

# Release of CO<sub>2</sub> and CH<sub>4</sub> from small wetland lakes in western Siberia

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## ABSTRACT

CO<sub>2</sub> and CH<sub>4</sub> fluxes were measured from three small wetland lakes located in the middle taiga and forest tundra zones on West Siberian Lowlands (WSL), the world's largest wetland area. Fluxes were measured during summer 2005 using floating chambers and were validated against the thin boundary layer model based on the relationship between gas exchange and wind speed. All studied lakes were supersaturated with CO<sub>2</sub> and CH<sub>4</sub>, and acted on a seasonal basis as sources of these greenhouse gases to the atmosphere. Daily mean CO<sub>2</sub> fluxes measured with chambers ranged from near the zero to 3.1 g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> and corresponding CH<sub>4</sub> fluxes from 1.1 to 120 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. CH<sub>4</sub> ebullition (0.65–11 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) was detected in two of the lakes. Total carbon evasion from the studied lakes during the active season was 23–66 g C m<sup>-2</sup>, of which more than 90% was released as CO<sub>2</sub>-C. The carbon loss per unit area from the studied lakes was of similar magnitude as previously reported values of net carbon uptake of Siberian peatlands. This emphasizes the importance of small water-bodies in the carbon balance of West Siberian landscape.

## 1. Introduction

West Siberian Lowlands (WSL) is the largest wetland area of the world. The relatively humid, cool climate accompanied by poor drainage across the flat terrain has maintained the accumulation of organic matter and development of an important carbon pool. The total area of  $0.6 \times 10^6$ – $1.0 \times 10^6$  km<sup>2</sup> of the West Siberian peatlands contains a carbon pool of 51–70 Pg C (Pg = 10<sup>15</sup> g) (Yefremov and Yefremova, 2001; Sheng et al., 2004). These values represent 18–30% of the area and 11–15% of the peat carbon pool of all boreal and subarctic peatlands (Gorham, 1991). Extensive wetland areas, such as WSL, are also characterized by high amount of small and shallow wetland lakes and ponds, whose role in the regional carbon cycle is poorly understood.

Although the global lake area is only 2.8% of the non-oceanic land area (Downing et al., 2006), lakes play an important role in the processing and transport of terrestrially fixed carbon to the atmosphere. Heterotrophic metabolism that produces CO<sub>2</sub> and CH<sub>4</sub> in the water column and in lake sediments is largely

driven by carbon input from surrounding terrestrial ecosystems (Kling et al., 1991; Cole et al., 1994; Algesten et al., 2005; Kortelainen et al., 2006). Aerobic decomposition of allochthonous and autochthonous carbon is the main process producing CO<sub>2</sub> in the lakes, while CH<sub>4</sub> is an important end-product in anaerobic decomposition. Carbon not only enters lakes as organic matter, but also as free CO<sub>2</sub> and CH<sub>4</sub> in stream waters draining the surrounding terrestrial ecosystems, especially in peatland dominated catchments (Billett et al., 2004). It has been estimated that  $513 \times 10^{12}$  g of CO<sub>2</sub> (Cole et al., 1994) and  $8$ – $48 \times 10^{12}$  g of CH<sub>4</sub> (Bastviken et al., 2004) are annually released from the world's lakes to the atmosphere. Thus, lakes should be taken into account in the regional carbon budgets in landscapes that are rich in lakes.

Recent studies have suggested that the importance of small lakes and ponds in the global lake area and further, in the global carbon cycle might have been underestimated (Downing et al., 2006; Kortelainen et al., 2006). Small wetland lakes and ponds that are often abundant in peatland areas can have high CO<sub>2</sub> and CH<sub>4</sub> emissions (e.g. Hamilton et al., 1994; Riera et al., 1999). Moreover, it has been shown that the small areas of high CH<sub>4</sub> emissions, such as wetland ponds, can largely contribute to the landscape-scale CH<sub>4</sub> budgets in wetland regions and create a

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major uncertainty in the areal CH<sub>4</sub> emission estimates (Bubier et al., 2005).

The aim of this study was to determine the magnitude of CO<sub>2</sub> and CH<sub>4</sub> fluxes from small wetland lakes typical of WSL during summer season. To our knowledge, no data on the exchange of CO<sub>2</sub> and CH<sub>4</sub> in West Siberian lakes has been previously published. Diffusive CO<sub>2</sub> and CH<sub>4</sub> fluxes from the lakes were determined with two methods: (i) the floating chamber technique and (ii) the thin boundary layer (TBL) model. In addition, CH<sub>4</sub> ebullition was measured with funnel gas collectors.

