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Release of CO_2 and CH_4 from lakes and drainage ditches in temperate wetlands

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Abstract Shallow fresh water bodies in peat areas are important contributors to greenhouse gas fluxes to the atmosphere. In this study we determined the magnitude of CH_4 and CO_2 fluxes from 12 water bodies in Dutch wetlands during the summer season and studied the factors that might regulate emissions of CH_4 and CO_2 from these lakes and ditches. The lakes and ditches acted as CO_2 and CH_4 sources of emissions to the atmosphere; the fluxes from the ditches were significantly larger than the fluxes from the lakes. The mean greenhouse gas flux from ditches and lakes amounted to 129.1 ± 8.2 (mean ± SE) and 61.5 ± 7.1 mg m^{-2} h^{-1} for CO_2 and 33.7 ± 9.3 and 3.9 ± 1.6 mg m^{-2} h^{-1} for CH_4, respectively. In most water bodies CH_4 was the dominant greenhouse gas in terms of warming potential. Trophic status of the water and the sediment was an important factor regulating emissions. By using multiple linear regression 87% of the variation in CH_4 could be explained by PO_4^{3-} concentration in the sediment and Fe^{2+} concentration in the water, and 89% of the CO_2 flux could be explained by depth, EC and pH of the water. Decreasing the nutrient loads and input of organic substrates to ditches and lakes by for example reducing application of fertilizers and manure within the catchments and decreasing upward seepage of nutrient rich water from the surrounding area will likely reduce summer emissions of CO_2 and CH_4 from these water bodies.

Keywords Biogeochemistry · Drainage ditches · Greenhouse gas emission · Nutrients · Peatlands · Temperate lakes

Introduction

Freshwater bodies such as ditches, streams, wetlands and lakes contribute appreciably to the processing of carbon and its transport to the atmosphere (e.g. Bastviken et al. 2004; Walter et al. 2006; Wang et al. 2006). It has been estimated that lakes annually emit 8–48 Tg methane (CH_4), which is 6–16% of the global natural CH_4 emissions (Bastviken et al. 2004; St. Louis et al. 2000), and 513 Tg C carbon dioxide (CO_2) (Cole et al. 1994). Saarnio et al. (2009) have

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estimated that large lakes alone account for 24% of all wetland CH_4 emissions in Europe. It has been shown that small water bodies also significantly contribute to the landscape-scale CH_4 budgets in wetland regions (e.g. Schrier-Uijl et al. 2009a, b; Juutinen et al. 2009; Repo et al. 2007; Walter et al. 2007; Roulet and Moore 1995). Yet though it is likely that both lakes with organic-rich sediment and also eutrophic ditches contribute especially significantly to regional greenhouse gas balances, they are poorly studied and very little is known about their underlying biogeochemical processes (Saarnio et al. 2009).

CO_2 is produced by respiration in sediments and throughout the water column and can also be a product of biological processes in the sediment (Fig. 1). As CO_2 is highly soluble, high concentrations can accumulate near the sediment/water interface, which results in oversaturation and release to the atmosphere. It has been suggested that the transport of dissolved organic carbon (DOC) from terrestrial environments is an important source of carbon in aquatic environments. If this is the case, lakes in organic-rich peatlands have larger CO_2 fluxes than lakes in mineral catchments (Rantakari and Kortelainen 2005; Huttunen et al. 2002).

CH_4 emission is the balance of two counteracting processes: methanogenesis in anoxic conditions and the oxidation of the generated CH_4 (Minkkinen and Laine 2006; Bastviken et al. 2002), (Fig. 1). CH_4 is a major product of carbon metabolism in lakes; its production depends on the availability of alternative electron acceptors such as O_2, NO_3^-, Fe^{3+} and $SO4^{2-}$ (van Bodegom and Scholten 2001). After these electron acceptors have been used up, CH_4 production becomes the dominating degradation process of organic matter and is the terminal microbial process in the anaerobic degradation of organic matter. The CH_4 travels from the sediment through the water column to the atmosphere and on the way it can be oxidised into CO_2 (Whiting and Chanton 2001). Most of the CH_4 that remains unoxidised will be emitted by diffusive flux to the atmosphere.

The underlying microbial processes affecting CO_2 and CH_4 production and emission are regulated by variables such as sediment and water temperature, oxygen availability, organic matter availability and composition, sediment and water chemistry, the presence of electron acceptors (redox conditions), pH, electrical conductivity (EC) and factors such as water depth and lake size (e.g. Stadmark and Leonardson 2005; Juutinen et al. 2009; Repo et al. 2007; Frei et al. 2006; Loeb et al. 2007; Casper et al. 2003).

Most of the freshwater lakes in the Netherlands are in peat areas, are very shallow (<2 m), and were created by large-scale dredging and removal of peat during the early seventeenth century (Gulati and van Donk 2002). They vary in area, depth, hydrology and physico-chemical characteristics, but most of them are eutrophic, due to the application of fertilisers and manure within their catchments, the oxidation of peat and the upward seepage of nutrient-rich water from the surrounding area. Drainage since the Middle Ages has resulted in the typical landscape of narrow fields separated by drainage ditches (Fig. 2).

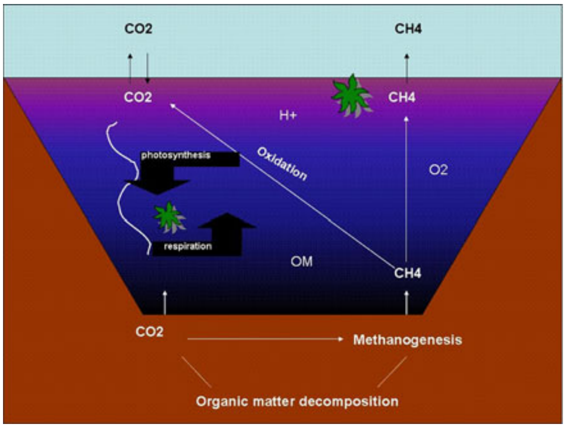


Fig. 1 Simplified illustration of CO_2 and CH_4 dynamics in water bodies, with *OM* organic matter

Fig. 2 The typical Dutch peat area landscape of lakes and narrow fields separated by drainage ditches (TOP10vector Product information 2007)

Shallow fresh water bodies are not very well understood in terms of their greenhouse gas emissions and have not been incorporated in previous regional or global greenhouse gas budgets. The emission from these water bodies is probably high and poses an international rather than a domestic problem because in many lowland regions of Europe agriculture continues to contribute appreciably to the nutrient loading of lakes and ditches (Gulati and van Donk 2002; Lamers et al. 1998).

