spring GHG emission

The method used to estimate the potential spring emission is shown in Fig. 5. Depending on when the ice break-up began (as recorded by the ice survey program over the last 30 years), the spring emission period in the studied areas was supposed to last around one month, from 15 May to 15 June (Demarty et al., 2009). Between these dates, fluxes were assumed to follow a linear trend. Maximum potential gas partial pressures were used to calculate the maximum potential fluxes at the beginning of the degassing period, and baseline gas partial pressures were used to calculate potential

fluxes during the following period (summer and fall; Table 5). Averaging these two fluxes, we obtained a mean daily spring flux, which was used to compute an annual carbon budget.

3.6 Annual cumulative diffusive fluxes

The annual CO_2 flux was calculated as the sum of the daily fluxes between two spring emission periods (for example, 15 May 2007 to 15 May 2008): we added 31 days of mean spring flux (see Table 5), 214 days of mean ice-free period flux (summer, fall and beginning of winter) and 120 days without flux (ice-cover period). The resulting annual CO_2 evasions for the water bodies studied are presented in Table 6. Spring CO_2 fluxes represented 16% (Yasinski Lake) to 52% (Mistumis Lake) of the annual CO_2 diffusive emission, and spring CH_4 fluxes represented 5% (Eastmain 1 reservoir) to 18% (Clarkie Lake) of the annual CH_4 diffusive emission.

4 Discussion

This study is the first to provide a multi-annual estimation of spring GHG diffusive fluxes for both reservoirs and nearby lakes. The potential spring GHG flux of lakes and reservoirs is commonly calculated as the difference between the accumulated amount of gas (using the potential GHG storage of water bodies integrated versus depth to obtain the concentration per square metre) and the amount of gas at atmospheric equilibrium (around 14 µmol, or 385 ppm; 1998; Striegl et al., 2001; Huttunen et al., 2004; Duchemin et al., 2006), rather than the actual gas concentration, thereby overestimating annual CO_2 emissions and the winter contribution. Indeed, CO_2 supersaturation is described for lakes around the world (Cole et al., 1994; Del Giorgio et al., 1999; Duarte and Prairie, 2005) as well as for reservoirs (Tremblay et al., 2005; Demarty et al., 2009), meaning that their CO_2 concentration is generally above atmospheric concentration for most of the year. That is why, in this article, we advocate using the difference in water

surface partial pressure between the beginning and end of the ice-cover period as a more relevant method to estimate spring diffusive fluxes.

Two main assumptions were made regarding the estimation of maximum dissolved gas partial pressures before ice break-up. First, our field measurements showed dissolved CO_2 accumulation between January and March, thus suggesting that ice formation over the entire water body took a few weeks (from November to the beginning of January). This is confirmed by the La Grande and Eastmain ice survey, which indicates that the ice reached a thickness of about 20 cm by the end of December. For safety reasons, field campaigns could only be conducted between early January and the end of March, when the ice was at least 20 cm thick. Therefore, the baseline fluxes, averaging summer, fall and early winter fluxes (end of December), were used to compute a conservative annual flux. Because of the lack of data between September and ice cover, we could not examine the importance of the fall turnover, which is known to affect CO_2 and CH_4 fluxes. However, for Eastmain 1 reservoir, data from September 2008 (Fig. 3) showed that CO_2 and CH_4 partial pressures did not increase with depth, so that the fall turnover should not have led to an increase in fluxes; we consequently assume that the use of an ice-free mean (baseline) is acceptable. Second, we assumed a linear trend in CO_2 accumulation under the ice from January until the beginning of the spring emission period and a linear decrease during spring. These trends are supported by the results obtained from automated systems providing continuous pCO_2 and pCH_4 measurements at Eastmain 1 and Robert-Bourassa reservoirs since 2007 (Demarty et al., 2009). We observed that the spring emission period began around one month after the start of the ice break-up, thus showing that both the ice break-up and, most importantly, spring water column mixing (and the subsequent modification of the concentration gradient, see Eq. 6) are responsible for spring emissions.

