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Soil-derived trace gas fluxes from different energy crops – results from a field experiment in Southwest Germany

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Abstract

Willow coppice, energy maize and Miscanthus were evaluated regarding their soil-derived trace gas emission potential involving a nonfertilized and a crop-adapted slow-release nitrogen (N) fertilizer scheme. The N application rate was 80 kg N ha^{-1} yr^{-1} for the perennial crops and 240 kg N ha^{-1} yr^{-1} for the annual maize. A replicated field experiment was conducted with 1-year measurements of soil fluxes of CH_4, CO_2 and N_2O in weekly intervals using static chambers. The measurements revealed a clear seasonal trend in soil CO_2 emissions, with highest emissions being found for the N-fertilized Miscanthus plots (annual mean: 50 mg C m^{-2} h^{-1}). Significant differences between the cropping systems were found in soil N_2O emissions due to their dependency on amount and timing of N fertilization. N-fertilized maize plots had highest N_2O emissions by far, which accumulated to 3.6 kg N_2O ha^{-1} yr^{-1}. The contribution of CH_4 fluxes to the total soil greenhouse gas subsumption was very small compared with N_2O and CO_2. CH_4 fluxes were mostly negative indicating that the investigated soils mainly acted as weak sinks for atmospheric CH_4. To identify the system providing the best ratio of yield to soil N_2O emissions, a subsumption relative to biomass yields was calculated. N-fertilized maize caused the highest soil N_2O emissions relative to dry matter yields. Moreover, unfertilized maize had higher relative soil N_2O emissions than unfertilized Miscanthus and willow. These results favour perennial crops for bioenergy production, as they are able to provide high yields with low N_2O emissions in the field.

Keywords: CH_4, CO_2, maize, Miscanthus, N_2O, nitrogen fertilization, perennial crops, willow

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Introduction

The agricultural sector is the second-largest emission source of greenhouse gases (GHG) in the European Union, including CO_2, CH_4, N_2O and fluorinated GHGs. More than half of these emissions originate from agricultural soils (UNFCCC, 2010b). The most important GHGs from terrestrial ecosystems are water vapour (H_2O) and the trace gases carbon dioxide (CO_2), methane (CH_4) and nitrous oxide (N_2O) (Arneth *et al*., 2010). In agricultural soils, if plant respiration is disregarded, CO_2 is mainly produced by the decomposition of organic carbon which is mostly derived from fresh plant litter or was accumulated in the soils over long time periods (Dolman, 2008). Carbon turnover in soils reaches equilibrium if external conditions remain stable, therefore notable amounts of CO_2 emissions exceeding carbon inputs by plant litter derive only from land use changes or changes in cropping management. The production of N_2O is associated with the microbial soil N turnover processes of nitrification and denitrification, both requiring the presence of either nitrate (NO_3^-) or ammonium (NH_4^+) and organic N if heterotrophic nitrification prevails. Thus, N_2O production is highly affected by N-fertilization. Methane fluxes from soils are the result of simultaneously occurring production (methanogenesis) or oxidation processes (methanotrophy), processes which are linked to predominantly anaerobic or aerobic conditions, respectively (Conrad, 1996). However, agricultural soils in Western Europe are considered overall as small sinks of CH_4 (Boeckx & Van Cleemput, 2001).

Quantification of soil trace gas emissions on national and international scales is currently based on activity data, for example the amount of fertilizer use, which are multiplied with specific emission factors to derive national emission estimates (IPCC, 2006; UNFCCC, 2010a). Since the emission of trace gases from agricultural soils is strongly influenced by many factors, like crop, soil type, pH, carbon content, tillage practices, temperature and moisture, it is important to obtain qualified field measurements at as many sites as possible to further improve estimates of GHG emissions for a variety of sites differing in environmental conditions and

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field management. Thus, field measurements are indispensible to provide data related to regional conditions and crops, which can be taken into account when comparing different production strategies in terms of their sustainability with regard to the climate system (Kiese *et al.*, 2005).

The expansion of energy crops as a measure to combat climate warming through replacing fossil fuels by bioenergy is currently discussed controversially. Major energy crops like maize require an intensive fertilization with N and maize cultivation may negatively affect the soil humus balance (Dittert & Mühling, 2009). Moreover, due to the emission of N_2O resulting from the necessary N fertilizer use, the entire greenhouse gas balance may even be negative, i.e. net GHG emissions from biofuels may be higher than those arising from the use of fossil fuels (Crutzen *et al.*, 2008). Perennial energy crops are seen as viable alternatives, since they accumulate high amounts of biomass while requiring limited resources, specifically N-fertilization (Heaton *et al.*, 2008; Rowe *et al.*, 2009). The aim of this study was to compare the annual energy crop maize with the perennial crops Miscanthus and willow in terms of soil emissions of CO_2, N_2O and CH_4 throughout 1 year. To assess the effect of N input to the system, two levels of N fertilization for each crop were considered, i.e. an unfertilized control and a crop-specific N fertilization, based on the principle of replacing N losses due to biomass harvested. The applied fertilization regimes represent recommendations for best management practice for bioenergy crop production in southwest Germany as provided by governmental agricultural advisors (LTZ, 2010a, b, 2011). Providing crop-adapted N fertilization was chosen to compare yields and emissions under practice conditions to gather information on how these bioenergy crops may have an impact on the environment if they are implemented on a large scale. Since annual maize cropping has a higher need for N input than the perennial crops, it was assumed that this system is more susceptible to N losses, thus leading to higher emissions of N_2O. Furthermore, it was assumed that fertilization will result in higher soil CO_2 fluxes due to the stimulation of root growth and soil microbial activity. With respect to CH_4 we hypothesized that the CH_4 sink strength of soils planted with perennial crops (willow, Miscanthus) should be higher compared with plots planted with maize and that fertilization will negatively affect the oxidation capacity of soils for atmospheric CH_4.

Material and methods

Site description and experimental design

The field experiment was located in Southwest Germany at the research station Ihinger Hof (48.75°N and 8.92°E, 480 m asl) of the University of Hohenheim. Mean air temperature for the period 1999–2009 was 8.3 °C and mean annual precipitation was 691 mm. The soil is a Haplic Luvisol with a silt clay texture overlaid by loess loam. In 2002, a field trial with different bioenergy cropping systems was established; three representatives were selected for this study. The trial is a complete split-plot design with main plots for the cropping systems divided into subplots with two different N-application levels. The nutrients P, K and Mg were fertilized in a long-term concept which aims for soil concentrations of 20, 20 and 15 mg (100 g soil)$^{-1}$, respectively. Measurements of soil trace gas fluxes were conducted with three replications; each subplot had an area of 160 m².

