Title
Greenhouse gas emissions in biogas production systems

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Introduction
Concerns about the long-term sustainability of fossil fuel consumption and of greenhouse gas (GHG) emissions from fossil sources brought renewable energy crops to the forefront. However now it is discussed whether GHG emissions during agricultural production of bio-energy crops would negate GHG emission savings (Crutzen et al. 2008). Biogas production from organic residues and/or plant biomass is one of the most efficient production lines among these upcoming technologies, but substantial amounts of fermentation effluent (biogas waste) remain after fermentation which may serve as valuable nutrient source for agricultural production but may also lead to new GHG emissions (Clemens et al. 2006). CO₂, CH₄, and N₂O are the major greenhouse gases produced in soils, and with the exception of wetland crops such as lowland rice, N₂O is the most important greenhouse gas that is emitted from agro-ecosystems, as it has a 300-fold higher global warming potential than CO₂ (IPCC 2007). The two biological processes of nitrification and denitrification are responsible for its production (Weier et al. 1993).

In most agricultural landuse systems organic and inorganic fertilizers are the major contributors to anthropogenic N₂O emissions (Mosier et al. 1998). Chemical and physical properties of organic and inorganic fertilizers largely affect their microbial turnover, and improved knowledge of soil and fertilizer interactions may serve to understand and eventually reduce fertilizer-borne N₂O emissions. Often in Western Europe, energy crops are grown with high external inputs including organic or inorganic nitrogen fertilizers, and also biogas fermentation remains (biogas waste). Biogas waste and other organic fertilizers contain large amounts of available carbon, nitrogen and other nutrients that accelerate soil microbial activity. In previous works we have shown that denitrification is the dominant microbial process producing N₂O in the initial period after application of biogas waste and other organic manures (Senbayram et al. 2009). As denitrification produces N₂O and N₂ at different rates it is important to consider effects of organic fertilizers on soil microbial processes and those on greenhouse gas production specifically.

Therefore the present study aimed at evaluating two favourable biogas crops in two agro-ecological regions of Northern Germany for their productivity and their environmental impact. It focused on nitrous oxide, carbon dioxide, and methane fluxes from soil under various nitrogen fertilizer rates and different fertilizer types. The objectives of our study were i) to obtain year-round trace gas flux data of two biogas crops in two agro-ecosystems in Northern Germany, and ii) to evaluate effects after application of biogas waste as manure in comparison to other organic and mineral fertilizers.

Material and Methods
A 2-year field experiment was conducted at two sites with different soil type and soil fertility but similar temperate maritime climate (ø 684 mm, 9.0 °C). The soil type at site ‘Karkendamm’ was a gleyic Podzol, derived from glacio-fluvial deposits of the last glaciation, with a texture being dominated by sand. Site ‘Hohenschulen’ had a Luvisol with sandy loam texture. As silage maize is currently the standard crop grown for biogas fermentation purposes, we compared silage maize to alternative bioenergy-crops. Thus at site ‘Hohenschulen’ we evaluated trace gas fluxes from silage maize and wheat (whole crop) - Italian ryegrass (catch crop) rotation that was grown in parallel plots (12 x 12 m) with both main crops in both years. At site ‘Karkendamm’ maize monoculture was compared to a four-cut silage grass system. We evaluated three levels of nitrogen fertilizer rates (0, 120, 360 kg N ha⁻¹ for maize and wheat; 0, 160, 480 kg N ha⁻¹ for grassland), with the highest level - chosen for modeling purposes - being clearly beyond
recommended rates. Three forms of fertilizers/manures were given: calcium ammonium nitrate, cattle / pig slurry, biogas waste. Fertilizers were applied in 2 to 4 split dressings. Pre-emergence manure dressings were immediately incorporated into the soil; later dressings were applied using trailing hoses only.