Except at Duncan Lake, we observed CO_2 accumulation under ice cover for both lakes and reservoirs that were studied. The spring daily rates of CO_2 increase under ice cover presented in Table 4 were then calculated in terms of concentration rather than partial pressure (calculation according to Eqs. 1 and 4, considering a surface

temperature of 0.1 °C, based on field observations). Rates varied between 0.3 and $1 \mu g \, C \, L^{-1} \, h^{-1}$ at Yasinski Lake and Eastmain 1 reservoir, respectively. In their review about respiration in lakes, Pace and Prairie (2005) summarize field observations of planktonic respiration, which varies between $0.4 \mu g \, C \, L^{-1} \, h^{-1}$ and $81 \mu g \, C \, L^{-1} \, h^{-1}$

5 (mean: $7.9 \mu g \, C \, L^{-1} \, h^{-1}$); temperature is presented as an important factor influencing these respiration rates. Interestingly, the CO_2 increase rates observed under ice in our systems are comparable to the lower range reported by Pace and Prairie (2005). This strongly suggests that CO_2 increase under ice is due to bacterial respiration under the influence of cold temperatures, concomitant with low primary production (which

10 hides the respiration rate in warmer conditions). This idea is supported by the study of Striegl et al. (2001), which demonstrated that high dissolved pCO_2 prior to ice melt was related to bacterial respiration. Moreover, the highest pCO_2 values and winter CO_2 increase rates were observed at Eastmain 1 reservoir. This was expected, since it is a young reservoir flooded four years ago (in 2006), and the flooding of large quan-

15 tities of organic matter is known to lead to an increase in dissolved pCO_2 through an increase in bacterial activity (Tremblay et al., 2005; Tadonkélé et al., 2005). After an initial peak, generally reached within the first years after flooding, CO_2 fluxes and CO_2 concentrations decrease, reaching values comparable to natural aquatic ecosystems within a period of around 10 years (Tremblay et al., 2005). After this transition period,

20 CO_2 emissions are related to carbon entering the reservoir through runoff from the watershed or autochthonous primary production (Marty et al., 2005; Matthews et al., 2005). In the case of Eastmain 1 reservoir, the return to natural aquatic ecosystem values (i.e., values in the same range as nearby lakes) indeed occurred the third year after flooding (2008) (Tremblay and Bastien, 2009).

25 In accordance with the fact that most of the CH_4 produced in the anoxic sediment may have been oxidized at the sediment-water interface (Frenzel et al., 1990), and that CH_4 escaping from the sediment-water interface would have been slowly oxidized in the water column (Wang et al., 1996), no clear CH_4 accumulation was observed under the ice. Huttunen et al. (2004) also observed no CH_4 accumulation under ice cover in

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a small boreal lake during three consecutive late winters. Similar results were observed in four reservoirs in Manitoba over three years and in three reservoirs in Québec over 18 months (Demarty et al., 2009). Finally, a recent study by Juutinen et al. (2009) showed that CH_4 concentration in the surface and hypolimnetic waters is negatively correlated with oxygen and lake depth and area, whatever the season. It is therefore not surprising to observe low winter CH_4 concentration in the large reservoirs studied.

Our results showed that diffusive CH_4 emissions were not of concern in the boreal systems studied, and tally with the findings of Juutinen et al. (2009) and Kortelainen et al. (2006), who report CH_4 and CO_2 releases of $0.588\,mg\,C\,m^{-2}$ Lake $area^{-1}$ ($49\,mmol\,m^{-2}$ Lake $area^{-1}$ in the text) and $42\,mg\,C\,m^{-2}$ Lake $area^{-1}$, respectively, for Finnish lakes. Our results contradict the conclusion suggested by Duchemin et al. (2006) who, for their study, collected very few samples in very shallow (<2 m), small impounded bays with limited water exchange with the main reservoir water body. Shallow areas in reservoirs should be the most favorable environment for CH_4 production with warmer water temperatures (>15 °C in summer). However, most of the shallow areas in reservoirs are situated around the edge of the reservoir, where the organic matter is eroded to the mineral horizon or bedrock by wind, ice action and water level fluctuation related to electricity generation, generally within the first five years (Tremblay et al., 2005). Therefore, even in shallow areas, sites of CH_4 production might be sparse and the results reported by Duchemin et al. (2006) are thus representative of particular shallow or near-shore areas of the reservoir.

