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# Changes of methane and nitrous oxide emissions in a transition bog in central Germany (German National Park Harz Mountains) after rewetting

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**Abstract** During the last decades, various renaturation programmes have been initialized to recover nutrient sink and ecological functions of peatlands by rewetting. Rewetting, however, often results in the formation of hotspots for methane (CH<sub>4</sub>) emissions and in temporal dieback of local vegetation. The present study aimed at quantifying changes of CH<sub>4</sub> and nitrous oxide (N<sub>2</sub>O) emissions in a peatland currently under continuous rewetting conditions. Emissions where studied at a permanently flooded site and a non-flooded peat site with fluctuating water tables by using common closed chamber method. The

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Department of Soil Physics, Helmholtz Centre for Environmental Research-UFZ, Theodor-Lieser-Str. 4, 06120 Halle (Saale), Germany permanently flooded site revealed extremely high CH<sub>4</sub> emissions (up to 1195 mg C m<sup>-2</sup> d<sup>-1</sup>) which were positively correlated with temperature, nutrient content, dissolved organic carbon and nitrogen concentration of the peat soil water. In contrast, the non-flooded peat site, with lower and fluctuating water tables (WT), showed significantly lower CH<sub>4</sub> emissions and an increasing trend of CH<sub>4</sub> release associated with a generally increasing WT caused by the progressing rewetting process. Lower N<sub>2</sub>O emissions (<24 µg N m<sup>-2</sup> d<sup>-1</sup>) were observed at the flooded site. By contrast, the non-flooded peat site with fluctuating WT showed significantly higher N<sub>2</sub>O emissions (up to 4178 µg N m<sup>-2</sup> d<sup>-1</sup>), in particular at high temperatures during summer time. The present results indicate

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that permanently flooded conditions during rewetting processes might cause higher  $CH_4$  emissions compared to fluctuating WT which in contrast might enhance N<sub>2</sub>O emissions. In total, however, no decreasing trend for  $CH_4$  emissions throughout the five-year renaturation period could be found. At least for N<sub>2</sub>O we observed a decreasing trend during rewetting.

**Keywords** Climate change · Methane · Nitrous oxide · Renaturation · Vegetation shifts · Wetland

#### Introduction

Natural peatlands represent a tremendous sink of atmospheric carbon dioxide (CO<sub>2</sub>) with an accumulation of about 500  $\pm$  100 Gt of the global soil carbon (Yu 2012). Drainage of peatlands for agriculture and forestry use resulted in an accelerated mineralisation of stocked carbon (C) and led to a substantial loss of C and nitrogen in form of CO2 and nitrous oxide (N2O), respectively, of globally 2-3 Gt CO<sub>2</sub>-eq per year (Joosten and Couwenberg 2009). During the last four decades, rewetting programmes were initialized in Central Europe aiming at reducing greenhouse gas (GHG) emissions and thus, to recover the nutrient sink and ecological functions of pristine peatlands (e.g. Drösler 2005). It has been shown recently that rewetted peatlands may evolve to highly dynamic systems characterised by an increased mobilisation of nutrients, dissolved organic carbon (DOC) and elevated emissions of methane (CH<sub>4</sub>) and CO<sub>2</sub> as a result of rapid plant dying and decomposition (Zak et al. 2015). Extremely high CH<sub>4</sub> emissions from rewetted peatlands have been observed and attributed to enhanced C and nutrient availability via the supply of fresh labile organic substrates resulting from the die-off of plants in formerly non-flooded areas (Wilson et al. 2009; Hahn et al. 2015). Afterwards prevailing plant species revealed a major impact on the CH<sub>4</sub> release from rewetted peatlands. First, they are assumed to stimulate CH<sub>4</sub> formation due to root exudation of low-molecular organic compounds from living plants (Hines et al. 2008). Second, they provide freshly reduced C from senescent plant biomass (Lawrence et al. 2013). And third, they permit transport of CH<sub>4</sub> from the anaerobic root zone directly into the atmosphere, by bypassing the aerobic methane-oxidizing peat layers via their welldeveloped aerenchyma tissue (Whalen 2005). Concomitantly also CH<sub>4</sub> oxidation could occur in the oxygenated zone surrounding the plant roots (Chanton et al. 2002). In addition, the litter quality, in terms of microbial usability of a potential substrate, may differ substantially between plant species (Lai 2009), resulting in differences of CH<sub>4</sub> emissions between various plant species (Zak et al. 2015). It was shown that  $CH_4$ production by decomposition of plant litter with higher nitrogen to C content is supported whereas plant litter composition rich in lignin and polyphenols suppress CH<sub>4</sub> production (Freeman et al. 2001; Thangarajan et al. 2013). Thus, differences in CH<sub>4</sub> emissions between rewetted peat sites can be expected due to differences in plant litter quality of the prevalent plant species.

In contrast to CH<sub>4</sub>, rewetted and natural peatlands are generally characterised by low fluxes of N<sub>2</sub>O, due to anoxic conditions, which inhibit nitrification processes and cause nitrate limitation at high water saturation (Drösler 2005; Beyer and Höper 2015). Heterotrophic denitrification represents the most important N<sub>2</sub>O source in water-logged peat soils (Regina et al. 1996). It was observed previously that N2O produced from fertilized nitrate via denitrification was stored in the water phase of water-logged peat soils and later on released to the atmosphere only by falling water tables (Aerts and Ludwig 1997; Tauchnitz et al. 2015). As a consequence temporarily strong N<sub>2</sub>O bursts can be expected in rewetted peat sites which are characterized by considerably fluctuating water tables. In contrast to CH<sub>4</sub> permanently flooded conditions therefore appear to be at least favourable to decrease N<sub>2</sub>O emissions to near zero (Beyer and Höper 2015).

Most rewetting studies in temperate regions were performed in formerly agriculturally used fens with vegetation dominated by hydrophytes. In temperate regions, however, bogs with an oligotrophic vegetation have scarcely been studied so far. Therefore, the present study aimed at quantifying  $CH_4$  and  $N_2O$ emissions of a drained transition bog (currently undergoing a rewetting process) over a period of four years by using common closed chamber technique. The main objectives of the study are:

(i) Examining changes in the  $CH_4$  and  $N_2O$  release in the course of progressing rewetting,

- (ii) Evaluating differences of the  $CH_4$  and  $N_2O$  emission patterns between (a) permanently flooded conditions and (b) non-flooded conditions with fluctuating water table (mostly below the peat surface),
- (iii) Identifying main drivers of  $CH_4$  and  $N_2O$  release at the study site.

