

Nitrous oxide and methane fluxes of a pristine slope mire in the German National Park Harz Mountains

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Abstract Pristine peatlands covered by Histosols (bogs and fens) with high water table and a restricted oxygen (O₂) availability are known to have low emissions of nitrous oxide (N₂O) but may be a significant source for atmospheric methane (CH₄) which are both important greenhouse gases. For the first time N₂O and CH₄ fluxes of a pristine slope mire in the German Harz Mountains have been monitored. Previously reported peatlands are characterised by anaerobic conditions due to high water table levels. Slope mires monitored here receive O₂ through slope water inflow. Gas fluxes have been monitored deploying closed chamber method on a central non-forested area and a forested area at the periphery of

the slope mire. By means of groundwater piezometers water table levels, ammonium and nitrate contents as well as hydro-chemical variables like oxygen content and redox potential of the mire pore water have been concurrently measured with trace gas fluxes at both monitoring sites of the slope mire. The slope mire took up small amounts of atmospheric methane at a rate of -0.02 ± 0.01 kg C ha⁻¹ year⁻¹ revealing no significant difference between the forested and non-forested site. Higher uptake rates were observed during low water table level. In contrast to pristine peatlands influx of oxygen containing pore water into slope mire does limit reduction processes and resultant CH₄ emission. N₂O fluxes of the forested and non-forested sites of the slope mire did not differ and amounted to 0.25 ± 0.44 kg N ha⁻¹ year⁻¹. Higher emissions were observed at low water table levels and during thawing periods. In spite of favourable conditions N₂O fluxes of the slope mire have been comparable to those of pristine peatlands.

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Introduction

Natural peatlands, covered by Histosols (FAO/ISRIC/ISSS 1998), are important sinks for carbon (C) and nitrogen (N) due to prevailing permanent anaerobic

conditions and resultant low rates of decomposition (Moore et al. 2004). They represent only about 3% ($3.88\text{--}4.38 \times 10^6 \text{ km}^2$) of the world's area but comprise up to 30% of their C and N reserves (Martikainen et al. 1993). Thus, peatlands play an important role in the global C and N cycle and they may also contribute significantly to the production and consumption of the greenhouse gases nitrous oxide (N_2O ; Martikainen et al. 1993; Regina et al. 1996; Regina et al. 2004) and methane (CH_4 ; Augustin et al. 1996; Khalil 1999) but have not been considered in respective global estimates (Brumme et al. 2005) due to large uncertainties resulting from the diversity of peatlands.

The global warming potentials (GWP) of nitrous oxide (N_2O) and methane (CH_4) are 310 and 21 times, respectively, than that of CO_2 during a 100 year time scale (IPCC 2007). N_2O is mainly produced in soils due to autotrophic nitrification and heterotrophic denitrification (Davidson 1991). Virgin peatlands with natural high water table levels show low N_2O fluxes or consumption of N_2O due to limitation of oxygen and hence absence of nitrification resulting in low NO_3^- concentrations (Martikainen et al. 1993; Augustin et al. 1996; Regina et al. 1996; Aerts 1997; Brumme et al. 1999).

The greenhouse gas CH_4 is produced during the anaerobic decomposition processes in anoxic parts of peats by methanogenic bacteria whereas it is oxidized in the presence of free oxygen by methanotrophic bacteria (Joabsson et al. 1999). Pristine peatlands are a significant source of CH_4 to the atmosphere. Three emission pathways have been reported for CH_4 : molecular diffusion of dissolved CH_4 , ebullition (bubbling) of CH_4 and transport of CH_4 through plant roots and shoots (Tokida et al. 2005).