Potential spring GHG diffusive emissions from lakes (presented in Table 5) are in the same order as those measured in a boreal lake by Huttunen et al. (2004; 103 to $128\,g\,CO_2eq\,m^{-2}$). Spring GHG emissions represented 16% to 52% of annual GHG emissions, and are higher than those reported by Duchemin et al. (2006) for shallow areas, where spring GHG diffusive fluxes represented only 7% of the annual flux at Robert-Bourassa reservoir. Our results also demonstrated that, generally speaking, >95% of annual GHG diffusive emissions from the studied systems are carbon dioxide.

5   Summary

We have provided the results of exhaustive surveys of GHG partial pressure and diffusive fluxes in both lakes and reservoirs in northwestern Québec, which allowed us to reliably estimate spring GHG fluxes from a few assumptions.

Spring GHG diffusive emissions represented a higher proportion of annual GHG emissions than what was previously proposed, with CO_2 being largely responsible for the total annual diffusive emissions.

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Table 1. Description of study site.

Study Site		Latitude	Longitude	Surface (km^2)	Mean depth (m)	Maximum depth (m)
Eastmain River watershed area	Eastmain-1 Reservoir (created in 2005)	52.19° N	75.05° W	603	16	63
	Mistumis Lake	52.16° N	76.16° W	4	5	12
	Clarkie Lake	52.23° N	75.47° W	24	6	18
La Grande River watershed area	Robert Bourassa Reservoir (created in 1979)	53.78° N	77.53° W	2875	20	91
	Yasinski Lake	53.28° N	77.53° W	46	8	25
	Duncan Lake	53.49° N	77.89° W	96	9	35

Table 2. Sampling periods and number of sampled stations for each water body studied.

Systems	Sampling periods	Number of sampling stations		
		Eastmain 1 R.	Mistumis L.	Clarkie L.
Eastmain River	10 to 22 Jul 2006	41	3	2
	18 Sep to 4 Oct 2006	42		
	20 to 27 Mar 2007	39	3	
	5 to 21 Jul 2007	38	3	3
	14 to 26 Jan 2008	42	3	3
	26 Mar to 5 Apr 2008	35	3	2
	3 to 22 Jul 2008	57	3	3
	15 to 22 Sep 2008	27	3	2
		Robert Bourassa R.	Yasinski L.	Duncan L.
La Grande River	25 Feb to 4 Mar 2006	17	1	3
	4 to 11 Apr 2006	29	3	4
	24 to 27 Jul 2006	14	2	2

Table 3. Mean surface pCO_2 and pCH_4 (±standard deviation) measured at Eastmain 1 reservoir, Mistumis Lake, Clarkie Lake, Robert-Bourassa reservoir, Duncan Lake and Yasinski Lake.

Field campaigns	Eastmain-1 R.		Mistumis L.		Clarkie L.		Robert Bourassa R.		Duncan L.		Yasinski L.	
	pCO_2	pCH_4	pCO_2	pCH_4	pCO_2	pCH_4 ppm	pCO_2	pCH_4	pCO_2	pCH_4	pCO_2	pCH_4
Mar 2006	–	–	–	–	–	–	870±186	–	650±80	–	750	–
Apr 2006	–	–	–	–	–	–	803±204	59±189	580±20	9±1	810±20	20±17
Jul 2006	2230±563	125±151	565±30	33±5	558±13	–	560±149	–	540±80	–	661±24	–
Sep 2006	2181±485	83±65	–	–	–	–	–	–	–	–	–	–
Mar 2007	2798±708	20±31	1441±47	8±2	–	–	–	–	–	–	–	–
Jul 2007	1333±317	–	568±47	–	496±42	–	–	–	–	–	–	–
Jan 2008	1211±194	40±82	856±25	17±2	716±31	11±1	–	–	–	–	–	–
Mar 2008	2529±796	287±982	1533±268	19±9	975±171	13±2	–	–	–	–	–	–
Jul 2008	1025±361	58±56	620±73	25±19	507±34	9±2	–	–	–	–	–	–
Sep 2008	1340±459	38±38	454±18	24±7	425±4	17±5	–	–	–	–	–	–