### Materials and methods

#### Study site

Field experiments were performed in the German National Park Harz Mountains (central Germany) in a transition bog ecosystem (650–670 m a.s.l) in direct vicinity to the Mt. Brocken (1141 m a.s.l.; 51°48'N, 10°37'E). A detailed description of the study site was previously given by Osterloh et al. (2016). Climate conditions are characterised by a long-term (1961–1990) mean annual temperature of 5.3 °C and a mean annual precipitation of 1278 mm (data from German Weather Service).

Prior to rewetting the study site was drained by several ditches and planted with spruces for forestry use in the 1980s. Rewetting was initiated in 2005 and continued in 2009 and 2010 by closing the ditches and redirecting a small stream into the central peatland area.

For emission analyses two different study sites were chosen within the peatland. One site was characterised by permanently flooded conditions (i.e. water table (WT) above the soil surface since August 2009). Consequently, vegetation changed from grass dominated to sedge-grass-moss dominated structures and plant productivity increased simultaneously.

The second study site was rewetted since June 2010, non-flooded and characterised by a fluctuating WT. The vegetation is characterised by an open young spruce stand rich in *Vaccinium myrtillus* and mosses like *Polytrichum formosum* and *Pleurozium schreberi* but rare in wetland plant species. Rewetting caused a die-off of typical acidic forest plant species followed by a spreading of fen indicator species (namely *Carex canescens, Agrostis canina, Molinia caerulea* and *Sphagnum spec.*).

The peat soil layers of both sites are 0.7-1.1 m thick. Dry density is in the range of  $0.15-0.22 \text{ g cm}^3$ .

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Both peat sites are characterised by mesotrophic conditions with C:N ratios ranging from 22 to 29. Nitrate contents are low with a maximum of 10 mg g<sup>-1</sup> dry matter of soil. Detected pH values are in a range of 3.4–3.9 and represent strong acid conditions.

### Field sampling and analysis

At each sampling site five polyethylene (PE)-tubes (30 cm diameter, 30 cm height) were pushed 15 cm into the peat soil layer acting as the emission chambers frame. From June 2010 to October 2014 gas release was measured weekly and biweekly (in some cases monthly) by the closed chamber method. Gas measurements were omitted during the winter time (December to February) in case of thick snow covering. For gas sampling, a peat soil column was covered with a dark, opaque PE-chamber (31.5 cm in diameter, 15 cm height) and sealed against the atmosphere by a rubber strip. Chamber then remained closed for two hours while gas samples (aliquot of 100 ml) were transferred by a polypropylene syringe via a three-way valve and a needle from the chamber headspace into a septum-capped vial (20 ml) every 20 min (Tauchnitz et al. 2015). Before gas sampling, all vials were flushed with oxygen to prevent contamination from previous samplings. Gas samples were analysed within a few days for CH<sub>4</sub> and N<sub>2</sub>O by a specially configured gas chromatograph (GC 14B, Shimadzu, Japan) equipped with a flame ionization detector (FID for CH<sub>4</sub>) and an electron capture detector (ECD for N<sub>2</sub>O) (see Segschneider et al. 1996). Emission rates (E) of CH<sub>4</sub> and N<sub>2</sub>O ( $\mu g m^{-2}$  $h^{-1}$ ) were calculated according to Eq. 1 with  $\Delta C$ ( $\mu$ g N per norm liter) as the amount of CH<sub>4</sub> and N<sub>2</sub>O emitted into a normalized gas volume between  $t = 0 \min$  and t = 20, 40, 60, 80, 100 and 120 min, respectively, and h (current champers height) multiplied by 10 and divided by the time in order to correct for the current chamber volume, the covered area and the corresponding closure time. Prior to the calculation of the volume normalized gas concentrations analysed gas concentration was always corrected to standard temperature (i.e. 273 K) and standard pressure (i.e. 1013.25 hPa).

$$E = \Delta C_t \cdot h \cdot 10/t \tag{1}$$

Daily emission rates between the measurement campaigns were obtained by linear interpolation and were used to calculate yearly balances.

Soil temperatures were measured with a puncture thermometer (stainless steel sensor, VWR International GmbH) in 5 and 10 cm depths concomitantly to the gas measurements. In addition to gas sampling WT, redox potential (Eh, Hach Lange GmbH, Germany) as well as oxygen level (O2, Hach Lange GmbH, Germany) was monitored simultaneously by groundwater piezometers placed 20 cm deep in the peat soil layer adjacent to the chambers. Furthermore, water samples were taken and analysed for ammonium (NH<sub>4</sub><sup>+</sup>, photometric, Thermo Electron Corporation, Germany), nitrate, sulfate  $(NO_3^-, SO_4^{2-}, ion chro$ matography, Metrohm GmbH & Co. KG Filderstadt, Germany), DOC (Total Carbon Analyser, NPOC method, Shimadzu TOC-V<sub>CSH</sub>, Japan), iron (Fe, flame atomic absorption spectrometry, Thermo Electron Corporation, Germany) as well as pH (potentiometric, WTW GmbH, Germany) and electric conductivity (EC, conductimetrically, WTW GmbH, Germany). Total N and C contents of plants were determined by high temperature combustion method (Elementar Analysensysteme GmbH, Germany). Total P contents of biomass were analysed by inductively coupled plasma optical emission spectrometry (Ultima 2, Horiba Jobin–Yvon). For analyses of dissolved CH<sub>4</sub> concentration, peat soil water was sampled according to Tauchnitz et al. (2015) and measured by a Headspace GC (HSS 86.50, DANI, Italy) coupled to a GC system (GC 14B, Shimadzu, Japan) equipped with a Porapak Q column (80/100 mesh, 6L, 2 mm ID) and an ECD.