About 16% of the total area (41,864 km^2) of the Netherlands is covered by water, mostly classified as wetland (Gulati and van Donk 2002) and with over 300,000 km of drainage ditches. Much of this wetland is in peat areas. It is very important to quantify how these wetlands contribute to the greenhouse gas balance and which factors regulate the emission. In this study we focus on the high-emitting temperate lakes and drainage ditches in peat areas in the Netherlands and on many variables that can alter the emission of CH_4 and CO_2. The two aims of this study were (1) to quantify CH_4 and CO_2 fluxes from shallow lakes and drainage ditches in the Netherlands during a 3-week period in the summer season and (2) to identify the factors that regulate the emissions of CH_4 and CO_2 from lakes and ditches.

Materials and methods

Study sites

Measurements were performed in a 3-week period between June 16th and July 6th in the summer of 2009 in 5 shallow fresh water lakes and 14 drainage ditches at 7 locations in peat areas in the Netherlands (Fig. 3).

The 5 lakes are located in peat areas in the Netherlands and differ in trophic status (de Haan et al. 1993) and depth (Table 1). L4 and L5 are located in the east of the Netherlands where the subsoil consists of mesotrophic to oligotrophic sedge-peat overlying sand. The other lakes are located in the southwest of the Netherlands where the subsoil consists of eutrophic to mesotrophic reed-sedge peat and alder carr peat.

Drainage ditches at 7 locations in different peat areas in the Netherlands were sampled. They differed in trophic status and water depth (Table 2). At each location 2 connected ditches were sampled and because there were no significant differences between them related to water quality they were treated as 1 location in the analyses. All the drainage ditches sampled contained some aquatic vegetation.

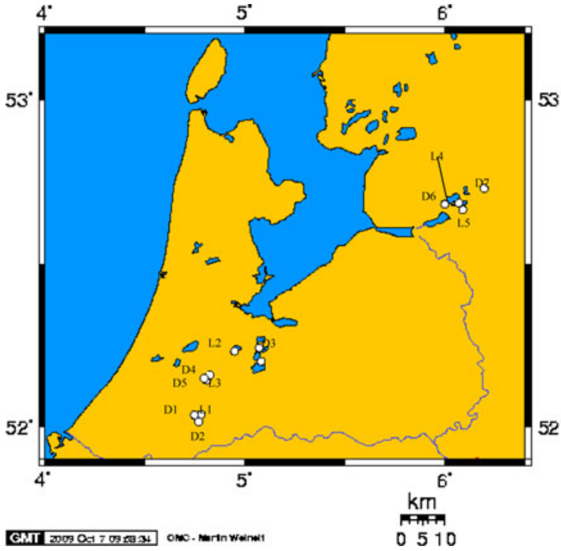


Fig. 3 Geographical distribution of the 5 sampled lakes (L1–L5) and 14 sampled drainage ditches (at 7 locations) (D1–D7) in peat areas in the Netherlands

Measurements

Flux measurements and calculation of fluxes

Detailed measurements of CH_4 emission and CO_2 emission were performed with floating chambers from a dinghy at different locations in the lakes and drainage ditches. We measured the emissions from each lake on two different days. On each of these days we measured at three different locations per lake, and repeated the measurements five times at each location. This yielded 30 measurements per lake. Each ditch was sampled on 1 day in the 3-week period, with 8 replicates per ditch. This yielded 16 measurements per location in the two connected ditches. All measurements were performed between 10.30 and 14.30 h. Data quality was assessed and outliers resulting from disturbances were removed from the dataset. Emissions of CH_4, CO_2 and N_2O were determined using a closed dark chamber method and a Photo Acoustic Field Gas Monitor (INNOVA 1412 sn, 710-113, ENMO services, Belgium) connected to a PVC chamber by Teflon tubing (e.g. van

Table 1 Characteristics of the lakes sampled

Lake	Abbreviation	Trophic status	Lake size (ha)	Average depth (m)
Reeuwijkse plas	L1	Eutrophic	927	2.1
Vinkeveense plas	L2	Eutrophic	1,079	2.4
Nieuwkoopse plas	L3	Eutrohpic	676	2.5
Belterwiede	L4	Mesotrophic to eutrophic	613	1.8
Schutsloterwiede	L5	Mesotrophic to eutrophic	141	1.2

Table 2 Characteristics of the drainage ditches

Drainage ditch	Abbreviation	Trophic status	Ditch width (m)	Mean water depth (m)
Oukoop	D1	Eutrophic	8	0.25
Stein	D2	Mesotrophic	6	0.28
Horstermeer	D3	Eutrophic	3	0.90
Drie berken	D4	Mesotrophic	3	0.87
Koole	D5	Mesotrophic	3	0.80
Sint Jans	D6	Eutrophic	2	0.28
Doosje	D7	Mesotrohpic	6	0.43

Huissteden et al. 2005; Hendriks et al. 2007). Fluxes of N_2O appeared to be too low to detect with the gas analyzer, therefore the N_2O flux measurements were not included in the analyses. Samples were taken from the headspace of this closed cylindrical dark chamber (30 cm diameter, 25 cm height). Gas samples were taken every minute during a 5-min period and every single measurements was checked on linearity of the build up of the gas concentration in the chamber. This check eliminated about 30% of the measurements. The slope dC/dt of the gas concentration curve at time $t = 0$ was estimated using linear regression (e.g. van Huissteden et al. 2005; Schrier-Uijl et al. 2009b). A small fan was installed in the chamber to homogenise the inside air and a water lock was used to control pressure in the chamber. We used a floater to place the chamber onto the water surface, carefully avoiding the effect of pressure differences and the disturbance of the water surface (for details, see Schrier-Uijl et al. 2009a). Since the gas monitor software does not compensate fully for cross-interference of CO_2 and water vapour at high concentrations, air was led through glass tubes filled with silica gel and soda lime before it entered the gas analyser, to remove water vapour. To cross-validate the chamber-based measurements, we also performed eddy covariance measurements on L1 at the same time and location and compared these with the chamber measurements within the footprint of the system. The eddy covariance system was located along a boardwalk in L1 and the footprint of the mast was on the lake. Within this footprint chamber measurements were performed on the lake during a period of 4 h. The two independent methods had previously been compared at different temporal scales in a heterogeneous landscape of fields and ditches (Schrier-Uijl et al. 2009b; Kroon et al. 2007).

