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Special issue article**Annual balances of CH₄ and N₂O from a managed fen meadow using eddy covariance flux measurements**P. S. KROON^{a,b}, A. P. SCHRIER-UIJL^c, A. HENSEN^a, E. M. VEENENDAAL^c & H. J. J. JONKER^b

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Summary

Annual terrestrial balances of methane (CH₄) and nitrous oxide (N₂O) are presented for a managed fen meadow in the Netherlands for 2006, 2007 and 2008, using eddy covariance (EC) flux measurements. Annual emissions derived from different methods are compared. The most accurate annual CH₄ flux is achieved by gap filling EC fluxes with an empirical multivariate regression model, with soil temperature and mean wind velocity as driving variables. This model explains about 60% of the variability in observed daily CH₄ fluxes. Annual N₂O emissions can be separated into background emissions and event emissions due to fertilization. The background emission is estimated using a multivariate regression model also based on EC flux data, with soil temperature and mean wind velocity as driving variables. The event emissions are estimated using emission factors. The minimum direct emission factor is derived for six fertilization events by subtracting the background emission, and the IPCC default emission factor of 1% is used for the other events. In addition, the maximum direct emission factors are determined for the six events without subtracting the background emission. The average direct emission factor ranges from 1.2 to 2.8%, which is larger than the IPCC default value. Finally, the total terrestrial greenhouse gas balance is estimated at 16 Mg ha⁻¹ year⁻¹ in CO₂-equivalents with contributions of 30, 25 and 45% by CO₂, CH₄ and N₂O, respectively.

Introduction

Peatland ecosystems cover approximately 3% of the global land surface and have evolved as globally important sinks of atmospheric carbon dioxide (CO₂) since the last ice age. This is the result of their ability to accumulate more organic matter through photosynthesis than is released through respiration, mainly as a result of high water tables and therefore anaerobic conditions. The carbon (C) accumulated in peatlands is equivalent to almost half the total atmospheric content (Drösler *et al.*, 2008). Peatlands have helped to remove significant amounts of CO₂ from the atmosphere over the last 10 000 years. However, peatlands are not just acting as a sink for CO₂. The wet and anaerobic conditions can lead to formation of the important greenhouse gas methane (CH₄).

Over the last few centuries, many peatland areas have been converted into agricultural land by artificially lowering the water table. As a result peatlands have become a strong source of CO₂ and nitrous oxide (N₂O) due to an increase in peat oxidation and fertilizer application (Schothorst, 1977; Langeveld *et al.*, 1997). Peatland restoration by elevating the water level and decreasing the agricultural intensity may return peatland areas to a sink of CO₂ and also modify the emissions of CH₄ and N₂O (e.g. Van den Bos *et al.*, 2003). All three greenhouse gases should thus be taken into account to assess the total effect of restoration on the greenhouse gas (GHG) balance.

In order to compile the full GHG balance and to understand the processes that affect this balance, long-term (multiple-year) measurements are needed covering all three species. There are only a few studies published with annual records of CO₂, CH₄ and N₂O in peatlands drained for agriculture (e.g. Maljanen *et al.*, 2003; Regina *et al.*, 2004, 2007; Veenendaal *et al.*, 2007). Most

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studies only assess annual emissions of CO₂ (e.g. Veenendaal *et al.*, 2007), and the published annual estimates of CH₄ and N₂O have uncertainties that can exceed 50% (e.g. Flechard *et al.*, 2007a). The relatively large uncertainties in CH₄ and N₂O are due to a combination of complexity of the source (i.e. spatial and temporal variation), limitations in the measurement equipment and the methodology used to quantify the emissions. High-frequency micrometeorological methods are good candidates for determining integrated emission estimates on a hectare scale that also have continuous time coverage. Instrumentation has recently been proven to be suitable for CH₄ and N₂O eddy covariance (EC) flux measurements (e.g. Eugster *et al.*, 2007; Kroon *et al.*, 2007; Neftel *et al.*, 2007; Hendriks *et al.*, 2008).

The main objective of this study is to obtain annual terrestrial CH₄ and N₂O balances from an intensively managed site on peat grassland in the Netherlands over 2006, 2007 and 2008 using EC flux measurements. The terrestrial emission estimates include emissions from fields, ditches and ditch edges. Different methods are compared to estimate the annual emissions. The magnitude and the uncertainty in the annual terrestrial CH₄ emission measured by EC are compared with the annual emission based on static chamber measurements (Schrier-Uijl *et al.*, 2009a). In addition, the direct emission factor EF₁ (ratio of emitted N₂O-N to applied nitrogen (N)) is compared with the default IPCC value of 1% (IPCC, 2006). Finally, a first estimate of the total terrestrial GHG balance is made using the CH₄ and N₂O balances derived from this study and the CO₂ annual balance given in Veenendaal *et al.* (2007). This GHG balance is compared with the terrestrial GHG emissions measured at other peatlands.

Experimental site and climatic conditions

The measurements were performed at an intensively managed dairy farm area. This farm is located at Oukoop near the town of Reeuwijk in the Netherlands (52°02'11"N, 4°46'49"E). The site has peat soil with a clayey peat or peaty clay surface layer of about 0.25 m on 12 m eutrophic peat deposits. Average C and N contents in the top 0.20 m of the soils are 24 and 2.4%, respectively (Veenendaal *et al.*, 2007). Ryegrass (*Lolium perenne*) is the most dominant grass species, with often co-dominant rough bluegrass (*Poa trivialis*) (Veenendaal *et al.*, 2007). The site consists of three main landscape elements according to microtopography and soil moisture conditions: permanently water-filled ditches, almost saturated ditch edges and the relatively dry field area. Ditches, ditch edges and fields account for 16, 5 and 79% of the average footprint area of the EC flux system (Schrier-Uijl *et al.*, 2009b). The contribution over the three different landscape elements is approximately equal for all wind directions. The mean elevation of the polder is between 1.6 and 1.8 m below New Amsterdam Reference water level (NAP; also referred to as sea level). The ditch water level in the polder is being kept at -2.39 m NAP in winter and -2.31 m NAP in summer (Veenendaal *et al.*, 2007). The groundwater depth varies from 0.70

Table 1 Climatic and management characteristics of the Oukoop site in the Netherlands

	2006	2007	2008
Mean annual temperature at 10 m / °C ^a	11.1	11.1	10.6
Mean WFPS ^b at 10 cm depth / %	83	91	88
Mean WFPS ^b at 30 cm depth / %	97	98	99
Annual precipitation / mm ^a	767	1087	736
Cow manure application / kg N ha ⁻¹	253	262	414 ^c
Artificial fertilizer application / kg N ha ⁻¹	100	95	70

^aTemperature and precipitation data were made available by KNMI.

^bWFPS: water-filled pore space.

^cFarmer stopped the agricultural activities and emptied slurry storage.

to 0.15 m below field level and perched water tables occur after heavy rain.

Cow manure and artificial fertilizer were applied about two to six times a year in the period from February to October. There were four harvest events each year. In 2008 the fertilization protocol was adapted from October because the farmer stopped the agricultural activities at this peatland. A summary of the main climatic and management characteristics of the site is given in Table 1.

