Biogeosciences

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

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Abstract. Surface water pCO_2 and pCH_4 measurements were taken in the boreal zone of Québec, Canada, from summer 2006 to summer 2008 in Eastmain 1 reservoir and two nearby lakes. The goal of this follow-up was to evaluate annual greenhouse gas (GHG) emissions, including spring emissions (N.B. gross emissions for reservoir), 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 estimate of spring diffusive emissions as the ice breaks up. We clearly demonstrated that in our systems, diffusive CH_4 flux (in terms of CO_2 equivalent) were of minor importance in the GHG emissions (without CH_4 accumulation under ice), with diffusive CO_2 flux generally accounting for more than 95% of the annual diffusive flux. We also noted the extent of spring diffusive CO_2 emissions (23% to 52%) in the annual carbon 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 with forest or oceans. However, in a recent review, Cole et al. (2007) demonstrated 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 (i.e., biomass production, emissions, storage) 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). Lakes thus clearly appear to be sources of carbon emissions to the atmosphere.

In the nineties, direct diffusive flux measurements from natural water bodies and diffusive flux calculations from partial pressure measurements were carried out 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 statements of Rudd et al. (1993) and St. Louis et al. (2000) about the potential of reservoirs to be net greenhouse gas (GHG) emitters, the techniques developed for GHG measurements in natural systems have been applied to reservoirs. 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, 2006; Houghton et al., 2001).

Reservoirs can emit GHGs by three major pathways, the first two of which are also observed in natural water bodies: (1) diffusion at the reservoir surface (Huttunen et al., 2003; Rosa et al., 2004), (2) bubbles produced at the sediment-water interface that migrate through the water column and into the atmosphere (Huttunen et al., 2003; Abril et al., 2005), and (3) downstream emissions, which include degassing (diffusive emissions in the turbulent waters downstream of reservoirs, called degassing emissions to differentiate them from the diffusive emissions at the reservoir's

Table 1. Description of study site.

Study site		Latitude	Longitude	Surface (km^2)	Residence time (month)	Mean depth (m)	Maximum depth (m)
Eastmain River watershed	Eastmain 1 reservoir (created in 2005)	52.19° N	75.05° W	603	2.3	16	63
	Mistumis Lake	52.16° N	76.16° W	4	N/A	5	12
	Clarkie Lake	52.23° N	75.47° W	24	N/A	6	18

surface) and diffusion and bubbling in the river downstream of the generating station (Soumis et al., 2004; Roehm and Tremblay, 2006). Tremblay et al. (2005), Tremblay and Bastien (2009) and Tremblay et al. (2010) have shown that in Eastmain 1 reservoir, diffusive emissions comprising over 95% of total gross emissions, with degassing and bubble emissions representing less than 5% of the total. Therefore, in this study we have focused on diffusive flux.

In northern temperate and boreal regions, seasonal variations in diffuse emissions of carbon (CO_2 and CH_4) from freshwater ecosystems are difficult to measure directly, because the ice cover during winter prevents flux measurements from being taken with floating chambers. Also, continuous monitors, such as eddy covariance towers, are still controversial, because of possible interference with, for example, the surrounding forest (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, in certain conditions, CO_2 and CH_4 can accumulate under ice (Riera et al., 1999; Kortelainen et al., 2000; Striegl et al., 2001) and be released as diffusive flux on spring ice break-up (Michmerhuizen and Striegl, 1996; Huttunen et al., 2004; Duchemin et al., 2006).

The objectives of our study were to (1) present a follow-up from 2006 to 2008 of GHG partial pressures and flux measured at different seasons in the Eastmain River watershed area; (2) address the question of gas accumulation under ice cover (increase in partial pressure) in both lakes and reservoir by comparing results of winter and summer field surveys; and (3) propose a way to estimate annual gross diffusive GHG flux from seasonal field campaign measurements. We present a comparison of gross diffusive emissions from Eastmain 1 reservoir and two nearby lakes to document the effect of anthropogenic reservoir creation on aquatic GHG emissions (as performed by Huttunen et al., 2003, for European reservoirs).

2 Methodology

2.1 Study sites

Eastmain 1 reservoir, Mistumis Lake and Clarkie Lake were sampled in the Eastmain River watershed (Table 1), in the boreal zone, northwestern Québec, Canada (Fig. 1). 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. The Eastmain River's catchment is dominated by coniferous forest, shallow podzolic and peat soils, and igneous bedrock. The aquatic systems studied are oligotrophic, with an overall low primary production (Planas et al., 2005). They are partially to totally covered by ice from about 15 December to 15 May (according to Hydro-Québec surveys). For safety reasons, winter field surveys could only be done between early January and the end of March, when the ice was at least 20 cm thick.

Eastmain 1 reservoir was four years old at the time of the study, and it has been observed that it can take around 10 years after reservoir creation for the labile flooded organic matter to decompose and for diffusive CO_2 flux to return to levels observed in natural lakes before reservoir creation (Chartrand et al., 1994; Tremblay et al., 2005). Accordingly, higher CO_2 emissions were expected in Eastmain 1 reservoir than in the natural lakes studied.