In 2009, trace gas measurements started in the plots with the crops energy-maize (*Zea mays* 'Mikado'), Miscanthus (*M.* × *giganteus)* and willow (*Salix schwerinii* × *viminalis* 'Tora'). The measurements included maize-plots with an annual N-fertilization of 0 (unfertilized) and 240 kg N ha^{-1} yr^{-1} and plots of willow and Miscanthus with fertilization rates of 0 and 80 kg N ha^{-1} yr^{-1}. All plots were fertilized with the ammonium-stabilized N-fertilizer Entec 26 (K+S Nitrogen GmbH, Mannheim, Germany), containing 7.5% nitrate-N, 18.5% ammonium-N and 13% sulphur. The nitrification inhibitor 3,4-dimethylpyrazole phosphate (DMPP) delays the bacterial nitrification of ammonia to nitrate in the soil by depressing the activity of autotrophic nitrifiers for a time period of 4–10 weeks (Zerulla *et al.*, 2001; Díez-López *et al.*, 2008).

The energy maize was cropped in a maize-after-maize system. The measurements started in late 2009 after the maize was harvested. The sowing date in 2010 was April 23, 1 week after the one-time N-application in the fertilized plots. Herbicides were applied once, early after emergence. Harvest date was October 27, 2010, followed by soil tillage with a rotary harrow on October 5. Miscanthus plots were planted in 2002 and thereafter harvested each year in late winter or early spring. During the period of the gas flux measurements, the Miscanthus plots were harvested once at March 26, 2010. Fertilizer was applied on April 9, 2010 before the new sprouts emerged. No weed control was carried out during the period of the measurements. The willow short rotation coppice was also established in 2002. After 1 year, harvest took place every third year. The last harvest was February 2, 2009, 1 year before the gas flux measurements started. Fertilizer was applied on April 21, 2010. No weed control was carried out during the period of the experiment. Soil analysis was carried out prior to trace gas measurements. Soil organic carbon (SOC) concentrations of the 0–30 cm soil layer were between 0.9% and 1.4%. Lowest SOC concentrations were found in the unfertilized maize plots (0.9%), whereas highest SOC concentrations were measured in the unfertilized Miscanthus plots (1.4%). These two variants had significantly different SOC concentrations all other variants did not differ significantly. The soil acidity ranged between pH 6.38 and pH 6.71 for all plots, the N-fertilized plots having slightly lower pH values than the unfertilized plots; however, the difference was not significant (Table 1).

Measurement of trace gas fluxes

The measurements of CO_2, CH_4 and N_2O fluxes between the soil and the atmosphere were conducted using the static chamber

Table 1 Soil properties of the investigated plots (soil depth 0–30 cm; ±standard error; N = 3 composite samples; soil NO_3^- content measured 1 week before fertilization in Miscanthus and maize and 2 months after fertilization in willow; soil texture as average of all plots)

Crop	N-fertilization (kg N ha^{-1} yr^{-1})	SOC (%)	pH	Bulk density (g cm^{-3})	Soil NO_3^- (kg ha^{-1})
Maize	0	0.9 ± 0.06	6.7 ± 0.01	1.3 ± 0.04	3.4
	240	1.0 ± 0.03	6.5 ± 0.05	1.3 ± 0.04	23.8
Miscanthus	0	1.4 ± 0.34	6.7 ± 0.02	1.5 ± 0.06	1.7
	80	1.2 ± 0.07	6.4 ± 0.08	1.5 ± 0.08	2.8
Willow	0	1.1 ± 0.07	6.7 ± 0.03	1.4 ± 0.03	2.2
	80	1.1 ± 0.12	6.6 ± 0.08	1.4 ± 0.03	2.6
Soil texture	Sand (%)	Silt (%)	Clay (%)		
	2.1 ± 0.04	69.7 ± 0.19	28.2 ± 0.19		

method (Hutchinson & Mosier, 1981). Measurements started on November 30, 2009 and were carried out once a week until December 1, 2010, except between June 7 and July 18, when samples were taken twice a week. After application of slow-release fertilizer, first flux measurements were done within the following 6 days. Overall 18 frames (56.5 × 36.5 × 15.5 cm) – three opaque chambers per crop and treatment – were inserted approximately 10 cm into the soil not covering the crop stalks. Frame positions were chosen in such a way that soil conditions and weed content of the plots were covered representatively. During measurement, the frames were closed with complementary opaque chambers which were clamped on the frame. Rubber seals ensured a gas-tight closure of the chambers, which then had a volume of approximately 0.05 m^3 depending on the depth of the frame in the soil. Gas samples were withdrawn with syringes and via septum from the chamber headspaces. In total, four gas samples of 60 ml volume each were taken, right after closure and then every 15 min thereafter. The gas samples were transferred into 30 ml vacuum vials and analysed within the following days by gas chromatography (SRI 810C, SRI Instruments, Torrance, CA, USA). A flame ionization detector was used to analyse the CO_2 and CH_4 content in the samples. Prior to detection, CO_2 was reduced to CH_4 using a methanizer. N_2O was analysed with a ^{63}Ni electron capture detector. Detection limit for N_2O concentration changes in sample air at ambient atmospheric N_2O concentrations was approximately 10 ppbv N_2O h^{-1}, which is equivalent to a N_2O flux of approximately 3 μg N_2O-N m^{-2} h^{-1}. Detection limits for CH_4 and CO_2 were 3.7 μg N_2O-N m^{-2} h^{-1} and 1 mg CO_2-C m^{-2} h^{-1} For further details on analytical conditions, see e.g. Kiese *et al.* (2003). Fluxes were calculated from linear changes in gas concentrations with time, since there was no proof that temporal changes in headspace concentrations deviated from linearity as e.g. discussed by Kroon *et al.* (2008).

Calculation of a trace gas emission subsumption

To assess the soil GHG subsumption for the different bioenergy cropping systems, a subsumption was calculated taking into account solely soil fluxes of CH_4 and N_2O. CO_2 was excluded from this subsumption, since the measuring design did not allow distinguishing between heterotrophic and autotrophic respiration or plant litter inputs. Since weeds were also covered by the chambers, the CO_2 gas fluxes rather refer to ecosystem respiration. Cumulative annual fluxes were obtained by linear interpolation between sampling times. CH_4 and N_2O subsumptions were transformed in global warming potentials (GWP), which are calculated in CO_2 equivalents according to IPCC (2007). The GWP of CH_4 was calculated with a factor of 25, N_2O was calculated with a factor of 298 and CO_2 as the reference was maintained with no calculation factor. Furthermore, the mean yield of each cropping system was related to the respective soil N_2O emission subsumption. Yields of perennial crops often have the appearance of a saturation curve, with low yields in the establishment period and high yields until exhaustion. For this reason, the mean yield of the previous 5 years (2005–2009) was taken as bases for calculations.

Soil temperatures and moisture

Following gas flux measurements soil temperatures were taken in a depth of 10 cm three times for each cropping system, using a portable thermocouple (Testo 925, Lenzkirch, Germany). Furthermore, volumetric soil moisture between 0 and 16 cm was measured with a portable rod probe (Trime FM with P3 probe IMKO GmbH, Ettlingen, Germany). Soil moisture values were converted into water filled pore space (WFPS) by the following formula:

$$\mathrm{WFPS}(\%) = \frac{\mathrm{Vol}(\%)}{1 - \frac{\mathrm{bd}(\mathrm{g\ cm^{-3}})}{2.65(\mathrm{g\ cm^{-3}})}}$$

where bd is bulk density, Vol is volumetric water content and 2.65 is the density of quartz.