Instrumentation and methodology

Instrumentation

Two separate EC systems were used; one was used for CO₂ and latent heat fluxes (Veenendaal *et al.*, 2007), and the other one, with a quantum cascade laser (QCL), for CH₄ and N₂O fluxes (Kroon *et al.*, 2007). Both masts of 3 m height were positioned about 5 m apart in the middle of the field. Terrain around the towers was flat and free of obstruction for at least 600 m in all directions, except for the 2 × 2 × 2 m container positioned downwind in which the QCL spectrometer was placed. CO₂ and latent heat fluxes were derived from an EC system consisting of a Campbell SCAT C3 sonic anemometer (Campbell Scientific, Logan, UT, USA) and a Licor 7500 open-path infrared gas analyser (LICOR, Lincoln, NE, USA). Wind speed, air temperature and CH₄ and N₂O concentrations were measured with a system consisting of a three-dimensional sonic anemometer and a QCL spectrometer (model QCL-TILDAS-76, Aerodyne Research Inc., Billerica, MA, USA).

Two sonic anemometer types were used, a WMPRO (Gill Instruments, Lymington, UK) from 26 October 2007 to 9 July 2008, and during the rest of the measurement period an R3 (Gill Instruments, Lymington, UK). The sonic anemometer data and the QCL spectrometer output were logged using the RS232 output and processed using a data acquisition program developed at ECN, following the procedures of McMillen (1988). A more detailed explanation of this measurement set-up can be found in Kroon *et al.* (2007).

Soil measurement sensors included soil heat flux plates (HPF01, Campbell Scientific), soil temperature sensors at depths of 0.02, 0.04, 0.08, 0.16 and 0.32 m (Campbell Scientific) and soil moisture probes by volume at depths of 0.10, 0.20 and 0.30 m

(Theta probes ML 2x; Delta T Devices, Burwell, UK). Additional micrometeorological observations were used from the Cabauw site, which is located about 14 km from Oukoop. These data were made available by the Royal Netherlands Micrometeorological Institute (KNMI).

Methodology

Emission measurements. The expression for the net ecosystem exchange of CH₄ (F_{CH_4}) and N₂O (F_{N_2O}) is derived from the tracer conservation equation (e.g. Aubinet *et al.*, 2000). Assuming horizontal homogeneity and a flat terrain within the averaging time of 30 minutes, the net ecosystem exchange F_{wc} consists of two contributions, the storage term St_c and the eddy covariance flux term EC_{wc} , and is given by:

$$\underbrace{\int_0^h \overline{S_c} dz}_{F_{wc}} = \underbrace{\frac{\overline{c_i} - \overline{c_{i-1}}}{T_{av}} h}_{St_c} + \underbrace{\overline{w'c'}|_{z=h}}_{EC_{wc}} \quad (1)$$

where h is the measurement height in m, S_c the source/sink term in ppb s⁻¹, c the gas concentration in ppb (nmol mol⁻¹), i the flux number, T_{av} the averaging time in s and w the vertical wind velocity in m s⁻¹. We calculate the storage term St_c using the average values of CH₄ and N₂O at 3 m height over each 30 minutes period.

We determine the EC flux EC_{wc} by:

$$EC_{wc} = \chi_{cal} \chi_{res} EC_{wc}^{meas} + \chi_{cal} \chi_{Webb}, \quad (2)$$

with EC_{wc}^{meas} the measured EC flux in ppb m s⁻¹, χ_{cal} the calibration correction, χ_{res} the frequency response correction and χ_{Webb} the Webb-correction. More information about the correction process is given in Kroon *et al.* (2010a,b) and in the supporting information in Appendix S1 in the online version of this paper.

Annual terrestrial balances. The annual terrestrial emission of CH₄, E_{CH_4} , in kg CH₄ ha⁻¹ year⁻¹ is given by:

$$E_{CH_4} = \left(\frac{16}{12}\right) \left(\frac{1 \times 10^4}{1 \times 10^{12}}\right) \sum_{i=1}^N F_{CH_4_i} T_{av}, \quad (3)$$

where F_{CH_4} is the emission in ng C m⁻² s⁻¹, the factor (16/12) is the conversion from kg C to kg CH₄, the factor 1×10^4 is used for the conversion from m⁻² to ha⁻¹ and the factor 1×10^{12} for the conversion from nanograms to kilograms, and N represents the number of time slots within a year that is equal to T_{year}/T_{av} .

Below we distinguish three methods to deal with data gaps in CH₄ flux data. Method 1 is based on the average CH₄ EC flux where the average flux is extrapolated to an annual value, in other words the data gaps are filled with the average emission. Method 2 uses all available EC flux measurements and the remaining data gaps are filled by an empirical multivariate regression. Method 3 is

totally based on the empirical regression. We derive this regression using the statistical method ANOVA.

The annual terrestrial emission of N₂O E_{N_2O} in kg N₂O ha⁻¹ year⁻¹ consists of a background emission level on which emission peaks are superimposed. These peaks are related to manure and fertilizer application (direct agricultural soil emission). The background emission level derived from the EC data includes a component that is caused by atmospheric N deposition. Outside the measurement fetch indirect N₂O emission is caused by nitrogen emitted from the field as NH₃ or leached to the ditches–lakes–rivers and finally to the sea. These two processes should be taken into account in the full terrestrial N₂O balance.

Consequently, the annual emission of N₂O E_{N_2O} in kg N₂O ha⁻¹ year⁻¹ is given by:

$$E_{N_2O} = E_{EC} + E_1 + E_d, \quad (4)$$

with E_{EC} the emissions measured by the EC flux technique, E_1 the indirect emissions due to leaching and runoff and E_d the indirect emissions due to deposition. The contribution E_1 is estimated using the IPCC methodology (IPCC, 2006):

$$E_1 = \left(\frac{44}{28}\right) (N_{synth} + N_{cow}) \text{Frac}_{leach} \text{EF}_5, \quad (5)$$

where the factor (44/28) is used for the conversion from kg N to kg N₂O and N_{synth} the annual amount of synthetic fertilizer in kg N ha⁻¹ year⁻¹, N_{cow} the annual amount of applied cow manure in kg N ha⁻¹ year⁻¹, $\text{Frac}_{leach} = 0.3$ and $\text{EF}_5 = 0.75\%$. The contribution E_d is partly based on the IPCC methodology (IPCC, 2006):

$$E_d = \left(\frac{44}{28}\right) \left(\left(\sum_{m=1}^{N_{event}} N_{fert_m} (1 - k_m) \right) - \overline{N}_d \right) \text{EF}_4, \quad (6)$$

where N_{event} is the number of fertilization events, N_{fert} is the amount of applied fertilizer in kg N ha⁻¹, k is equal to 0.9 for synthetic fertilizer or 0.8 for organic fertilizer (IPCC, 2006), \overline{N}_d is the average annual N deposition in the Netherlands, which equals to 30.8 kg N ha⁻¹ year⁻¹ (Ruiter *et al.*, 2006), and $\text{EF}_4 = 1\%$ (IPCC, 2006).