Variables measured

At each lake and drainage ditch we measured water temperature and pH at two depths (10 and 30 cm and at 25 cm depth in D1 and D2), dissolved oxygen at 10 cm intervals from the water surface to the sediment surface, and the EC at 10 cm depth. Oxygen, pH, temperature and EC were measured with an HQ multiprobe with a luminescent dissolved oxygen sensor (Hach Company, Loveland, Colorado, USA). The variables investigated in the ditches were the dissolved CH_4 concentrations at the water surface, the middle of the water column and in the water immediately above the sediment. Samples for dissolved methane analysis were taken using an airtight 20 ml glass syringe at three depths in the water column: at the sediment surface, at the water surface, and at a depth half-way in between. The water samples were transferred into airtight glass Exetainers® (Labco, high Wycombe, UK) containing 120 µl $ZnCl_2$ to halt biological processes; to prevent air bubbles being trapped in these vials they were filled to overflowing before being capped. The samples were stored in water at 20°C until analysis. Dissolved methane was measured by membrane inlet mass spectrometry (MIMS) (Lloyd and Scott 1983) using an OmniStar™ Gas Analysis System (Pfeiffer Vacuum, Asslar, Germany), equipped with a quadrupole QMS 200 mass spectrometer with a Channeltron detector (Burle Industries). The MS was operated by Quadstar 32-bit software for data acquisition. The sample was pumped through a water bath at 20°C before passing through silicon membrane tubing in which gases were released to the MS. An inlet as described by Kana (1994) was used for the analysis, but without using a cryotrap, as this would have frozen out the methane. Instead, to prevent confounding effects of water vapour, the inlet at the MS side was heated to 180°C. Methane was measured at mass to charge ration (*m/z*) of 15, as a pre-calibration experiment had shown that this gave the most reliable results. Concentrations of methane were calculated by comparing the ion current at *m/z* 15 of the sample to the ion current at *m/z* 15 of air-saturated water at 20°C.

The water in each lake was sampled at three locations with 3 replicates (mixed sample). The water in each ditch was sampled at two locations with 3 replicates (mixed sample). Undisturbed sediment samples were taken from the sediment top layer (upper 10 cm) by means of a plastic cup perforated with holes 2 cm apart at the end of a length-adjustable pipe.

Two of the three water samples were filtered immediately with a Whatman 0.45 µm cellulose membrane filter (Whatman International Ltd, Maidstone, England); the third sample was not filtered. All samples were transported in coolers and stored frozen (−20°C) until analyses. The unfiltered water samples were analysed for organic matter (OM) content, %C, %N, Chlorophyll-*a* content, total N and total P; the filtered samples were analysed for $NO_3^- + NO_2^-$, NH_4^+, SO_4^{2-}, Fe^{2+} and PO_4^{3-} using a SAN^{plus} autoanalyzer (Skalar Analytical, Breda, the Netherlands). Dissolved organic carbon (DOC) and dissolved inorganic carbon (DIC) were measured in filtered samples using a carbon analyser. Total N and total P were measured using a SAN^{plus} auto analyser with laser destructor. All these samples were measured in duplicate. Chlorophyll-*a* content in unfiltered samples from the microcosms was measured using a phyto-PAM fluorometer (Heinz Waltz GmbH, Effeltrich, Germany). For the sediment samples a $CaCl_2$ extraction was used to obtain the available PO_4–P, NH_4–N and NO_3–N, and an ammonium oxalate extraction was used to obtain the active form of Fe.

Data analysis

Correlations between the measured variables and fluxes of CO_2 and CH_4 were first tested by using Pearson correlation analysis. Data were tested for normality. We used stepwise, multiple linear regression analyses to quantify the relationships between environmental variables and fluxes of CO_2 and CH_4 (SPSS 15.0). The variables that significantly enhanced the emissions of CO_2 and CH_4 were selected and were used to build regression models. Differences in the fluxes and variables between and within lakes and ditches were tested using one-way ANOVA (SPSS 15.0).

Results

Climatic variables

During the sampling period the mean day air temperatures ranged from 15 to 25°C, the average temperature at the surface of the water bodies studied ranged from 19.2 to 25.4°C and the wind speed at 3 m above water level ranged from 2.1 to 4.5 m s^{-1} (Table 3).

Characteristics of lakes and drainage ditches

The lakes and drainage ditches were humic, shallow and nutrient-rich. The sediment in D6 and D7 had the lowest organic matter content because these two ditches are located in an area with shallow peat on sand. The EC in all the lakes and ditches sampled ranged from 269–866 µS cm^{-1}; the pH ranged from 6.8–9.0, with the highest values in the lakes (Table 4).

Emissions to the atmosphere

Lakes and drainage ditches studied acted as sources of CO_2 and CH_4 emissions to the atmosphere

Table 3 Average (mean ± SD) water temperatures at 10 cm depth (T_{10} in °C), at 30 cm depth (T_{30} in °C) and average wind speed (U in m s^{-1}) during

	Mean T_{10}	Mean T_{30}	Mean U
D1	20.1 (± 0.1)	19.5 (± 0.9)	4.1
D2	25.4 (± 2.5)	25.2 (± 2.4)	3
D3	19.2 (± 0.6)	18.4 (± 0.4)	2.7
D4	25 (± 0.5)	25.8 (± 0.3)	3
D5	22.3 (± 0.3)	22 (± 0.1)	4.6
D6	20.2 (± 0.7)	19.1 (± 1.4)	2.1
D7	22.8 (± 0.2)	21.8 (± 1.2)	3.9
L1	23.6 (± 3.0)	23.7 (± 0.5)	3
L2	21.4 (± 1.1)	20.3 (± 0.5)	3.6
L3	23.3 (± 1.1)	23.2 (± 1.1)	3
L4	23.2 (± 1.4)	22.7 (± 1.2	3.9
L5	25 (± 1.2)	24.9 (± 0.6)	3.9

For the location codes see Tables 1 and 2

(Figs. 4, 5), except for L1 where a small uptake of CO_2 was measured. The mean release of both gases to the atmosphere was significantly higher from the ditches than from the lakes ($P < 0.001$).

The contribution of CO_2 emission compared to CH_4 emission in terms of warming potential is given in Fig. 6, where CH_4 fluxes have been transformed to CO_2 equivalents (CH_4 is 23 times as potent as CO_2).