Previous studies have pointed out that both groundwater table levels and temperature mostly affected the fluxes of trace gases in peatlands (Augustin et al. 1996; Koponen and Martikainen 2004). The few studies of greenhouse gas emissions from virgin peatlands have been conducted in classical bogs and fens without permanent water inflow (e. g. Martikainen et al. 1993; Regina et al. 1996; Heikkinen et al. 2002). Reports of N_2O and CH_4 fluxes from slope mires, a peatland type widespread in mountain forests are lacking. Globally slope mires represent 1% of the world's peatlands comprising an area of about 4 million hectare (Joosten, 2007, personal communication). Solely the different movement of soil water in slope

mires should change controlling factors of gas formation and resulting greenhouse gas fluxes. We hypothesize that slope mires release more N_2O because of increased influx of O_2 which inhibits N_2O reductase and concurrently restricts N_2 formation. The influx of O_2 might adversely affect CH_4 formation and might favour CH_4 oxidation resulting in lower CH_4 emission from slope mires than that of pristine bogs and fens.

To test these hypotheses we conducted a field experiment in a pristine slope mire at the Harz Mountains with the following objectives:

1. To quantify and to compare fluxes of N_2O and CH_4 in mire zones at a central non-forested and a peripheral forested area representing different nutrient status, water table levels and oxygen input caused by slope water inflow
2. To monitor N contents and other relevant features of mire pore water in order to identify relationships to the production and consumption of N_2O and CH_4
3. To find out seasonal dynamics of N_2O and CH_4 fluxes

Materials and methods

Study site

The study was conducted in the Harz mountains, central Germany (federal state Saxony-Anhalt), close to the highest peak of the Harz mountain (Mt Brocken, 1,142 m a.s.l., $51^\circ 48' \text{N}$, $10^\circ 37' \text{E}$) at an elevation of 843–860 m a.s.l. Springs of the Ilse stream characterize features of the studied area (Böhlmann et al. 2005). Average annual precipitation (1951–1980) amounts to 1,609 mm and mean annual temperature is $+2.8^\circ \text{C}$ (Wegener and Kison 2002). The annual bulk N deposition at the mire was about $51 \text{ kg ha}^{-1} \text{ year}^{-1}$ in 2003 of which $25 \text{ kg ha}^{-1} \text{ year}^{-1}$ was deposited as nitrate (Böhlmann 2004).

The investigated mire comprising Fibric Histosols covers a slope of 0.015 km^2 which is part of a catchment of 0.13 km^2 . The mire was formed about 1,000 years ago (^{14}C analysis, Böhlmann 2004) on granite debris of the Mt Brocken. The thickness of the peat layer is up to 1.4 m. The largest part of the slope mire is a swamp overgrown with spruce plants (0.01 km^2) and a *Calamagrostio villosae-Piceetum-*

community. Vegetation of the central part of the mire is dominated by a *Eriophorum angustifolium*-community and is mostly free of spruce trees (Böhlmann et al. 2005).

Field sampling and analysis

Fluxes of N_2O and CH_4 were measured biweekly with the closed chamber method during the study period that lasted from 19 June 2002 to 9 July 2003. Measurements have not been made at days with high snow cover during winter from November to March. For flux measurements, 10 chambers consisting of PVC rings (30 cm height, 30 cm diameter) were installed into the peat and remained throughout the entire study period. Four chambers were set up at the peripheral forested site and six chambers at the central non-forested site of the slope mire. Gas samples were taken at 0, 20 and 40 min after closing the chambers with a lid using evacuated glass flasks (100 ml). Before sampling, air pressure of the glass flasks was checked by pressure sensor (Loftfield et al. 1997). Air temperature was measured at a height of 2 m and soil temperatures were measured in 2.5, 5, 10 cm soil depth during gas sampling periods. The gas samples were analysed within few days by an automated gas chromatographic system consisting of a computer-controlled gas chromatograph (Carlo Erba) equipped with a flame ionization detector (FID) for CH_4 , an electron capture detector (ECD) for N_2O measurements, and an autosampler for 64 sample flasks (Loftfield et al. 1997).

Peat samples were taken at the beginning of measurements in June 2002 next to the chambers from a depth of 0–20 cm. Air-dried peat samples were analysed for total C (C_t) and N (N_t) with an elemental analyzer (Hanau, Germany). The pH of the peat samples was determined after shaking with 0.01 M $CaCl_2$. NH_4^+ and NO_3^- of the peat samples have been extracted with a 2 M KCl-solution and analysed with a continuous-flow-analyser (Skalar, Netherlands) using the indophenol-blue method.