## 2. Material and methods

### 2.1. Study sites

The studied lakes are situated in two subarctic peatland areas on WSL. The southern study area (60°59'N, 70°10'E; 35–37 m a.s.l.) (referred to hereafter as 'MTaiga') is located in the middle taiga zone about 65 km east from Khanty-Mansiysk, the capital of Khanty-Mansiysk autonomous region, Russia. The climate in the region is continental with long and cold winter, warm summer and moderate annual rainfall of 550 mm. Mean temperature is 18.2 °C in July and –18.6 °C in January (Goddard Institute for Space Studies, 2006). Permafrost is absent in MTaiga. The dominating peatland types in the area are ombrotrophic pine bogs, ridge–hollow complexes and oligotrophic through-flow fens. In MTaiga, gas fluxes were measured on a shallow wetland lake ('MTlake') and a small wetland pond ('MTpond'). The ~1.5 m deep MTlake has an area of 23 ha and a flat, peaty bottom. Two sites in the lake were studied: one near the shore (Site 1) and one in the middle of the lake (Site 2). The ~1 m deep MTpond has an area of 0.5 ha. It is situated in a poor through-flow fen and is partly covered by *Menyanthes trifoliata* stands. In MTpond, we selected one measuring site in the middle of the pond. Concentrations of total organic carbon (TOC) and total phosphorus (tot-P), analysed once during the season, were 10 mg l<sup>-1</sup> and 6 µg l<sup>-1</sup> in MTlake and 17 mg l<sup>-1</sup> and 5 µg l<sup>-1</sup> in MTpond, respectively. Fluxes were measured seven times in MTlake and six times in MTpond between 3 July and 6 September 2005.

The northern study area (65°52'N, 74°58'E; 70–75 m a.s.l.) (referred to hereafter as 'FTundra') is located in the forest tundra zone characterized by discontinuous permafrost (permafrost thickness <100 m). It is situated about 85 km west from the city of Novy Urengoy in Yamalo-Nenets autonomous region, Russia. FTundra has continental climate and moderate annual rainfall of 440 mm. July is the warmest month with a mean temperature of 16.3 °C, and January is the coldest with a mean temperature of –24.7 °C (long-term climatic data from Tarko–Sale (64°55'N, 77°49'E)) (Goddard Institute for Space Studies, 2006). Landscape in FTundra is dominated by flat, frozen palsas with lichens and dwarf shrubs alternating with wet sedge-*Sphagnum* hollows. In FTundra, flux measurements were conducted on a 7 ha lake

with a flat, peaty bottom and a depth of 1.2–1.4 m ('FTlake'). TOC concentration in FTlake was 11 mg l<sup>-1</sup> and tot-P concentration 44 µg l<sup>-1</sup>. The two sites, Site 1 near the shore and Site 2 in the middle of the lake, were visited during two campaigns that took place in early July and late August 2005.

### 2.2. Surface water CO<sub>2</sub> and CH<sub>4</sub> concentrations

Surface water was sampled for dissolved CO<sub>2</sub> and CH<sub>4</sub> during chamber measurements (see below). Water samples were taken from a depth of 10 cm with a polypropylene syringe and shaken vigorously for 2 min with a headspace with known CO<sub>2</sub> and CH<sub>4</sub> concentration. CO<sub>2</sub> and CH<sub>4</sub> concentrations in headspace gas after shaking were analysed in a field laboratory on a gas chromatograph (Crystall 5000, JSC SBD Chromatek, Russia), equipped with a flame ionization detector (FID) for CH<sub>4</sub>. CO<sub>2</sub> was determined with FID after conversion to CH<sub>4</sub> by a methanizer. The GC had Porapak N (2 m × 2 mm, mesh size 80/100) and HayeSep N (2 m × 2 mm, mesh size 80/100) columns. Injector, column and detector temperatures were 325, 60 and 150 °C, respectively. Argon (30 ml min<sup>-1</sup>) was used as a carrier gas. Analysis time was 2 min.

Dissolved CO<sub>2</sub> and CH<sub>4</sub> concentrations in water samples were calculated with Henry's law taking into account the temperature dependence of solubility (Lide and Fredrikse, 1995). The determined CO<sub>2</sub> and CH<sub>4</sub> concentrations were compared with the theoretical concentrations in equilibrium with the atmosphere. The atmospheric concentrations used in the calculation were the seasonal mean concentrations of CO<sub>2</sub> and CH<sub>4</sub> in ambient air, sampled with polypropylene syringes at 1 m above the lake surface during the chamber measurements. The atmospheric CO<sub>2</sub> and CH<sub>4</sub> concentrations (mean ± SD) were 400 ± 60 and 2.2 ± 0.5 ppm in MTaiga (*n* = 20) and 420 ± 60 ppm and 2.3 ± 0.5 ppm in FTundra (*n* = 7), respectively. There were not consistent differences in atmospheric concentrations between the lake sites.

### 2.3. Chamber measurements

A closed floating chamber was used to measure diffusive CO<sub>2</sub> and CH<sub>4</sub> emissions from the lakes during daytime (Lambert and Fréchet, 2005). Our plexiglass chamber, equipped with a styrofoam float ensuring a floating depth of 5 cm, had dimensions of 60 × 60 × 30 cm (length × width × height) and a headspace volume of 70 dm<sup>3</sup>. A pressure compensation tube and sampling tube connected to a three-way stop-cock (Connecta, BOC Ohmeda AB, Sweden) were installed through a rubber septum in an inlet on the top of the chamber. Four gas samples were taken from the chamber headspace at 7 min intervals during a measuring period of 28 min with 35 or 60 ml polypropylene syringes (Terumo Europe, Belgium). Prior to sampling, the sampling tube was flushed with the chamber air several times.

