

Gas chromatographic studies on soil emission of nitrous oxide in dependence on nitrogen fertilising

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Abstract

The emissions of greenhouse gases and other gases, which affect the environment, influence the environmental balance of renewable raw materials. Fertiliser-induced emissions of nitrous oxide depend on the type and level of fertilisation. Different emission rates have been reported in literature in dependence on the type of fertiliser, type of soil, type of cultivated plants, and climatic conditions, ranging from less than 0.1 % and up to about 2.5 % for N₂O-N in relation to fertiliser-N. When the environmental impacts of agriculture are evaluated, this conversion factor plays an extraordinary role, especially in the climatic evaluation of the non-food sector, as it is the case in Germany (UBA 1993 and UBA 1999).

In the study reported here, soil emissions were measured in an experimental field, where different short rotation crops were cultivated for use as biofuels. The experimental field was separated into 40 plots. The plots were fertilised with different nitrogen levels (125 kg N ha⁻¹ a⁻¹, 75 kg N ha⁻¹ a⁻¹ and plots with no fertilisation). Gas samples of 100 ml were taken from cover boxes with a volume of 64 litres four times per week and analysed by means of a gas chromatograph.

The soil emission of nitrous oxide showed a considerable variability. The accumulated mean values of fertilised and non-fertilised plots for rye and for orchard grass resulted in an annual emission of 170 mg N₂O m⁻² (1.1 kg N₂O-N ha⁻¹). In case of plots with triticale, 155 mg N₂O m⁻² (1.0 kg N₂O-N ha⁻¹) was found. The lowest emissions were observed in plots with poplars (120 mg N₂O m⁻²; 0.8 kg N₂O-N ha⁻¹) and willows (90 mg N₂O m⁻²; 0.6 kg N₂O-N ha⁻¹). The difference between background emissions (plots with no fertilisation) and emissions from fertilised plots led to an N₂O-N emission factor of 0.2 %.

Introduction

Nitrous oxide (N₂O) is produced by micro-organisms during denitrification of nitrate (NO₃⁻) and nitrification of ammonium (NH₄⁺). In soil, mineralisation of humus and microbial conversion of organic substance generate nitrate and ammonium. These soil processes have been studied and results were published for several decades (e.g. Papen 1990, Swert 1996, Vermoesen 1996). Nitrogen fertilisers increase the soil concentration of nitrate and/or ammonium. Then, fertiliser-enhanced emissions may occur. Different emission rates have been reported in literature in dependence on the type of fertiliser, type of soil, type of cultivated plants, and climatic conditions. There is a wide span of emission rates between less than 0.1 % and up to about 2.5 % for N₂O-N with reference to the nitrogen content of the fertilisers (e.g. Wintzer 1993, Beese 1994, IPCC 1995 and 1996). All recent work regarding national inventories of emissions of nitrous oxide is based upon a linear relation between soil emissions and fertiliser input (Emission N₂O-N = background + k * fertiliser-N). This relation was first published by Bouwman (1989), and was derived by regression analysis of available data a decade ago. The discussions on the background emissions and on the conversion factor k for direct emissions are still going on, the span of consideration for k is from 0.25 % for soils with low emissions up to k = 2.25 % for soils with high emissions (OECD 1995, IPCC 1995 and 1996). Moreover, this factor is one basic value in the IPCC guidelines for N₂O inventories. Following the IPCC methodology, agricultural emissions for any country in the world can be estimated using FAO data as input (Kroeze and Moiser 2000). When the cultivation of energy plants is evaluated regarding environmental effects, this conversion factor plays an extraordinary role. This is one main point in the ongoing controversial discussion in Germany, triggered by two UBA reports.