## Statistical analyses

Statistical analyses were performed using SPSS (vers. 22). Levene-test was used to examine data sets concerning their equality of variances. Since data were not normally distributed the non-parametric Mann–Whitney-U-test was applied to check for significant differences between peat site locations and years, respectively. Spearman correlation coefficients were calculated to test correlations between gas fluxes and environmental as well as nutrient conditions (e.g. WT,  $NH_4^+$ -,  $NO_3^-$ -contents, etc.).

# Results

Weather and hydrological parameters

Annual precipitation ranged between 974 mm (2011) and 1428 mm (2010) during the study period and was, with the exception of 2010, below the long-term mean (1278 mm) of the study site (Table 1). Annual mean temperature was in a range of 4.6  $^{\circ}$ C (2010) to 7.3  $^{\circ}$ C (2014).

Water tables were significantly different between the two sampling sites. At the flooded peat site a permanently high WT with an annual variation of 2.5 cm (2012) to 4.3 cm (2014) above the soil surface was observed. WT at this site showed very low temporal variations (Table 1). In contrast, the recently rewetted non-flooded peat site showed a significantly lower WT (Table 1) with annual means in the range of 11.7 cm (2011) to 1.8 cm (2014) below the soil surface and high temporal variations. WT decreased markedly during summer with lowest value of 33 cm below the soil surface observed at the end of September 2011. In the same year, lowest annual mean WT coincided with the lowest annual precipitation of the whole study period. Nonetheless, in total an increasing trend of the WT was observed for the non-flooded peat site accompanied by decreasing temporal variations, presumably due to progressive rewetting (Table 1).

Characteristics of the peat soil water

Median redox potentials of the flooded and non-flooded peat site were 187 and 259 mV, respectively, with significant differences between both sites (Table 2).

 $\rm NH_4^+$  concentrations and EC of the peat soil water differed significantly between both sites with higher values at the non-flooded site (Table 2).  $\rm NO_3^-$  concentrations of the peat soil water were low at both sites with median values of <0.1 mg N L<sup>-1</sup> and maximal concentrations of 1.5 mg N L<sup>-1</sup> (observed during strong snow melt coinciding with high precipitation in April 2013; Tables 1, 2). DOC concentrations of the peat soil water (Fig. 1) showed no significant differences between both sites with median values of 21 mg L<sup>-1</sup> (flooded) and 26 mg L<sup>-1</sup> (non-flooded). Most notable was a distinct temporal variation in DOC concentrations with highest values during summer.

Year	P (mm)	T (°C)	ΜT	, (cm)																		
			Floc	oded									-non-	flooded								
			п	Mean	SD	Min	Q25	Med	Q75	Max	IJ	U2	u	Mean	SD	Min	Q25	Med	Q75	Max	IJ	U2
2010	1428.3	4.6	13	2.9	0.5	1.9	2.7	3.0	3.2	3.5	а	q	12	-3.3	3.2	-8.9	-5.1	-2.9	0.0	0.5	bc	а
2011	973.7	6.8	15	3.0	0.9	1.4	2.4	3.1	3.6	4.2	а	q	16	-11.7	8.4	-32.8	-15.6	-9.4	-6.6	-2.1	а	а
2012	1130.7	5.9	8	2.5	0.8	1.3	1.7	2.7	3.1	3.3	а	q	11	-7.7	7.4	-20.8	-11.1	-5.0	-2.1	-0.3	ac	а
2013	1152.5	5.3	Ζ	3.1	0.5	2.1	3.0	3.3	3.5	3.6	а	q	10	-2.4	3.8	-12.0	-2.4	-1.8	0.2	0.9	q	а
2014	1202.0	7.3	9	4.3	0.5	3.4	4.1	4.3	4.6	4.9	q	q	9	-1.8	1.7	-4.3	-2.6	-1.9	-1.1	0.8	$\mathbf{bc}$	а
	1177.44	6.0	49	3.1								q	56	-5.4								а

**Table 1** Annual precipitation (P), air temperature (T) and water table (WT) related to soil surface from the flooded and non-flooded peat sites during the study period (June

### Nutrient contents of biomass

N contents of biomass at the flooded and non-flooded site were in the range of 1.4-3.9% and 1.1-4.1%, respectively (Table S-1). C:N ratios of biomass ranged between 11 and 31 (flooded) and 11 and 43 (non-flooded) and P contents between 0.1 and 0.4% (flooded) and 0.07 and 0.4% (non-flooded). During the study period N and P contents significantly increased, while C:N ratios of biomass decreased at both site.

# CH<sub>4</sub> emissions

Highest CH<sub>4</sub> emissions were measured at the permanently flooded peat site ranging from 1.5 to 1195 mg C  $m^{-2} d^{-1}$  (Fig. 2; Table S-2).

In addition, a pronounced inter-annual and spatial variation of  $CH_4$  emissions was found. Highest  $CH_4$  release was generally observed during summer associated with high temperatures. Overall,  $CH_4$  emissions at the flooded site showed no significant trend during the whole study period (Fig. 2; Table S-2). Significantly lower  $CH_4$  release was observed at the non-flooded site where emission rates largely tended towards zero. Nonetheless, also higher rates could be detected (up to 571 mg  $CH_4$ – $C m^{-2} d^{-1}$ ), but mainly at the end of the study period where WT fluctuations diminished and WT remained constantly close the soil surface (Fig. 2).

Mean annual CH<sub>4</sub> emissions of the permanently flooded and non-flooded site accounted for  $505 \pm 414 \text{ kg C} \text{ ha}^{-1} \text{ a}^{-1}$  and  $68 \pm 60 \text{ kg C} \text{ ha}^{-1} \text{ a}^{-1}$ , respectively.

# N<sub>2</sub>O emissions

N<sub>2</sub>O emissions showed significant differences between both peat sites (p < 0.05, n = 49, Man-Whitney-U-test). High N<sub>2</sub>O release rates of maximal 4178 µg N m<sup>-2</sup> d<sup>-1</sup> were found at the non-flooded peat site (Fig. 3; Table S-3) within the first years of the study with highest emissions during summer coinciding with low WT and high temperatures.