The annual emission of N₂O, E_{EC} , can be derived in a similar way to the annual emission of CH₄ when no data gaps are available. The annual emission is then given by:

$$E_{EC} = \left(\frac{44}{28}\right) \left(\frac{1 \times 10^4}{1 \times 10^{12}}\right) \sum_{i=1}^N F_{N_2O_i} T_{av}, \quad (7)$$

where F_{N_2O} is the N₂O emission in ng N m⁻² s⁻¹.

However, the data coverage is hardly ever 100%. In that case, we distinguish two contributions in the annual N₂O emission, E_{EC} , the background emissions E_{bgnd} and the emissions due to fertilizer application E_{fert} . The background emission is defined as the N₂O emission that occurs when no fertilizer is applied. We estimate

the background fluxes F_{bgnd} using a multivariate regression model based on all EC fluxes, excluding EC fluxes measured around a management event. This background emission is probably larger than the true background emission (unmanaged situation) because it will include emissions from the accumulated N in the soil due to previous fertilization events. The background emission level also includes the emission due to atmospheric N deposition on the field. Therefore, we use an adapted direct emission factor EF_1^{min} for estimating the direct emissions due to fertilizing application. This factor is calculated by:

$$\text{EF}_1^{\text{min}} = \left(\frac{1 \times 10^4}{1 \times 10^{12}} \right) \frac{\sum_{i=1}^N (F_{\text{N}20_i} - F_{\text{bgnd}_i}) T_{\text{av}}}{k N_{\text{fert}}} \times 100\%, \quad (8)$$

with N the number of time slots within a fertilization event that is equal to $T_{\text{event}}/T_{\text{av}}$, N_{fert} the amount of applied fertilizer in kg N ha^{-1} , and k equal to 0.9 for synthetic fertilizer or 0.8 for organic fertilizer (IPCC, 2006). The factor k is used to account for the amount of N that volatilizes as NH_3 and NO_x (IPCC, 2001, 2006). The total annual N_2O emission E_{EC} is then derived from:

$$E_{\text{EC}} = \left(\frac{44}{28} \right) (E_{\text{bgnd}} + E_{\text{fert}}), \quad (9)$$

where the factor $(44/28)$ is used for the conversion from kg N to $\text{kg N}_2\text{O}$. The background emission E_{bgnd} in $\text{kg N ha}^{-1} \text{ year}^{-1}$ is:

$$E_{\text{bgnd}} = \left(\frac{1 \times 10^4}{1 \times 10^{12}} \right) \sum_{i=1}^N F_{\text{bgnd}_i} T_{\text{av}}, \quad (10)$$

where N represents the number of time slots within a year that equals to $T_{\text{year}}/T_{\text{av}}$ and the fertilizer emission E_{fert} in $\text{kg N ha}^{-1} \text{ year}^{-1}$ is:

$$E_{\text{fert}} = E_{\text{direct}} = \sum_{m=1}^{N_{\text{event}}} \text{EF}_{1_m}^{\text{min}} k_m N_{\text{fert}_m}, \quad (11)$$

where E_{direct} indicates the direct N_2O emissions due to fertilizing and N_{event} is the number of fertilization events.

However, we realize that the emission factor EF_1 used in the inventory reports is higher than EF_1^{min} because the real background emission is between $0 \text{ ng N m}^{-2} \text{ s}^{-1}$ and the measured background emission. To check the reliability of the IPCC default value of 1%, we compare this default value with an emission factor range that is set from EF_1^{min} to EF_1^{max} , where EF_1^{max} is determined by:

$$\text{EF}_1^{\text{max}} = \left(\frac{1 \times 10^4}{1 \times 10^{12}} \right) \frac{\sum_{i=1}^N F_{\text{N}20_i} T_{\text{av}}}{k N_{\text{fert}}} \times 100\%, \quad (12)$$

with N the number of time slots within a fertilization event that is equal to $T_{\text{event}}/T_{\text{av}}$.

In this study, we use three methods to derive the annual terrestrial N_2O emission $E_{\text{N}20}$. Method 1 uses the average measured N_2O flux over the available EC fluxes within a year and this value is extrapolated to an annual E_{EC} value and the indirect annual emissions E_1 and E_d are added. In method 2, we first determine for each day number (i.e. from 1 to 365) the average N_2O emission $\overline{F}_{\text{N}20}$ over the 3 years, while the data gaps are filled by the last available daily average. The annual emission E_{EC} is then calculated using Equation (7) and the contributions E_1 and E_d are again added. Method 3 uses Equation (9) for E_{EC} , where we determine the emission factor EF_1^{min} for each available measured fertilization event with Equation (8) and we use an emission factor of 1% for the remaining fertilization events. The total terrestrial N_2O emission is again obtained by adding E_{EC} to E_1 and E_d . In addition, we compare the direct emission factors with the 1% factor to check whether it is reasonable to use this value for the events that are not available.

We finally derive the total terrestrial GHG emission E_{GHG} in $\text{kg CO}_2\text{-equivalents ha}^{-1} \text{ year}^{-1}$ by:

$$E_{\text{GHG}} = E_{\text{CO}2} + 25E_{\text{CH}4} + 298E_{\text{N}20}, \quad (13)$$

with $E_{\text{CO}2}$ the annual terrestrial CO_2 emission in $\text{kg CO}_2 \text{ ha}^{-1} \text{ year}^{-1}$. The factors 25 and 298 are the global warming potential of CH_4 and N_2O over a 100-year time horizon, respectively, in relation to CO_2 (IPCC, 2007).

Results and discussion

Data coverage

The EC flux measurement set-up was installed for 1 week in February 2006, and from April 2006 to October 2008. The data coverage is 48% over the measurement periods ($n = 20\,957$ 30 minutes EC fluxes); data losses occurred due to problems with the QCL (28%), computer (9%), automatic liquid nitrogen filling system (6%) and sonic anemometer (2%). The relatively high data losses with the QCL were due to it being at the development stage at the beginning of the measurement period. The footprint check and non-steady-state tests result in data rejection of 19 and 23% for CH_4 and N_2O , respectively. We correct the EC fluxes for systematic errors using Equation (2). The total CH_4 and N_2O flux F_{wc} are determined by adding the storage term to the EC flux for each 30-minute period (Equation (1)).

It is often stated that the measured total flux can be underestimated under conditions of low turbulence. A common procedure for CO_2 fluxes is to investigate the effect of the friction velocity u_* on the measured night-time CO_2 fluxes. CO_2 fluxes are then rejected below a threshold u_* for which the average fluxes decrease significantly (e.g. Nieveen *et al.*, 2005; Wohlfahrt *et al.*, 2005; Veenendaal *et al.*, 2007). All these studies were focused on night-time fluxes.

In order to compare the night- and daytime u_* dependence of CH_4 and N_2O fluxes, we select the data with shortwave radiation of 0 W m^{-2} for night-time and shortwave radiation greater than

50 W m⁻² for daytime. The fluxes are additionally divided into groups, with air temperature between 0 and 10°C, and 10 and 20°C. The fluxes in each group are sorted by u_* and pooled into groups of 50 fluxes each. This leads to a similar behaviour for night- and daytime u_* dependence of CH₄ and N₂O (Figure 1). The fluxes show a marked decrease for $u_* < 0.07$ m s⁻¹, which is comparable with the threshold $u_* = 0.1$ m s⁻¹ of CO₂ fluxes measured at the same site (Veenendaal *et al.*, 2007). For $u_* > 0.45$ m s⁻¹, the average CH₄ and N₂O EC fluxes show a small increase that is not caused by higher temperatures.