Two to three replicate chamber measurements were made at each lake site on each sampling occasion. Air temperature inside the chamber, water temperature at 10 cm depth and air temperature at 1 m height were recorded during each measurement with a digital thermometer. Wind speed at a height of 1 m was measured with a hand-held anemometer several times during the 28 min measurement. Precipitation at the study sites was recorded using a rain gauge (Davis 7852 Rain Collector II, Davis, MD, USA) and a HOBO MicroStation Data Logger (HOBO H21-002, Onset Computer Corporation, MA, USA). The CO<sub>2</sub> and CH<sub>4</sub> concentrations in gas samples were analysed with the GC as described above. Gas fluxes were calculated from the linear change in gas concentration in chamber headspace with time. As a general rule, the regression lines with  $r^2 > 0.70$  were accepted. However, small fluxes from  $-1$  to  $1 \text{ mg CH}_4 \text{ m}^{-2} \text{ d}^{-1}$  and  $-0.1$  to  $0.1 \text{ g CO}_2 \text{ m}^{-2} \text{ d}^{-1}$  were included in the dataset regardless of their  $r^2$ -values. In this range  $r^2$ -values were limited by the accuracy of the measuring system and did not necessarily reflect the quality of the measurement. Thus, rejection of the small fluxes would have biased the results.

#### 2.4. TBL-model

CO<sub>2</sub> and CH<sub>4</sub> fluxes were calculated from local wind speed and surface water gas concentrations with the two-layer model described by Liss and Slater (1974), also known as the TBL model. Gas exchange between air and water can be described with the following equation:

$$F = k(c_{\text{sur}} - c_{\text{eq}}), \quad (1)$$

where  $c_{\text{sur}}$  is the gas concentration in surface water ( $M = \text{mol l}^{-1}$ ),  $c_{\text{eq}}$  is the gas concentration in surface water in equilibrium with the atmosphere ( $M$ ) and  $k$  is the gas exchange constant ( $\text{cm h}^{-1}$ ).  $k$  was calculated from local wind speed using two published relationships between wind speed and gas exchange (Cole and Caraco, 1998; Crusius and Wanninkhof, 2003), both determined for low wind environments. Crusius and Wanninkhof (2003) presented a bilinear relationship between wind speed and gas exchange:

If the wind speed is  $< 3.7 \text{ m s}^{-1}$ ,

$$k_{600}(\text{cm h}^{-1}) = 0.72\mu_{10} \quad (2)$$

and

if the wind speed is  $\geq 3.7 \text{ m s}^{-1}$ ,

$$k_{600}(\text{cm h}^{-1}) = 4.33\mu_{10} - 13.3, \quad (3)$$

where  $\mu_{10}$  = the local wind speed normalized to a height of 10 m ( $\text{m s}^{-1}$ ) and  $k_{600}$  = the gas transfer velocity for SF<sub>6</sub>, normalized to a Schmidt number ( $Sc$ ) of 600. The second relationship was the power relationship from Cole and Caraco (1998), where  $k$  at zero wind speed is  $\sim 2 \text{ cm h}^{-1}$ :

$$k_{600} = 2.07 + 0.215 \times \mu_{10}^{1.7} \quad (4)$$

Average wind speed at 1 m during the chamber measurement was converted to a height of 10 m according to Crusius and Wanninkhof (2003):

$$\mu_{10} = 1.22 \times \mu_1, \quad (5)$$

where  $\mu_{10}$  and  $\mu_1$  are the wind speeds at 10 and 1 m height. The equation is valid under the assumptions of a neutrally stable boundary layer, a logarithmic wind profile and a drag coefficient at 10 m height of  $1.3 \times 10^{-3}$  (Crusius and Wanninkhof, 2003).

The Schmidt number ( $Sc$ )—the ratio of kinematic viscosity of water and the diffusion coefficient—is different for different gases and depends on temperature. Schmidt numbers can be used to calculate gas transfer velocities for different gases from  $k_{600}$  values with the following equation (Jähne et al., 1987):

$$k_{\text{gas}}(\text{cm h}^{-1}) = k_{600} \left( \frac{Sc_{\text{gas}}}{600} \right)^x \quad (6)$$

The power dependence,  $x$ , is dependent upon the roughness of the water surface. A  $-0.67$  power dependence was used if the wind speed (at 1 m) was  $< 3 \text{ m s}^{-1}$ , while the value of  $-0.5$  was used if the wind speed was  $> 3 \text{ m s}^{-1}$  (Crusius and Wanninkhof, 2003).  $Sc$  for CO<sub>2</sub> and CH<sub>4</sub> were calculated using in situ temperatures according to Wanninkhof (1992):

$$Sc_{\text{CO}_2} = 1911.1 - 118.11t + 3.4527t^2 - 0.041320t^3, \quad (7)$$

$$Sc_{\text{CH}_4} = 1897.8 - 114.28t + 3.2902t^2 - 0.039061t^3, \quad (8)$$

where  $t$  is temperature in situ ( $^{\circ}\text{C}$ ).

After calculation of  $k_{600}$  values from wind speed,  $k_{\text{CO}_2}$  and  $k_{\text{CH}_4}$  were obtained using eq. (6) and  $Sc$  values for CO<sub>2</sub> and CH<sub>4</sub>.

#### 2.5. CH<sub>4</sub> ebullition

Submerged funnel gas collectors (see Huttunen et al., 2001) were used for continuous monitoring of CH<sub>4</sub> ebullition in the lakes. The gas collectors consisted of a 20 cm diameter funnel connected via a PVC-tube to a graduated 60 ml polypropylene syringe, which was equipped with a three-way stop-cock for sampling. Three to four replicate gas collectors were placed randomly in the vicinity of each lake site on 7 July 2005 in MTlake and MTpond and on 8 July 2005 in FTlake. Bubble gas volume in the gas collectors was read and samples were taken for analysis of CH<sub>4</sub> concentration three times during the summer in MTaiga and once in FTundra. The sampling interval was 5–30 d in MTaiga and 48 d in FTundra. The CH<sub>4</sub> flux by ebullition was calculated from the volume of the released bubble gas and the bubble CH<sub>4</sub> concentration analysed on GC-FID as described above.