Statistical analysis

Mean values for each variant and measurement date were calculated from untransformed data using the mixed-model procedure in SAS (Statistical Analysis System 9.2, SAS Institute Inc., Cary, NC, USA). Since in the case of CO_2 and N_2O, untransformed data did not show a normal distribution, a Box-Cox transformation was conducted for CO_2-data and a $\log_{10}$ transformation for N_2O data before analysis of variance of the whole measurement period. Mean values of the whole measurement period of CO_2 and N_2O are presented as back-transformed values to provide the data in a comparable dimension. The analysis of variance of the whole measurement period of CH_4 data, however, was performed with untransformed data. To evaluate the relationship between soil temperature and moisture, regression analysis was performed using the regression procedure in SAS including all measured values. Graphics were created with Origin 7.0 (OriginLab Corporation, Northampton, MA, USA).

Results

Soil temperature and soil moisture

Soil temperatures measured in a depth of 10 cm during autumn and winter were mostly lower in maize plots than in plots planted with Miscanthus and willow. In summertime, however, maize plots showed the highest temperatures with a maximum of 24.7 °C on July 2 which was significantly higher than in the other crops, whereas for Miscanthus and willow coppice plots maximum soil temperatures were 19.9 and 20.0 °C, respectively. Significant differences between the different bioenergy crops were also found to WFPS. With a mean of 54.0%, WFPS in maize plots was considerably lower throughout the year than in Miscanthus plots with 73.9% and willow plots with 64.7% (Fig. 1).

Soil CO_2 fluxes

Since chambers also covered weeds, the measured values present the ecosystem respiration at ground level; however, soil CO_2 emission is used in the following. At all plots soil CO_2 emissions showed a clear seasonal pattern, with minimum values in winter and spring and maximum values during summer (Fig. 2). Significant differences between the different cropping systems regarding magnitude and timing of maximum and minimum CO_2 emissions could be observed during the year. Soil CO_2 emissions from maize plots started to rise late during springtime compared with those of Miscanthus and willow. However, with row closure in late June, the emissions increased sharply and spiked out the other two cultures with the highest mean emission of 206.3 mg C m^{-2} h^{-1} on July 16 (Fig. 2). Although the mean soil CO_2 emissions in the Miscanthus plots were generally comparable with those of the willow plots during late winter, a distinctive emission peak of 80.7 mg C m^{-2} h^{-1} was observed 4 days following the harvest of Miscanthus on March 26. Thereafter, soil emissions dropped to only 25.6 mg C m^{-2} h^{-1} within 1 week. During June, the mean CO_2 emissions in the Miscanthus plots were higher than those of the two other cultures. From July on, the maize plots had the highest soil CO_2 emissions until the emissions of the three crops equalled in September. The soil CO_2 emissions in the willow plots increased very fast during April to 132.9 mg C m^{-2} h^{-1} on May 12. Afterwards the emissions remained relatively stable until end of August when the CO_2 emissions in all soils started to decrease.

The analysis of variance showed a significant effect of N-fertilization only for maize plots, with a higher mean rate of 38.5 in the N-fertilized plots compared with 25.3 in the unfertilized plots. Throughout the entire observation period, soil CO_2 emissions from the fertilized maize plots were higher than those of the unfertilized control (Fig. 2). Miscanthus plots had a mean CO_2 emission of 50.0 and 47.8 mg C m^{-2} h^{-1} for N-fertilized and unfertilized plots, respectively. These rates do not differ significantly from the mean CO_2 emissions of the willow plots, which were 47.3 and 48.1 mg C m^{-2} h^{-1} for the N-fertilized and unfertilized plots, respectively. Both maize variants, however, had a significantly lower annual CO_2 emission than both of the Miscanthus and willow variants.

Using data from all plots, soil CO_2 emissions were highly positively correlated to soil temperature ($P < 0.0001$, $r^2 = 0.55$) and soil water content ($P < 0.0001$, $r^2 = 0.11$).

Soil CH_4 fluxes

Over the entire observation period CH_4 exchange rates between the soil and the atmosphere ranged from −74.6 to +63.2 μg C m^{-2} h^{-1}. On average, all unfertilized plots with bioenergy crops were a weak sink for atmospheric CH_4. Lowest mean net CH_4 uptake was measured for unfertilized maize (−2.6 μg C m^{-2} h^{-1}) and Miscanthus plots (−2.9 μg C m^{-2} h^{-1}), whereas the mean net CH_4 uptake for unfertilized willow plots was significantly higher (−6.0 μg C m^{-2} h^{-1}).

N-fertilization of the different crops had no significant effect on CH_4 fluxes, though there was a tendency for positive average fluxes in the fertilized plots of maize and Miscanthus with 0.9 and 0.6 μg C m^{-2} h^{-1}, respectively. This indicates that N-fertilization turned soils from being a weak net sink towards a weak net source. In contrast, N-fertilization of the willow plots resulted in an increased average CH_4 uptake of −8.1 μg C m^{-2} h^{-1}.

The CH_4 fluxes were significantly correlated to measured soil temperature ($P = 0.006$), though the coefficient of determination was very low ($r^2 = 0.008$). The values of WFPS were not significantly correlated to the CH_4 fluxes. Thus, CH_4 fluxes did not show a clear