Sampling dates between 2006 and 2008 and the number of sampling stations (visited once per sampling period) for each water body are described in Table 2. When Eastmain 1 reservoir was created, three types of ecosystems were flooded: forest (57%), lakes and river (25%), and peatlands (18%). For the purposes of the Eastmain 1 reservoir GHG survey program, a large number of sampling stations were chosen to cover the spatial variability of the flooded landscape (Fig. 1, all campaigns together) and to allow net emission calculation (Tremblay et al., 2010). At least three sampling stations were chosen above each flooded lake or peatland of interest. As well, several sampling stations were chosen above the flooded riverbed. Two or three sampling stations were chosen per lake studied, with one station in deep water. Locations remained the same during each field survey. The number of stations sampled depended on weather conditions (especially
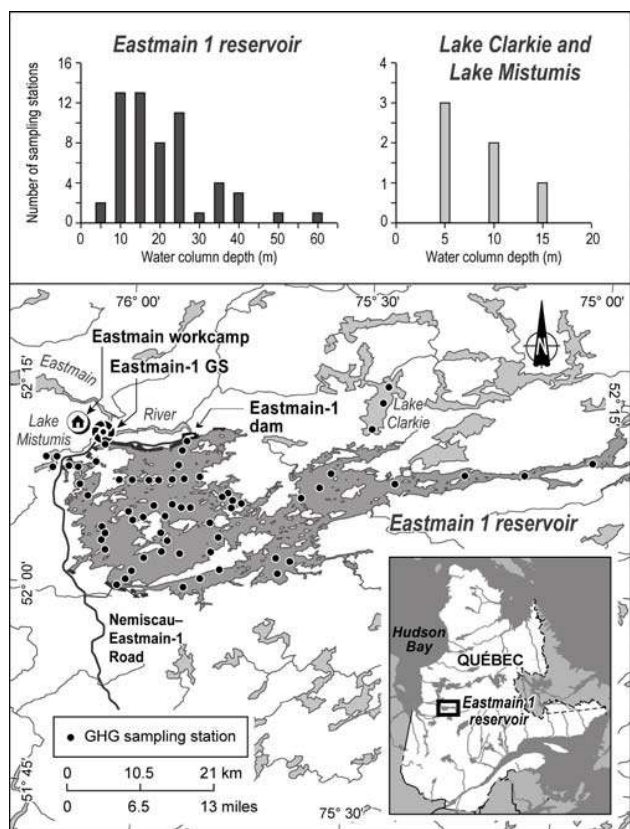
Fig. 1. Map of sampling stations in Eastmain River watershed; graphs representing distribution of sampling depths.

in winter), with at least one station per sampling section (e.g., one flooded lake of interest).

Temperature and dissolved oxygen profiles determined during the field surveys showed that there was no water column stratification (neither thermocline nor oxycline), whatever the season (Demarty et al., 2008)

2.2 Spatial coverage of CO$_2$ and CH$_4$ partial pressures

The partial pressure of CO$_2$ and CH$_4$ (pCO$_2$ and pCH$_4$) was measured at the water surface (0.1 m) for all sampling stations at Eastmain 1 reservoir, Mistumis Lake and Clarkie Lake. Also, at Eastmain 1 reservoir, pCO$_2$ and pCH$_4$ profiles were produced for 11 sampling stations in March 2007, 17 stations in January 2008, 2 stations in March 2008, 13 stations in July 2008 and 13 stations in September 2008. Stations were chosen to represent the reservoir's spatial variability and according to weather conditions. At Mistumis Lake, pCO$_2$ and pCH$_4$ profiles were produced at one station during each field survey. Three sampling depths were chosen according to the station's water depth, with one station at the bottom of the water column and the others at one third and two thirds of the water column's depth.

To measure pCO$_2$, water was sampled with a peristaltic pump and surface water pCO$_2$ was measured in situ with a nondispersive infrared (NDIR) sensor (EGM-4 manufactured by PP Systems) coupled with a gas exchanger (Membrana Celgard). Ten consecutive measurements (one per minute) were averaged to obtain the pCO$_2$ at each station (mean variation coefficient of 1.3%).

To determine pCH$_4$, three 30-mL water 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. Partial pressure of CH$_4$ in the headspace (pCH$_4$ HS) was quantified by means of a gas chromatograph (with a flame ionization detector) within 24 h. Partial pressure before equilibration (pCH$_4 f$) was determined using Eqs. (1) and (2):

$$p\text{CH}_4 f = \frac{(p\text{CH}_4\,\text{HS} \times K_{\text{H Eq.}}) + (\text{HSR} \times \frac{(p\text{CH}_4\,\text{HS} - p\text{CH}_4 i)}{V_\text{m}})}{K_{\text{H Sample}}} \quad (1)$$

where HSR is the headspace ratio (here equal to 1); pCH$_4 i$ is equal to 0, since the only gas in the air inside the syringe before equilibration was N$_2$; V_m is the molar volume (according to Avogadro's law); $K_{\text{H Eq.}}$ and $K_{\text{H Sample}}$ are the gas partition constants at equilibrium (20 °C) and at sampling temperature, respectively, calculated according to Lide (2007):

$$\ln K_\text{H}(\text{CH}_4) = -115.6477 + \frac{155.5756}{(T_\text{K}/100)} + 65.2553$$
$$\times \ln(\frac{T_\text{K}}{100}) - 6.1698 \times (\frac{T_\text{K}}{100}) \quad (2)$$

where T_K is the temperature in kelvin. K_H (CH$_4$) is in molar fraction atm^{-1} (Lide, 2007) but converted to mole L^{-1} atm^{-1} using the following factor, ($\frac{1000\,\text{g L}^{-1}\,[\text{water density}]}{18.0153\,\text{g/mole [water molecular weight]}}$). The field pCH$_4$ was obtained by averaging the results from three sampling syringes.