When dividing the fluxes into air-temperature classes of 2°C and three soil moisture classes, a similar pattern is found for which the fluxes decrease for $u_* < 0.07$ m s⁻¹ and increase for $u_* > 0.45$ m s⁻¹. Gu *et al.* (2005) suggested that there are two thresholds for CO₂ night-time fluxes, u_{*L} and u_{*H} , delimiting three zones in the u_* range, with a decrease of flux below u_{*L} and an increase above u_{*H} . Fluxes below u_{*L} would be lower due to storage increase and fluxes above u_{*H} are higher due to pressure-pumping effects (Gu *et al.*, 2005; Flechard *et al.*, 2007b). Flechard *et al.* (2007b) also showed that the addition of soil storage change to the flux removed a large fraction of the u_* dependence, which suggests that no u_* filtering should be applied to avoid possible double-counting. Therefore no u_* filtering is applied on the EC flux data set in this study.

Another point of concern is related to the uncertainty in a 30-minute CH₄ and N₂O EC flux, which can be larger than the flux itself (Kroon *et al.*, 2010a). This uncertainty is mainly caused by the random one-point uncertainty (e.g. Businger, 1986). To reduce the uncertainty, averages over longer time spans than 30 minutes can be used because the random uncertainty decreases with the number of 30-minute EC fluxes. Therefore, we assess the suitability of using daily average emissions instead of 30-minute F_{wc} values. It is justified to use daily average emissions when the data coverage is 100%, when there is no diurnal variation or when the number of night and day fluxes balances.

Several studies investigated the diurnal cycle of CH₄ and N₂O emissions (e.g. Mikkilä *et al.*, 1995; Duan *et al.*, 2005; Hendriks *et al.*, 2008, for CH₄; and, for example, Skiba *et al.*, 1996; Crill *et al.*, 2000; Maljanen *et al.*, 2002 for N₂O). For both gases some studies have observed a diurnal cycle and some studies have not. For CH₄, the diurnal pattern is maybe dependent on the vegetation type (e.g. Mikkilä *et al.*, 1995; Duan *et al.*, 2005). In this study, we check the diurnal cycles using the whole data set (Figure 2). A diurnal pattern is observed for CH₄, with larger CH₄ fluxes in the late afternoon, while no clear pattern is found for N₂O. Consequently, we evaluate further the diurnal variation of N₂O for days with high emissions or high temperatures; however, we still did not observe a clear diurnal pattern. The average soil

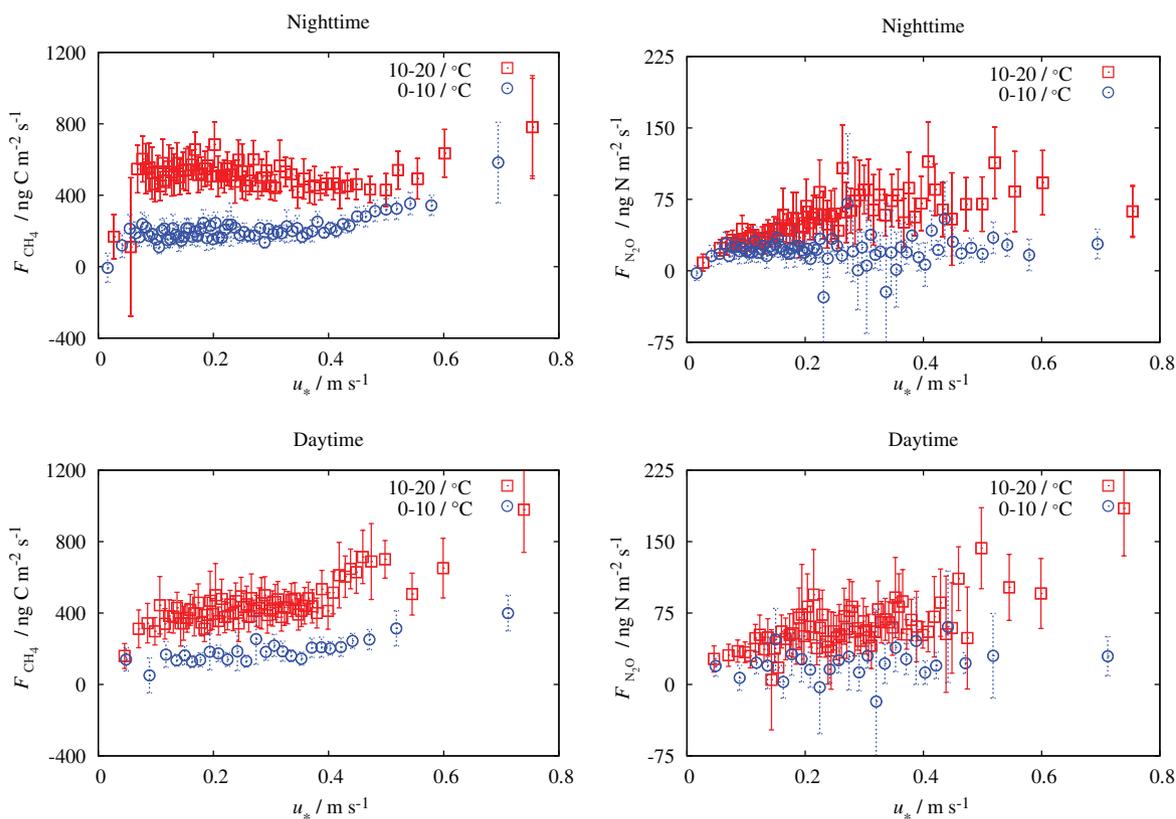


Figure 1 Average CH₄ fluxes (left) and N₂O fluxes (right) as function of friction velocity u_* . Data are split into night- and daytime, and into two temperature classes. The data are pooled into groups of 50 fluxes each. Error bars show the uncertainty in the average CH₄ and N₂O flux given by the standard error, $SE = 2\sigma/\sqrt{N}$.