The German Federal Environmental Agency (Umweltbundesamt - UBA) twice published (UBA 1993 and UBA 1999) studies on the environmental impacts of biofuel for diesel engines. According to these reports (1993 - "Eco-balance for rape-oil"; 1999 - "Current evaluation of the use of rape-oil/RME in comparison to diesel fuel"), nitrous oxide (N₂O) is the one main source for negative contributions to the environmental balance of diesel fuel substitutes. In the study of 1993, nitrogen fertiliser-induced emission of N₂O-N were assumed to be in the range from 2% to 3%, likely nearer to 3%. Since N₂O is a greenhouse gas and contributes to global warming about 250 times more effectively than carbon dioxide (CO₂), even low N₂O emissions can counterbalance the CO₂ advantage of biofuels. Another environmental impact of N₂O, damaging the ozone layer, was an essential part of

the report of 1999. Therefore, in the course of the last decade, several authors started research on the emission of N_2O in dependence on soil type, fertilisation and cultivated plants (e.g. Heinemeyer et al. 1995&1998, Hellebrand&Scholz 1997, Teepe 1999). Measurements of the N_2O fluxes from forest soils located in an area of intensive animal breeding showed that the fluxes were in the range of $1-11.5 \text{ g } N_2O-N \text{ ha}^{-1} \text{ d}^{-1}$ ($0.3-4 \text{ kg } N_2O-N \text{ ha}^{-1} \text{ a}^{-1}$). The effect of water-filled pore space (WFPS) of the mineral layer on the N_2O flux was highly significant. The effect of the moisture content of the organic layer was less pronounced. The lowest N_2O emission was found for the moisture combination of 15% for WPS and 100% for the organic layer. The highest emission occurred for the highest moisture levels of both layers (Vervaeet et al. 2000). The effect of compost and nitrate additions on N_2O emissions was studied in anaerobic soil incubations. For a loamy sand, an immediate and high N_2O production was observed in all treatments as long as nitrate was available. The loamy sand produced higher amounts of N_2O than the loamy soil (De Wever et al. 2000). During nitrification, nitric oxide (NO) is emitted to the atmosphere in addition to N_2O . Until now, there is a large uncertainty in the NO emission rate from cultivated soils (Beese 1994). Tsuruta and Akiyama (2000) studied the NO and N_2O emissions from upland soils with the application of different types of nitrogen fertiliser. The total emission rate of NO-N was in the range 0.1 % and 0.5 % of the total nitrogen applied. Additionally, the emission ratio NO/ N_2O showed a wide span between 0.1 up to 15 in dependence on the type of fertiliser and moisture content.

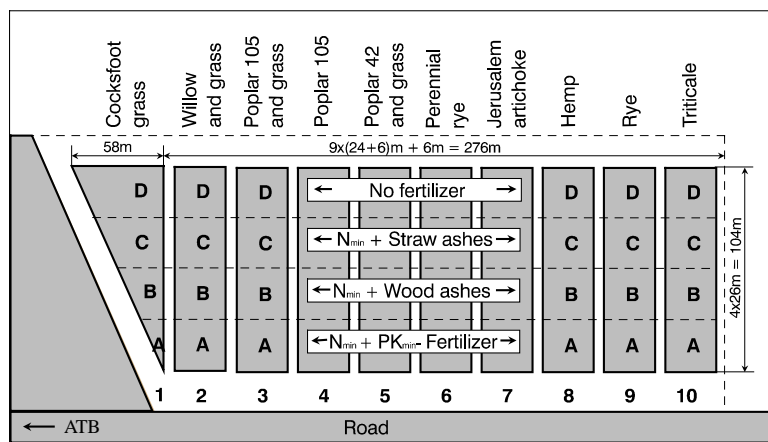


Fig. 1 Experimental field for cultivation of energy plants. Plots with different plants (1 ... 10) and different type of fertilising (A ... D) are indicated.