All trace gas flux measurements were done with the closed-chamber technique (Hutchinson and Mosier 1981). At the onset of the experiment, basal PVC rings (60 cm in diameter x 10 cm height) were pressed into the soil (5 cm depth) of all plots. For the measurements, PVC chambers (60 cm diameter x 25 cm height) were sealed onto the basal rings with butyl rubber band and gas samples were taken with 12 mL evacuated Exetainers (Labco, High Wycombe, UK) 0, 20 and 40 minutes after chamber closure. For the cutting of the grass, basal rings were removed and re-installed afterwards. Trace gas fluxes were monitored daily after fertilizer applications with successive expansion of the sampling intervals to up to one week. Gas concentrations in triplicate samples were analyzed by FID/ECD gas-chromatography following a set-up modified after Loftfield et al. (1996). After each N application, top soil samples (15cm depth) were taken daily for a period of 1 week, followed by sampling once a week until the end of the experiment. For the analysis of soil mineral N, soil samples were extracted with 2M KCl solution (1:4 w/v) for 1 h. The extracts were filtered using Whatman 602 filter paper and stored at -20°C until analysis. The concentrations of NH$^+_{4}$ and NO$_3^-$ in soil extracts were measured colorimetrically using a TRAACS 800 autoanalyzer (Bran and Luebbe, Norderstedt, Germany). We used time domain reflectometry (TDR) technique (Dobson et al. 1985) to measure soil moisture dynamics. Soil temperature (5, 10 cm depth), air temperature (30 cm above surface), and precipitation (100 cm above surface) were automatically logged as daily means/sums at the meteorological station.

Cumulative N$_2$O emissions were calculated by linear interpolation between measured daily fluxes. Emission rates were expressed as arithmetic means of the three replicates and log-transformed for statistical analysis. Tukey's HSD post-hoc test was used to reveal significant pairwise differences among treatments. Statistical analyses were done using SPSS version 13.0 (SPSS Inc., Chicago, IL, USA), with $p < 0.01$.

**Results and Discussion**

Greenhouse gas emissions of all biogas crops were strongly dominated by N$_2$O emissions. There were very short CH$_4$ emission events immediately after application of slurry and biogas waste, but these small fluxes were attributable to physical volatilization of dissolved CH$_4$. N$_2$O flux patterns in all crops usually followed fertilizer application events at both sites. However, in spring 2007 we observed a pronounced delay due to very dry soil conditions at both field sites (Fig. 1, showing site Karkendamm only). In 2008, more evenly distributed rainfall caused significantly higher N$_2$O emission at both sites. In 2007 the greatest daily N$_2$O flux with grassland soil was 0.1 kg N$_2$O-N ha$^{-1}$ day$^{-1}$ whereas it was 0.17 kg N$_2$O-N ha$^{-1}$ day$^{-1}$ in 2008, starting immediately after fertilizer application. Consequently, as there was much more rainfall in the 2008 growing period compared to 2007 especially in early spring, the level of N$_2$O emission in 2008 was 27% higher. The great relevance of high soil moisture on N$_2$O losses has been reported earlier, as the main factors driving the denitrification process are redox potential, substrate and oxygen diffusion which strongly depend on water availability and the water-/air-filled pore space in soil (Dobbie and Smith 2001). At both of our study sites significant N$_2$O fluxes were always observed earlier in maize compared to the other crops tested. Flux patterns furthermore indicated that the later onset of soil water consumption by transpiration and of mineral N uptake by maize contributed to 20 – 30 % higher N$_2$O fluxes in maize compared to the
other tested crops (Fig 2). Consistently soil moisture and soil mineral N concentrations were significantly higher in maize soil compared to both, grassland and wheat soil in early spring. Thus, apart from soil moisture effects we anticipate that the great nitrate uptake potential of wheat and particularly grass that has been reported for growing swards in spring (Stevens and Laughlin 1997) contributed to lower N\textsubscript{2}O fluxes with wheat and grass in spring. Surprisingly in our study more than 70% of the N\textsubscript{2}O emissions occurred after the third fertilizer dressing in grassland soil (July-Sep.) whereas the latter contributed only 15-25% in maize soils. Furthermore, soil moisture was slightly higher in grassland soils than in maize soils in this period. During winter time we did not observe significant N\textsubscript{2}O emission in any treatment. This result does not agree with findings by Lampe et al. (2006) (at a neighbouring grassland stand with autumn grazing) and Kammann et al. (1998). Here they reported that 15-41% of the annual N\textsubscript{2}O emitted during winter on grassland. However, almost no direct N\textsubscript{2}O emission over both winter periods (2007-2009) and at both field sites cannot be explained at present.