### 3. Results and discussion

#### 3.1. CO<sub>2</sub> and CH<sub>4</sub> concentrations in surface water

All sites were supersaturated with CO<sub>2</sub> and CH<sub>4</sub> during open water conditions, a phenomenon typical of northern lakes (e.g. Kling et al., 1992; Cole et al., 1994). The surface water gas concentrations did not differ significantly between the two sites in MTlake or FTlake (*t*-test). Dissolved CO<sub>2</sub> concentration in surface water, [CO<sub>2</sub>], ranged from 22 to 160 μM in the taiga lakes, being 1.3 to 11 times higher than the theoretical atmospheric equilibrium concentration. Daily [CO<sub>2</sub>] tended to be higher in MTpond (92 ± 53 μM, mean ± SD) than in MTlake (51 ± 29 μM). In FTlake, [CO<sub>2</sub>] ranged from 36 to 115 μM (60 ± 15 μM, mean ± SD), which was 1.6–5.7 times the equilibrium concentration.

In MTaiga, surface water CH<sub>4</sub> concentrations peaked during midsummer and were low in the early and later parts of the season. [CH<sub>4</sub>] ranged from 0.066 to 7.8 μM in MTaiga, being 18 to over 2400 times the concentration in equilibrium with the atmosphere. As in the case of [CO<sub>2</sub>], higher [CH<sub>4</sub>] peaks were found in MTpond than in MTlake, while the mean concentrations in MTpond (2.6 ± 2.6 μM) and MTlake (0.28 ± 0.32 μM) were nearly similar. In FTlake, [CH<sub>4</sub>] ranged from 0.079 to 0.60 μM (0.29 ± 0.17 μM, mean ± SD), being 18–150 times the equilibrium concentration.

In shallow lakes, such as the lakes of this study, the water column is regularly mixed during the open-water period preventing any long-term seasonal stratification. However, diurnal stratification and mixing due to the difference between day and night temperatures can occur in shallow lakes and result in changes in the diurnal course of surface water gas concentrations (Ford et al., 2002). This can bias the flux estimates, if the fluxes are measured only once during daytime (Ford et al., 2002). The effect of daily mixing on surface water CO<sub>2</sub> and CH<sub>4</sub> concentrations, and further on the corresponding fluxes, was not accounted for in this study, but should be considered in future studies on shallow lakes.

#### 3.2. Chamber fluxes

All studied lakes acted as sources of CO<sub>2</sub> to the atmosphere during the open water season, indicating high carbon input from the surroundings in relation to the photosynthetic activity in the lake. In the whole data, net CO<sub>2</sub> uptake was found only in MTlake on 31 August, when the surface water CO<sub>2</sub> concentration in the lake was close to the equilibrium concentration with the atmosphere and the CO<sub>2</sub> was flux slightly negative (−0.07 g CO<sub>2</sub> m<sup>−2</sup> d<sup>−1</sup>). Lakes that are sources of CO<sub>2</sub> on annual basis may show CO<sub>2</sub> uptake during summer months, if the CO<sub>2</sub> concentration in the surface water drops below the atmospheric equilibrium due to CO<sub>2</sub> consumption by photosynthesis (Striegl and

Michmerhuizen, 1998; Riera et al., 1999). The mean CO<sub>2</sub> emission from MTpond, averaging 1.6 ± 0.9 g CO<sub>2</sub> m<sup>−2</sup> d<sup>−1</sup> (mean ± SD), was significantly higher than that from MTlake (0.5 ± 0.4 g CO<sub>2</sub> m<sup>−2</sup> d<sup>−1</sup>) (*t*-test, *p* < 0.05) (Table 1). At the MTaiga sites, the highest CO<sub>2</sub> emission rates were measured in the beginning of August. Mean CO<sub>2</sub> emission from FTlake was 1.5 ± 0.9 mg CO<sub>2</sub> m<sup>−2</sup> d<sup>−1</sup> (Table 1).

In MTlake, seasonal mean CH<sub>4</sub> flux was 8.1 ± 8.7 mg CH<sub>4</sub> m<sup>−2</sup> d<sup>−1</sup> (mean ± SD) (Table 1). Highest instantaneous CH<sub>4</sub> emissions were measured from MTpond, where the daily mean CH<sub>4</sub> emission ranged from 12 to 120 mg CH<sub>4</sub> m<sup>−2</sup> d<sup>−1</sup> (41 ± 41 mg CH<sub>4</sub> m<sup>−2</sup> d<sup>−1</sup>) (Table 1). However, the difference in CH<sub>4</sub> flux between MTlake and MTpond was not significant (*t*-test). High CH<sub>4</sub> flux rates from MTpond located in a through-flow fen can be explained by decomposition of underlying peat deposits (see Hamilton et al., 1994) and continuous water-flow from surrounding peatlands. *M. trifoliata* stands growing on the edges of MTpond may also promote CH<sub>4</sub> production by supplying sediment methanogenesis with fresh organic carbon (e.g. Whiting and Chanton, 1993). In FTlake, CH<sub>4</sub> emissions varied between 4.1 and 9.4 mg CH<sub>4</sub> m<sup>−2</sup> d<sup>−1</sup> averaging 6.1 ± 2.5 mg CH<sub>4</sub> m<sup>−2</sup> d<sup>−1</sup> (Table 1). Although the near-shore sites in MTlake and FTlake showed higher peak CO<sub>2</sub> and CH<sub>4</sub> emissions than the sites in the middle of the lakes, the differences between the sites within either of the lakes were not significant (*t*-test).