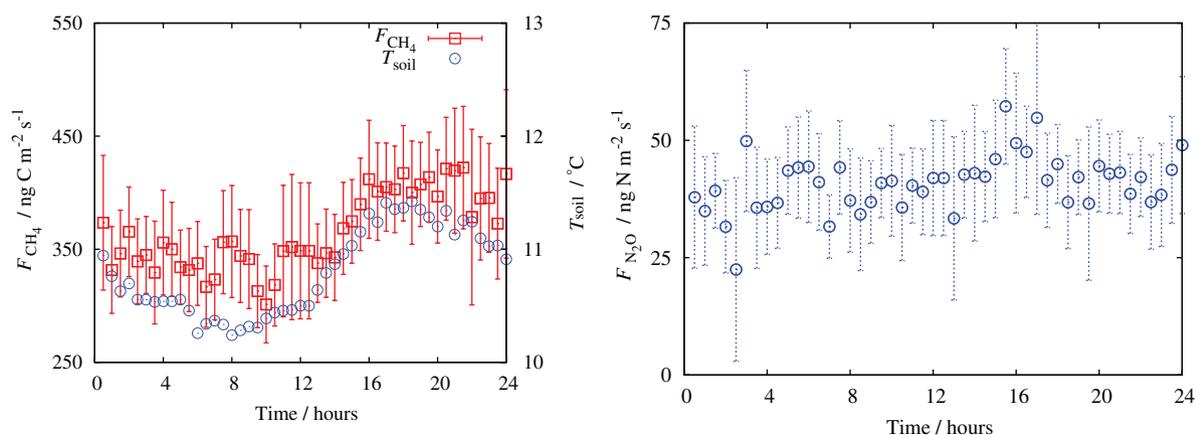


Figure 2 Diurnal pattern of CH₄ fluxes and soil temperature (left) and N₂O fluxes (right) at a peat area in the Netherlands. Each point represents the average of about $n = 320$ 30-minute EC fluxes for CH₄ and N₂O, and the average of about 1000 30-minute T_{soil} values. The error bars indicate the uncertainty in the average given by the standard error, $\text{SE} = 2\sigma/\sqrt{N}$.

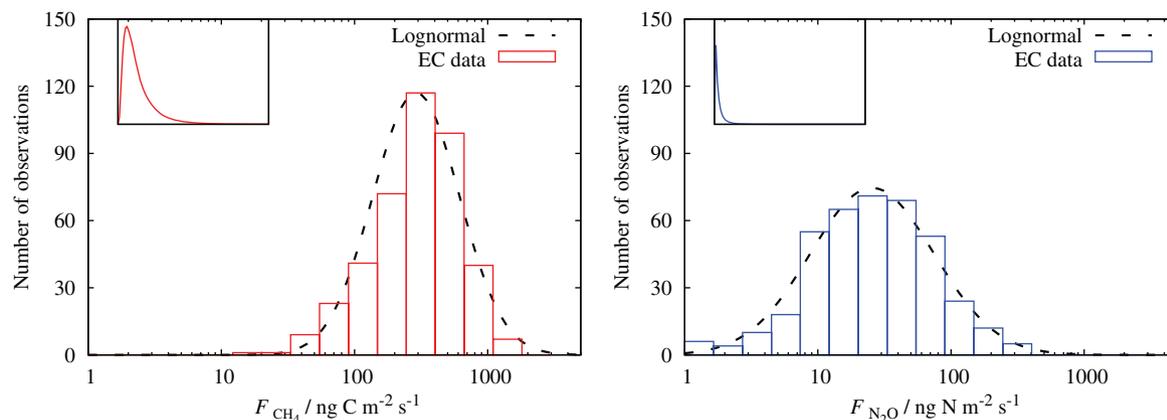


Figure 3 Distribution of daily average CH₄ (left) and N₂O fluxes (right) with the lognormal distribution. The insert shows the shape of the distribution on a linear scale.

temperature, in Figure 2, shows the same diurnal pattern as CH₄, supporting the suggestion that soil temperature is an important driver for CH₄ fluxes, which is also found in other studies (e.g. Hendriks *et al.*, 2007). The absence of a clear diurnal pattern for N₂O allows for the use of daily average values for N₂O for further analyses. For CH₄, however, daily averages can only be used when balanced numbers of day and night observations are available, which is valid for this study.

Consequently, we calculate daily averages for days with more than 12–30-minute EC fluxes balanced over day and night, which lead to 119, 106 and 188 daily CH₄ fluxes and 117, 102 and 186 for N₂O in 2006, 2007 and 2008, respectively. The average daily CH₄ and N₂O flux and standard deviation are 368 and 251 ng C m⁻² s⁻¹, and 42 and 52 ng N m⁻² s⁻¹, respectively. The fluxes of both gases are approximately lognormally distributed (Figure 3). The geometric average and geometric standard deviation are defined by $\mu_{\text{Geo}} = e^{\mu}$ and $\sigma_{\text{Geo}} = e^{\sigma}$, where μ and σ are the mean and standard deviation of the logarithms of

the observed flux values, which resemble a normal distribution. The geometric average indicates the emission that occurred most often. When omitting a small number of negative values (<1%) from the data set, the geometric average fluxes in the lognormal distribution are 291 ng C m⁻² s⁻¹ and 26 ng N m⁻² s⁻¹ for CH₄ and N₂O, respectively. In all further analyses, we do not omit the negative numbers because uptake fluxes of CH₄ and N₂O could really occur.

Annual terrestrial CH₄ balance

A regression model is often used to fill the data gaps and to derive annual balances. Several studies have used a regression with temperature as the only driving variable, explaining about 90% of the variation (Hargreaves *et al.*, 2001) or more commonly lower values of about 30% (Hendriks *et al.*, 2007). The uncertainty in annual emissions based on these models is sometimes even larger than 50%.

In this study, we perform a step-wise multivariate regression analysis using the daily CH₄ values for which data are available for all possible regression parameters ($n = 265$). The correlation of F_{CH_4} and $\ln(F_{\text{CH}_4})$ is tested for air temperature (T_{air}), soil temperature (T_{soil}), rain intensity (R), wind velocity (U), air pressure (P_{air}) and soil moisture (θ). We do not include water table information because Schrier-Uijl *et al.* (2009a) showed that the CH₄ emissions did not correlate with the (actively managed) water table at our study site. The Pearson correlations (r) appear to be somewhat more significant for $\ln(F_{\text{CH}_4})$ than for F_{CH_4} with the different variables (Table 3). Significant correlations ($P < 0.05$) are found for $\ln(F_{\text{CH}_4})$ with T_{air} , T_{soil} , P_{air} , θ and R . In these cases r values are larger than those derived from static chamber measurements at the same site (Schrier-Uijl *et al.*, 2009a). The best regression is found by including T_{soil} and U , while adding other variables does not improve the regression. The regression of $\ln(F_{\text{CH}_4})$ versus T_{soil} and U explains about 60% of the variability in the observed daily CH₄ values ($R^2 = 0.63$; $P < 0.001$). Adding soil moisture does not improve the regression, which is a remarkable result. However, this can be explained by the fact that the effect of temperature on CH₄ fluxes is larger than the effect of soil moisture. In addition, the parameters soil moisture and temperature are significantly anti-correlated (r for soil moisture at 30 cm depth and T_{soil} is -0.82 and $P < 0.001$). In other words, the CH₄ fluxes are higher for higher soil temperatures, but at the same time higher soil temperatures lead to lower soil moisture contents.

A correlation between CH₄ fluxes and U has also recently been reported by, for example, Wille *et al.* (2008) and Schrier-Uijl *et al.* (2009b). They stated that higher CH₄ fluxes occur from water bodies at high wind velocities. These emissions are maybe due to the turbulence-induced ebullition. In addition, Frohling & Crill (1994) indicated that ebullition could be triggered by changes of air pressure. This could explain the correlation between air pressure and CH₄ fluxes. Next to the enhanced emissions from water, enhanced fluxes from the field could occur at higher mean wind velocities due to the soil storage effect discussed in this study.