Nonetheless,  $N_2O$  emissions showed a decreasing trend associated with increasing WT (with exception of the sampling date 25.07.2013). Furthermore, decreasing inter-annual variation of  $N_2O$  emission was observed. High  $N_2O$  release rates occurred in

Parameter	Floo	oded						Non	-flooded					
	n	Mean	SD	Min	Med	Max	U	n	Mean	SD	Min	Med	Max	U
pH (mg $L^{-1}$ )	67	4.3	0.4	3.7	4.3	5.3	b	66	4.2	0.4	3.6	4.3	5.2	a
EC ( $\mu$ S cm <sup>-1</sup> )	67	59.1	10.1	40.5	58.0	92.9	а	66	65.9	15.4	47.3	62.1	115.6	b
$O_2 (mg L^{-1})$	64	3.7	2.7	0.4	2.9	12.1	а	62	3.2	1.5	0.6	3.0	6.4	a
T (°C)	64	8.4	5.0	0.0	9.5	17.6	а	62	8.7	4.7	0.8	9.6	18.6	a
Eh (mV)	61	152	111	-126	187	338	а	60	221	120	-96	259	358	b
$SO_4^{2-}$ (mg L <sup>-1</sup> )	67	7.10	4.48	0.25	6.85	17.62	а	66	6.64	3.14	0.33	6.47	13.21	a
Fe (mg $L^{-1}$ )	57	1.93	1.77	0.11	1.15	7.00	а	56	1.61	1.34	0.18	1.10	5.07	a
$NO_3-N (mg L^{-1})$	67	0.16	0.26	0.00	0.03	1.46	а	66	0.08	0.09	0.00	0.04	0.35	a
$NH_4-N (mg L^{-1})$	67	0.43	0.33	0.10	0.32	2.05	а	66	0.53	0.35	0.09	0.44	2.20	b
DON (mg $L^{-1}$ )	61	0.87	0.64	0.00	0.70	3.03	а	59	0.92	0.67	0.12	0.71	2.89	a
DOC (mg $L^{-1}$ )	61	32.18	24.73	4.41	21.07	105.87	а	59	34.75	26.70	4.27	25.91	99.58	a
DOC:DON	60	48	64	9	36	496	а	58	54	82	15	37	622	а

**Table 2** Peat soil water properties from the flooded and non-flooded peat sites during the study period (June 2010–October 2014), different letters indicate significant differences (p < 0.05, Man-Whitney-U-Test) between sites (U)

*EC* electric conductivity; *T* water temperature; *Eh* redox potential; *DON* dissolved organic nitrogen; *DOC* dissolved organic carbon; *SD* standard deviation; *Min* Minimum; *Max* Maximum; *Med* median; *Q*25, 25%-quantile, *Q*75, 75%- quantile

2010, 2011, and 2012 with annual median values of 193, 836 and 415  $\mu$ g N m<sup>-2</sup> d<sup>-1</sup>, respectively. By contrast, markedly lower N<sub>2</sub>O loss was observed in the study years 2013 and 2014 which were, however, only significant in 2014 (p < 0.05, n = 49, Man-Whitney-U-test).

At the permanently flooded peat site N<sub>2</sub>O emissions were generally low with a maximal N<sub>2</sub>O release of 200 µg N m<sup>-2</sup> d<sup>-1</sup>. For 63% of the sampling dates no significant N<sub>2</sub>O release (i.e. >24 µg m<sup>-2</sup> d<sup>-1</sup>) could be detected, whereas an uptake of N<sub>2</sub>O was observed at numerous sampling dates. Contrary to the non-flooded peat site N<sub>2</sub>O release of the flooded site showed very low inter-annual and spatial variations. Median annual N<sub>2</sub>O values indicate no significant trend throughout the study period (Fig. 3). Annual N<sub>2</sub>O emission of the permanently flooded site accounted for  $-0.03 \pm 0.1$  kg N ha<sup>-1</sup> a<sup>-1</sup>. At the non-flooded site significantly higher emissions of  $1.6 \pm 1.5$  kg N ha<sup>-1</sup> a<sup>-1</sup> were found (p < 0.05, n = 4, Man-Whitney-U-test).

# Discussion

## CH<sub>4</sub> emission rates and associated drivers

Highest  $CH_4$  emissions of up to 1195 mg C m<sup>-2</sup> d<sup>-1</sup> were observed at the permanently flooded peat site

(Fig. 2; Table S-2). Similar results for flooded peatlands have been reported elsewhere (Augustin and Joosten 2007; Hahn-Schöfl et al. 2011; Hahn et al. 2015). High emissions are caused by rapid plant dieoff and related decomposition processes along with the flooding and thus, providing fresh and easily degradable organic substrates and therefore enhanced C and nutrients availability for methanogenic microorganism (Wilson et al. 2009; Hahn et al. 2015). It was stated elsewhere that the first stage of litter break down during flooding subsequent to die-back of plants resulted in high mass loss of plant nutrient stock (e.g. Aerts and De Caluwe 1997), hydrolysis of polyphenols and other organic substances of mostly low molecular weight, such as carbohydrates and amino acids (Maie et al. 2006). This is supported by the increased mobility of DOC observed during the rewetting process at this site (Osterloh et al. 2016). A positive correlation between CH<sub>4</sub> emissions, which are known as a good proxy for biomass production (Strack et al. 2004), and DOC concentrations in peat soil water were additionally found at the flooded peat site (Table 3).

It was also shown recently that rewetted sites at the studied peatland are characterised by a more unbalanced DOM quality with higher fluorescence and humification indices (HIX), indicating a higher level of aromatic DOM and a stronger humification Fig. 1 Temporal course of soil and water temperature (T) and dissolved organic carbon (DOC) from the flooded and non-flooded peat sites (n = 4) during the study period (March 2010–December 2014); within the DOC diagrams *black lines* indicate means, *box* represent 25-, 50 and 75%-quantiles, *dots* are outliers



compared to pristine sites (Herzsprung et al. 2017; Table 4). These findings are well in agreement with Kalbitz et al. (2000), who reported changes of DOM quality during peat degradation with higher amounts of aromatic moieties in degraded peatlands. A high content of aromatic components might contradict with the idea of enhanced C availability via easily degradable organic substrates derived from submerged plants which was previously discussed (e.g. Zak et al. 2015). However rewetted sites are characterised by large seasonal and spatial variations in DOM quality (Herzsprung et al. 2017), indicating a high dynamic of rewetted systems.