Different land use changed the fluxes of the greenhouse gases N_2O and CH_4 (Teepe 1999). Teepe studied soil emissions from four sites (poplars, rape, oaks, and fallow land) by gas flux chambers and a gas chromatograph (GC). The results of this two-year study were based upon weekly measurements. Great variations and a high dynamic for N_2O -emissions were observed. The lowest emissions, in the range $0.2 - 0.6 \text{ kg } N_2O-N \text{ ha}^{-1} \text{ a}^{-1}$, were observed for poplars. Fallow land, rape fields and oak forest had a higher level of N_2O -emissions between $1.0 \text{ kg } N_2O-N \text{ ha}^{-1} \text{ a}^{-1}$ and $2.5 \text{ kg } N_2O-N \text{ ha}^{-1} \text{ a}^{-1}$. Additionally, the type of soil determined the N_2O soil emissions. On average, having the same crop rotation, 1.5% of fertiliser-N escaped as N_2O -N from sandy loam, whereas the emission from loamy silt were only 0.8% (Heinemeyer et al. 1995, Schmädeke 1998). Since the N_2O emission factors depends on local conditions, the aim of this study was to find out results typical for loamy sand, the type of soil, which is prevalent in the German State of Brandenburg.

Materials and Methods



Fig. 2 Gas flux chamber.

The measurements were performed in an experimental field. The soil had sufficient homogeneity. The mean and standard deviations (s) of 40 soil samples were (Pagel 1995, Scholz et al. 1999): clay content 6.4 % (s: 1.3 %), organic carbon content 0.91 % (s: 0.14 %) and pH value 6.0 (s: 0.34). The field had been subdivided into 40 plots (624 m^2 each, except Orchard grass - also called Cocksfoot grass; Fig. 1). Ten different plant varieties or plant combinations were arranged as columns, and the different kinds of fertilisation were realised in four rows. There were plots without fertilisation (D), and plots with different levels of nitrogen fertilising (A: $150 \text{ kg } N \text{ ha}^{-1} \text{ a}^{-1}$; B and C: $75 \text{ kg } N \text{ ha}^{-1} \text{ a}^{-1}$) supplemented by PK-fertiliser (A), wood ashes (B), and straw ashes (C).

In the years 1997 and 1998, utilising gas flux chambers and FT-IR gas analysis (Hellebrand & Scholz, 1997&1998), soil emission measurements were performed once a week. Since 1999, the emission measurements have been performed four times a week by means of an automated GC (Loftfield 1992&1997). On measuring days, gas samples were taken from 25 gas flux chambers at different plots (cover boxes, Fig. 2).

The gas flux chambers (volume 64 dm³) had a volume to area ratio of V/A = 0.315 m. Two evacuated gas samplers (100 cm³ bottles with taps) were connected with each box. The first was opened, when the box was put on the water-sealed ring on the soil and the second one after about 100 minutes' collecting time. Then the boxes were removed and the samplers were connected with the GC-injection control system. In the course of the automatic GC-measurement the samplers were checked, the GC calibrated and the concentrations determined. Automated GC analysis improves the accuracy. The GC uses an ECD detector for N₂O and CO₂ concentration determination, for CH₄ an FID detector is applied. 64 samples can be analysed in one computer-controlled run. From the sample volume, 3 cm³ are injected. The N₂O detection limit is better than 5 ppb (5 · 10⁻⁹). At atmospheric mixing ratios, the coefficient of variation is 1.2% for N₂O measurements and 2.1% for CH₄ measurements. The standard deviation for complete flux determination (gas sampling and concentration measurement) is 0.6 µg m⁻² h⁻¹ (50 g ha⁻¹ a⁻¹) for CH₄ and 0.17 µg m⁻² h⁻¹ (15 g ha⁻¹ a⁻¹) for N₂O.