Table 4. Daily rates of under-ice pCO_2 increase and maximum potential pCO_2 reached by 15 May at Eastmain 1 reservoir and Mistumis Lake.

Study site	n	Spring daily rates of pCO_2 increase ppm d^{-1}	SD ppm d^{-1}	R^2	Maximum potential pCO_2 reached (15 May)
Eastmain-1 R. 2007	122	8	17	0.18	3154
Eastmain-1 R. 2008	116	17	14	0.59	3285
Mistumis L. 2007	7	12	1	0.99	1967
Mistumis L. 2008	9	11	6	0.83	2026
Clarkie L. 2008	8	5	4	0.65	1196
Robert Bourassa R. 2006	60	4	6	0.52	987
Yasinski L. 2006	6	2	1	0.85	875

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Table 5. Spring CO_2, CH_4 and GHG fluxes. The beginning and end of the degassing periods are assumed to be around 15 May and 15 June, respectively.

Study site	Degassing Period	CO_2 Flux (mmole m^{-2} d^{-1})	CH_4 Flux (mmole m^{-2} d^{-1})	Mean CO_2 spring flux (mmole m^{-2} d^{-1})	Mean CH_4 spring flux (mmole m^{-2} d^{-1})	Mean GHG spring flux (mmole CO_2eq m^{-2} d^{-1})	Total GHG spring flux (g CO_2eq m^{-2})
Eastmain-1 R.	Beginning – 2007	131	0.017	86	0.017	86	117
	End – 2007	40	0.017				
	Beginning – 2008	173	0.059	108	0.036	108	147
	End – 2008	43	0.012				
Mistumis L.	Beginning – 2007	75	0.010	45	0.010	45	61
	End – 2007	15	0.010				
	Beginning – 2008	98	0.036	53	0.043	53	73
	End – 2008	9	0.049				
Clarkie L.	Beginning – 2008	49	0.023	27	0.019	27	37
	End – 2008	5	0.015				
Robert Bourassa R.	Beginning – 2006	34	0.110	22	0.110	22	30
	End – 2006	10	0.110				
Yasinski L.	Beginning – 2006	28	0.035	21	0.035	21	29
	End – 2006	15	0.035				

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Table 6. Ice-free period, spring and annual CO_2, CH_4 and GHG emissions for the water bodies studied.