We tested the correlations between the measured environmental parameters and chamber fluxes in order to clarify, how well the gas emissions of our study lakes could be explained with the factors accounted for by the TBL-model (wind speed, gas concentration in the surface water) (Table 1; Fig. 1). Surface water CH<sub>4</sub> concentration was a good predictor of the temporal variation observed in chamber CH<sub>4</sub> flux in taiga lakes (Pearson correlation coefficient *R* = 0.929, *p* < 0.01, *N* = 7 in MTlake and *R* = 0.997, *p* < 0.01, *N* = 6 in MTpond) (Table 1; Fig. 1B). Although CO<sub>2</sub> flux also tended to increase with surface water CO<sub>2</sub> concentration, the correlation was not significant (Table 1). However, CO<sub>2</sub> flux correlated significantly with surface water CH<sub>4</sub> concentration (*R* = 0.818, *p* < 0.05, *N* = 7 in MTlake and *R* = 0.883, *p* < 0.05, *N* = 5 in MTpond) (Table 1; Fig. 1A). The strong correlation between CO<sub>2</sub> flux and CH<sub>4</sub> concentration is probably a consequence of the similar seasonality in CO<sub>2</sub> and CH<sub>4</sub> dynamics and does not necessarily imply a more direct relationship between the two variables. The highest concentrations of these gases as well as the highest flux rates were measured in taiga lakes in the beginning of August. This is an indication of high microbial activity during midsummer supported by high temperatures and supply of fresh substrates from surrounding peatlands. Gas fluxes followed the seasonal pattern in water temperature in all studied lakes, although the correlation between water temperature and gas flux was statistically significant only for CO<sub>2</sub> flux in MTlake (*R* = 0.884, *p* < 0.01, *N* = 7) (Table 1; Fig. 1D).

**Table 1.** The seasonal mean CO<sub>2</sub> fluxes (g m<sup>-2</sup> d<sup>-1</sup>) and CH<sub>4</sub> fluxes (mg m<sup>-2</sup> d<sup>-1</sup>) at the study sites (measured with chambers) and the Pearson correlations of the daily mean fluxes with the surface water gas concentrations, wind speed, water temperature and precipitation during the measurement. Mean fluxes for MTlake and FTlake were calculated averaging the daily mean values of two sites within the lakes. Positive flux values indicate release from the lake to the atmosphere

	MTlake CO <sub>2</sub> flux	CH <sub>4</sub> flux	MTpond CO <sub>2</sub> flux	CH <sub>4</sub> flux	FTlake CO <sub>2</sub> flux	CH <sub>4</sub> flux
Mean	0.5	8.1	1.6	41	1.5	6.1
Median	0.4	4.1	1.5	24	1.4	5.3
SD	0.4	8.7	0.9	41	0.9	2.5
Min	-0.07	1.1	0.9	12	0.5	4.1
Max	1.3	23.1	3.1	120	2.6	9.4
N <sup>a</sup>	7	7	5	6	4	4
Pearson correlations:						
[CO <sub>2</sub> ]	0.622	0.527	0.851	0.532	0.238	0.056
[CH <sub>4</sub> ]	0.818*	0.929**	0.883*	0.997**	0.746	0.670
Wind speed	0.892**	0.741	0.183	-0.129	-0.152	-0.103
T <sub>water</sub>	0.884**	0.501	0.372	0.608	0.929	0.927
Precipitation	-0.157	-0.053	- <sup>b</sup>	- <sup>b</sup>	0.974*	0.999**
CO <sub>2</sub> flux	1	0.656	1	0.853	1	0.971*
CH <sub>4</sub> flux	0.656	1	0.853	1	0.971*	1

<sup>a</sup>Number of the measuring days.

<sup>b</sup>Precipitation during the measurements was zero.

\*Correlation is significant at the 0.05 level.

\*\*Correlation is significant at the 0.01 level.

Wind significantly enhanced the CO<sub>2</sub> release in MTlake ( $R = 0.892$ ,  $p < 0.01$ ,  $N = 7$ ), despite the wind speeds over the lake during the measurements were mostly low ( $< 4 \text{ m s}^{-1}$ ) and gas exchange has generally been considered to be independent of wind in low-wind environments (e.g. Cole and Caraco, 1998) (Table 1; Fig 1C). On the other hand, an increasing trend in gas exchange with wind has been found also at wind speeds  $< 3.7 \text{ m s}^{-1}$  (Crucius and Wanninkhof, 2003) or even  $< 1.0 \text{ m s}^{-1}$  (Matthews et al., 2003). In MTpond gas fluxes were independent of wind speed (Table 1). It has to be noted here that the chamber itself can alter the conditions at the water surface by increasing the turbulence in low-wind environments (Matthews et al., 2003) and sheltering the water in windy conditions (Duchemin et al., 1999), which may distort the actual wind-gas exchange dependence occurring on free water surface.