Groundwater piezometers were placed in the peat layer adjacent to the chambers. The groundwater piezometers were used to measure water table levels (WT), redox potentials (Eh, WTW, Germany), and oxygen levels (O_2 , WTW, Germany) at the time of gas measurements. Measurements were performed at a depth of 30 dcm. Furthermore, water samples were

collected and analysed for ammonium, nitrate, pH (WTW, Germany) and electric conductivity (EC) (WTW, Germany).

Statistical analyses

All statistical analyses were performed using a SAS software package [SAS Institute (1999–2001), Inc., Cary, USA, Release 8.2] and Statistica for Windows v. 5.1 (StatSoft, Inc. 1996, Tulsa). Pearson correlation coefficients were calculated to elucidate relationships between gas fluxes, soil temperature and variables of mire pore water. One-way analysis of variance (ANOVA) has been used to test significance of differences between gas fluxes at the forested and non-forested part of the mire. Data were tested for normality and equality of variance before statistical analyses.

Results

Soil temperature, water table, peat soil, mire pore water

Thickness of peat differed significantly ($p < 0.05$, $n = 4$, Table 1) between the central and peripheral site of the mire being 105 and 56 cm respectively. Top layer (0–20 cm) of peat at both locations showed similar pH, bulk density, total N and C:N ratio while nitrate content was higher in the forested area of the mire ($p < 0.05$, $n = 4$, Table 1).

Table 1 Characteristics of the peat soil (*Fibric Histosol*) at the forested peripheral and the non-forested central area of the slope mire (0–20 cm depth, $n = 4$)

	Forested area	Non-forested area
Peat layer thickness (cm)	56.2±5	105±6*
Bulk density ($g\ cm^{-3}$)	0.14±0.08	0.06±0.01
pH	3.9±0.2	3.9±0.03
EC ($\mu S\ cm^{-1}$)	191±17	122±39
NH_4 ($mg\ N\ kg^{-1}$)	65.6±3	96.3±51
NO_3 ($mg\ N\ kg^{-1}$)	24.1±15	4.2±1*
N_t (%)	1.6±0.2	1.4±0.2
C:N	24±0.2	29±4.8

Data are means ± SD (standard deviation)

* $p < 0.05$ (Tukey Test)

Not being different between soil depths soil temperatures ranged from -0.1 to 19.6°C for the dates monitored (Fig. 1a). Water table levels differed significantly between the forested and non-forested areas ($p < 0.05$, $n = 16$, Fig. 2a). At the peripheral forested area, water table levels varied between -1 and -10 cm below the mire surface and between -4 and -27 cm at the central non-forested part of the mire. The highest water table levels were found during snow melting in April 2003, the lowest in July 2002.