The empirical regression of F_{CH_4} against T_{soil} and U is given by:

$$F_{\text{CH}_4} = \exp(3.796(\pm 0.105) + 0.136(\pm 0.006)T_{\text{soil}} + 0.113(\pm 0.016)U), \quad (14)$$

with F_{CH_4} in $\text{ng C m}^{-2} \text{s}^{-1}$, T_{soil} at 4 cm depth in $^{\circ}\text{C}$ and U at 3 m in m s^{-1} . In further research, the regression could possibly be improved by adding management information. For example, Kroon *et al.* (2007) and Schrier-Uijl *et al.* (2009a) showed that CH₄ emissions are larger after cow manure application. The regression-based emissions F_{CH_4} and their uncertainties are represented together with the daily measured emissions and their uncertainties in Figure 4.

The three methods to estimate the annual CH₄ balances (see methodology section) result in average annual emissions over the period 2006 to 2008 of 157 ($\pm 33\%$), 165 ($\pm 17\%$) and

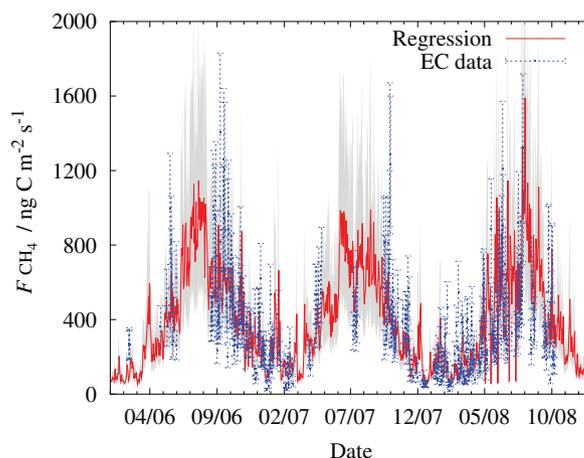


Figure 4 Measured and modelled CH₄ emissions in Oukoop in 2006, 2007 and 2008. The model is a multivariate regression based on EC flux measurements. Emissions are given together with their uncertainty; the grey surface represents the uncertainty range of the empirical regression.

Table 2 Annual terrestrial CH₄ emission in $\text{kg CH}_4 \text{ ha}^{-1} \text{ year}^{-1}$ and its relative uncertainty, Oukoop, the Netherlands

	2006	2007	2008	Average
Method 1 ^a	194 ($\pm 54\%$)	140 ($\pm 63\%$)	138 ($\pm 53\%$)	157 ($\pm 33\%$)
Method 2 ^b	176 ($\pm 30\%$)	169 ($\pm 31\%$)	149 ($\pm 26\%$)	165 ($\pm 17\%$)
Method 3 ^c	172 ($\pm 37\%$)	166 ($\pm 37\%$)	155 ($\pm 37\%$)	164 ($\pm 21\%$)
Method 4 ^d	203 ($\pm 48\%$)	162 ($\pm 60\%$)	146 ($\pm 60\%$)	170 ($\pm 32\%$)

^aAverage EC flux is extrapolated.

^bBased on EC flux measurements; the remaining data gaps are filled by a multivariate regression model.

^cBased on a multivariate regression model derived from EC flux measurements only.

^dBased on multivariate regression model derived from static chamber measurements only (Schrier-Uijl *et al.*, 2009a).

164 ($\pm 21\%$) $\text{kg CH}_4 \text{ ha}^{-1} \text{ year}^{-1}$ for methods 1, 2 and 3, respectively (Table 2). The terms within brackets denote the relative uncertainties, which are calculated by the methods described in Appendix S1. The three estimates compare well, and these annual estimates are in good agreement with the annual estimate of 170 ($\pm 32\%$) $\text{kg CH}_4 \text{ ha}^{-1} \text{ year}^{-1}$ determined by Schrier-Uijl *et al.* (2009a) using static chamber measurements at the same site. The estimate by method 2 has the smallest uncertainty and has therefore been used in the rest of this study. The uncertainty in the estimate by method 2 is much smaller than the uncertainty in the estimate by static chamber measurements. In addition, much more accurate estimates can be derived from EC flux measurements for measurement sets with larger data coverage. If data coverage above 80% is attained, the uncertainty in the annual balances could be even smaller than 10% (based on Kroon *et al.*, 2010a).

This agricultural peatland is thus a serious source for CH₄ emissions, in contrast to other published studies that indicate

a small source (Regina *et al.*, 2007), or even a sink (Van den Pol-van Dasselaar *et al.*, 1999). The difference can be explained mainly by the ability of the EC flux method to include the CH₄ emission from the ditches and ditch edges at our site. Schrier-Uijl *et al.* (2009a) indicated that about 70% of the emissions are caused by the ditches and ditch edges at our site. This means that only about 50 kg CH₄ ha⁻¹ year⁻¹ is emitted by the field. The remaining difference between the northern peat grassland emissions given in Regina *et al.* (2007) can be explained by the temperature difference. Van den Pol-van Dasselaar *et al.* (1999) indicated an uptake; however, their emission values are only based on field emissions. Consequently, it is important that emissions of ditch and ditch edges are taken into account when they occur within the investigated area.

Annual terrestrial N₂O balance

The annual terrestrial N₂O emission E_{N_2O} and its relative uncertainty are derived from three methods (see methodology section and Appendix S1). We estimate an average annual N₂O emission E_{EC} of 21 (±36%) kg N₂O ha⁻¹ year⁻¹ with method 1 and of 18 (±21%) kg N₂O ha⁻¹ year⁻¹ with method 2. The average annual N₂O emissions E_1 and E_d are both estimated at 1 (±50%) kg N₂O ha⁻¹ year⁻¹. This leads to an average annual terrestrial N₂O emission E_{N_2O} of 23 (±31%) kg N₂O ha⁻¹ year⁻¹ and 20 (±19%) kg N₂O ha⁻¹ year⁻¹ with methods 1 and 2, respectively. For method 3, we first determine the background fluxes, F_{bgnd} , and the emission factors EF_1^{min} (see Equation (9)). The background fluxes are determined using a multivariate regression model based on EC flux data, excluding the EC fluxes around fertilization events. We select the N₂O EC fluxes for which all possible driving variables are available ($n = 252$). The correlations of F_{N_2O} and $\ln(F_{N_2O})$ are tested for air temperature (T_{air}), soil temperature (T_{soil}), rain intensity (R), wind velocity (U), air pressure (P_{air}) and soil moisture (θ) (Table 3). The best linear regression is found by including T_{soil} and U ($R^2 = 0.29$ and $P < 0.001$) and is given by:

$$F_{bgnd} = \exp(1.777(\pm 0.200) + 0.122(\pm 0.012)T_{soil} + 0.063(\pm 0.030)U), \quad (15)$$

with F_{bgnd} in ng N m⁻² s⁻¹, T_{soil} the soil temperature at 4 cm depth in °C and U the mean wind velocity at 3 m in m s⁻¹. The annual background emissions E_{bgnd} and their relative uncertainties are then estimated at 18 (±71%), 17 (±71%) and 17 (±71%) kg N₂O ha⁻¹ year⁻¹ for 2006, 2007 and 2008, respectively, corresponding to an average background N₂O flux F_{bgnd} of about 35 ng N m⁻² s⁻¹. These values are larger than the background emission previously reported in Velthof *et al.* (1997) for peat soils in the Netherlands (5.3 ± 5.2 kg N₂O-N ha⁻¹ year⁻¹). This is explainable because they derived the estimates from unfertilized and non-grazed cut grassland sites.