To obtain mean profiles of the water bodies, for each field surveys of Eastmain 1 reservoir, results from same sampling depth were averaged (different sampling station were thus considered as replicates at the reservoir/lake scale). This method of averaging data gives a rough snapshot of the state of the whole reservoir, but does not provide a fine description of the processes at work. We found that the means calculated at each sampling depth had an average coefficient of variation of 20%. At Mistumis Lake, three stations were sampled and a GHG profile was produced at a single station (explaining the absence of an error bar in Fig. 4a and c).

Table 2. Sampling periods and number of GHG (and water quality for Eastmain 1 reservoir and Mistumis Lake) sampling stations for each water body studied.

System	Sampling periods	Ice/Ice-free	Number of sampling stations		
			Eastmain 1 reservoir	Mistumis Lake	Clarkie Lake
Eastmain River	10–22 July 2006	Ice-free	41	3	2
	18 September–4 October 2006	Ice-free	42		
	20–27 March 2007	Ice	39	3	
	5–21 July 2007	Ice-free	38	3	3
	14–26 January 2008	Ice	42	3	3
	26 March–5 April 2008	Ice	35	3	2
	3–22 July 2008	Ice-free	57	3	3
	15–22 September 2008	Ice-free	27	3	2

2.3 Time series of pCO$_2$ and pCH$_4$ and temperature from Eastmain 1 reservoir

Automated GHG systems (monitors) have been in place at Eastmain-1 generating station (GS) since 2006 to obtain GHG partial pressure time series. The continuous gas monitor is modeled after Carignan's design (1998) and built of commercial components. Every three hours, CO_2 and CH_4 are measured by two different sensors (LICOR LI-820 NDIR sensor and Neodym Panterra metal oxide semiconductor sensor, respectively) on a gas stream that has been equilibrated with the source water (Demarty et al., 2009). Measurements taken at a single sampling station with monitors installed in the GS (Québec and Manitoba, Demarty et al., 2009) have been shown to be representative of conditions in the whole reservoir. Generally, no CH_4 accumulation under ice cover has been observed, contrary to CO_2 (see Fig. 2 for the example of results from Eastmain-1 GS GHG monitor). Trends observed from GHG monitoring measurements were used to determine the trends between field surveys and to calculate maximum pCO$_2$ reached before ice break-up and flux accordingly (Fig. 3 and Sects. 3.4 and 3.5).

2.4 Calculation of CO$_2$ and CH$_4$ flux

We calculated mean pCO$_2$ and pCH$_4$ for ice-free periods, and thereby estimated mean flux using corresponding mean water temperatures (data from automated systems at Eastmain-1 GS) and wind speeds (data from Nemiscau Airport meteorological station, 69 km away from the study area).

According to MacIntyre et al. (2005), flux calculations require conversion of 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), using Eqs. (3) and (4) (Morel, 1982; Anderson, 2002):

$$CO_2wc = K_H \times CO_2wp \qquad (3)$$
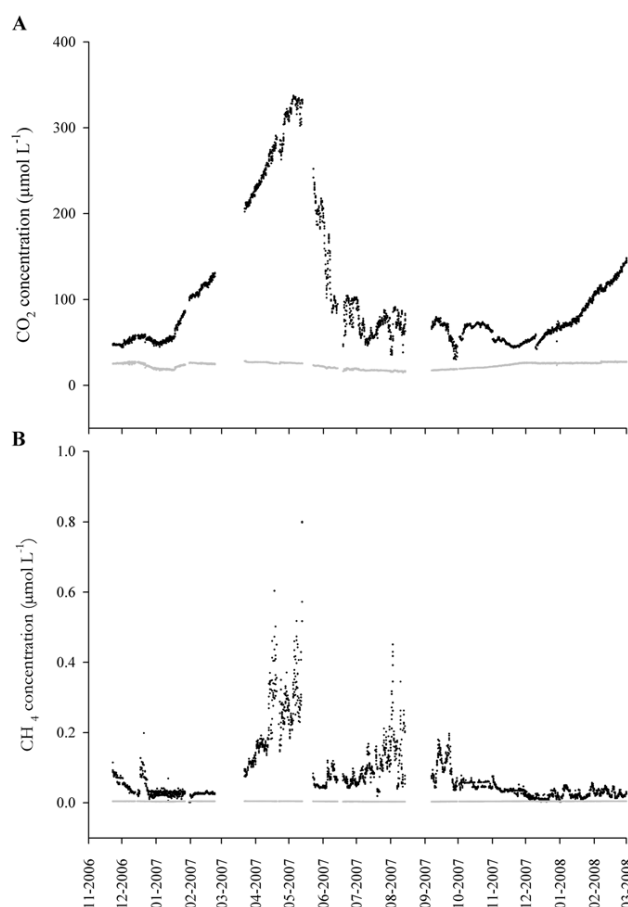
Fig. 2. Time series of CO_2 and CH_4 concentrations measured by GHG monitors at Eastmain-1 generating station, 2006–2008.
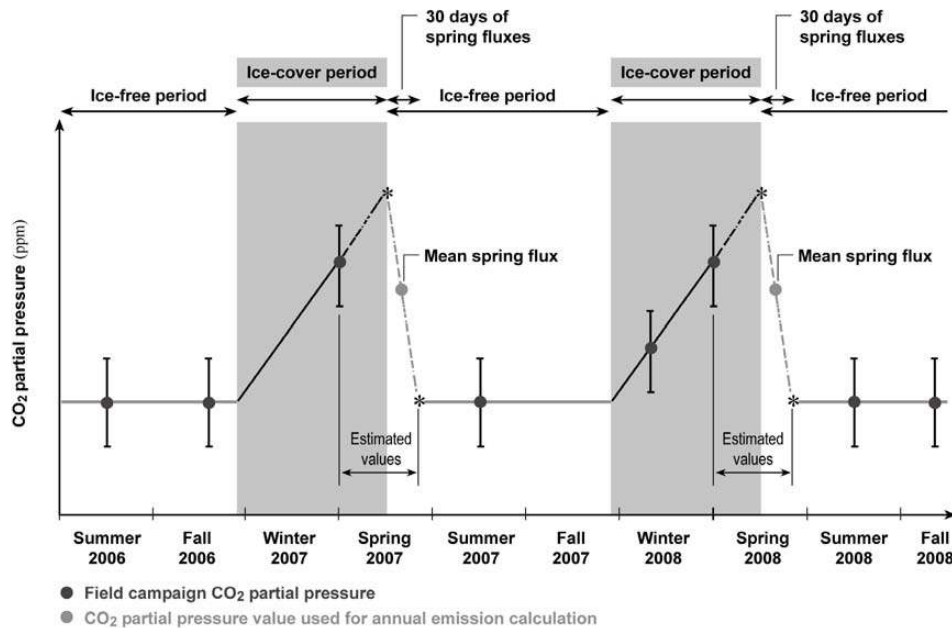
Fig. 3. Schematic representation of estimated annual emissions based on seasonal field surveys for CO_2-supersaturated systems. Dotted line represents partial pressure of CO_2 at water surface in equilibrium with atmosphere.