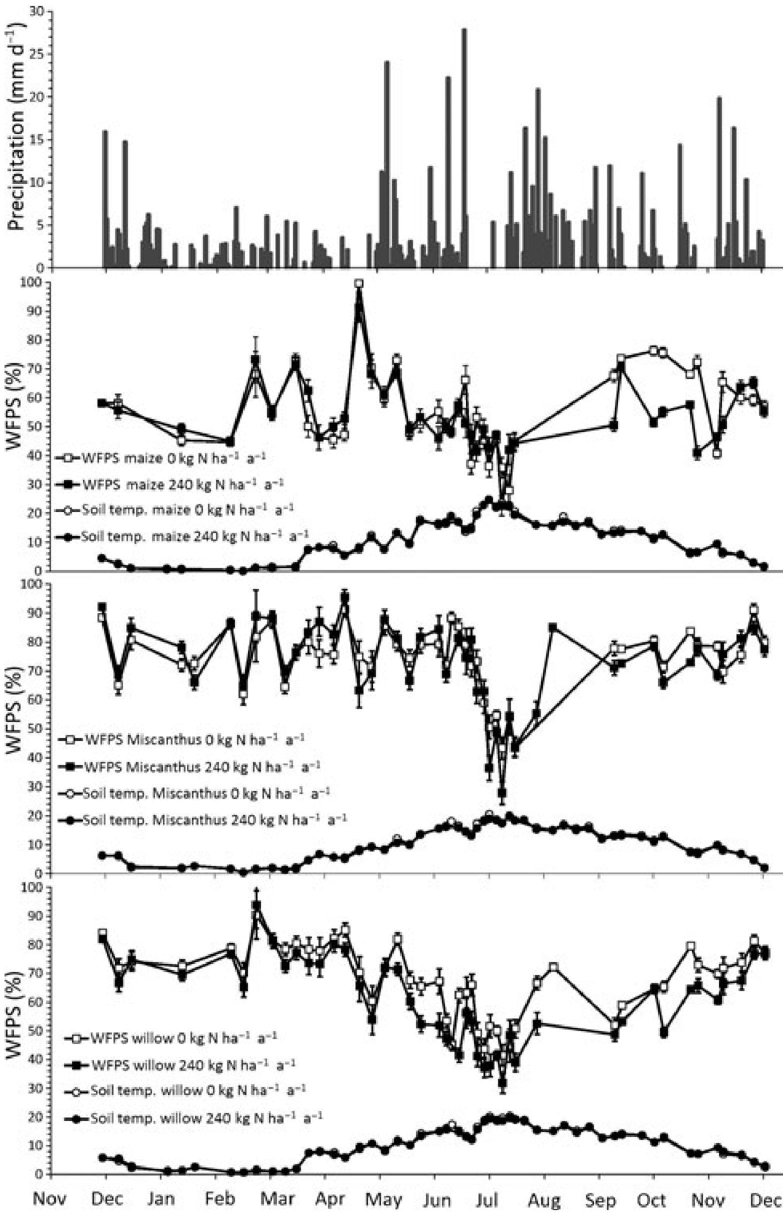

Fig. 1 Precipitation, soil temperatures in 10 cm depth (°C) and water filled pore space (WFPS) in 0–16 cm depth (%) in maize, Miscanthus and willow plots between October 2009 and December 2010. Closed symbols: N-fertilized plots; open symbols: non N-fertilized plots. Given are mean values ± SE of three replicated measurements.

seasonal trend but were oscillating around zero while being below zero more often (Fig. 3).

Soil N_2O fluxes

In the maize plots, mean soil N_2O fluxes ranged between −5.4 and 111.0 μg N m^{-2} h^{-1}. From the beginning of the measurements on November 30, 2009, to February 23, 2010, N-fertilized maize plots mostly had higher soil N_2O emissions (5.3–49.1 μg N m^{-2} h^{-1}) than unfertilized maize plots (0.1–5.2 μg N m^{-2} h^{-1}). During late winter up to April 12, the soil N_2O emissions of both maize variants were approximately the same, staying in a range between −3.9 and 6.1 μg N m^{-2} h^{-1}. Following fertilization on April 15, mean soil N_2O emissions increased and peaked on June 19 with a mean of 111.0 μg N m^{-2} h^{-1}. Subsequently, mean soil N_2O emissions of the N-fertilized maize plots decreased with the absence of rainfall in late June and beginning of July but increased again to values of up to 84.5 μg N m^{-2} h^{-1} in August after rainfall events. Subsequently, emissions decreased down to a minimum of

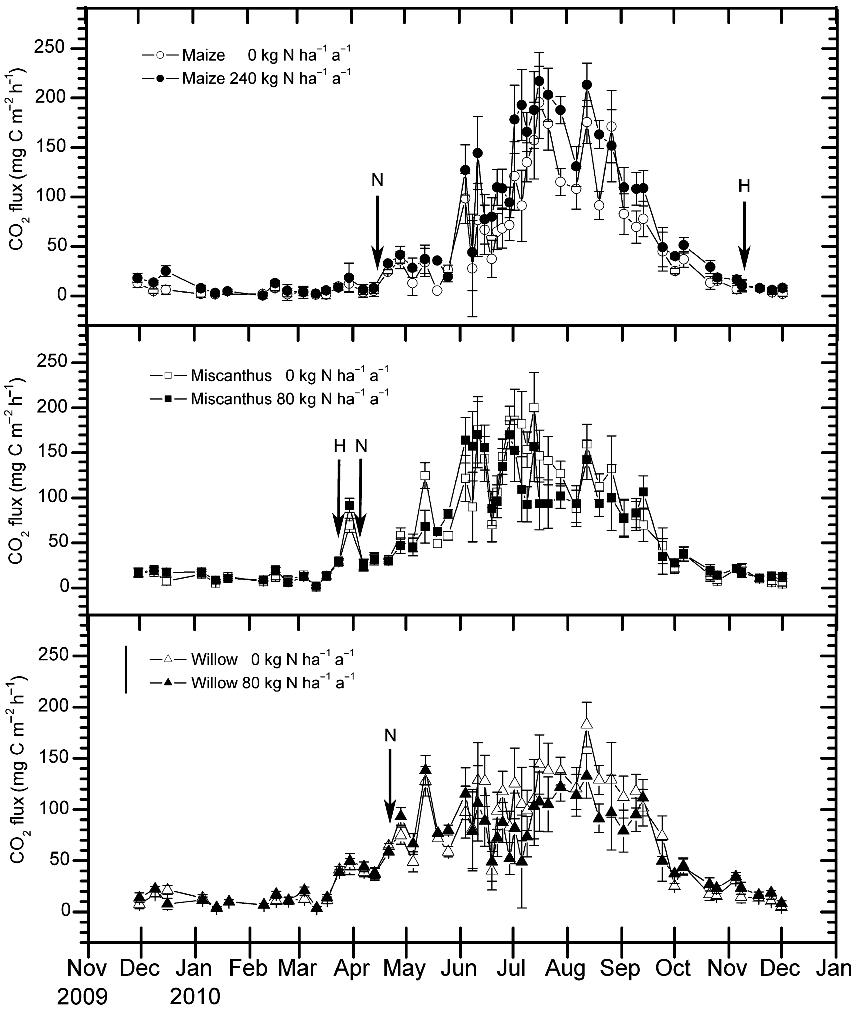

Fig. 2 Soil CO_2 fluxes in maize, Miscanthus and willow; arrows with the letter N indicate N-fertilization events, arrows with the letter H indicate harvest events. Fluxes are the mean of three independent measurements (± SE).

4.9 μg N m^{-2} h^{-1} on November 25, but stayed above the unfertilized plots, where mean emissions did not exceed 14.3 μg N m^{-2} h^{-1} during the whole year.

Before harvest on March 26, soil N_2O emissions for the plots planted with Miscanthus were close to the detection limit. Following harvest, soil N_2O fluxes in the N-fertilized plots increased to a mean of 13.6 and 9.2 μg N m^{-2} h^{-1} on the following measurement dates. N-fertilization on April 9 resulted in peak emission of N_2O (140.4 μg N m^{-2} h^{-1}) on April 13. In the following 5 weeks, emission fell below 50 μg N m^{-2} h^{-1}, but spiked once more on May 25 with a mean rate of 131.9 μg N m^{-2} h^{-1}. Subsequently, the rates decreased to 9.4 μg N m^{-2} h^{-1} and declined further during the following 4 weeks. They finally aligned to the mean values of the not N-fertilized plots at the beginning of July. By contrast, the unfertilized plots did not show higher soil N_2O emissions than 4.4 μg N m^{-2} h^{-1} during the entire measurement period (Fig. 4).