A positive correlation was found between  $CH_4$ emissions and EC, as well as DON concentrations of peat soil water at the permanently flooded site, confirming the positive effect of nutrient availability on microbial activity (Table 3). Note that decomposing microorganisms depend on N for their anabolism, thus increasing N concentrations induced by higher litter input suggest enhanced microbial activity (Tremblay and Benner 2006). Therefore,  $CH_4$  **Fig. 2** Temporal course of the CH<sub>4</sub> emissions and water table (WT) from the flooded and non-flooded peat sites (n = 5) during the study period (June 2010–October 2014), *dashed line* in the WT diagram represent soil surface; *black lines* in the emission diagrams indicate means, *box* represent 25-, 50 and 75%-quantiles, *dots* are outliers



production induced by fresh plant litter input during the rewetting processes might differ substantially between various plant species depending on their litter quality (Lai 2009). Zak et al. (2015) obtained a 70-times higher CH<sub>4</sub> production under the hydrophyte *Ceratophyllum demersum* (C:N ratio: 14, N content: 2.8%, P content: 0.68%) compared to *Carex riparia* (C:N ratio: 35, N content: 1.6%; P content: 0.06%). In the present study the C:N ratios, as well as the N and P contents of main plant species at the flooded site (e.g. *Carex canescens*, Table S-1) are in the range of those reported by Zak et al. (2015). Also Bohdálková et al. (2013) presented highest  $CH_4$  emissions from an oligotrophic peat bog in the Czech Republic liked to the occurrence of *Carex*. The high  $CH_4$  production rate under *Carex* species (and other vascular plants) could be attributed to their relatively fast decomposition rates compared to moss species (Lahio 2006). In

Fig. 3 Temporal course of the N<sub>2</sub>O emissions and water table (WT) from the flooded and non-flooded peat sites (n = 5) during the study period (June 2010– October 2014), *dashed line* in the WT diagram represent ground surface; *black lines* in the emission diagrams indicate means, *box* represent 25-, 50 and 75%quantiles, *dots* are outliers



agreement with other studies (e.g. Aerts and Ludwig 1997) we observed a high spatial variability of  $CH_4$  emissions with coefficients of variation often exceeding 100% within the replicates (Fig. 2; Table S-2). Similar to our results a high inter-annual variation of  $CH_4$  emissions was recognized in recent rewetting studies (Koebsch et al. 2013; Günther et al. 2015). Overall, highest  $CH_4$  release occurred at high temperatures during summer months, as supported by the

positive correlation between  $CH_4$  emissions and soil temperature at the permanently flooded site. Also Audet et al. (2013) and Beyer and Höper (2015) found increased  $CH_4$  production rates under warmer conditions. Additionally, a significant negative correlation between  $CH_4$  emissions and  $SO_4^{2-}$  concentrations of peat soil water was observed at the flooded peat site (Table 3). This finding confirms previously published results by Audet et al. (2013).  $CH_4$  is produced under

Table 3 Spearman	Parameter		Spearm	an corre	lation coe	fficient	s			
between methane $(CH_4)$ and			N <sub>2</sub> Ο (μ	$g m^{-2} d$	$ ^{-1})$		CH <sub>4</sub>	$(\mu g \ m^{-2})$	d <sup>-1</sup> )	
nitrous oxide (N <sub>2</sub> O) emissions from the flooded			Flooded	1	Non-flo	oded	Floo	ded	Non-flo	oded
and non-flooded peat sites	WT (cm)		0.25		-0.63**	*	-0.2	26	0.11	
and relevant parameters	Soil-T <sub>5 cm</sub> (°C)		-0.27		0.49**	*	0.5	51**	-0.01	
	Soil-T <sub>10 cm</sub> (°C)		-0.29*		0.54**	*	0.5	56**	0.04	
	Water-T <sub>20 cm</sub> (°C)	1	-0.27		0.54**	*	0.5	53**	0.08	
	$O_2 (mg L^{-1})$		0.34*		-0.35*		-0.0	50**	-0.02	
	Eh (mV)		0.24		0.07		-0.3	37**	-0.26	
	рН		0.08		-0.45**	*	-0.4	47**	0.26	
WT water table related to	EC ( $\mu$ S cm <sup>-1</sup> )		0.00		0.27		0.3	37**	-0.40*	*
soil surface; Soil-/Water-	$SO_4^{2-}$ (mg L <sup>-1</sup> )		0.34*		-0.39**	*	-0.7	74**	-0.10	
$T{cm}$ soil/water	Fe (mg $L^{-1}$ )		-0.17		0.63**	*	0.5	51**	0.05	
temperature in cm depth;	NO <sub>3</sub> –N (mg $L^{-1}$ )		0.37*	*	-0.09		-0.2	26	-0.14	
electric conductivity: DON	$NH_4-N (mg L^{-1})$		0.23		0.26		-0.2	22	-0.09	
dissolved organic nitrogen;	DON (mg $L^{-1}$ )		-0.15		0.66**	*	0.4	45**	0.09	
DOC dissolved organic	DOC (mg $L^{-1}$ )		-0.19		0.67**	*	0.5	56**	0.04	
* $p < 0.05$ ; ** $p < 0.01$	Date		-0.15		-0.27		-0.	14	0.45*	*
Table 4       Humification	Study sites	n	ніх				Humic-l	ike fluores	scence	
index (HIX) and humic-like	Study sites		<u></u>							
fluorescence of peat soil			Med	Mın	Max	U	Med	Mın	Max	U
pristine and rewetted peat	All values									
sites during sampling time	Pristine site	10	6.1	4.7	6.9	а	9797	8978	12031	а
of May 2013–April 2014;	Rewetted site	40	11.6	4.0	23.9	b	8081	2295	30544	а
significant differences	Summer values (M	/lay–Oo	ctober)							

Med media	an; M	in
minimum;	Max	maximum

(p < 0.05, Man-Whitney-

U-Test) between sites (U)

strictly anaerobic conditions by methanogenic archaea species as a result of fermentation of organic compounds (acetate) or reduction of CO<sub>2</sub> by H<sub>2</sub> (Le Mer and Roger 2001). The negative effect of  $SO_4^{2-}$ can be explained by the competition between  $SO_4^{2-}$ reducing bacteria, using  $SO_4^{2-}$  as an electron acceptor, and methanogens regarding the utilization of H<sub>2</sub> and acetate (Conrad 2007). Note that methanogenesis might be also inhibited in the presence of NO<sub>3</sub><sup>-</sup> and iron (Fe(III)) as electron acceptors (Le Mer and Roger 2001). However, we could not confirm this latter suggestion. Contradicting, a positive correlation between CH<sub>4</sub> emissions and Fe concentrations of peat

Pristine site

Pristine site

Rewetted site

Rewetted site

5

20

5

20

Winter values (November-April)

5.9

13.2

6.2

9.0

4.7

9.8

5.7

4.0

6.8

23.9

6.5

13.6

а

b

а

а

soil water was found, which was assumed rather a result of the interaction between DOC and Fe due to the formation of chelate complexes (Osterloh et al. 2016).