Lakes

The emission of CO_2 from the lakes ($n = 93$) ranged from −6.0 to 123.9 mg m^2 h^{-1} and CH_4 emission ($n = 96$) ranged from 1.4–18.1 mg m^2 h^{-1}. The CO_2 fluxes from L1 were significantly lower than those from the other lakes ($P < 0.01$); the CO_2 fluxes from L2 were significantly higher than those from L3, L1 and L5 ($P < 0.05$). The highest CH_4 emission was measured from L5 and the lowest from L2, but the differences were not significant. The lakes acted as sources of emissions of both gases, except for L1 that acted as a very small sink for CO_2. In terms of warming potential, in 3 lakes the dominant emitted greenhouse gas was CH_4 and in 2 lakes it was CO_2 (Fig. 6).

Ditches

The emission of CO_2 from the drainage ditches ($n = 80$) ranged from 69.6 mg m^2 h^{-1} to 199.0 mg m^2 h^{-1} and CH_4 emission ($n = 79$) ranged from 1.2 to 39.3 mg m^2 h^{-1}. The CO_2 emission from D3 was significantly higher than the fluxes from D7, D1 and D5. The highest CH_4 emission was measured from D4 and the lowest from D7, but the CH_4 fluxes did not differ significantly because there was great variability among the ditches. In all ditches except D6 and D7, the dominant greenhouse gas in terms of

Table 4 General characteristics of the lakes and ditches sampled, with standard deviations for lake/ditch depth, EC, pH and percentage organic matter in the sediment

Site	Lake size (ha) or Ditch width (m)	Sediment type	Mean depth (cm)	Mean EC (µS cm^{-1})	Mean pH	Organic matter sediment (% dry weight)
L1	927	Peat	209.2 (± 42)	508.7 (± 53)	9.0 (± 0.3)	46.63 (± 9.5)
L2	1079	Peat	240.0 (± 42)	866.0 (± 13)	8.4 (± 0.1)	36.89 (± 12.9)
L3	676	Peat	253.3 (± 71)	392.5 (± 2)	8.2 (± 0.1)	63.08 (± 14.1)
L4	613	Peat on sand	178.0 (± 27)	398.0 (± 9)	8.4 (± 0.1)	58.72 (± 3.4)
L5	141	Peat on sand	120.0 (± 21)	393.0 (± 15)	8.4 (± 0.2)	59.81 (± 5.8)
D1	8	Peat	25.0 (± 3)	523.0 (± 38)	9.0 (± 0.0)	55.94 (± 3.0)
D2	6	Peat	27.5 (± 9)	269.3 (± 46)	6.8 (± 0.3)	57.37 (± 7.9)
D3	3	Peat	90.0 (± 14)	662.0 (± 48)	7.0 (± 0.4)	37.77 (± 3.7)
D4	3	Peat	86.7 (± 8)	386.7 (± 2)	7.2 (± 0.3)	74.22 (± 0.8)
D5	3	Peat	80.0 (± 17)	387.0 (± 2)	7.7 (± 0.1)	73.89 (± 0.8)
D6	2	Peat on sand	27.5 (± 3)	350.3 (± 26)	7.2 (± 0.3)	9.53 (± 3.1)
D7	6	Peat on sand	42.5 (± 3)	395.5 (± 6)	8.1 (± 0.3)	2.65 (± 2.4)

Lake area was determined from Top10vector maps: TDN (2006)

Fig. 4 Mean CO_2 fluxes (mg m^{-2} h^{-1}) and their standard deviations in the sampling period for the ditches (D1–D7) and lakes (L1–L5) at different locations in peat areas in the Netherlands. Positive flux values indicate release from the water to the atmosphere

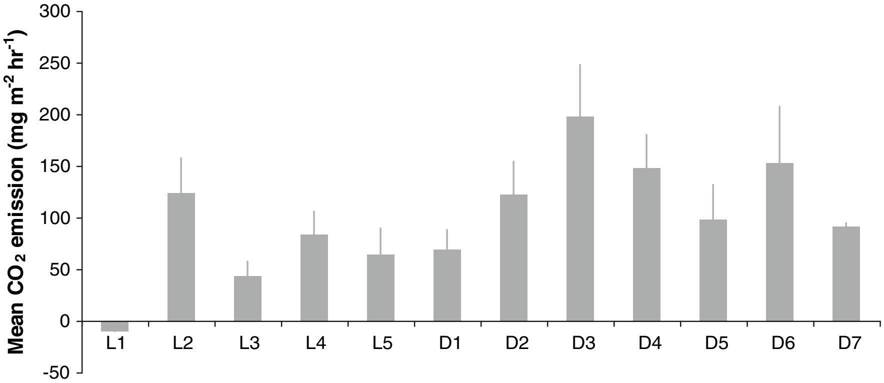


Fig. 5 Mean CH_4 fluxes (mg m^{-2} h^{-1}) with their standard deviations for the ditches (D1–D7) and lakes (L1–L5) at different locations in peat areas in the Netherlands. Ditches were sampled on 1 day ($n = 16$ per location) in the period 16 June–6 July 2009; Lakes were sampled during 2 days in the same period ($n = 24$ per location). Positive flux values indicate release from the water to the atmosphere

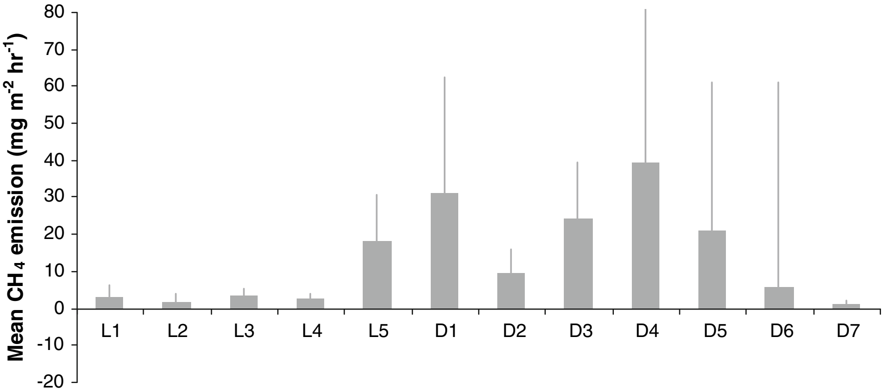
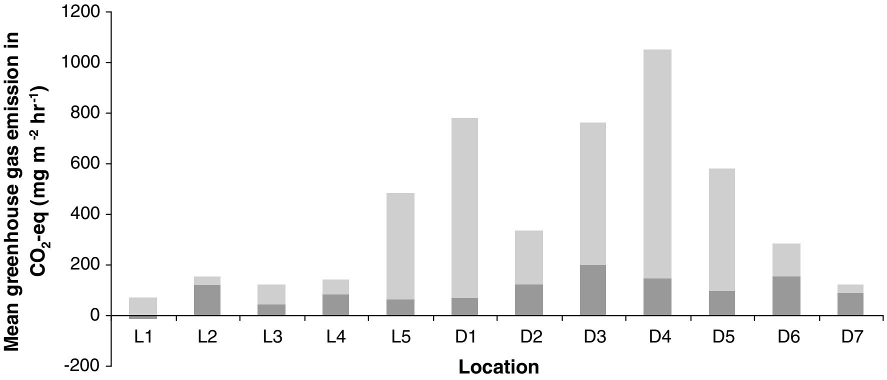