Compared with the maize and Miscanthus plots, soil N_2O emissions in the willow plots stayed roughly constant throughout the entire measuring period (range: −7.1 and 6.7 μg N m^{-2} h^{-1}). N-fertilized plots tended to be a weak source of N_2O, whereas unfertilized plots tended to be weak net sinks for N_2O. N-fertilization on April 21 did not result in a marked increase in N_2O fluxes.

Statistical analysis showed no significant effect of N-fertilization on the soil N_2O emissions of willow plots during the course of 1 year. However, a highly significant effect of N-fertilization on soil N_2O emissions was found for Miscanthus and maize cropping. N fertilization with 240 kg of slow release N-fertilizer ha^{-1} resulted in a mean soil N_2O emission of 17.9 μg N m^{-2} h^{-1} in the maize plots, compared with 1.0 μg N m^{-2} h^{-1} in the unfertilized maize plots. For Miscanthus, N application of 80 kg ha^{-1} increased mean N_2O fluxes to 4.3 μg N m^{-2} h^{-1}, compared with −1.0 μg N m^{-2}

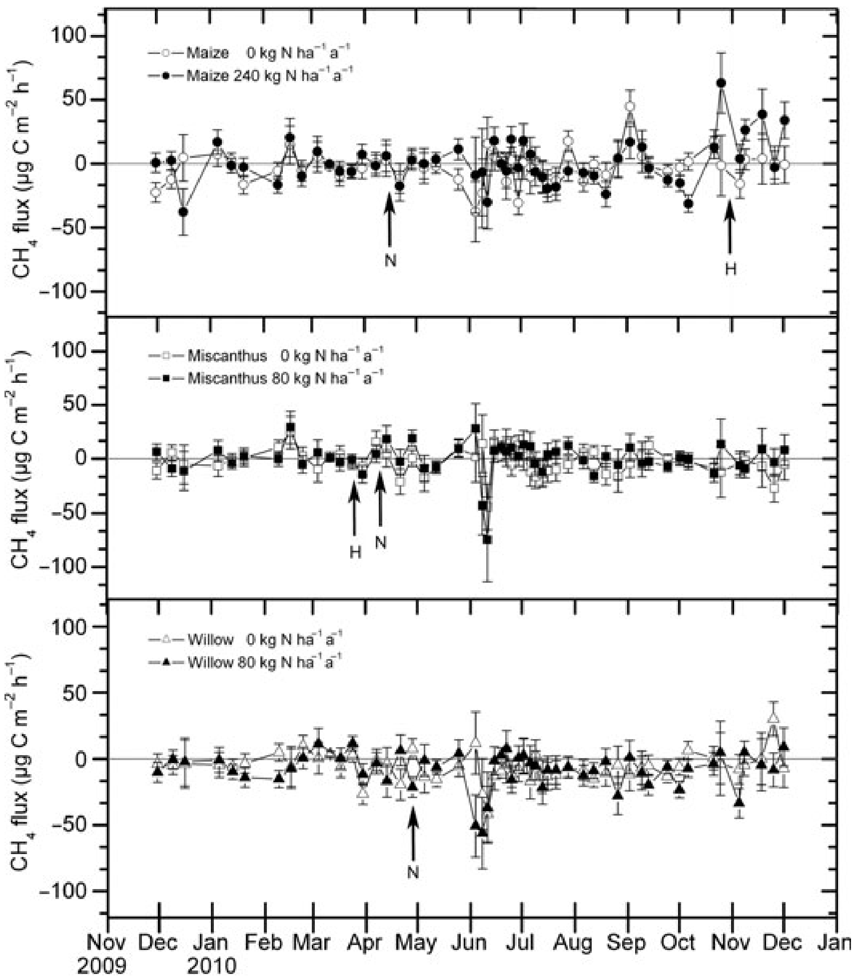

Fig. 3 Soil CH_4 fluxes in maize, Miscanthus and willow; arrows with the letter N indicate N-fertilization events, arrows with the letter H indicate harvest events (error bars are standard errors). Given are mean values (± SE) of three independent flux measurements.

h^{-1} for the unfertilized plots. Only in the willow plots N application of 80 kg N ha^{-1} did not change N_2O fluxes significantly (N-fertilized: -0.2 µg N m^{-2} h^{-1}, unfertilized: -0.4 µg N m^{-2} h^{-1}). Linear regression analysis showed a significant correlation of soil N_2O emissions with temperature ($P < 0.0005$), although the coefficient of determination was only $r^2 = 0.014$. Surprisingly, soil moisture and soil N_2O emissions were not significantly correlated in this experiment.

Soil greenhouse gas emission subsumption

For the calculation of the soil greenhouse gas subsumption, the GWP approach was followed. Annual soil CO_2 emissions totalled 13 172–18 737 kg CO_2 ha^{-1} yr^{-1}. However, soil-derived CO_2 emissions were not incorporated in the GWP-subsumption since net CO_2 exchange depends on CO_2 inputs by plant residues (West & Marland, 2002). Furthermore, soil CO_2 efflux is highly affected by root respiration (Kuzyakov *et al.*, 2003). Thus, the main proportion of the soil CO_2 efflux is contemporarily fixed CO_2 from the atmosphere. Since CO_2 fixation was not measured in this experiment, CO_2 emissions were excluded from the soil GHG subsumption. With -15 to 1063 kg CO_2-equivalents, the contribution of N_2O to the soil gas subsumption was more important than that of CH_4 (-22 to 1 kg CO_2-equivalents). The smallest GWP was found for unfertilized Miscanthus plots, since soil CH_4 and soil N_2O flux rates both were negative. Highest soil GHG emissions were found in N-fertilized maize plots, due to relatively high soil N_2O emissions (Table 2).