In FTlake, all the measured environmental variables—CO<sub>2</sub> and CH<sub>4</sub> concentrations in the surface water, wind speed, water temperature and precipitation—showed only minor variation between the measurements. CO<sub>2</sub> and CH<sub>4</sub> fluxes correlated positively with each other ( $R = 0.971$ ,  $p < 0.05$ ,  $N = 4$ ) and with precipitation during the chamber measurements ( $R = 0.974$ ,  $p < 0.05$ ,  $N = 4$  for CO<sub>2</sub>;  $R = 0.999$ ,  $p < 0.01$ ,  $N = 4$  for CH<sub>4</sub>) (Table 1). The enhancement of gas exchange by rain, caused by increased turbulence and bubble formation in the surface water has been demonstrated in laboratory experiments and in field studies (Ho et al., 1997; Cole and Caraco, 1998; Ho et al., 2000) and it can be important especially in low wind environments,

where the turbulence is low. However, with the limited amount of data, it remains uncertain if the low precipitation during our measurements (or, some other factor associated with the rainy conditions) could have affected the gas exchange in FTlake.

### 3.3. Comparison between measured and modelled fluxes

The fluxes measured with the chambers and calculated with the TBL-model were generally in good agreement in all studied lakes and the two models predicting gas exchange gave similar results (Fig. 2). The correlation between the measured and modelled CH<sub>4</sub> fluxes was highly significant regardless of which of the two wind-gas flux relationships was used (Fig. 2). Moreover, the tundra CH<sub>4</sub> fluxes fitted well to the same regression plot with the taiga results (Fig. 2). In the case of CO<sub>2</sub> fluxes, the correlation between the measured and modelled fluxes was lower, but it increased markedly if the FTlake data were left out ( $R = 0.413$  versus 0.671 with the bilinear relationship;  $R = 0.545$  versus 0.827 with the power relationship) (Fig. 2). In FTlake, the measured CO<sub>2</sub> fluxes were notably higher than the modelled ones (Fig. 2), reason for which remains unclear.

Looking at all data, CO<sub>2</sub> emissions calculated with the bilinear relationship (eqs 2 and 3; Crucius and Wanninkhof, 2003) did not differ significantly from the chamber results ( $t$ -test). When the power relationship (eq. 4; Cole and Caraco, 1998) was used, the TBL-model gave higher CO<sub>2</sub> flux values than the chamber method ( $t$ -test,  $p < 0.05$ ,  $N = 18$ ) (Fig. 1).

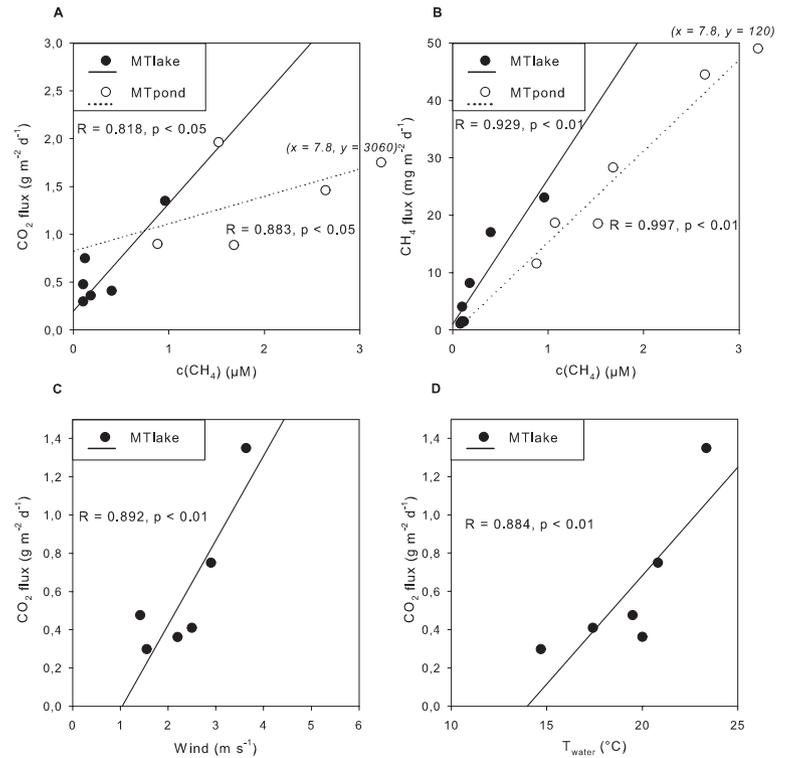


Fig. 1. The statistically significant correlations of the chamber CO<sub>2</sub> fluxes ( $\text{g m}^{-2} \text{d}^{-1}$ ) and CH<sub>4</sub> fluxes ( $\text{mg m}^{-2} \text{d}^{-1}$ ) with the surface water CH<sub>4</sub> concentration ( $\mu\text{M} = \mu\text{mol l}^{-1}$ ), wind speed ( $\text{m s}^{-1}$ ) and water temperature ( $^{\circ}\text{C}$ ) in MTaiga lakes ( $R$  = Pearson correlation coefficient). In plots **A** and **B**, one distant data point has been left out from the plot (co-ordinates presented in italics), but it has been included in the regression.