Fig. 6 Contribution to greenhouse warming of CO_2 emission (*dark grey*) compared to CH_4 emission (*light grey*) in lakes and ditches, given in CO_2 equivalents



warming potential was CH_4. All ditches acted as sources of emissions of both gases.

Cross-validation

Large-scale CH_4 flux measurements by eddy covariance were performed on one of the lakes (L1) to cross-validate flux values from this homogeneous landscape on a diurnal base. A cross-validation of chamber based CH_4 and CO_2 values and eddy covariance based values is also performed earlier for a more heterogeneous peat area (Schrier-Uijl et al. 2009b). Details for the used eddy covariance instruments have been reported in Veenendaal et al. 2007 and Kroon et al. 2007. In this study, CH_4 fluxes within the footprint of the eddy covariance system were 5.8 ± 3.26 (mean ± SD, $n = 24$) measured by chambers compared to 4.6 ± 1.3 measured by eddy covariance over a 4-h period. It would be of great interest in the future to also use eddy covariance to capture temporal variability of greenhouse gas fluxes (CH_4 and CO_2) from water bodies and to explain more of the measured variability.

Dissolved oxygen and dissolved CH_4

Typical vertical profiles of oxygen saturation during the measurements are shown in Fig. 7.

On average, the lakes had a higher O_2 saturation than ditches. In both types of waterbody, oxygen saturation decreased only slightly at the top of the water column, which suggests that there was hardly any respiration by aquatic organisms. Deeper in the water column the oxygen saturation fell rapidly to values close to 0% just above the sediment. Of the lakes, L2 had the highest O_2 saturation throughout the profile, and of the ditches D3 and D6 had the lowest O_2 saturation.

Dissolved CH_4 concentrations were measured at three depths: at the top and middle of the water column and just above the ditch sediments. In all the ditches the dissolved CH_4 concentrations increased with depth (Fig. 8).

Concentrations of dissolved CH_4 (μg l^{-1}) in the water of ditches at the top, middle and bottom of the water column. The y-axis shows the depth (cm).

D1 and D3 had a high dissolved CH4 concentration and also a high CH_4 emission (Fig. 5). None of the following variables correlated significantly with the dissolved CH_4 in the ditch water immediately above the sediment or with the difference between dissolved CH_4 concentration at the water surface and at the sediment surface: nutrient content (NO_3^-, NH_4^+, Fe, PO_4^{3-}); sediment oxygen demand (SOD); O_2 saturation of the water, organic matter content (% organic matter, %N, %C); amount of green algae and plants. We did not find any significant correlation between dissolved CH_4 concentration at the water surface and CH_4 release to the atmosphere. The oxygen saturation at the sediment surface correlated negatively with CH_4 emission to the atmosphere ($P = 0.065$).

The variables measured and their correlation with CH_4 and CO_2 emission

Climate, depth, EC and pH

Climatic conditions in the 3-week sampling period were stable. No significant correlation was found between the CO_2 and CH_4 fluxes and the temperature or the wind velocity. Neither the depth of water in the ditches (range 0.28–0.90 m) or the depth of water in the lakes (range 1.20–2.53 m) correlated significantly with CO_2 or CH_4 release to the atmosphere, although the deepest lakes tended to have the lowest CH_4 and CO_2 emissions. A positive correlation was found between EC and CO_2 flux and a significant negative correlation was found between CO_2 emission and the pH of the water ($r = -0.81$; $P = 0.001$). Though the correlation between CH_4 emission and pH was also negative, it was not significant ($r = -0.23$; $P = 0.41$).

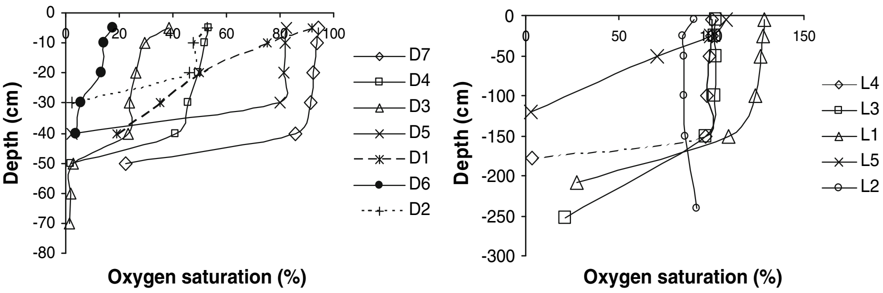


Fig. 7 Oxygen saturation (%) in the water columns of lakes (*right*) and drainage ditches (*left*) at different depths in the water column at the time of the measurements

Fig. 8 Concentrations of dissolved CH_4 (μg l^{-1}) in the water of ditches at the *top, middle and bottom* of the water column. The y-axis shows the depth of the ditch (cm)

Nutrients and organic matter in water and sediment

The percentage of N measured in the lake sediments was significantly positively correlated with the release of CO_2 ($P < 0.01$); in ditches, the %N and the %OM measured in the sediments were significantly positively correlated with the release of CH_4 ($P = 0.02$ and $P = 0.05$, respectively). The lowest organic matter contents of the sediments were found in D6, D7 and L2 (Table 4), which had the lowest CH_4 fluxes (Fig. 5). In this study neither the DIC nor the DOC correlated significantly with the CO_2 flux or the CH_4 flux.