Discussion

To our knowledge this is the first study comparing unfertilized and fertilized variants of three different common energy crops – i.e. maize, Miscanthus and

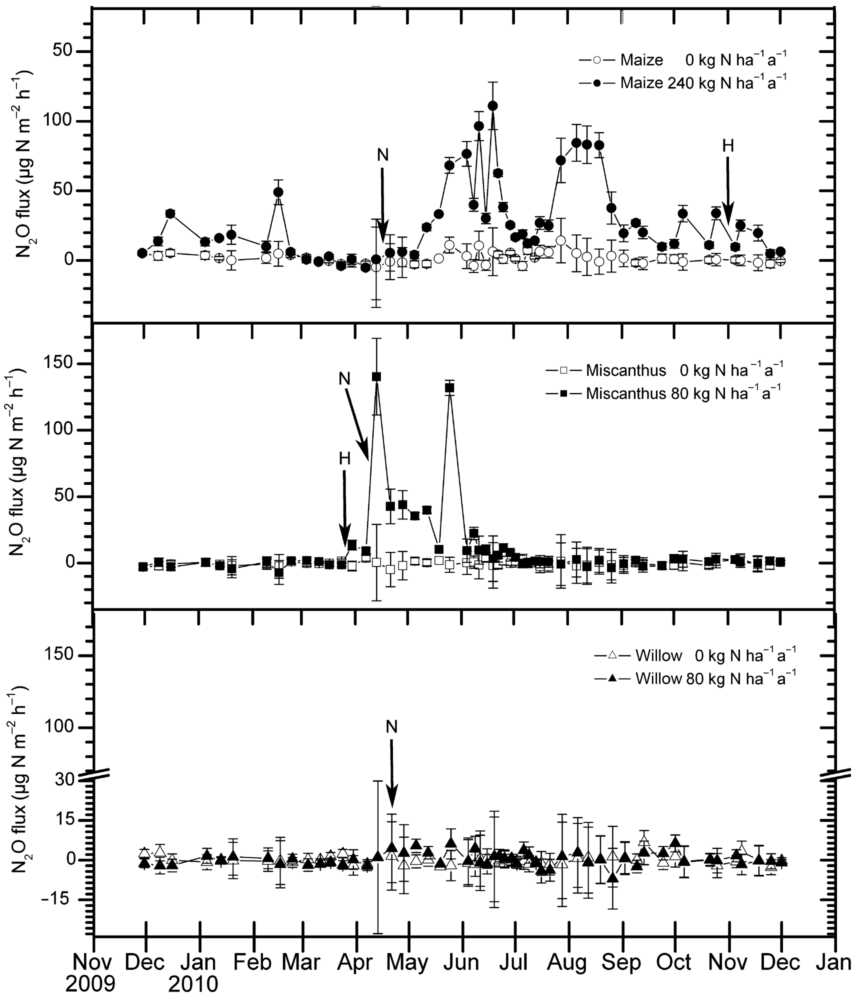

Fig. 4 Soil N_2O fluxes in maize, Miscanthus and willow; arrows with the letter N indicate N-fertilization events, arrows with the letter H indicate harvest events. Fluxes are the mean of three independent measurements (± SE).

willow – with regard to soil GHG fluxes at one site with comparable soil characteristics simultaneously.

During the course of 1 year, mean soil CO_2 emissions ranged from 0.3 to 217.1 mg C m^{-2} h^{-1}. A seasonality of soil CO_2 emissions – mainly determined by changes in soil temperatures (r^2 = 0.55) – could be observed in all plots. A clear correlation between soil temperature and soil CO_2 emissions was also reported in previous experiments in temperate agricultural soils (Parkin & Kaspar, 2003; Amos *et al.*, 2005; Regina & Alakukku, 2010; Sainju *et al.*, 2010). Soil water content also positively affected soil CO_2 emissions (r^2 = 0.11), which had been similarly reported by Sainju *et al.* (2010) and Jabro *et al.* (2008). According to the Arrhenius kinetics, reaction rates are dependent on temperature; however, enzyme-catalysed reactions also depend on substrate availability, which in this case is organic carbon concentration (Davidson & Janssens, 2006). SOC was lower in the maize plots, indicating that maize growing could be inferior to the tested perennial crops regarding the humus balance of the soil. Since SOC content was higher in the plots of perennial crops, higher soil CO_2 emissions measured in the plots of the perennial crops compared with maize plots had probably their source in carbon which was previously accumulated by the crops (Zan *et al.*, 2001). Post & Kwon (2000) calculated mean SOC accumulation of grassland and forest established on agricultural land to be 33.2 and 33.8 g C m^{-2} yr^{-1}, respectively. Unfertilized maize plots had the lowest SOC content and showed the lowest soil CO_2 emissions during 1 year. N-fertilized maize plots showed highest soil CO_2 emissions during summertime, exceeding 200 mg C m^{-2} h^{-1} at three dates, whereas Miscanthus and willow plots showed higher soil CO_2 emission rates during autumn and winter, but especially during springtime in March and April. One factor for the more constant soil CO_2 emissions of the perennial crops could be the soil coverage throughout

Table 2 Summation of annual soil-derived trace gas fluxes from three energy crops with two N-fertilization rates [global warming potential (GPW) of CH_4: 25 and N_2O: 298, according to (IPCC, 2007)]

Cropping system	CO_2 (kg CO_2 ha^{-1} yr^{-1})	CH_4 (kg CH_4 ha^{-1} yr^{-1})	N_2O (kg N_2O ha^{-1} yr^{-1})	GWP CH_4 (kg CO_2-eq ha^{-1} yr^{-1})	GWP N_2O (kg CO_2-eq ha^{-1} yr^{-1})	GWP: CH_4 + N_2O (kg CO_2-eq ha^{-1} yr^{-1})
Maize 0 kg N ha^{-1} yr^{-1}	13 172.2	−0.2	0.2	−5.6	68.1	**62.5**[a]
Maize 240 kg N ha^{-1} yr^{-1}	18 136.7	0.0	3.6	−0.1	1062.7	**1062.6**[b]
Miscanthus 0 kg N ha^{-1} yr^{-1}	17 942.0	−0.3	0.0	−7.2	−14.6	**−21.8**[a]
Miscanthus 80 kg N ha^{-1} yr^{-1}	17 222.0	0.0	1.4	0.8	419.3	**420.1**[c]
Willow 0 kg N ha^{-1} yr^{-1}	18 373.0	−0.6	0.0	−13.9	−0.4	**−14.3**[a]
Willow 80 kg N ha^{-1} yr^{-1}	16 801.3	−0.9	0.0	−22.3	14.4	**−8.0**[a]

Different letters indicate significant differences ($P < 0.05$).

the year which provides leaf litter input over long periods and leads to less fluctuating temperatures and more stable soil moisture conditions. The measurements of temperature in a depth of 10 cm and the soil moisture measurements between 0 and 16 cm showed that maize plots had a higher fluctuation in soil temperatures and lower water content throughout the year. A comparatively high soil CO_2 peak was detected 4 days after the Miscanthus harvest on March 26 in both N-fertilized and unfertilized plots. This effect could be caused by the high amount of leaf litter which dropped to the ground during harvest procedure. Beuch *et al.* (2000) measured harvest residues between 0.7 and 3.1 t DM ha^{-1} at different sites. Harvest could have also stimulated fine root growth, which is well documented for other perennial crops (Idol *et al.*, 2000; Mello *et al.*, 2007). Furthermore, sunlight was able to reach the soil surface and the litter after harvest, initiating activity of microorganisms by increased temperatures. Soil temperature at 10 cm depth, however, did not differ significantly from maize and willow plots. By contrast, harvest of maize on October 27 had no direct effect on soil CO_2 emissions. Since litterfall during maize harvest did not occur in notable amounts, an increased substrate supply did not occur in this case.