9981

15873

9613

6111

9185

7406

8978

2295

12031

30544

10527

10190

а

a

а

а

Significantly lower CH<sub>4</sub> emissions were detected at the non-flooded peat site with fluctuating WT compared to the permanently flooded site (Fig. 2). Note that the WT level is one of the key factors of CH<sub>4</sub> emissions in peatlands determining the thickness of the aerobic soil layer where CH4 oxidation takes place (Lai 2009; Wilson et al. 2009). In contrast to Jungkunst et al. (2008) who found a non-linear correlation between WT position and CH<sub>4</sub> emissions, no significant correlation was detectable in the present

study at the non-flooded peat site (Table 3). Nevertheless, higher CH<sub>4</sub> emissions coincide with higher WT throughout the whole study period (Fig. 2). Differences between the flooded and non-flooded peat site were found to be concerning CH<sub>4</sub> concentrations within the peat soil water (Table 5). In addition, no significant correlation between temperature and relevant hydro-chemical parameters to CH<sub>4</sub> emission could be seen at the non-flooded site with fluctuating WT, in contrast to the permanently flooded site (Table 3), supporting the assumption of differences in CH<sub>4</sub> production conditions between the both sites. A very effective way to channel large amounts of CH<sub>4</sub> from the anaerobic zone directly into the atmosphere, i.e. bypassing the aerobic methane-oxidizing peat layers, is a plant mediated transport through the aerenchyma of aquatic vascular plants (Whalen 2005). However, such an effect, mainly induced by wetland plants (reed plant species), are not relevant for the here presented data due to their absence on both study sites.

In 2010 (beginning of the study period), the nonflooded peat site showed significantly lower CH<sub>4</sub> concentrations in depths of 30 and 60 cm compared to the permanently flooded peat site. Already Chanton and Whiting (1995) described that at high rates of methanogenesis supersaturation of CH<sub>4</sub> might occur in the porewater of deeper anaerobic peat layers and hence, gas bubbles are formed when the partial pressure of all dissolved gases in solution exceeds the hydrostatic pressure in soil water. These bubbles accumulate within the peat profile and can be released suddenly in case of a rapid drop of atmospheric pressure, a lowering of hydrostatic pressure (e.g. via decreasing WT), a rise in temperature or mechanical stress by e.g. walking on the peat surface (Tokida et al.

**Table 5** Dissolved methane (CH<sub>4</sub>) concentrations in peat soil water in 30 and 60 cm depth from the flooded and non-flooded peat sites during sampling time of September 2010; different

2007). It is well known that the process of ebullition may contribute significantly to the total  $CH_4$  fluxes in peatlands (Tokida et al. 2007). It can be assumed that the high inter-annual and spatial variation of  $CH_4$  emissions observed in our study largely resulted from ebullition events.

# N2O emissions rates and associated drivers

Generally, low N<sub>2</sub>O emissions were observed at the permanently flooded peat site, which is in line with results from previous studies reporting negligible N<sub>2</sub>O release from rewetted peatlands (Drösler 2005; Beyer and Höper 2015; Günther et al. 2015). Note that heterotrophic denitrification should be the major process of N<sub>2</sub>O formation in peatlands (Regina et al. 1996). In fact, a weak positive correlation between  $N_2O$  release at the flooded peat site and  $NO_3^$ concentration in the peat soil water was observed indicating that N<sub>2</sub>O formation mainly depended on the availability of NO<sub>3</sub><sup>-</sup> as a source for heterotrophic denitrification (Table 3). It could therefore be assumed that the process of denitrification is still active at this peat site, even at very low soil pH, as long as  $NO_3^-$  is available (Tauchnitz et al. 2015). Actually Rückauf et al. (2004) stated that  $N_2$  should be the predominant product of denitrification in water-saturated peatlands. This could explain the extremely low N<sub>2</sub>O release rates observed at the permanently flooded peat site. The permanent flooding could limit the mobility of N<sub>2</sub>O in the sense of moving towards the atmosphere and hence N<sub>2</sub> production is favoured (i.e. accumulation and consumption of N<sub>2</sub>O) over N<sub>2</sub>O release (Jørgensen et al. 2012). Previous studies already revealed an accumulation of N2O in peat soil

letters indicate significant differences (p < 0.05, Man-Whitney-U-Test) between sites per depth (U)

Sampling depth (cm)	CH	4 <b></b> C μg 1	$L^{-1}$													
	Flo	oded							Nor	n-flooded	ł					
	n	Mean	SD	Min	Q25	Med	Q75	Max	n	Mean	SD	Min	Q25	Med	Q75	Max
30	27	4730	1563	1845	3322	4828	5896	7422	19	204	74	61	149	205	255	337
U		b								a						
60	27	4374	1486	1942	3227	4338	5248	7617	19	232	135	21	126	194	300	516
U		b								a						