$$CH_4wc = K_H \times CH_4wp \tag{4}$$

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

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

where K_H (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 kelvin (Weiss, 1974).

Then, CO_2 and CH_4 flux was calculated using Eq. (6) (MacIntyre et al., 1995),

$$Flux = k_x(Cw - Ca) \tag{6}$$

with Ca 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}):

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

where x is equal to 0.66 for wind speed ≤ 3 m s^{-1} and is equal to 0.5 for wind speed >3 m s^{-1}; Sc is the Schmidt number for CO_2 or CH_4, which is dependent on temperature (T, in kelvin) 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):

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

The flux obtained from Eq. (6) is converted from moles to grams. 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):

$$GHG\ Flux(CO_2eq) = CO_2flux + 25 \times (CH_4\ flux) \tag{11}$$

3 Results

3.1 Interannual variation and comparison of reservoir and nearby lakes

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

Mean summer water surface pCO_2 measured at Eastmain 1 reservoir decreased from 2006 to 2008, with 2205 ppm, 1269 ppm and 1126 ppm in 2006, 2007 and 2008, respectively. Mean winter water surface pCO_2 was higher and not significantly different in 2007 and 2008 (2798 ppm in March 2007 and 2529 ppm in March 2008; t-test, $p > .05$). So a winter increase in partial pressure does not appear to

Table 3. Mean surface pCO$_2$ and pCH$_4$ ($\pm$ standard deviation) measured at Eastmain 1 reservoir, Mistumis Lake, Clarkie Lake, Robert Bourassa reservoir, Duncan Lake and Yasinski Lake. The number of replicates corresponds to the number of sampling stations given in Table 2.

Field surveys	Eastmain 1 reservoir		Mistumis Lake		Clarkie Lake	
	pCO$_2$	pCH$_4$	pCO$_2$	pCH$_4$	pCO$_2$	pCH$_4$
	ppm					
July 2006	2230$\pm$563	125$\pm$151	565$\pm$30	33$\pm$5	558$\pm$13	–
September 2006	2181$\pm$485	83$\pm$65	–	–	–	–
March 2007	2798$\pm$708	20$\pm$31	1441$\pm$47	8$\pm$2	–	–
July 2007	1333$\pm$317	–	568$\pm$47	–	496$\pm$42	–
January 2008	1211$\pm$194	40$\pm$82	856$\pm$25	17$\pm$2	716$\pm$31	11$\pm$1
March 2008	2529$\pm$796	287$\pm$982	1533$\pm$268	19$\pm$9	975$\pm$171	13$\pm$2
July 2008	1025$\pm$361	58$\pm$56	620$\pm$73	25$\pm$19	507$\pm$34	9$\pm$2
September 2008	1340$\pm$459	38$\pm$38	454$\pm$18	24$\pm$7	425$\pm$4	17$\pm$5

affect summer trends, which indicate that the limiting processes for CO$_2$ production are not the same in winter (temperature limits the decomposition of organic matter) and in summer (organic matter availability would be the limiting factor, decreasing over time). For Mistumis and Clarkie Lakes, summer and winter values were not different from year to year (ANOVA and t-test, $p > .05$).

Winter and summer surface pCO$_2$ measured at Eastmain 1 reservoir are higher than pCO$_2$ measured at the two lakes nearby (ANOVA, $p < .05$).