In our study CH_4 fluxes were generally low. In general, unfertilized plots were net sinks for atmospheric CH_4, with average annual uptake rates in the range of −6.0 to −2.0 μg C m^{-2} h^{-1}. For Miscanthus and maize, N fertilization turned soils to weak net sources on average (0.6–0.9 μg C m^{-2} h^{-1}). The observed range of fluxes is in good agreement with previous studies for other arable soils, further confirming that managed soils are not a major sink for atmospheric CH_4 (e.g. Hütsch, 2001; Smith & Conen, 2004). The somewhat higher uptake rates for willow in our study could also be expected, since management operations are only performed every 3 years compared with Miscanthus and maize plots which are either harvested annually or even ploughed and tilled. This is in line with the study of Priemé *et al.* (1997) who showed that CH_4 uptake may recover slowly if disturbance intensity is weakening, although a full recovery to rates of CH_4 uptake observed for natural systems may take decades up to a century. The observed slight inhibition CH_4 uptake by fertilization, turning plots of maize and Miscanthus from weak net sinks to weak net sources may be explained by a switching of function of the CH_4 monooxygenase to oxidize ammonia rather than CH_4 in the presence of N as discussed by Acton & Baggs (2011) or Bedard & Knowles (1989).

During the course of 1 year, soil N_2O fluxes ranged between −7.1 and 140.4 μg N m^{-2} h^{-1}. The observed soil N_2O fluxes differed remarkably between the crops and the N-fertilization regimes. For maize, higher soil N_2O emissions were observed in fertilized plots between December and February 23. Before measurements started in late November, harvest and soil tillage had taken place. Since the dry-matter yield in the N-fertilized plots was three times higher than in the unfertilized plots (22.6 and 6.4 t DM yr^{-1}), much more root biomass could be assumed to have been left in the soil of the N-fertilized plots. Turnover of plant roots can lead to increased soil N_2O emissions and therefore be a plausible cause for the elevated emissions in the N-fertilized plots until late winter (Beck & Christensen, 1987). Interestingly, soil N_2O emissions did not increase significantly right after N-fertilization of maize took place on April 15 and sowing on April 23. When the temperatures remained above 10 °C after May 5, soil N_2O emissions in the N-fertilized maize plots began to increase rapidly. Since the N fertilizer had the nitrification blocker DMPP added, a delayed and prolonged nitrification (Zerulla *et al.*, 2001) is the most likely explanation for the observed time lag between N fertilizer application and peak N_2O emissions. In contrast to soil CO_2 emissions, soil N_2O emissions decreased with row closure of maize in late June, but increased again 4 weeks later to above 50 μg N m^{-2} h^{-1} at the end of July. Li *et al.* (2004) found indications that soil N_2O fluxes are influenced by light availability. Foereid *et al.* (2010) also found an increased decomposition of plant material in combination with exposure to light, which may explain the initial decrease of soil N_2O emissions after the soil was totally shadowed by the plants. However, since during this period, i.e. late June to beginning of July, no rainfall was recorded, and the decreased soil N_2O emissions are most likely a consequence of reduced soil moisture. Over the whole year, mean soil N_2O emissions in the N-fertilized plots were 17.9 μg N m^{-2} h^{-1}, which was significantly higher ($P < 0.001$) than in the unfertilized plots (1.0 μg N m^{-2} h^{-1}).

The N-fertilized Miscanthus plots received an annual N fertilizer input of 80 kg N ha^{-1}, and therefore a considerably lower N input than the N-fertilized maize plots receiving 240 kg N ha^{-1} yr^{-1}. Thus, it is not surprising that mean soil N_2O emissions were considerably lower in the N-fertilized Miscanthus plots than in the N-fertilized maize plots. Both Miscanthus N-fertilization rates showed soil N_2O fluxes close to zero in most periods of the year. However, 4 days following harvest on March 26, soil N_2O emissions increased from −1.2 to 13.5 μg N m^{-2} h^{-1} in the N-fertilized plots. Since N-fertilized plots had accumulated 33% more above ground biomass than the unfertilized plots (27.1 and 20.4 t DM ha^{-1}) but only 4% more was harvested (18.1 and 18.7 t DM ha^{-1}), the amount of plant residues remaining on the field was more than three times higher on the N-fertilized plots (8.4 and 2.3 t DM ha^{-1}). Plant litter which had fallen to the ground during harvest and its initial decomposition was the probable cause for the increased soil N_2O emission 4 days later on the N-fertilized plots. This interpretation is in line with previous studies which also observed increased soil N_2O emissions following residue input (Huang *et al.*, 2004; Novoa & Tejeda, 2006). The very high soil N_2O emissions after N fertilization in springtime in Miscanthus compared with those in maize may be a result of the fertilization technique in both crops. N-fertilizer in maize plots had been incorporated into the soil after application, which has partly prevented the N from initial gaseous losses. In Miscanthus plots, however, the N-fertilizer was left on top of the soil where it was susceptible to gaseous losses (Khalil *et al.*, 2009). Similar results were also found by Jørgensen *et al.* (1997), who measured significant soil N_2O emissions after fertilization of Miscanthus. A technique avoiding surface fertilization of Miscanthus after resprouting in springtime, i.e. an incorporation of fertilizer into the soil, would probably reduce the observed soil N_2O emissions considerably.

In the willow plots, N fertilization had no significant effect on the overall mean soil N_2O emission rate during the year. Only a slightly increased soil N_2O emission in the fertilized plots was observed in the first 5 weeks after N-fertilization on April 21. In other experiments, Hellebrand *et al.* (2008) and Kavdir *et al.* (2008) found slightly increased soil N_2O emissions in willow plots with N fertilization, but these were comparably low and much smaller than N_2O emissions of annual crops with the same amounts of N-fertilizer. In these articles, it was assumed that soil disturbance led to higher mineralization rates which could explain the low soil N_2O emissions in the willow plots. Nitrogen use efficiency is also reported to be higher in perennial crops, leading to decreased gaseous losses (Kavdir *et al.*, 2008). In the case of this particular experiment, N fertilization of willow took place on April 21, which is later in the vegetation period than the fertilization dates of maize (April 15) and Miscanthus (April 9). Furthermore, the soil in the willow plots was covered with grasses and herbs which were in active growing stage when the fertilizer was applied. It is likely that the applied N was taken up very efficiently by the vegetation, which includes willow and weeds, compared with the other crops where no plant cover was present at the time of fertilization. In all crops, the measurements of soil temperature and soil moisture could only explain soil CO_2 fluxes satisfactorily. Even though soil CH_4 and soil N_2O fluxes were significantly correlated with soil temperature, less than 10% ($r^2 < 0.1$) of the variation in fluxes could be explained by these factors (Table 3). Apparently other factors like the mineralization of the fertilizer play a role which, however, could not been measured in this study.