SD standard deviation; Min minimum; Max maximum; Med median; Q25, 25%-quantile; Q75, 75%- quantile

water after  $NO_3^-$  addition, which was then released to the atmosphere only at falling WT (e.g. Tauchnitz et al. 2015). Conversely, Krüger et al. (2014) found a positive response of rising WT on N<sub>2</sub>O emission during rewetting of a temporally dry wetland in Southern Africa. These high N<sub>2</sub>O emissions after short-term changes in WT were explained by a flushing out of N2O which had accumulated in deeper zones. In contrast to the flooded site significantly higher N<sub>2</sub>O emission rates were observed at the nonflooded site associated with lower WT and considerable WT fluctuations (Table 1). Similar results concerning a significant response of N<sub>2</sub>O release on WT fluctuations were reported previously (Aerts and Ludwig 1997; Tauchnitz et al. 2015). It was pointed out that N<sub>2</sub>O fluxes of water-logged soils are (besides the availability of  $NO_3^{-}$ ) mainly controlled by the balance of production and consumption of N<sub>2</sub>O and by convective or diffusive transport (e.g. Well et al. 2003). As mentioned above  $N_2O$  accumulates in the saturated zone when NO<sub>3</sub><sup>-</sup> was added to a rewetted peat soil. This accumulation then occurs under constant high WT conditions due to a transport limitation towards the atmosphere. A falling WT leads then to a release of accumulated N2O based on a significant decrease of diffusional transport limitations, i.e. a direct connection of the soil gas phase to the atmosphere (e.g. Tauchnitz et al. 2015). A significant negative correlation between WT and N<sub>2</sub>O emissions was detected at the non-flooded peat site (Table 3), which proves the impact of falling WT on N<sub>2</sub>O release. However it is also known that N<sub>2</sub>O emission of waterlogged soils can be promoted by plants through rapid gas phase diffusion in the aerenchyma of wetland plants (reed plant species) (Jørgensen et al. 2012). But such species were not present at our study site.

In addition to the above mentioned negative correlation between WT and N<sub>2</sub>O emissions, a positive relationship between soil (and water) temperature and N<sub>2</sub>O emissions was found for the non-flooded peat site (Table 3). It is known from other studies (e.g. Silvan et al. 2002) that microbial processes (such as denitrification) are favoured by warm conditions and falling WT. This framework may support NO<sub>3</sub><sup>-</sup> formation via nitrification processes, causing high N<sub>2</sub>O release during summer at simultaneously wet conditions. However, increased N<sub>2</sub>O emissions during freeze–thaw-cycles as shown e.g. by Jungkunst et al. (2006), could not be confirmed here. Moreover, a

significant positive correlation between N<sub>2</sub>O release and DOC concentrations of the peat soil water was observed at the non-flooded peat site (Table 3). Since most denitrifying bacteria couple organic C oxidation and  $NO_3^-$  oxidation to gain energy (Knowles 1982), it is obvious that a high supply of readily biodegradable organic C would favour denitrification. The impact of a high availability of organic C on N<sub>2</sub>O release was already shown in other studies (e.g. Eickenscheidt et al. 2014). The positive relationship between N<sub>2</sub>O emission and dissolved organic N (DON) concentrations of peat soil water at the non-flooded peat site furthermore indicates a stimulation of microbial activity due to increasing amounts bioavailable N (Glatzel et al. 2006).

Nitrate contents of the peat soils at both peat sites were low and showed no significant differences among each other. Furthermore no differences in NO<sub>3</sub><sup>-</sup> contents of peat soil and peat soil water during the study period could be identified (unpublished data), which may indicate an enhanced NO<sub>3</sub><sup>-</sup> availability via nitrification. We therefore suggest that NO<sub>3</sub><sup>-</sup> mainly originates from atmospheric N deposition. This assumption is supported by the atmospheric NO<sub>3</sub><sup>-</sup> deposition of 18 kg N ha<sup>-1</sup> a<sup>-1</sup> observed at the study site (Tauchnitz et al. 2010). Hill et al. (2016) also found a positive correlation between potential denitrification and atmospheric NO<sub>3</sub><sup>-</sup> input for a bog watershed. Another pathway might be the NO<sub>3</sub><sup>-</sup> transport via slope water into the bog (Tauchnitz et al. 2010).

# Evolution of $CH_4$ and $N_2O$ emission in the course of rewetting

The annual CH<sub>4</sub> emission accounted for  $68 \pm 60$  and  $505 \pm 414$  kg C ha<sup>-1</sup> a<sup>-1</sup> at the non-flooded and flooded peat site, respectively. Thus, a global warming potential over 100 years (GWP100) of 1428 (non-flooded site) and 10605 kg CO<sub>2</sub>-eq. ha<sup>-1</sup> a<sup>-1</sup> (flooded site) can be derived considering a 21-fold higher global warming potential of CH<sub>4</sub> compared to CO<sub>2</sub> (IPCC 1996). The results are in line with previous studies of rewetted temperate peatlands showing a wide range of -0.75 to 3525 kg C ha<sup>-1</sup> a<sup>-1</sup> (Meyer et al. 2001; Hahn et al. 2015) with the highest CH<sub>4</sub> release observed in a flooded coastal fen in Northeast Germany (Hahn et al. 2015).

At the permanently flooded peat site CH<sub>4</sub> release remained high during whole study period but revealed a small decreasing trend within five years after flooding. Similar high CH<sub>4</sub> emissions, recently after flooding were found in fens in Northeast and Northwest Germany where CH<sub>4</sub> was presumably emitted from a newly formed litter layer derived by dead plant residues (Meyer et al. 2001; Günther et al. 2015; Hahn et al. 2015). In contrast, Drösler (2005) showed markedly lower CH<sub>4</sub> fluxes of a recently flooded bog heathland in South Germany which in part remained below common CH<sub>4</sub> emissions of natural bog sites. After Höper et al. (2008) degradation of fen mires after flooding occurs in three different phases with different characteristics. The first initial phase is characterised by extremely high CH<sub>4</sub> emissions associated with a low net CO<sub>2</sub> uptake, resulting in a negative climate effect. In the second phase a strong reduction of CH<sub>4</sub> emissions occurs accompanied by an increase of CO<sub>2</sub> uptake and thus, a slightly positive climate effect. Whereas in the third phase low CH<sub>4</sub> release and low net  $CO_2$  uptakes can be expected (Asaeda et al. 2002). Until yet little is known about the duration of those phases because most rewetting studies are based on data gathering shortly after rewetting. Beyer and Höper (2015) observed high CH<sub>4</sub> emissions with interannual variations at two flooded sites of a bog in Northwest Germany even 30 years after rewetting. Drösler (2005) reported that after 10 years of renaturation the optimal conditions for low greenhouse gas emissions had not been achieved. Besides that, Günther et al. (2014) found that after 15 years of rewetting the net greenhouse gas balance of a fen in Northeast Germany was similar to those of pristine fens. Note that the present study represents the initial phase of rewetting.