Study site and period	CO₂				CH₄				GHG	
	Ice free period CO₂ emission	Spring CO₂ emission	Annual CO₂ emission	Spring flux/ annual flux	Ice free period CH₄ emission	Spring CH₄ emission	Annual CH₄ emission	Spring flux/ annual flux	Annual GHG emissions	CO₂ contribution
	mmol m⁻²			%	mmol m⁻²			%	g CO₂eq m⁻²	%
Eastmain-1 R. 2007–2008	8747	2654	11 401	23	7	1	7	7	509	98.6
Eastmain-1 R. 2008–2009	7374	3348	10 723	31	20	1	21	5	493	95.6
Mistumis L. 2007–2008	3248	1392	4640	30	2	0	3	12	207	98.8
Mistumis L. 2008–2009	1528	1658	3186	52	8	1	10	13	150	93.5
Clarkie L. 2008–2009	903	834	1737	48	3	1	3	18	80	95.9
Robert Bourassa R. 2006–2007	2208	676	2884	23	26	3	29	11	156	81.2
Yasinski L. 2006–2007	3448	664	4112	16	8	1	9	11	190	95.1
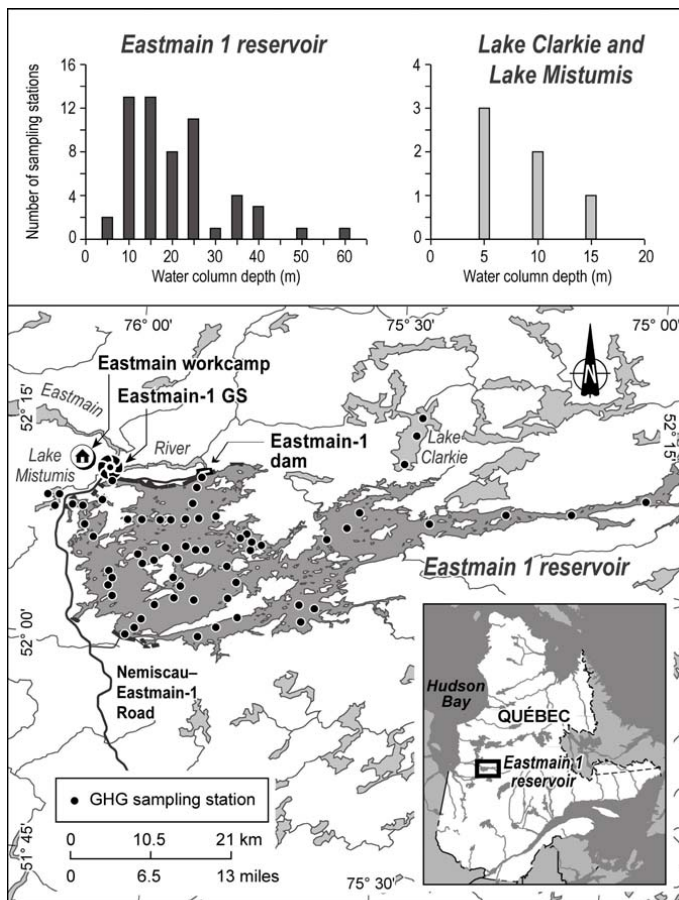
Fig. 1. Map of sampling stations in the Eastmain River area; graphs representing the distribution of sampling depths.
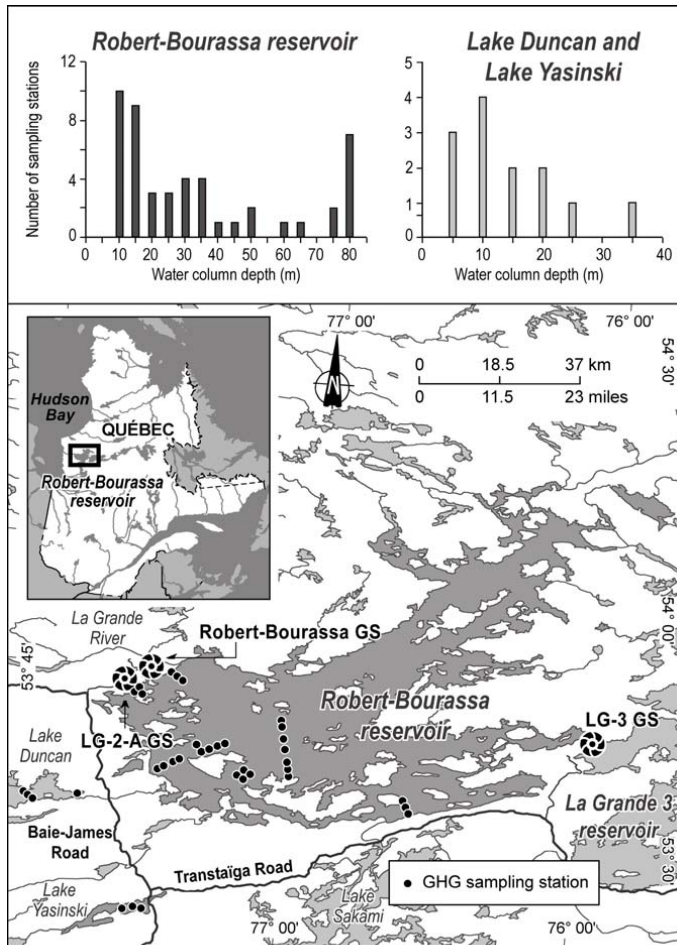
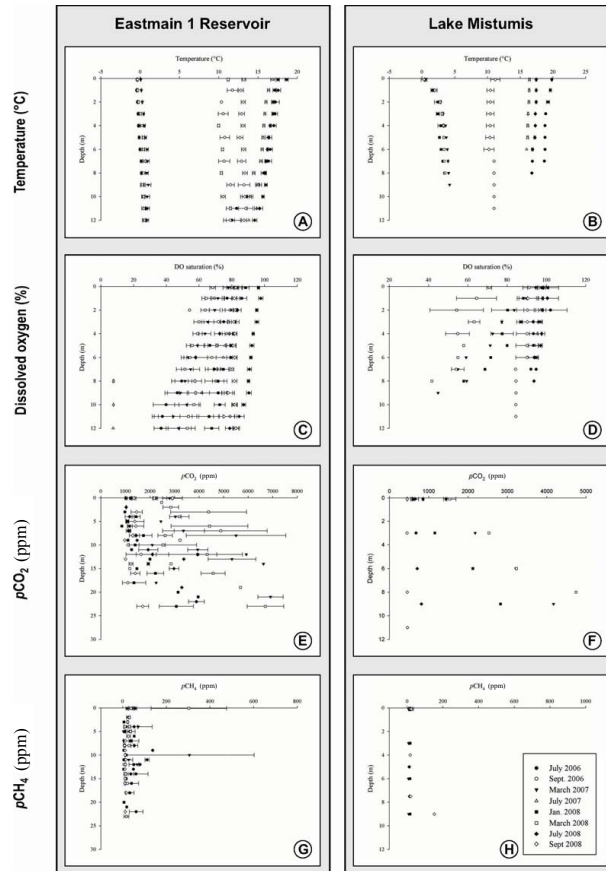**Fig. 2.** Map of sampling stations in the La Grande River area; graphs representing the distribution of sampling depths.