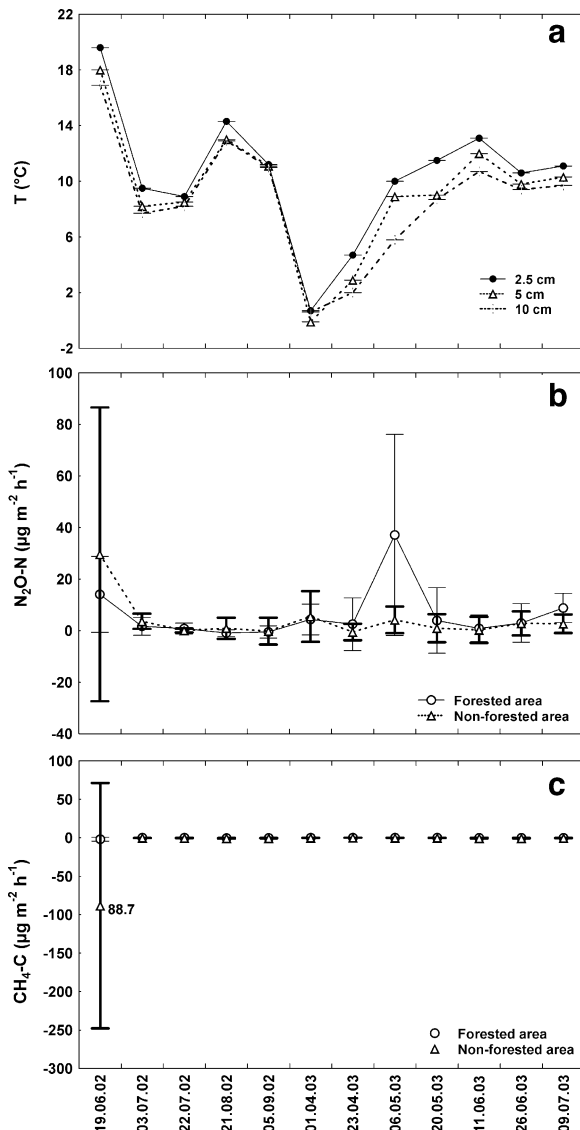


Fig. 1 a–c: Seasonal pattern of soil temperatures ($^{\circ}\text{C}$) (a) and gas fluxes of N_2O (b) and CH_4 (c) ($\mu\text{g m}^{-2} \text{h}^{-1}$) at the forested and non-forested monitoring sites of the slope mire (means with standard deviations)

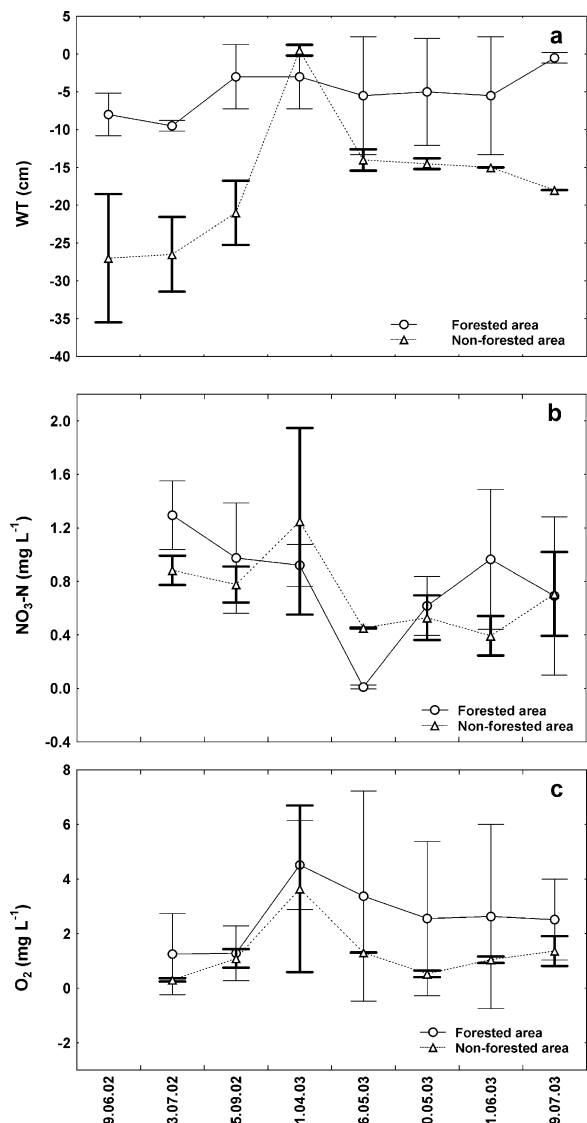


Fig. 2 a–c: Seasonal patterns of water table levels (WT, cm) (a), nitrate concentrations (mg L^{-1}) (b) and oxygen concentrations (mg L^{-1}) of the mire pore water at the forested and non-forested monitoring sites of the slope mire (means with standard deviations)

Pattern of pore water characteristics followed the temporal variation of the water level. Values of O_2 being low during low water table levels in summer increased with water table in winter and peaked in April. On average the redox potential was 150 mV and 14 mV and O_2 content averaged 2.6 mg L^{-1} and 1.4 mg L^{-1} at the forested and non-forested area, respectively. Resulting from a considerable spatial variation of O_2 and Eh in pore water (Table 2) these values did not significantly differ. Values of pH,