In contrast to the permanently flooded peat site, a small increasing trend of  $CH_4$  emissions was observed at the non-flooded site with fluctuating WT, associated with an increasing WT and observed dieback of plants, which was also reported by Hahn et al. (2015).

Different approaches were discussed to mitigate  $CH_4$  emissions of flooded fen peatlands. When keeping the WT on a constant level, with a threshold value for the mean annual water level of about 10 cm below the soil surface, high  $CH_4$  emissions during rewetting could be prevented (Höper et al. 2008). Other wetland studies have demonstrated that an aerobic layer of 20–30 cm is sufficient to ensure low  $CH_4$  emissions (Koebsch et al. 2013) which are, however, unsuitable for establishing *Sphagnum* 

species. Zak et al. (2015) suggested the removal of upper degraded peat layers in order to minimize nutrient availability. As another possibility with the same goal biomass harvesting, which could be used then for energetic purposes, was proposed (Günther et al. 2015).

The annual N<sub>2</sub>O emissions of the permanently flooded and non-flooded peat site accounted for  $-0.03 \pm 0.1$  and  $1.6 \pm 1.5$  kg N ha<sup>-1</sup> a<sup>-1</sup>, respectively. Thus, a GWP100 of -9.3 and 496 kg CO<sub>2</sub>eq. ha<sup>-1</sup> a<sup>-1</sup> can be expected when considering a 310-fold higher GWP of N<sub>2</sub>O compared to CO<sub>2</sub> (IPCC 1996). The results are in line with previous studies of restored temperate peatlands showing a range of -0.42 to 3.2 kg N ha<sup>-1</sup> a<sup>-1</sup> (e.g. Meyer et al. 2001; Beyer and Höper 2015) with highest N<sub>2</sub>O release reported for rewetted fens.

At the permanently flooded peat site no significant trend in N<sub>2</sub>O emission was observed during the whole study period (five years after rewetting started). Gas measurements started one year after rewetting already at permanent water saturation, which resulted in negligible N<sub>2</sub>O emissions. These results were confirmed by other rewetting studies who reported very low N<sub>2</sub>O fluxes and even a net sink function of flooded peatlands (Meyer et al. 2001; Drösler 2005; Günther et al. 2015). The non-flooded peat site with fluctuating WT also showed no significant trend in N<sub>2</sub>O emission throughout the study period. Nevertheless, annual differences with significantly lower N<sub>2</sub>O release rates towards the end of study could be observed, most likely due to less intensive WT fluctuations and generally higher WT levels. Particularly in the first two years, parallel to the progressing rewetting, N<sub>2</sub>O emissions reached their maximum (up to 4178 µg N  $m^{-2} d^{-1}$ ), associated with a significantly decreasing WT during summer. These high emissions did not occur as strong three years after rewetting. However, one significant N2O outlier with a release rate of 11454  $\mu$ g N m<sup>-2</sup> d<sup>-1</sup> could be observed on 25th of July 2013 (Fig. 3), which was presumably caused by optimal conditions for N<sub>2</sub>O formation, i.e. high soil temperatures (15.2 °C in 5 cm depth) combined with a WT just slightly below the peat soil surface (-2 cm)and a weak precipitation event (4 mm) shortly before gas measurements.

A weak negative trend of  $N_2O$  emissions at the nonflooded peat site with lower and fluctuating WT was observed since 2012 (Fig. 3). Drösler (2005) showed lower N<sub>2</sub>O release rates of maximal 672  $\mu$ g N m<sup>-2</sup>  $d^{-1}$  at a bog in South Germany flooded parallel to the gas measurements. However, the mean water table was thereby 44.5 cm above the peat soil surface with low WT fluctuations. Meyer et al. (2001) reported N<sub>2</sub>O emissions near zero and even an uptake of N<sub>2</sub>O at a recently flooded (parallel to gas measurements) shallow grassland fen in Northwest Germany with a maintained WT of -10 cm. At an also lately, but less rewetted site (WT -30 cm below soil surface) they observed slightly higher N<sub>2</sub>O emissions. Also Augustin and Joosten (2007) described a rewetted grassland fen in Northeast Germany as a weak source of N2O  $(0.7 \text{ kg N ha}^{-1} \text{ a}^{-1})$ . In general, it is well known that frequently alternating wet-dry-cycles may enhance N<sub>2</sub>O emissions (e.g. Harrison-Kirk et al. 2013). Concerning the impact of N<sub>2</sub>O emissions during rewetting a fast adjustment of permanent high WT near the peat soil surface (i.e. a permanent flooding) should be preferred. This will have, however, negative consequences with respect to CH<sub>4</sub> release which will then significantly increase.

# Conclusions

The present study showed an increasing trend of CH<sub>4</sub> release and decreasing N2O emissions during rewetting. Permanent flooded conditions caused the highest CH<sub>4</sub> release. Thus, the present results reveal that not only flooded fens with high nutrient availability of decomposed plant litter but also bog can contribute to very high CH<sub>4</sub> emissions when kept under permanently flooded conditions. Approaches to mitigate CH<sub>4</sub> emissions from rewetted peatlands as discussed in literature (e.g. removal of upper degraded peat soil layer to minimize nutrient availability or biomass harvesting) are not realizable in nature reserves as studied here. Also the maintenance of a constant WT near 10 cm below the peat soil surface appears to be practically unfeasible due to spatial and seasonal variations at the large scale.

The main objective in the presented rewetting program was the recovery of ecological functions by establishing peat forming plants (*Sphagnum*) and hence, the long-term C sink function of the peatland. Present results have revealed that five years after rewetting no decreasing trend of  $CH_4$  emissions were detected. It could be expected that GHG emissions

will decrease in the long term after rewetting. However, in order to generate a better understanding of the functionality of restored peatlands and to improve the planning process of rewetting approaches long-term GHG emission monitoring and the development of vegetation patterns after rewetting are required. Furthermore, the impact of vegetation structure and quality of dissolved organic carbon on  $CH_4$  release should be addressed more in detail in further research.

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