We determine the emission factors EF_1^{min} for six fertilization events using Equation (8). The factor range is from 0.2 to 2.2% with an average and standard deviation of 1.2 and 0.8%, respectively, which is close to the IPCC default value of 1%. The IPCC default EF_1 value is therefore used for fertilization events for which no data are available. This leads to 5.2 (±50%), 5.3 (±50%) and 4.8 (±50%) kg N₂O ha⁻¹ year⁻¹ for 2006, 2007 and 2008, respectively. The total annual terrestrial N₂O emission estimated by method 3 is then derived from adding the fertilization event emission E_{fert} to the background emission E_{bgnd} , and the indirect emissions due to leaching E_l and deposition E_d (Equations 4–6 and 9). This results in annual terrestrial N₂O emissions E_{N_2O} of 25 (±52%), 24 (±51%) and 24 (±50%) kg N₂O ha⁻¹ year⁻¹ for 2006, 2007 and 2008, respectively. The terms within brackets denote the relative uncertainty in the annual N₂O emission, which is calculated by the algorithms described in Appendix S1. The average emission of method 3 is close to the average annual terrestrial N₂O emission of methods 1 and 2 (Table 4) and the average terrestrial N₂O emission is comparable to reported annual terrestrial emissions of other peat soils in the Netherlands (Langeveld *et al.*, 1997). A representation of the background emission, the annual average emission E_{EC} and the EC flux data is shown in Figure 5.

We cannot compare the annual terrestrial N₂O emission estimates derived from EC flux measurement with static chamber measurements because N₂O emissions were mostly too small to detect with the static chamber method used by Schrier-Uijl *et al.* (2009a,b). They used a closed dark chamber of 25 cm height from which air samples were taken at 1-minute intervals. Each flux measurement consisted of five point-measurements and the

Table 3 Pearson correlations (r) of CH₄ fluxes and N₂O background fluxes, Oukoop, the Netherlands. Fluxes have been measured by the EC flux technique and are correlated against soil temperature (T_{soil} at 4 cm depth), air temperature (T_{air}), atmospheric pressure (P_{air}), windspeed (U), volumetric soil moisture content (θ at 10 cm depth) and daily rainfall (R)

	N	T_{soil} / °C	T_{air} / °C	P_{air} / hPa	U / m s ⁻¹	$\theta_{10\text{ cm}}$ / m ³ m ⁻³	R / mm day ⁻¹
Ln(CH ₄)	265	0.749 ^a	0.688 ^a	-0.280 ^a	0.050	-0.510 ^a	0.193 ^b
CH ₄	265	0.714 ^a	0.648 ^a	-0.189 ^a	-0.065	-0.553 ^a	0.142 ^b
Ln(N ₂ O)	252	0.526 ^a	0.456 ^a	-0.141 ^b	-0.031	-0.401 ^a	0.112
N ₂ O	252	0.394 ^a	0.341 ^a	-0.165 ^a	-0.057	-0.367 ^a	0.130 ^b

^aCorrelation is significant at the 0.01 level.

^bCorrelation is significant at the 0.05 level.

Table 4 Annual terrestrial N₂O emission in kg N₂O ha⁻¹ year⁻¹ and its relative uncertainty, Oukoop, the Netherlands

	2006	2007	2008	Average
Method 1 ^a	30 (±58%)	20 (±57%)	19 (±47%)	23 (±31%)
Method 2 ^b	—	—	—	20 (±19%)
Method 3 ^c	25 (±52%)	24 (±51%)	24 (±50%)	24 (±28%)

^aAverage EC flux is extrapolated and the indirect emissions are added.

^bIntegrating average EC flux of each day number over the three years (i.e. from 1 to 365) while the data gaps are filled by the last available daily average. The indirect emissions are added.

^cBackground emissions are determined by a multivariate regression model based on EC flux data and the emissions due to fertilizing, leaching and deposition are added.

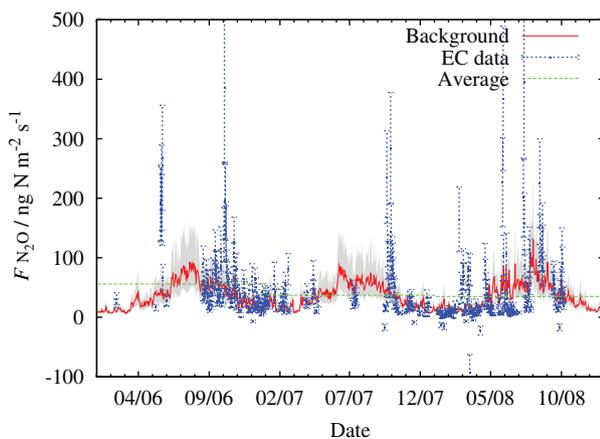


Figure 5 Measured N₂O emissions and modelled background N₂O emissions in Oukoop in 2006, 2007 and 2008. The model is a multivariate regression based on EC flux measurements. They are given together with their uncertainty; the grey surface represents the uncertainty range of the empirical regression. In addition, the average measured EC flux over each year is indicated.

concentrations were analysed by a photo-acoustic field gas monitor (INNOVA 1412 sn, 71-113, ENMO services, Turnhout, Belgium). The minimum detection limit of this static chamber system was

39 ng N m⁻² s⁻¹ and for the EC flux system 6 ng N m⁻² s⁻¹ (Kroon *et al.*, 2007). Thus, the use of this EC flux set-up was a big step forward in obtaining more accurate emission estimates. In addition, more accurate estimates can be obtained when the data coverage increases (Kroon *et al.*, 2010a).

After estimating the annual balances, we investigate in more detail the emission factor EF₁, which is used in the IPCC guidelines for estimating the direct N₂O emissions from agricultural soils. In this study, we calculate the possible EF₁ ranges for six fertilization events ranging from EF₁^{min} to EF₁^{max}. The minimum EF₁ has already been determined and ranges from 0.2 to 2.2%, with an average and standard deviation of 1.2 and 0.8%, respectively. The maximum EF₁ is estimated using Equation (12) and ranges from 0.9 to 4.6%, with an average and standard deviation of 2.8 and 1.9%. Thus, the average EF₁ range is from 1.2 to 2.8%, which is larger than the IPCC default EF₁ value of 1% (IPCC, 2006). Kuikman *et al.* (2006) have also reported a larger EF₁ than 1% for peat soils in the Netherlands, obtaining EF₁ values of 0.55, 1.24 and 3% for sand, clay and peat soils, respectively. This emphasizes the fact that separate parameterizations should be derived for each soil type.