There were no significant differences between pCH$_4$ measured at the reservoir and nearby lake surfaces in the different sampling periods, because of the high spatial variability observed in the reservoir (ANOVA, $p > .05$).

3.2 Dissolved gas profiles in Eastmain River watershed

Figure 4 shows the seasonal pCO$_2$ and pCH$_4$ profiles for Eastmain 1 reservoir and Mistumis Lake. For Mistumis Lake, an increase in pCO$_2$ with depth was observed for under-ice measurements, reflecting winter CO$_2$ buildup (Fig. 4a). 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. 4b), reflecting CO$_2$ accumulation under the ice.

At Mistumis Lake, there was no trend in pCH$_4$ with depth, except in September 2008 (Fig. 4c), where pCH$_4$ increased from 20 ppm at 7.5 m to 152 ppm at 9 m. Unfortunately, no other summer or fall data are available from a depth of 9 m, preventing us from reaching any conclusion about the potential recurrence of such an event.

At Eastmain 1 reservoir, there was no trend in pCH$_4$ with depth, except in March 2007 (Fig. 4d). During that survey, we measured pCH$_4$ from the surface to a depth of 21 m. Most of the pCH$_4$ profiles gave similar values from surface to bottom, with no CH$_4$ accumulation under the ice, but the trends were different at three of the seven sampling stations. Indeed, for stations KP 45-3 (at depths of 10 m and 13 m), KP 260-4 (at 21 m only) and Tower-3 (at 21 m only), high pCH$_4$ was measured (Fig. 4d), corresponding to low to very low dissolved oxygen saturation (50%, 15.5%, 16.7% and 21.6%, respectively, data from temperature and dissolved gas profiles not shown), thus suggesting CH$_4$ accumulation in deep zones. We cannot confirm a regular under-ice buildup of CH$_4$, since January 2008 profiles at the same stations showed no increase with depth and the same profiles are not available for the March 2008 survey. However, low dissolved oxygen saturation seen in March 2008 below a depth of 20 m (data not shown) may have led to similar favorable conditions for CH$_4$ production 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 high surface pCH$_4$ (1442 ppm, 2221 ppm and 5439 ppm), leading to the high mean surface partial pressure observed in Fig. 4d (303 ppm, standard deviation $\pm$ 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 survey. Accordingly, we will consider that there was no CH$_4$ accumulation under ice cover for either Eastmain 1 reservoir or Mistumis Lake, as already suggested by the GHG monitor data.

3.3 Estimate of maximum dissolved pCO$_2$ reached before ice break-up

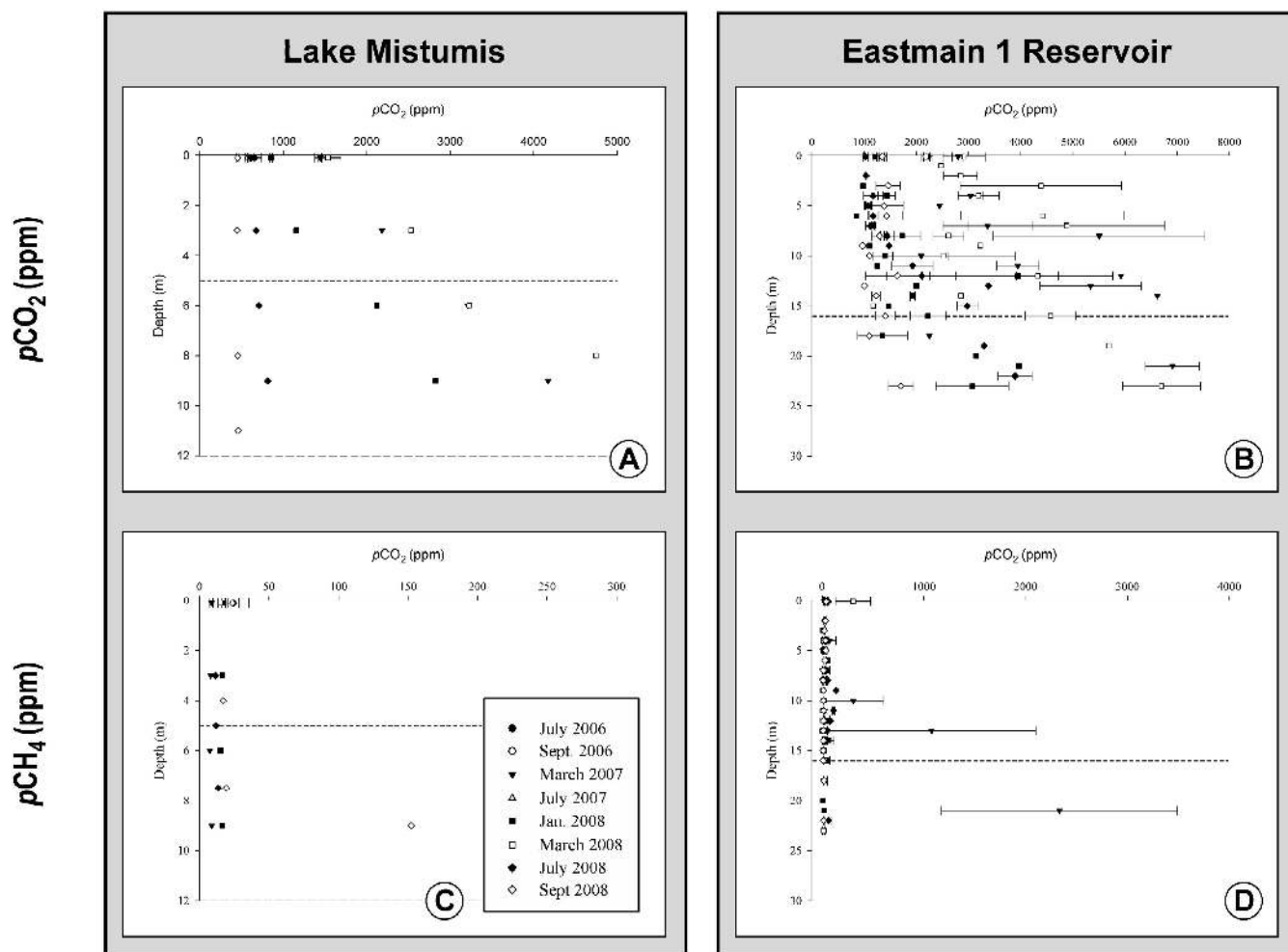CO$_2$ accumulation under ice cover has been observed in Eastmain 1 reservoir, Mistumis Lake and Clarkie Lake:

Fig. 4. Mean profiles of pCO_2 and pCH_4 for Eastmain 1 reservoir and Mistumis Lake. Bars represent standard errors. Dashed lines represent maximum depth (for Mistumis Lake) and dotted lines represent mean depth.

Table 4. Daily rates of under-ice pCO_2 increase and maximum potential pCO_2 reached by 15 May at study sites.

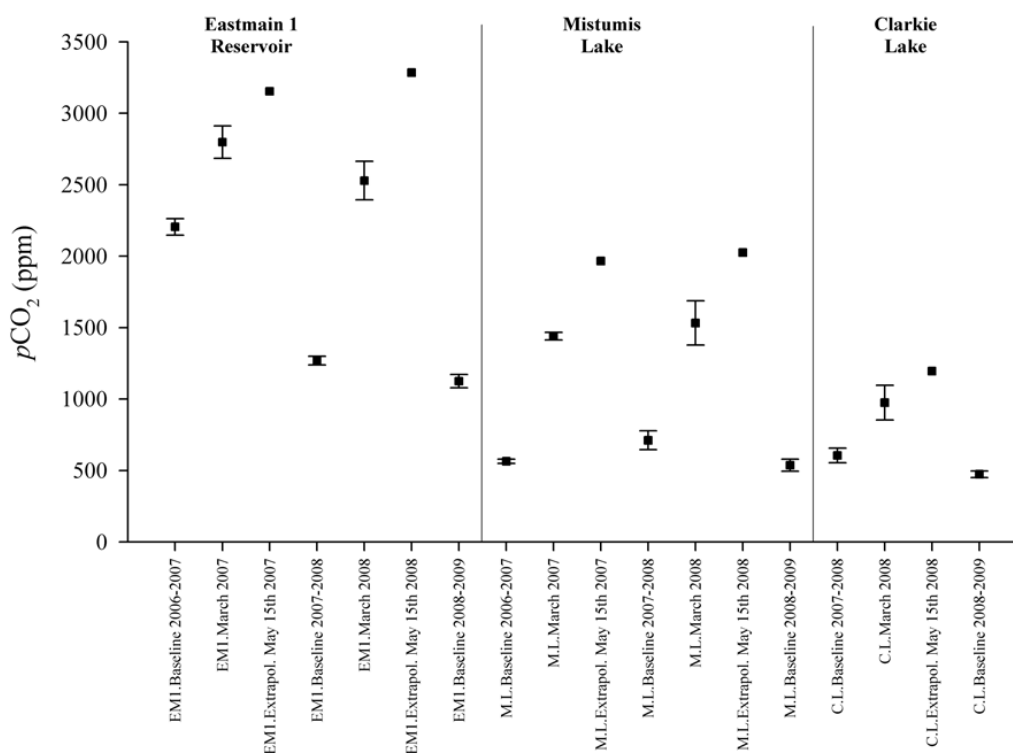| Study site	n	Springtime daily rates of pCO_2 increase ppm d^{-1}	SD ppm d^{-1}	R^2	Potential pCO_2 reached. 15 May (ppm)		
					Mean	Min.	Max.
Eastmain 1 res., 2007	122	7.91	16.79	0.18	3154	0	5168
Eastmain 1 res., 2008	116	16.80	14.26	0.59	3285	1574	4996
Mistumis Lake, 2007	7	11.68	1.02	0.99	1967	1844	2089
Mistumis Lake, 2008	9	10.95	5.60	0.83	2026	1354	2698
Clarkie Lake, 2008	8	4.90	4.00	0.65	1196	1085	2266

Fig. 5. Measured and extrapolated partial pressure of CO_2 for different systems studied. Bars represent standard errors. Baseline points represent mean values during ice-free periods.

spring values (i.e., in March–April) are clearly higher than what was measured during the other sampling period (ANOVA, $p < .05$; Table 4 and Fig. 5), whereas no differences in summer, fall and early winter (before January) mean dissolved pCO_2 was observed (ANOVA, $p > .05$). Taken together, the later data thus represent the baseline pCO_2 of the year (Fig. 5). A higher dissolved pCO_2 is observed in March than in January, and a linear increase of dissolved pCO_2 is expected under the ice cover from January to May (Fig. 3). 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 the two sampling dates, 15 January and 30 March; Table 4). Taking the date of 15 May as the beginning of the spring emission period (based on observations from automated systems), we thus estimated potential pCO_2 before the ice break-up (Table 4). From these calculations, it is clear that Eastmain 1 reservoir tends to present the highest pCO_2 before the spring emission period.