A soil-derived trace gas subsumption was calculated, though the measurements were not continuous and periods of about 1 week had to be interpolated between the measurements. Experiments comparing gas fluxes of automated and manual chambers in a rice-wheat rotation showed that sampling manual chambers overestimated N_2O fluxes by 18% and CO_2 fluxes by 31%. This difference was deduced to the fact that manual measurements took place during daytime neglecting diurnal temperature variations. No significant differences were found in terms of CH_4 fluxes. By contrast,

Table 3 Regression analysis between the measured trace gas fluxes of CH_4, CO_2, N_2O and soil temperature (°C in 10 cm depth) and soil moisture [WFPS (%) 0–16 cm depth], $P < 0.05$*, $P < 0.01$**

	Soil temperature		Soil moisture (WFPS)	
Trace gas	r^2	f(T)	r^2	f(M)
CH_4	0.008**	0.703 − 0.347T	0.004	−9.147 + 0.095M
CO_2	0.548**	−11.057 + 6.939T	0.112**	139.147 − 1.265M
N_2O	0.014**	2.443 + 0.420T	0.002	10.199 − 0.066M

automated chambers underestimated N_2O and CO_2 fluxes by 22% and 17%, respectively, due to a notable interference with the microclimate inside the chambers (Yao *et al.*, 2009). Also, Werner *et al.* (2006) calculated that the accuracy of weekly measurements would be reduced by 25% compared to 10 subdaily measurements in tropical forest systems. In this light, the soil-derived trace gas subsumption comprises considerable uncertainties. However, abating adjustments were not conducted, since possibly unrecognized temporal peaks of trace gas fluxes can be important for the total subsumption (Mogge *et al.*, 1998; Butterbach-Bahl *et al.*, 2004). Therefore, weekly measurements of soil trace gas fluxes – as performed in our study – may result in an underestimation of the overall flux rate of approximately 20–30% compared with subdaily flux measurements.

The subsumption of the sum of soil-derived trace gas emissions of CH_4 and N_2O showed the highest amounts in the N-fertilized maize system (Table 3). A relatively high soil N_2O emission compared with the other cropping systems was crucial for this result. In this experiment, soil N_2O emissions increased with the amount of N fertilization, which is also well documented in the literature (Jørgensen *et al.*, 1997; Weitz *et al.*, 2001; Hellebrand *et al.*, 2008; Kavdir *et al.*, 2008). N-fertilized maize has received 240 kg N ha^{-1} yr^{-1}, which was the highest amount of N in this experiment, thus relatively high soil N_2O emissions are coherent. The overall direct N_2O-N losses account for 0.95% of the added N by N-fertilization in maize, 1.1% in Miscanthus and an almost nondetectable loss of 0.04% in willow coppice (Table 4). The measured emissions in maize and Miscanthus match the default value of 1% which is proposed in the IPCC guidelines (IPCC, 2006). However, it should be noted that since N-fertilization in this experiment was conducted with enhanced-efficiency fertilizers containing the nitrification inhibitor DMPP an increased N_2O-N loss is likely to occur if conventional fertilizers are used. On the other hand, amounts of N applied in this experiment were with 240 kg N ha^{-1} slightly higher than the official recommendation for the study region, which is 200 kg N ha^{-1} (LTZ, 2011). Thus, a lower N fertilization rate would probably have resulted in reduced N_2O losses. A recently conducted meta-analysis of existing literature on the effect of enhanced-efficiency fertilizers on soil N_2O emissions (Akiyama *et al.*, 2010) concluded that the use of N-fertilizers with nitrification inhibitors does significantly reduce N_2O emissions in a range of 31–44%. The low N losses by N_2O emissions in willow coppice indicate an effective usage of added N in this cropping system. A high biomass yield is sought in crop systems for energy production; hence, trace gas emissions have to be put in proportion to the obtained yield. CH_4 fluxes in nonflooded agro-ecosystems can be only marginally influenced by management practices and do not play a decisive role in GHG emissions. Soil N_2O emissions, however, can be influenced directly by fertilization practices; furthermore, N_2O plays an important

Table 4 Mean yields of maize, Miscanthus and willow; up scaled soil N_2O emissions throughout 1 year, percentage of direct N_2O-N losses of fertilized nitrogen and soil N_2O emissions relative to yields (DM = dry matter)

Cropping system	Yield (t DM ha^{-1} yr^{-1})	N_2O emission (g N_2O ha^{-1} yr^{-1})	Perc. (%) of N_2O-N losses of NH_4^+-stabilized fertilizer [kg N_2O-N (kg N fertilized)$^{-1}$]	Relative N_2O emission [g N_2O (t DM yield)$^{-1}$]
Maize 0 kg N ha^{-1} yr^{-1}	8.4	229	–	27.2
Maize 240 kg N ha^{-1} yr^{-1}	20.2	3566	0.95	176.5
Miscanthus 0 kg N ha^{-1} yr^{-1}	16.8	−49	–	−2.9
Miscanthus 80 kg N ha^{-1} yr^{-1}	20.9	1407	1.12	67.3
Willow 0 kg N ha^{-1} yr^{-1}	7.1	−1	–	−0.2
Willow 80 kg N ha^{-1} yr^{-1}	14.4	48	0.04	3.3

role in the stratospheric ozone chemistry (Crutzen *et al.*, 2008). In this light, we related the mean yields of the different cropping systems to the soil N_2O subsumption to evaluate which system is favourable in terms of relative soil N_2O emissions. The N-fertilized maize cropping system showed the highest soil N_2O emissions per t DM yield. With 27 g N_2O t^{-1} DM^{-1}, the unfertilized maize also had relatively high soil N_2O emissions per DM yield compared with the other unfertilized crops. Very low relative soil N_2O emissions were found for unfertilized Miscanthus and for willow, irrespective if fertilized or not (Table 4). This result indicates a remarkable advantage of perennial energy crops in terms of N_2O avoidance compared with annual crops such as maize.

Conclusion

Data from this 1-year experiment showed a seasonal trend of soil CO_2 emissions, which was triggered mainly by soil temperature. Since the measuring design did not allow distinguishing between heterotrophic and autotrophic respiration or plant litter inputs, soil CO_2 emissions were not included in the trace gas emission subsumption. Soil N_2O emissions varied notably between the cropping systems dominantly influenced by N-fertilization. Over the course of 1 year, N-fertilized maize plots showed high soil N_2O emissions compared with the other cropping systems. The application scheme of N-fertilizer in Miscanthus, consisting of a broadband application on the soil surface in early spring, seemed to be unfavourable, since distinctive gaseous losses were observed in the following period. Smallest soil N_2O losses were observed for willow coppice, for both N-fertilized and unfertilized plots. This was attributed to a late N fertilizer application in conjunction with an intensely rooted top-soil. Soil CH_4 fluxes were mainly negative and had a minor effect on the trace gas subsumption. Soil N_2O emissions of the different cropping systems relative to yields showed a substantial benefit of the perennial crops. Unfertilized Miscanthus provided high yields of 16.8 t DM (mean of 5 years) connected with a negative N_2O subsumption; thus suggesting that an unfertilized or, in the long run, a very moderately N-fertilized Miscanthus planting is to be recommended as a bioenergy crop at the experimental site in terms of soil N_2O emissions.

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