Besides the clear effect of maize vs. other crops tested, our study revealed a strong effect of soil texture. Overall at site Hohenschulen with its loamy soil, N\textsubscript{2}O emissions were at least 3 times higher than in all crops and treatments examined at the site Karkendamm with sandy soil (Fig. 2). Both sites had similar climate thus higher N\textsubscript{2}O emission at site Hohenschulen can only be attributed to differences in soil properties. Soil physical properties, such as porosity, bulk density and soil aeration are important factors regulating N\textsubscript{2}O fluxes from soils (Skiba and Ball 2002; Pihlatie et al. 2004). Di and Cameron (2006) measured N\textsubscript{2}O fluxes from stony silt loam...
and fine sandy loam soil and applied urea and urine. In their study annual N₂O emissions were 1.6-fold higher with sandy loam soil compared to stony silt. In a similar way, Piilhatie et al. (2004) reported 2.5 and 4-fold higher N₂O emissions from clay and peat soil compared to a loamy sand in an incubation experiment. However, with respect to the texture effect it has to be stressed that at site ‘Karkendamm’ with its sandy soil, substantial nitrate leaching was found which highlights the relevance of indirect N₂O emissions, e.g. occurring in aquifers but resulting from leached NO₃⁻. Data on NO₃⁻ and NH₃ losses will be used for the assessment of indirect N₂O emissions that may compensate for lower direct N₂O emissions to some extent.

Figure 2. Cumulative first-year N₂O emission (March 2007 to February 2008) in Hohenschulen (A) and Karkendamm (B). Control without N; N1 and N2 = 120 and 360 kg N ha⁻¹ (maize, wheat), and 160 and 480 kg N ha⁻¹ (grassland). Means of 3 replications ± standard error.

N₂O emissions increased with increasing fertilizer rates at both field sites. In Karkendamm cumulative N₂O losses varied from 0.2 kg N₂O-N ha⁻¹ with unfertilized soils to 2.3 kg N₂O-N ha⁻¹ on the soils receiving highest N rates whereas it was 2 kg N₂O-N ha⁻¹ with unfertilized soils and 12 kg N₂O-N ha⁻¹ on the soils receiving highest fertilizer/manure N dressings in Hohenschulen. Over all treatments N₂O-N fluxes were equivalent to 0.5 % of the fertilizer N applied in Karkendamm and 1.7 % in Hohenschulen.

Finally the present study showed that there were similar nitrous oxide losses from soils supplied with biogas waste compared to mineral N and cattle or pig slurry. This finding is in
line with additional experiments conducted under controlled conditions, where there were no differences in N\textsubscript{2}O emissions after application of mineral N or biogas waste when the soil moisture was moderate (65 % WHC) (Senbayram et al. 2009). However, under conditions of high soil moisture (85 % WHC), biogas waste amended soil showed much higher emissions than soil supplied with mineral N. There, based on stable isotope labeling, we also reported that denitrification was the major process producing N\textsubscript{2}O in biogas waste or animal manure amended soil. This strengthens the view that all effects of N rates, fertilizer types and timing of fertilizer application on N\textsubscript{2}O emission very much depend on other variables that drive denitrification; i.e. soil physical and chemical properties and in particular the availability of water.

**Conclusions**

The present study provides a very good basis for the assessment of direct emissions of greenhouse gases from relevant biogas crops in North-West Europe. At the site with light-textured soil, low direct N\textsubscript{2}O emissions were observed. With this soil N\textsubscript{2}O emissions were 20-30% higher with maize, the most wide-spread crop used in biogas fermentation. However here maize also had 50-60% higher dry matter, hence energy production than grass. So maize showed a clearly greater potential than grass to replace fossil fuel at sites with sandy soil texture. This can at least partially compensate higher N\textsubscript{2}O emissions. In contrast with the loamy soil, wheat yielded only 15-25% less dry matter than maize whereas N\textsubscript{2}O emissions were 30-80% higher in maize. A specific difference in N\textsubscript{2}O emissions after use of animal manure or biogas waste has not been found in the present study. But the general high level of emissions after intensive fertilizer use at the loamy site stresses the need for fertilizer/manure technologies to reduce direct N\textsubscript{2}O emissions.

**References**