Table 2 Hydro-chemical variables of mire pore water at the forested peripheral and the non-forested central area of the slope mire during the study period (30 cm depth, $n=16$)

	Forested area	Non-forested area
Water table (cm)	-4.6 ± 2.8	$-16.7 \pm 5.9^*$
Temperature ($^{\circ}\text{C}$)	7.1 ± 3.2	10.2 ± 3.9
Eh (mV)	149 ± 130	14 ± 95
O_2 (mg L^{-1})	2.6 ± 2.2	1.4 ± 0.6
EC ($\mu\text{S cm}^{-1}$)	53 ± 11	$34 \pm 6^*$
pH	4.4 ± 0.5	4.5 ± 0.3
NH_4 (mg N L^{-1})	0.17 ± 0.1	0.13 ± 0.1
NO_3 (mg N L^{-1})	0.78 ± 0.4	0.71 ± 0.3
N_i (mg L^{-1})	1.83 ± 0.5	1.48 ± 0.4
TOC (mg L^{-1})	10.78 ± 9.6	15.37 ± 1.86

Data are means \pm SD (standard deviation)

* $p < 0.05$ (Tukey Test)

ammonium, total N and EC remained relatively constant during the study (not shown) while nitrate showed some variations with lowest values in May 2003 (Fig. 2b). In contrast to pH and values of ammonium and nitrate concentrations the conductivity of pore water differed significantly ($p < 0.05$, $n = 16$) between the forested and non-forested areas of the mire averaging 53 and 34 $\mu\text{S cm}^{-1}$ respectively.

N_2O

The N_2O emissions ranged between -0.9 and $+37.1 \mu\text{g m}^{-2} \text{h}^{-1}$ with peak emissions at the 19th June 2002 and 6th May 2003 (Fig. 1b).

No relationships were found between N_2O release of the mire and soil temperature, water table level, Eh and ammonium concentration. Negative correlations were observed between N_2O release and O_2 contents at the central part of the mire and between N_2O release and nitrate for the one measuring point only at the peripheral part of the mire (Table 3). Mean annual N_2O emissions did not differ significantly between the sites and amounted to $0.4 \pm 0.5 \text{ kg N ha}^{-1} \text{ year}^{-1}$ at the forested and 0.2 ± 0.4 at the non-forested site.

CH_4

The mire was found to be a CH_4 sink at almost all sampling dates (Fig. 1c). The annual CH_4 uptake of the mire adds up to $-0.02 \pm 0.01 \text{ kg ha}^{-1} \text{ CH}_4\text{-C}$. The highest uptake of $-88.7 \mu\text{g m}^{-2} \text{h}^{-1}$ occurred during low level of water table at the 19th June 2002. At all

other observation dates CH_4 fluxes of the mire varied around zero with CH_4 rates of -1.8 to $0.005 \mu\text{g m}^{-2} \text{h}^{-1}$. The exceptional singular high peak uptake does cause the significant correlation between CH_4 uptake and soil temperature (Table 3). None of the other variables revealed a significant correlation to CH_4 fluxes. CH_4 fluxes amounted to $-0.01 \pm 0.01 \text{ kg ha}^{-1} \text{ year}^{-1}$ at the forested and $-0.02 \pm 0.01 \text{ kg ha}^{-1} \text{ year}^{-1}$ at the non-forested areas and did not differ significantly.

Discussion

Temporal pattern of N_2O emission

Two N_2O emission peaks observed in this study could be classified as event-related emissions according to Brumme et al. (1999). One N_2O peak of $30 \mu\text{g m}^{-2} \text{h}^{-1}$ was measured at the central part of the mire on 19th June 2002 during low water table level. The second high N_2O release on 06th May 2003 followed a freezing–thawing periods. Increased N_2O emissions during the freezing–thawing cycles have been

Table 3 Pearson correlation coefficients between gas fluxes ($\mu\text{g m}^{-2} \text{h}^{-1}$), soil temperature ($^{\circ}\text{C}$), water table levels and hydro-chemical variables of mire pore water at the forested peripheral (P) and the non-forested central area of the slope mire (C)