The ammonium (NH_4^+) concentration in the water ranged from 0.1 to 478.6 μg NH_4–N l^{-1}: the highest concentrations were found in D3, D6 and L2. Ammonium concentration correlated positively with CO_2 emission ($r = 0.67$; $P < 0.05$). The NO_3^- concentration in the water was around 0 mg N l^{-1}—except for L2 and L4, where the mean concentrations were 0.43 and 0.12 mg N l^{-1}, respectively. In the sediment of the lakes and ditches the NH_4^+ concentrations ranged from 12.3 to 478.1 mg/kg dry weight with the highest concentrations in D4 (324.2 mg kg^{-1} dry weight) and D5 (478.1 mg kg^{-1} dry weight). The NO_3–N concentration ranged from 0.0 to 3.55 mg kg^{-1} dry weight, with the highest concentration in D4 and the lowest in L4. The only lake with high NO_3 concentrations in the water and sediment was L2: it was also the only lake where measurable N_2O emissions were observed (0.163 mg m^{-2} h^{-1}, $n = 23$). See Table 5 for the concentrations of NH_4^+ and table 6 for the concentrations of NO_3–N.

A weak negative correlation was found between the SO_4^{2-} concentrations in lake water (range 17.8–53.4 mg l^{-1}) and ditch water (range 3.4–47.1 mg l^{-1}) and CO_2 and CH_4 fluxes ($r = -0.43$, $P = 0.16$; $r = -0.1$, $P = 0.81$). The water of D3 had the lowest SO_4^{2-} concentrations of all the lakes and ditches sampled; this ditch had a high Fe concentration in its water and sediment (Table 6), which suggests the binding of SO_4^{2-} to iron. Overall, the SO_4^{2-} concentrations did not significantly differ between the ditches and lakes. The Pearson correlations between CO_2, CH_4 and Fe^{2+} in the water were weakly positive; the Fe^{2+} concentrations we measured ranged from 33.6 to 1,032 μg l^{-1}, with an average of 301.6 μg l^{-1}. Methane emission correlated significantly positively with the PO_4^{3-} concentration of the sediments of the lakes and ditches ($r = 0.77$, $P = 0.81$). The PO_4^{3-} concentration of the sediments correlated positively both with the Fe concentration and the SO_4^{2-} concentration of water. The total P concentration in the water correlated positively with CO_2 emission, indicating the high availability of organic substrates. The nutrient concentrations are given in Tables 5 and 6.

Multiple regression analyses

Multiple regression with stepwise elimination of variables showed that for summer CH_4 fluxes the

Table 5 Chemical composition of the water of lakes and ditches with means for sulphate (SO_4^{2-}), iron (Fe^{2+}), phosphate (PO_4^{3-}), ammonium (NH_4^+), nitrate (NO_3^-) and total P

Location	SO_4^{2-} (mg l^{-1})	Fe^{2+} (μg l^{-1})	PO_4^{3-} (μg P l^{-1})	NH_4^+ (μg N l^{-1})	NO_3^- (mg N l^{-1})	Ptot (μg P l^{-1})
D1	47.1	324.3	98	13.8	0.00	153.4
D2	16.6	864.5	33	0.1	0.00	66.3
D3	3.4	1032.0	291	478.6	0.00	515.7
D4	21.6	58.5	11	8.8	0.00	18.3
D5	22.1	26.7	24	9.2	0.00	15.0
D6	7.1	470.5	240	166.4	0.00	201.0
D7	19.2	146.7	45	3.8	0.00	15.8
L1	34.8	48	53	12.2	0.00	11.3
L2	53.4	59.7	11	76	0.42	14.0
L3	21.0	31.7	25	11.5	0.00	10.3
L4	18.2	113.5	10	15.8	0.12	12.6
L5	16.9	640.0	18	6.1	0.00	25.4

For location codes see Tables 1 and 2

Table 6 Means for Fe, PO_4–P, NH_4–N, NO_3–N in the top10 cm of the bottom sediments of the sampled lakes and drainage ditches

Location	Fe (mg kg^{-1})	PO_4–P (mg kg^{-1})	NH_4–N (mg kg^{-1})	NO_3–N (mg kg^{-1})
D1	2.7	8.6	154.9	0.4
D2	3.0	0.6	215.4	0.7
D3	7.0	0.3	187.8	0.9
D4	6	20.7	478.1	3.6
D5	3.1	8.8	323.6	0.5
D6	2.0	0.2	51.9	0.2
D7	0.8	0.1	12.3	0.1
L1	1.5	2.6	106.0	0.4
L2	1.7	3.3	8.0	1.2
L3	0.5	5.2	123.7	0.3
L4	5.7	0.5	86.7	0
L5	na	na	na	na

Table 7 Statistical details of the CH_4 and the CO_2 multiple regression models with PO_4 concentration in the sediment, Fe concentration in the water, depth, EC and pH as explanatory variables: R square of the model, significance of the model, Pearson correlations and significance of the separate variables

	R^2 model	P model	Pearson corr.			P parameter		
			$PO_{4sediment}$	Fe_{water}		$PO_{4sediment}$	Fe_{water}	
CH_4 model	0.87	0.000	0.77	0.19		0.000	0.003	
			Depth	EC	pH	Depth	EC	pH
CO_2 model	0.89	0.000	−0.62	0.17	−0.81	0.06	0.004	0.001

PO_4^{3-} concentrations in the sediment and the Fe^{2+} concentrations in the water explained 87% of the variation when Eq. 1 was used:

$$F_{CH_4} = -3.482 + 2.183[PO_4]_{sediment} + 22.116[FE]_{water} \quad (1)$$

where F_{CH_4} is the CH_4 flux (mg m^{-2} h^{-1}), $[PO_4]_{se\,dim\,ent}$is the PO_4–P concentration in the sediment (mg kg^{-1}) and $[FE]_{water}$ is the concentration of Fe^{2+} in the water (mg l^{-1}). Table 7 presents statistical details of the model. Fig. 9 shows the measured CH_4 fluxes versus the CH_4 fluxes in the sampled lakes and ditches, modelled by means of Eq. 1.By performing regression analyses with ditches only, the fit of the regression improved to $R^2 = 0.94$.