In addition, the emission factors EF₁ should be determined more accurately because the uncertainty in the direct N₂O emission from agricultural soils is a main contributor to the uncertainty in the total national GHG emission estimate (Maas *et al.*, 2008). The uncertainty is large due to large uncertainties in the measured N₂O emissions F_{N_2O} , the background emissions F_{bgnd} and the k factor. We recommend performing simultaneously continuous terrestrial measurements of N₂O and NH₃ at representative agricultural and managed sites at national and international scales. These measurements could be taken using micrometeorological techniques, like the EC flux technique. An overview is given of EF₁^{min}, EF₁^{max} and the micrometeorological properties of the six fertilization events in Table 5.

Annual terrestrial GHG balance

The total terrestrial GHG balance is estimated by Equation (13) and its uncertainty by Equation (A.7) (see Appendix S1).

Table 5 Emission factor range for 1-month periods following six fertilization events, Oukoop, the Netherlands

Date fertilizing	Cow manure N / kg ha ⁻¹	Artificial fertilizer N / kg ha ⁻¹	T_{soil_4} cm / °C	WFPS ₁₀ cm ^a / %	R / mm month ⁻¹	EF ₁ ^{min b} – EF ₁ ^{max c} / %
14 September 2006	55	—	16	85	47	1.9–4.6
15 September 2007	46	—	14	91	68	2.2–4.6
5 February 2008	138	—	3	96	32	0.6–1.0
3 April 2008	46	43	8	95	29	0.2–0.9
19 May 2008	83	27	15	77	48	0.5–1.2
30 June 2008	46	—	16	60	104	1.7–4.2

^aWFPS: water-filled pore space.

^bEF₁^{min} is the factor for direct N₂O emissions where the background emission derived from EC flux measurements is subtracted.

^cEF₁^{max} is the factor for direct N₂O emission where the background emission is set to 0 ng N m⁻² s⁻¹.

We use the annual CH₄ emission derived from method 2 and the annual N₂O emissions derived from method 3. The average annual CH₄ emission is 165 kg CH₄ ha⁻¹ year⁻¹ and the average annual N₂O is 24 kg N₂O ha⁻¹ year⁻¹. The annual terrestrial CO₂ emission, net ecosystem exchange (NEE), is taken from Veenendaal *et al.* (2007), who calculated a value of 134 g C m⁻² year⁻¹ for October 2004 to October 2005, which is equal to 4910 kg CO₂ ha⁻¹ year⁻¹. This leads to an average total terrestrial GHG emission of 16 (±15%) Mg ha⁻¹ year⁻¹ in CO₂-equivalents with 30, 25 and 45% from CO₂, CH₄ and N₂O, respectively (Figure 6). This means that the agricultural peatland is an important source for all three greenhouse gases.

To put this into the right perspective, it is important to realize that there are substantial additional sources due to the full range of agricultural activities on this peatland. If CO₂ emissions due to biomass removal (i.e. grass harvest) and farm emissions of CH₄ (i.e. enteric fermentation by dairy cows) are included, the total emission will increase by 10.5×10^3 kg CO₂ ha⁻¹ year⁻¹ (Veenendaal *et al.*, 2007) and 5911 kg CO₂ ha⁻¹ year⁻¹ (257 kg CH₄ ha⁻¹ year⁻¹) (Schrier-Uijl *et al.*, 2009a), respectively. The total balance will thus increase by even more than 250%, which emphasizes the importance of further research in which all GHG components will be taken into account.

We also compared the total field GHG emissions at this site (Oukoop) with the GHG emissions in a former agricultural site (Horstermeer), which was converted to a nature reserve 11 years ago by stopping intensive farming and raising the water table (Hendriks *et al.*, 2007). The terrestrial CO₂, CH₄ and N₂O emissions at the Horstermeer site are -11 403 kg CO₂ ha⁻¹ year⁻¹, 417 kg CH₄ ha⁻¹ year⁻¹ and negligible, respectively (Hendriks *et al.*, 2007). This leads to a total GHG uptake of -2 Mg ha⁻¹ year⁻¹ in CO₂-equivalents. Consequently, a transformation of an intensively agricultural site to a nature reserve will probably lead to a decrease in total GHG emission because the Oukoop site is a strong source of 16 Mg ha⁻¹ year⁻¹ in CO₂-equivalents. The

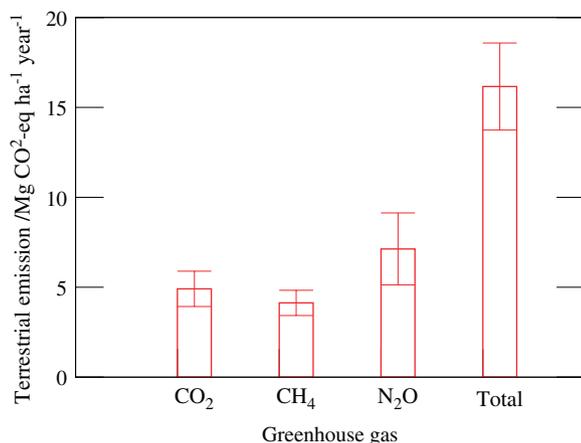


Figure 6 Annual terrestrial emissions of CO₂, CH₄ and N₂O and the sum of the emissions of these three gases in CO₂-equivalents ha⁻¹ year⁻¹ with their uncertainty measured at a managed peat area in the Netherlands.

CO₂ and N₂O emissions are thus much larger at the intensively managed Oukoop site than at the restored Horstermeer site. In addition, if we also include the farm-based emissions with the CH₄ emissions, the total CH₄ emissions at Oukoop will also be larger than the CH₄ emissions at the Horstermeer site (e.g. Schrier-Uijl *et al.*, 2009a).

Conclusions

The annual CH₄ and N₂O terrestrial balances were derived from a dairy farm on peat grassland in the Netherlands over 2006, 2007 and 2008, using EC flux measurements. The average terrestrial CH₄ balance was 165 kg CH₄ ha⁻¹ year⁻¹ and the average terrestrial N₂O balance 24 kg N₂O ha⁻¹ year⁻¹. The CH₄ estimate was more accurate than the estimate by static chambers and the N₂O estimate was a step forward because the background fluxes could not be detected by static chamber measurements using a photo-acoustic instrument at the same site.

Furthermore, we investigated the direct emission factor EF₁ for agricultural soils, which is used in the IPCC guidelines. We calculated the possible EF₁ range following six fertilization events, ranging from EF₁^{min} to EF₁^{max} because the real background emission was not known. The minimum EF₁ was determined by subtracting the background emission and the maximum EF₁ without subtracting the background emission. The average EF₁^{min} was 1.2% and the average EF₁^{max} 2.8%. Both values are larger than the IPCC default EF₁ value of 1%.

Finally, the total terrestrial GHG balance was estimated at 16 Mg ha⁻¹ year⁻¹ in CO₂-equivalents averaged over 2006, 2007 and 2008 with contributions of 30, 25 and 45% by CO₂, CH₄ and N₂O. This agricultural peatland was thus a serious source of all three greenhouse gases. The total emission would be even larger because the farm emissions and the biomass removal were not taken into account.

Supporting Information

The following supporting information is available for this article:

Appendix S1. Calculation method fluxes and uncertainties

Additional Supporting Information may be found in the online version of this article.

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