		$\text{N}_2\text{O-N}$	$\text{CH}_4\text{-C}$
Soil temperature, 2.5 cm	P	0.02	-0.42^*
	C	0.23	-0.30^*
Soil temperature, 5 cm	P	0.01	-0.41^*
	C	0.22	-0.29^*
Soil temperature, 10 cm	P	-0.12	-0.43^*
	C	0.22	-0.29^*
Water table ^a (cm)	P	-0.27	0.08
	C	-0.69	0.45
Eh (mV)	P	0.21	-0.13
	C	-0.39	0.72
O_2 (mg L^{-1})	P	-0.17	0.66
	C	-0.76^*	0.40
NO_3 (mg N L^{-1})	P	-0.79^*	0.04
	C	-0.38	0.55
NH_4 (mg N L^{-1})	P	-0.18	0.53
	C	-0.33	0.01
pH	P	0.14	-0.66
	C	0.18	-0.50

* $p < 0.05$ (Tukey Test)

^aWater table levels below the mire surface

reported by several other studies (Teepe et al. 2000; Koponen and Martikainen 2004; Koponen et al. 2006). Soil thawing has been suggested to favour denitrification processes due to high water saturation and limited O₂ diffusion (Teepe et al. 2000). Furthermore freezing of soils destroys microbial cells providing substrates for denitrifying microbes (Papen and Butterbach-Bahl 1999). Inhibition of N₂O reductase at low temperatures was discussed also as a reason for high N₂O release during freezing–thawing cycles (Melin and Nommik 1983). Several authors reported that increasing nitrate concentrations in liquid water during frost enhance N₂O production (Stähli and Stadler 1997).

With the exception of the event-related emissions N₂O fluxes remained low at both sites despite differences in water table levels. N₂O release of peatlands has been reported to be mainly affected by water table and nutrition status (Martikainen et al. 1993; Augustin et al. 1996; Regina et al. 1996; Brumme et al. 1999). Negative relationships between emitted N₂O amounts and water table levels have been established in various studies (Maljanen et al. 2003; Drösler 2005) due to increases of O₂ availability promoting nitrification in drier peat that either directly increased N₂O formation or produce NO₃⁻ as precursor of denitrification (Seitzinger 1994; Regina et al. 1996). Very high N₂O emissions between 25 and 72 kg N₂O-N ha⁻¹ within 46 weeks were observed in single chambers at a drained alder forest in Bornhöved, Germany (Brumme et al. 1999). N₂O emissions up to 27 kg N₂O-N ha⁻¹ year⁻¹ have been monitored at newly drained peatlands with a decreasing water table level down to -60 cm (Augustin et al. 1998). Similar drainage caused high N₂O releases up to 79 μg m⁻² h⁻¹ for a short time (Dorwick et al. 1999). Several authors observed annual N₂O emission rates of >1 kg N ha⁻¹ at mean water table levels of -23 to -50 cm (Regina et al. 1996; Augustin et al. 1998; Drösler 2005). Commonly found higher water table levels in our study might be the reason for the generally observed low N₂O emission.

Annual N₂O fluxes

Annual mean N₂O fluxes amounting to 0.25 kg N₂O-N ha⁻¹ did not statistically differ between the monitoring sites at the mire. These fluxes are in line with those reported for various studies of pristine bogs

and fens with high water table levels (Martikainen et al. 1993; Augustin et al. 1996; Regina et al. 1996; Drösler 2005). Hence our hypothesis that raising O₂ input through flowing water increases N₂O release from mire water below the water table level by decreasing N₂ formation could not be confirmed. Likewise assumptions that observed high O₂ concentrations in the pore water are assumed to restrict N₂ formation when nitrate is not limiting denitrification processes due to high atmospheric input (Davidson 1991; Granli and Bøckman 1994; Scholefield et al. 1997) have to be questioned. Denitrification processes resulting in N₂ formation in water and sediments require O₂ contents of <0.1 to a maximum of 0.8 mg L⁻¹ (Granli and Bøckman 1994; Mühlherr and Hiscock 1998; Venterink et al. 2003). Accordingly nitrate should not have been reduced to N₂ in large amounts in our slope mire during the study period since the oxygen contents mostly exceeded these values. In addition low pH of the mire pore water should have restricted N₂ formation. Acidity inhibits N₂O reductase and thus increases the N₂O:N₂ ratio (Granli and Bøckman 1994; Aerts 1997). At the prevailing hydro-chemical conditions of our slope mire formation of N₂O should be favoured compared to N₂. Various authors reported that groundwater was oversaturated with N₂O which emitted to the atmosphere when the water reached the surface (Mühlherr and Hiscock 1998; Hefting et al. 2006). N₂O captured in groundwaters below our slope mire is likely to be emitted when those groundwaters are rising to the surface in the spring which obviously requires further investigation.