For CO_2, a regression model with mean depth of lake or ditch water and the EC and pH of the water as independent variables explained up to 89% of the variation in summer CO_2 emission at the water–atmosphere interface when Eq. 2 was used:

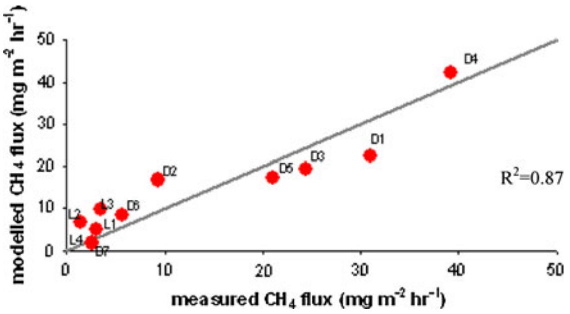


Fig. 9 Measured CH_4 fluxes versus modelled CH_4 fluxes (mg m^{-2} h^{-1}) based on a multiple regression model with PO_4–P concentration in the sediment and Fe^{2+} concentration in the water as explanatory variables. The *grey line* is the 1:1 line

$$F_{CO_2} = 477.359 - 0.213\text{depth} + 0.171\text{EC} - 54.4\text{pH} \quad (2)$$

where F_{CO_2} is the CO_2 flux, depth is the mean depth of the water in the sampled lake or ditch (cm), EC is the mean electrical conductivity and pH is the mean pH in the sampled lakes and ditches. Figure 10 shows

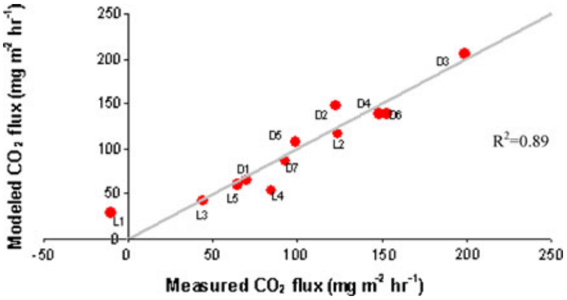


Fig. 10 Measured CO_2 flux versus modelled CO_2 flux (mg m^{-2} h^{-1}) based on a multiple regression model with mean depth of water, EC and pH as explanatory variables. The *grey line* is the 1:1 line

the measured CO_2 fluxes in lakes and ditches versus the modelled CO_2 fluxes by means of Eq. 2. Table 7 presents statistical details of the model.

Regression analyses on the data from the ditches only improved the predictive power of the regression to $R^2 = 0.91$.

Discussion

In this study we determined the magnitude of CH_4 and CO_2 fluxes from 12 water bodies in Dutch wetlands during a 3-week period in the summer season and studied the factors that might regulate emissions of CH_4 and CO_2 from these lakes and ditches. During this period the lakes and ditches acted as CO_2 and CH_4 sources of emissions to the atmosphere; the fluxes from the ditches were significantly larger. One lake (L1) was in equilibrium with the atmosphere in terms of CO_2 emission. Kosten et al. 2010 found that <10% of lakes worldwide are in equilibrium with the atmosphere in terms of pCO_2, and they found that most other lakes are CO_2 sources. Compared with other studies, the lake emissions founding our study were in the intermediate to high range (see Table 8 for CH_4 fluxes). For example, Rantakari and Kortelainen (2008) found CO_2 fluxes in the range 7.48–11.5 mg m^{-2} h^{-1} in 37 boreal Finnish lakes. The average CH_4 emission from our drainage ditches was higher than the lake fluxes found in other studies (Table 7). As the CH_4 emissions measured by the gas analyser within the footprint area of an eddy covariance system at location L2 agreed within the uncertainty limits with the EC system, we are confident that our measurement technique provided reliable flux estimates that are applicable to larger areas (Kroon et al. 2007; Schrier-Uijl et al. 2009b).

The temporal variability of emissions of CH_4 and CO_2 from water bodies is normally found to be related to temperature and wind velocity when measuring over longer time spans (e.g. Stadmark and Leonardson 2005; Frei et al. 2006; Hendriks et al. 2007; Repo et al. 2007; Schrier-Uijl et al. 2009a, b; Kroon et al., in press). However, a large part of the variability of fluxes cannot be explained by temperature or wind velocity only. Our results refer to data collected during summer, a period in which around 70% of the annual ditch emissions are generated. The study did not last long enough to include seasonal patterns of CH_4 and CO_2 production and emission. Diurnal stratification and mixing due to day–night temperature differences may bias flux estimates if the

Table 8 Comparison between the CH_4 emission rates in this study and the CH_4 emission rates reported in other studies

Reference	System	Location	Sampling period	Flux CH_4 (mg m^{-2} h^{-1})
Guerin and Abril 2007	Tropical lake	French Guiana	Late spring	4 ± 4.7
Juutinen et al. 2009	30 Eutrophic Boreal lakes	Finland	All seasons	Median of 0.137
Stadmark and Leonardson 2005	3 Ponds	South Sweden	Summer	10
Huttunen et al. 2002	Boreal lakes	Finland	Summer	1.0
Bastviken et al. 2004	11 Lakes	North America	Summer 2000	Range 0.15–3.2
Repo et al. 2007	3 Boreal lakes	Siberia	Summer	0.34
Present research	5 Temperate lakes	Netherlands	Early summer 2009	1.4–18.1, mean 5.0
Present research	7 Drainage ditches	Netherlands	Early summer 2009	1.2–39.3, mean 18.8

Mean CH_4 emission rates are in mg CH_4 m^{-2} h^{-1}. The period of sampling and the location are given

For units: mg CH_4 m^{-2} h^{-1} refer to mg CH_4 emitted per m^2 of water area per hour only measurements available are from the daytime (Repo et al. 2007). In Schrier-Uijl et al. (2009a, b) the diurnal variation of CH_4 fluxes over an area with fields and ditches was tested in October/November 2006. After correction for temperature dependency, the emission of CH_4 did not differ significantly between day and night. Nevertheless, there could be diurnal variation of fluxes from water bodies, because less oxygen will be produced at night, which will result in a lower redox potential and higher CH_4 production. In addition, less CO_2 will be taken up by aquatic plants at night, because then they are not photosynthesising. These effects should be considered when estimating annual fluxes from water bodies by using continuous measurements such as eddy covariance. In our study, only diffusive fluxes of CO_2 and CH_4 were measured; however, ebullition can also contribute to the emission of CH_4 from water bodies (Walter et al. 2006; Walter et al. 2007). While sampling ditches D5 and D4, we observed ebullition, so it is possible that we underestimated the release of CH_4 fluxes. In a summer study done by Repo et al. (2007) in Siberian water bodies, ebullition was observed in two of the three lakes sampled (depth <1.5 m) and accounted for 19–37% and 11–40% of the total CH_4 emissions from these two lakes.