3.4 Estimate of potential spring gross diffusive GHG emissions

The method used to estimate the potential spring gross diffusive emissions is shown in Fig. 3. 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 area was assumed to last around one month, from 15 May to 15 June. Between these dates, flux was assumed to follow a linear trend. Maximum potential partial pressures were used to calculate the maximum potential flux at the beginning of the spring emission period, and baseline partial pressures were used to calculate potential flux 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. Accordingly, over one month, mean GHG flux calculated ranged between 37 gCO₂eq m⁻² (Clarkie Lake) and 147 gCO₂eq m⁻² (Eastmain 1 reservoir, 2008).

3.5 Annual cumulative diffusive flux

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 emissions for the water bodies studied are presented in Table 6. Spring CO_2 flux represented 23% (Eastmain 1 reservoir) to 52% (Mistumis Lake) of annual diffusive CO_2 emissions, and spring CH_4 flux represented 5% (Eastmain 1 reservoir) to 18% (Clarkie Lake) of annual diffusive CH_4 emissions. Clarkie Lake had the lowest diffusive emissions. For

Mistumis Lake and Eastmain 1 reservoir, 2008–2009 emissions were lower than in the previous year. As this study is part of a long-term monitoring program, the observed trend merits further discussion with regard to he future data.

4 Discussion

This study is the first to provide a multiannual estimation of spring diffusive GHG flux for both reservoirs and nearby lakes.

Comparisons of reservoirs with nearby lakes are commonly found in the literature (Åberg et al., 2004; Harrison et al., 2009). The limnological features of reservoirs more than 10 years old are generally comparable to those of lakes in the same region (Schetagne, 1994). Old reservoirs also behave like lakes in terms of GHG emissions (Demarty et al., 2008). References are needed when studying the evolution of newly impounded reservoirs such as Eastmain 1. In the case of the Eastmain 1 reservoir GHG monitoring program, the emissions of several lakes were studied before the reservoir was created, and Mistumis Lake and Clarkie Lake were found to be representative of lakes in the area (Blais and Tremblay, 2007). Theyhave therefore been chosen as references for studies after creation of the reservoir.

4.1 Choice of partial pressures used to estimate spring flux

The potential spring GHG flux of lakes and reservoirs is commonly calculated as the difference between the amount of accumulated gas (using the potential GHG storage of water bodies to obtain the concentration per square metre) and the amount of gas at atmospheric equilibrium (i.e., 385 ppm for CO_2, 1998; Striegl et al., 2001; Huttunen et al., 2004; Duchemin et al., 2006). But many lakes (Cole et al., 1994; Del Giorgio et al., 1999; Duarte and Prairie, 2005) and reservoirs (Tremblay et al., 2005; Demarty et al., 2009) around the world are supersaturated with CO_2, meaning that the partial pressure of CO_2 is generally above atmospheric partial pressure for most of the year. It is therefore misleading to consider that the partial pressure of dissolved GHG will reach equilibrium with the atmosphere after spring emissions. Since the difference between the highest partial pressure of dissolved GHG reached under ice cover and equilibrium is greater than the actual partial pressure measured in late spring (see Fig. 3), the misconception leads to overestimates of spring (and annual) flux. That is why, in this paper, we felt that using the difference between partial pressure at the water surface at the beginning and end of the ice-cover period was a better method of estimating spring diffusive flux.

4.2 Assumptions regarding trends in partial pressure

Two main assumptions were made regarding the estimation of maximum partial pressure of dissolved gases before ice break-up. First, we averaged the results of the ice-free period (summer, fall and end of December) to obtain a baseline flux, and then computed 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 flux. However, for Eastmain 1 reservoir, data from September 2008 (Fig. 4) showed that partial pressure of CO_2 and CH_4 did not increase with depth, so fall turnover should not have led to an increased flux; we consequently assumed that the use of an ice-free mean as baseline would be 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 rely on the results obtained from monitors that have been measuring pCO_2 and pCH_4 time series at Eastmain-1 GS since 2006 and at other Québec and Manitoba GS (Demarty et al., 2009). In that paper, the authors showed that annual estimates of diffusive emissions from Eastmain 1 reservoir calculated either from GHG monitor data (time series) or by sampling surveys gave comparable results. We therefore decided to use the trends observed in Eastmain 1 reservoir to compute annual emissions from water bodies unequipped with GHG monitors, such as lakes.