Temporal pattern of CH₄ emissions

CH₄ fluxes of the mire didn't show any seasonal pattern. Hydro-chemical variables of the mire pore water did not explain fluxes except probably the high CH₄ uptake rates of 88 μg m⁻² h⁻¹ at the central part of the mire on the day of 19th June 2002. The high uptake occurred during low water table level of -27 cm and was assumed to be the result of CH₄ oxidation by aerobic methanotrophic bacteria when aerobic conditions in peat could be enhanced. Water table has been identified to be the major controlling factor of CH₄ emission rates in peatlands. With the water table up to 62% of the variability of CH₄ fluxes have been explained (MacDonald et al. 1998; Drösler 2005). Different water table levels have been reported

at which peatlands may change from CH₄ release to CH₄ uptake. Some authors reported a critical water table level of >−10 cm (Christensen et al. 2003; Drösler 2005). In contrast Augustin et al. (1996) observed still small CH₄ releases of 0.6–3.5 kg C ha^{−1} year^{−1} in drained mires with water table levels down to −60 cm. Beside water table levels plant species composition and cover of vascular plants are known to affect CH₄ release from peatlands. Plant species with aerenchymous tissue are able to transport CH₄ through the root surface without being oxidized by methanotrophic bacteria (Joabsson and Christensen 2001). Even though these CH₄ fluxes would be included in our measurements the mire showed no significant CH₄ release. Thus plant mediated CH₄ fluxes seems not to be important at the study site although our slope mire show a high cover of vascular plants. Similarly positive impact of available nutrients and labile carbon on the CH₄ release (Joabsson and Christensen 2001; Yavitt et al. 2005) could not be confirmed in our study.

Annual CH₄ fluxes

Annual CH₄ flux revealed an uptake of −0.02 kg CH₄-C ha^{−1} not differing between the forested and non forested parts of the slope mire. The small annual consumption of CH₄ is in contrast to reported CH₄ releases from peatlands with similar high water table levels (Augustin et al. 1996; Drösler 2005). Restricted oxygen diffusion causing strictly anaerobic conditions of Eh values lower than −250 mV and high thickness of peat enhancing anaerobic peat layer capable of CH₄ production were discussed to be reasons for high methane fluxes in peatlands with high water table levels (Mitsch and Gosselink 2000). The uptake of CH₄ at our slope mire might be the result of a restricted CH₄ production due to O₂ input through groundwater movement causing redox potential far above the necessary value for methane production. A weak positive correlation between water inflow and oxygen content in pore water ($R=0.52$, $p<0.05$, $n=16$) does indicate the importance of flowing groundwater for methane oxidation in slope mires. An additional explanation for low CH₄ emission rates might be the thin peat layer of our slope mire with low total production of methane not exceeding methane oxidation in the aerobic part of the slope mire.

Conclusions

In contrast to peatlands without permanent water inflow the studied slope mire showed oxygen influx through inflowing pore water restricting reduction processes. Compared to pristine peatlands the influx of oxygen is assumed to restrict CH₄ production of the slope mire explaining its weak CH₄ consumption. N₂O release from the slope mire didn't differ from those of reported peatlands with high water table levels and low oxygen availability. Nevertheless slope mires have a high potential for the N₂O formation due to available NO₃[−]. Our assumption that areas of slope mires with springs may be hot spots for N₂O releases requires further research.

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