The fact that the lakes and ditches acted as sources for CH_4 and CO_2 indicates that CO_2 production exceeded CO_2 uptake during photosynthesis by plants and that CH_4 production exceeded CH_4 oxidation. Our observation that the deeper, mostly less eutrophic lakes with low EC had the smallest fluxes agrees with findings reported for lakes in the boreal zone in Finland (depth range 3.8–26.5 m) (Juutinen et al. 2009). Deeper water bodies usually have less degradable organic matter and more oxidation of CH_4 than shallow lakes, because the transport pathway is longer (e.g. Borges et al. 2004). The EC, which is an indicator of trophic status in fresh water lakes, and the depth of the water body were two of the three significant predictors in the regression analyses for CO_2 fluxes.

The pH correlated negatively with emissions from both gases, yet at lower pH values ($pH < 7$) the correlations are usually positive (e.g. Inubushi et al. 2005). CO_2 enters the water as a result of the biological processes of organic carbon degradation and respiration by plants. In our ecosystems it is likely that through uptake of CO_2 by plants during the day in the growing season, HCO_3^- is transformed to CO_2, causing the HCO_3^- concentration to decline and diminishing the buffering effect. This reduced buffering effect can result in pH values above 9.0 and in a negative relation between CO_2 flux and pH. Incorporating pH in the regression equation for CO_2 significantly improved the equation's predictive power. In peat soils in temperate areas the optimum pH for methanogenesis is between 5.5 and 7.0, which explains the slightly negative correlation we found between CH_4 emission and pH (Le Mer and Roger 2001).

Water turbulence due to wind can increase mixing of oxygen in the water. This is illustrated by the higher oxygen concentration throughout the water column of L2, which had been subjected to high wind speeds on the day before sampling. The high O_2 saturation in L2 corresponded with higher CO_2 fluxes and very low CH_4 fluxes, illustrating the oxidation of CH_4 to CO_2. The opposite can be seen in D3, D6 and L5, where the oxygen concentrations at the water surface were low and the CH_4 fluxes were high, illustrating the low turnover of CH_4 carbon to CO_2. The fast decrease in dissolved CH_4 from the sediments to the water surface in D3 and D1 indicates that most of the dissolved CH_4 is oxidised during transport or passes through the water column and escapes to the atmosphere very quickly. As found in other studies, dissolved CH_4 poorly predicted the diffusive fluxes at the water–air interface (e.g. Huttunen et al. 2006; Juutinen et al. 2009). The factors responsible for this finding could be variation in duration of storage, release of CH_4 to the atmosphere and complex processes during transport of CH_4 through the water column (e.g. Kankaala et al. 2003).

The input of organic matter as a substrate in the lake or drainage ditch system increases the availability of substrates and this can increase the production of CO_2 and CH_4 (e.g. Casper 1992) and increases the possibility of minimising the competition for electron donors between methanogenesis and other anaerobic processes (Scholten et al. 2002; Scheid et al. 2003). As long as O_2 reaches the sediments, it will act as the primary oxidant of organic matter.

Permanently anaerobic conditions in the sediment may hamper nitrification of NH_4^+ to NO_3^-, but stimulate denitrification of NO_3^- to N_2 by microorganisms, leading to a high NH_4^+ to NO_3^- ratio, as was found in this study. Also, the greater availability of NH_4^+ compared to NO_3^- suggests the occurrence of dissimilatory reduction of NO_3^- to NH_4^+ (DNRA) under anaerobic conditions in these ditches. DNRA is likely to occur in the organic sediments that we sampled, as this process usually occurs at high carbon inputs (Burgin and Hamilton 2007). As our ditch systems did not contain much aquatic plant biomass, it is unlikely that the NO_3^- uptake by plants and algae was influential in the ditches. Other possible sources of NH_4^+ in the water could be cation exchange of adsorbed NH_4^+ by Fe^{2+} (but this only occurs at very high Fe^{2+} concentrations: Loeb et al. 2007), and leaching through groundwater from surrounding, managed agricultural areas. NH_4^+ inhibits methanotrophy and therefore may reduce CH_4 oxidation and increase its emission (Conrad and Rothfuss 1991), which may explain the positive correlation between the NH_4^+ and CH_4 fluxes in our study. The positive correlation of NH_4^+ with CO_2 emission is in line with the findings of other studies.

Our finding is that the two most significant predictors of CH_4 fluxes were the PO_4^{3-} concentration in the sediment of lakes and ditches and the Fe^{2+} concentration in the water of lakes and ditches. In anaerobic sediments, Fe^{3+} will be reduced to Fe^{2+}. At the sediment–water interface some of the Fe^{2+} will be oxidized to Fe^{3+} (how much depends on the oxygen concentration just above the sediment) and some of this will be released into the water. Thus a high concentration of Fe^{2+} in the water is related to anaerobic conditions. Both Fe concentration and SO_4^{2-} concentration correlate with PO_4^{3-} availability at the sediment–water interface. The PO_4^{3-} in sediments is bound to Fe^{3+} and when the Fe^{3+} is reduced to Fe^{2+}, PO_4^{3-} will be released to the water (e.g. Smolders et al. 2006; Smolders and Roelofs 1993).

Overall, a higher trophic status was positively correlated with summer emissions of CO_2 and CH_4, while the depth of the water and the pH were inversely correlated with CO_2 emission. It is therefore likely that decreasing the inputs of organic matter and nutrients (for example, by changing the management of the surrounding areas) will reduce emissions and that this effect will be strongest in drainage ditches.

Much of the uncertainty in flux estimates is due to temporal variation. So, also diurnal, seasonal, annual and inter annual variability must be studied in more detail to get insight in climatic responses, extreme drought/rainfall events and the influence of management in the surrounding catchments. In this respect, there is a need for long-term, continuous measurements of emissions (e.g. by eddy covariance).

Conclusion

The current study focused on emissions from temperate, shallow lakes ($n = 5$) and drainage ditches ($n = 14$) in agricultural peat areas in the Netherlands. It was found that in general, both these types of waterbodies are important sources of CO_2 and CH_4. The ditches had significantly higher CO_2 and CH_4 fluxes than the lakes. Trophic status was an important indicator of the magnitude of fluxes. 87% of the variation in the summer fluxes of CH_4 could be explained by PO_4^{3-} in the sediment and Fe^{2+} concentration in the water, and 89% of the CO_2 flux could be explained by water depth, EC and pH. Our results can be used to refine greenhouse gas emission inventories and to ascertain possible ways of reducing the release of CO_2 and CH_4 from water bodies to the atmosphere. Decreasing the nutrient loads and input of organic substrates to ditches and lakes will likely reduce summer emissions of CO_2 and CH_4 from these water bodies.

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