Fig. 3. Temperature, dissolved oxygen saturation, pCO_2 and pCH_4 mean profiles in East-main 1 reservoir and Mistumis Lake. Bars represent standard errors. Dashed lines represent maximum depth (for Mistumis Lake) and dotted lines represent mean depth.

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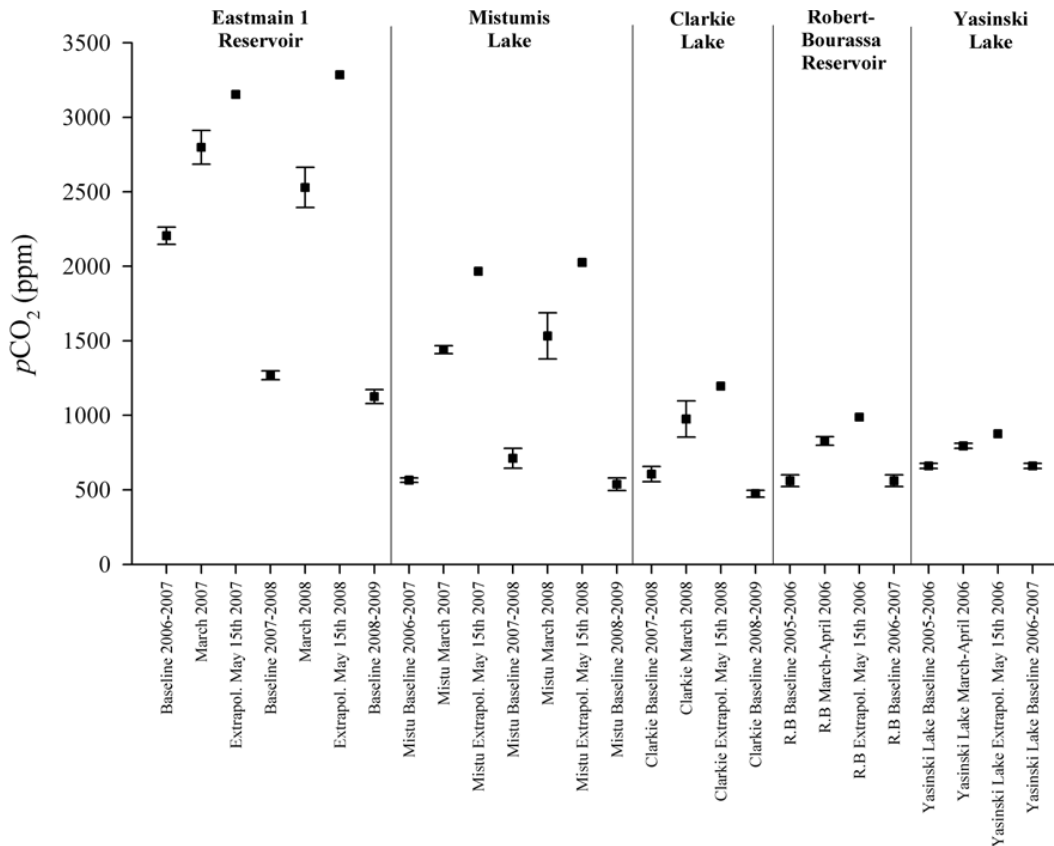
Fig. 4. Measured and extrapolated CO_2 partial pressure for the different systems studied. Bars represent standard errors. Baseline points represent mean values during ice-free periods.
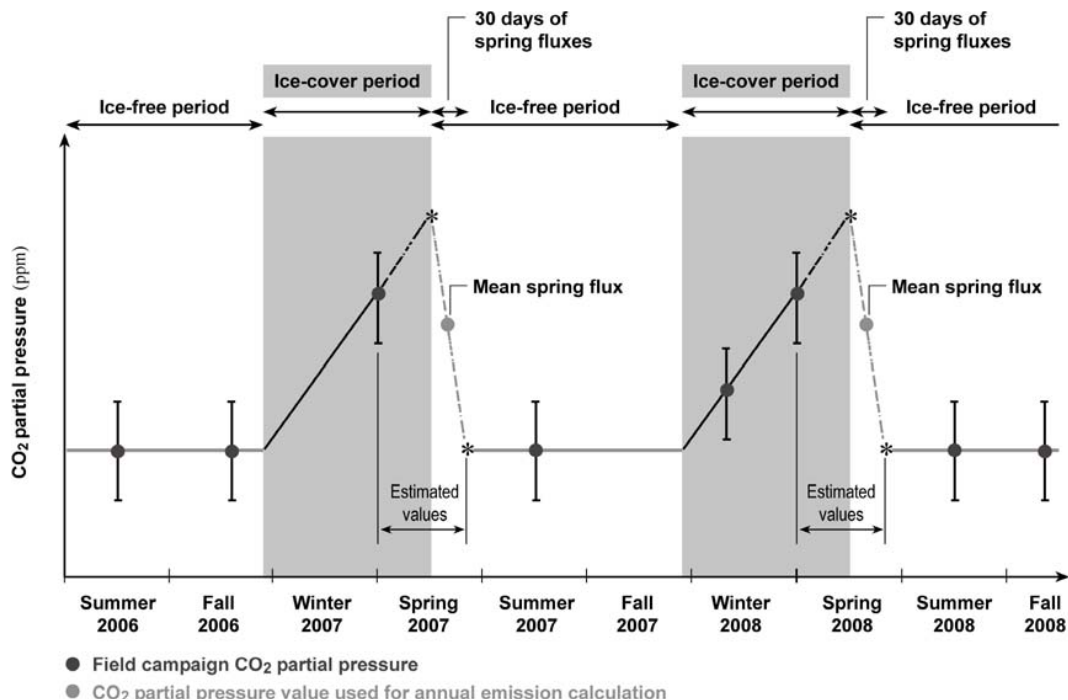
Fig. 5. Schematic representation of the estimation of annual emissions based on seasonal field campaigns.