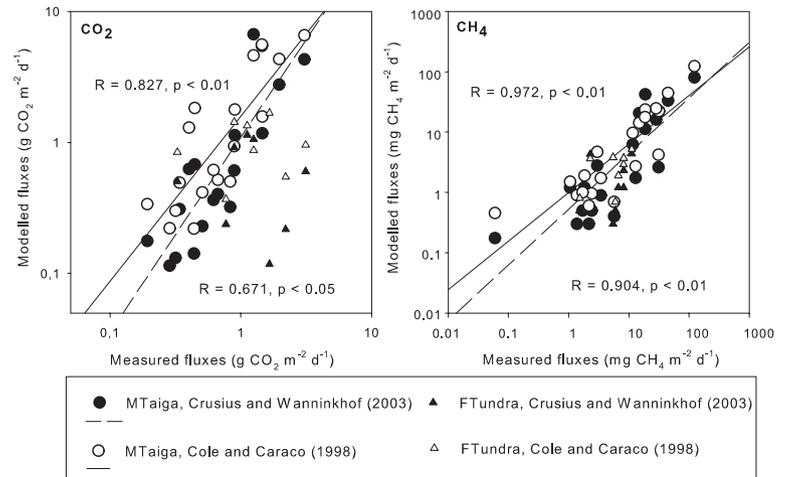


Fig. 2. The correlation between the modelled and measured daily mean CO<sub>2</sub> fluxes ( $\text{g m}^{-2} \text{d}^{-1}$ ) and CH<sub>4</sub> fluxes ( $\text{mg m}^{-2} \text{d}^{-1}$ ) at the studied lake sites ( $R$  = Pearson correlation coefficient). The regression lines presented in the figures are calculated for the fluxes at the MTaiga sites. Note the logarithmic scale in the figures.

Although 71–82% of the measured chamber CH<sub>4</sub> fluxes exceeded the corresponding modelled ones (depending on the used wind–gas flux relationship), the difference between the two methods was not statistically significant ( $t$ -test). The absolute value of difference between the modelled and measured CH<sub>4</sub> fluxes increased with the CH<sub>4</sub> flux, when the bilinear relationship (eqs 2 and 3; Crusius and Wanninkhof, 2003) was used in the model ( $R = 0.868$ ,  $p < 0.01$ ,  $N = 28$ ). For CO<sub>2</sub> this was the case with both of the two wind–gas flux relationships ( $R = 0.469$ – $0.600$ ,  $p < 0.05$ ,  $N = 27$ ): the higher the flux, the higher the difference between the results obtained by the floating chambers and the TBL-model.

Many studies have reported that floating chambers give higher values for gas exchange than, for example, the TBL-model or eddy covariance method (Duchemin et al., 1999; Eugster et al., 2003; Matthews et al., 2003). Chambers may overestimate the flux rates due to the disturbance of the surface boundary layer caused by chamber deployment (Matthews et al., 2003) or the unrepresentative sampling frequency (Ford et al., 2002; Eugster et al., 2003). On the other hand, the flux calculations based on wind speed may lead to underestimation in low wind environments, because with decreasing wind speeds the turbulence caused by precipitation (Ho et al., 1997; Cole and Caraco, 1998) and convective mixing due to heating and cooling at the

water surface (e.g. Eugster et al., 2003) become more important for the gas exchange than the effect of wind.

Despite the differences between the measured and modelled gas fluxes were sometimes quite high, the model was able to reveal a similar seasonal pattern as observed in the measured fluxes and to capture the differences between the lake sites. The advantages of TBL-model in comparison to the relatively time-consuming chamber method are obvious in remote areas rich in small lakes and ponds such as WSL. The modelling efforts enable spatially and temporally representative sampling regime, while less frequent chamber measurements can be used to verify the suitability of the model to the local conditions. Since a large part of the total CH<sub>4</sub> emissions from lakes can be released in gas bubbles (Chanton and Whiting, 1995), continuous measurements of ebullition should be used along with the TBL-method.

### 3.4. CH<sub>4</sub> ebullition

Gas collectors did not catch any gas bubbles in MTpond, but ebullition was found at both sites in MTLake and FTlake, although the peak values of diffusive (chamber) emissions in these lakes were lower than those in MTpond (Table 1). CH<sub>4</sub> concentrations in the gas bubbles ranged from 6 to 26% in MTLake and from 2 to 12% in FTlake, being low compared with the bubble gas concentrations commonly reported in other studies (e.g. Martens et al., 1992; Casper et al., 2000; Huttunen et al., 2001). Because the CO<sub>2</sub> concentrations in bubble gas were very low, typically 1–3%, the contribution of ebullition to the total CO<sub>2</sub> emissions was negligible (see also Huttunen et al., 2001).

The low rates of CH<sub>4</sub> ebullition associated with the low bubble CH<sub>4</sub> concentrations in our study are in accordance with the positive empirical relationship that has been established between the rate and the CH<sub>4</sub> content of ebullition (Martens et al., 1992). However, low CH<sub>4</sub> content in the bubbles could also result from dilution or CH<sub>4</sub> oxidation during the storage of the bubbles in the funnels (Chanton and Whiting, 1995) considering our long sampling intervals of 5–30 d in MTaiga and 48 d in FTundra. The mean CH<sub>4</sub> content in fresh bubbles released from MTLake by stirring the sediment,  $38 \pm 17\%$  (mean  $\pm$  SD;  $N = 10$ ), was higher than that in trapped bubbles. The range for the possible CH<sub>4</sub> ebullition rate was calculated using the concentrations in trapped bubbles (the lowest limit) and in fresh bubbles stirred from the sediment in MTLake (the highest limit). The time-weighted mean CH<sub>4</sub> ebullition from MTLake was 4.7–11 and 1.0–3.2 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> at Sites 1 and 2, respectively. The CH<sub>4</sub> ebullition from FTlake was 0.99–5.0 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> at Site 1 and 0.65–3.8 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> at Site 2.