4.3 Source of CO_2 increase under ice cover

We observed CO_2 buildup under ice cover at both the reservoir and lakes under study. We did not perform any experiments to examine heterotrophic CO_2 production under ice cover, but it is probably due to the bacterial decomposition of organic matter, rather than to an underground water source. To discuss this hypothesis, the spring daily rates of CO_2 increase under ice cover presented in Table 4 were calculated in terms of concentration rather than partial pressure, that is, using Eqs. (1) and (4), assuming a surface temperature of $0.1\,^\circ C$, based on field observations. Rates varied between 0.3 and 0.9 $\mu g C\,L^{-1}\,h^{-1}$ at Clarkie Lake and Eastmain 1 reservoir, respectively. In their review of 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 $\mu g C\,L^{-1}\,h^{-1}$ (mean: 7.9 $\mu g C\,L^{-1}\,h^{-1}$); temperature is presented as an important factor influencing respiration rates. Interestingly, the under-ice rates of CO_2 increase observed in our systems are comparable to the lower range reported by Pace and Prairie (2005). This strongly suggests that CO_2 increases under ice are due to bacterial respiration under the influence of cold temperatures, concomitant with low primary production, which hides the respiration rate in warmer conditions. This hypothesis is supported by Striegl et al. (2001), who demonstrated that high dissolved pCO_2 prior to ice melt

Table 5. Spring CO_2, CH_4 and GHG flux. 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 (mmolCO_2eq m^{-2} d^{-1})	Total GHG spring flux (gCO_2eq m^{-2})
Eastmain 1 reservoir	Beginning 2007 End 2007	131 40	0.017 0.017	86	0.017	86	117
	Beginning 2008 End 2008	173 43	0.059 0.012	108	0.036	108	147
Mistumis Lake	Beginning 2007 End 2007	75 15	0.010 0.010	45	0.010	45	61
	Beginning 2008 End 2008	98 9	0.036 0.049	53	0.043	53	73
Clarkie Lake	Beginning 2008 End 2008	49 5	0.023 0.015	27	0.019	27	37

Table 6. Ice-free period, spring and annual CO_2, CH_4 and GHG emissions for water bodies studied. Note that emissions from reservoirs are gross emissions.

Study site and period	CO_2				CH_4				GHG	
	Ice-free period CO_2 emissions	Spring CO_2 emissions	Annual CO_2 emissions	Spring flux/ annual flux	Ice-free period CH_4 emissions	Spring CH_4 emissions	Annual CH_4 emissions	Spring flux/ annual flux	Annual GHG emissions	CO_2 contribution
	mmol m^{-2}			%	mmol m^{-2}			%	gCO_2eq m^{-2}	%
Eastmain 1 res., 2007–2008	8747	2654	11 401	23	7	1	7	7	509	98.6%
Eastmain 1 res., 2008–2009	7374	3348	10723	31	20	1	21	5	493	95.6%
Mistumis Lake, 2007–2008	3248	1392	4640	30	2	0	3	12	207	98.8%
Mistumis Lake, 2008–2009	1528	1658	3186	52	8	1	10	13	150	93.5%
Clarkie Lake, 2008–2009	903	834	1737	48	3	1	3	18	80	95.9%

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 only four years ago (in 2006), and the flooding of large quantities 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 few after flooding, CO_2 flux and partial pressure decline, reaching values comparable to those of natural aquatic ecosystems within about 10 years (Tremblay et al., 2005). After this transition period, 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) occurred the third year after flooding (2008) (Tremblay et al., 2009).

4.4 CH_4 emissions

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, especially in 2008. In 2007, at three of the seven stations where pCH_4 was sampled, pCH_4 increased with depth, probably due to the weak oxygen concentration there, as observed by Kortelainen et al. (2000). The ancillary available information (e.g., sampling station location, type of flooded land) cannot explain the origin of the high pCH_4 observed at these stations. Huttunen et al. (2004) also observed no CH_4 accumulation under ice cover in 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 partial pressure of CH_4 in surface and hypolimnetic water is negatively correlated with oxygen and lake depth and area, whatever the season. This supports the observation of low winter partial pressure of CH_4 in the large reservoir studied.

We found that diffusive CH_4 emissions, in contrast to CO_2 emissions, were of little concern in the boreal systems studied. Thix finding tallies with those of Juutinen et al. (2009) and Kortelainen et al. (2006) with respect to Finnish lakes. They reported that CH_4 emissions amounted to 49 mmol m^{-2} (1.8% of total emissions) (Juutinen et al., 2009), vs. CO_2 emissions of only 3.5 mol m^{-2} (reported as 42 gC m^{-2} in Kortelainen et al., 2006).

Potential spring diffusive GHG emissions from lakes (given in Table 5) are comparable to those measured in a boreal lake by Huttunen et al. (2004; 103 to 128 gCO$_2$eq m^{-2}). Spring GHG emissions represented 23% to 52% of annual GHG emissions. We also did annual estimates of diffusive GHG emissions at Robert Bourassa reservoir (Québec, Canada) with data collected in 2006 (up to 29 sampling stations) and we found higher potential spring emissions (22% of annual emissions) than those reported by Duchemin et al. (2006) for shallow areas, whereas spring diffusive GHG flux amounted to only 7% of annual flux at Robert Bourassa reservoir. Our results also demonstrated that, generally speaking, over 95% of annual diffusive GHG emissions from the studied systems are carbon dioxide.

5 Summary

We have provided the results of surveys of partial pressure and diffusive flux of GHG in both lakes and a reservoir in northwestern Québec, which allowed us to reliably estimate spring GHG flux based on a few assumptions. The method we used to estimate spring emissions can be generally applied.

Spring diffusive GHG emissions accounted for a higher proportion of annual GHG emissions than what has been previously proposed, with CO_2 being largely responsible for total annual diffusive emissions.

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