### 3.5. Comparison with other studies

For better comparison of the CO<sub>2</sub> and CH<sub>4</sub> emissions with those measured from other northern lakes, the time-weighted seasonal average fluxes were calculated by multiplying the average chamber fluxes of two subsequent measuring days with the number

of days between them. The measuring interval varied from one to 22 d in MTaiga and from one to 47 d in FTundra. The CH<sub>4</sub> fluxes measured with floating chambers (diffusive flux) and bubble gas collectors (ebullition) were simply summarized to get the total CH<sub>4</sub> flux. Because of the good agreement between the measured and modelled CH<sub>4</sub> fluxes (Fig. 2), we could assume that the chambers did not receive significant amounts of CH<sub>4</sub> by ebullition, which is not accounted for in the TBL-model. The time-weighted average CO<sub>2</sub> emissions were 0.7, 1.9 and 1.6 g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> from MTLake, MTpond and FTlake, respectively, and the corresponding CH<sub>4</sub> emissions (by both diffusion and ebullition) were 18–23, 67 and 7.5–11 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>. The contribution of the ebullitive flux to the total seasonal CH<sub>4</sub> emission was 19–37% in MTLake and 11–40% in FTlake, while no CH<sub>4</sub> ebullition was observed in MTpond.

Mean CO<sub>2</sub> fluxes in our study were similar to those measured in 25 arctic lakes by Kling et al. (1992) (mean emission 0.9 g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup>). Although our study lakes released more CO<sub>2</sub> and CH<sub>4</sub> during summer season than, for example, two northern boreal mesotrophic ponds (3.5–7.6 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> and 0.02–0.5 g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup>; Huttunen et al., 2002), their emissions were low in comparison with 110–180 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup> and 3.7–11.0 g CO<sub>2</sub> m<sup>-2</sup> d<sup>-1</sup> reported for high boreal and low subarctic wetland ponds by Hamilton et al. (1994). The CH<sub>4</sub> emissions from our shallow WSL lakes were generally lower than the CH<sub>4</sub> fluxes of West Siberian peatlands as reported by Friberg et al. (2003) (136 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>) and Panikov et al. (1995) (28.9–683.7 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>), but similar as the CH<sub>4</sub> emissions measured in the middle and northern taiga peatlands by Naumov (2004) (0–33.7 mg CH<sub>4</sub> m<sup>-2</sup> d<sup>-1</sup>).

Carbon evasion to the atmosphere during the assumed active season (120 d in MTaiga and 100 d in FTundra), based on the measurements with chambers and bubble gas collectors, was 23–24 g C m<sup>-2</sup> from MTLake, 66 g C m<sup>-2</sup> from MTpond and 43–44 g C m<sup>-2</sup> from FTlake. CH<sub>4</sub> had only minor importance in the carbon budget of the study lakes. The contribution of CO<sub>2</sub> to the total carbon release was 93–94, 92 and 98–99% in MTLake, MTpond and FTlake, respectively. The carbon evasion during the active season from the West Siberian lakes we sampled was in the same range with the carbon release from Finnish lakes in the data of presented by Kortelainen et al. (2006) (21 g C m<sup>-2</sup> released as CO<sub>2</sub> during 4.5 months in spring and summer) and Huttunen et al. (2003) (1–61 g C m<sup>-2</sup> during the active season of 150 d). In the study of Kortelainen et al. (2006) on Finnish lakes, the carbon evasion decreased with the lake area being highest (twice the mean evasion of all lakes) in the lakes smaller than 0.1 km<sup>2</sup>, which supports the high carbon release found in the small lakes of our study.

The seasonal emission estimates reported in this article did not include potentially high episodic emissions at spring ice melt, which may contribute up to some tens of percents to the annual gas release (e.g. Michmerhuizen et al., 1996; Striegl and Michmerhuizen, 1998; Kortelainen et al., 2006). Thus, the

seasonal CO<sub>2</sub> and CH<sub>4</sub> emissions we measured are probably somewhat lower than the actual annual emissions including the cold part of the year.

The carbon loss by CO<sub>2</sub> and CH<sub>4</sub> evasion from the WSL lakes to the atmosphere, 24–66 g C m<sup>-2</sup>, is of a similar magnitude as the (opposite) net carbon uptake (43–64 g C m<sup>-2</sup>) measured in boreal Siberian peatlands during snow-free period (Arneeth et al., 2002), during summer (Friborg et al., 2003) or during the assumed active season of 120 d (Naumov, 2004). The high carbon loss from the studied lakes as CO<sub>2</sub> and CH<sub>4</sub> stresses the importance of small lakes in the processing and transport of terrestrially fixed carbon to the atmosphere and their impact on the areal carbon budgets in wetland regions rich in lakes and ponds, such as the WSL. An estimate of the total surface area covered by small water-bodies in WSL would be essential in further evaluation of the role of lakes in the carbon balance of West Siberian landscape.

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