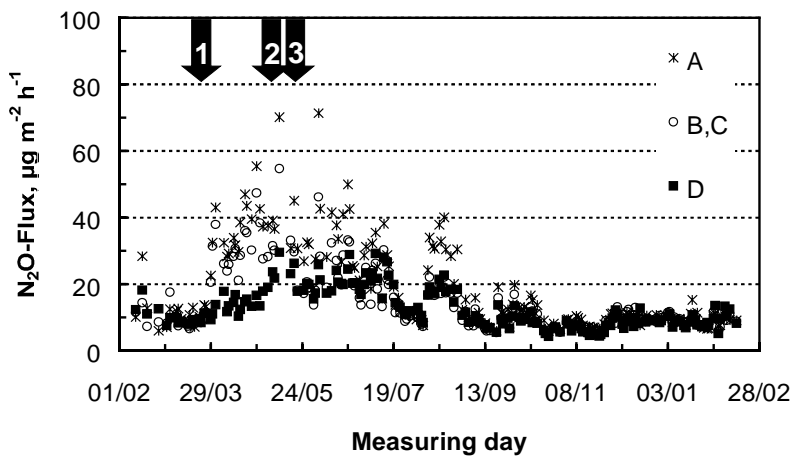


Fig. 3 Mean values of soil emission rates of N₂O from plots A (150 kg N ha⁻¹ a⁻¹), B & C (75 kg N ha⁻¹ a⁻¹), and D in the course of 1999/2000. Dates of fertilising are indicated by arrows.

- 1 - Fertilisation of A, B and C with 50 kg N ha⁻¹ on March 26, 1999
- 2 - Fertilisation of A with 50 kg N ha⁻¹ and B & C with 25 kg N ha⁻¹ on May 7, 1999
- 3 - Fertilisation of A with 50 kg N ha⁻¹ on May 18, 1999

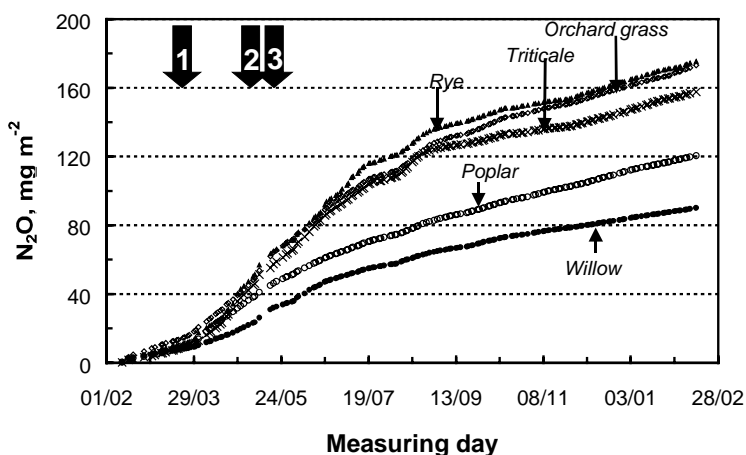


Fig. 4 Mean values (A, B, C, D) of accumulated N₂O-N soil emission from plant plots in the course of 1999/2000. Dates of fertilising are indicated by arrows (see Fig. 3).

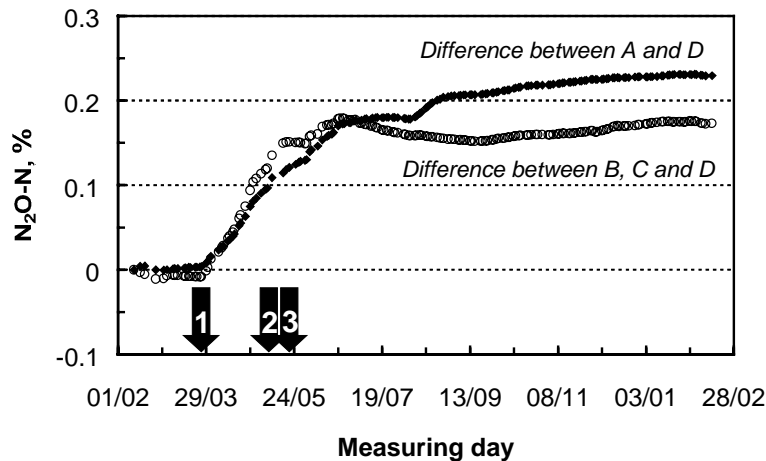
Results and discussion

In the first studies in 1997 and 1998, it was only possible to observe fertiliser-induced N₂O emissions in a qualitative way (Hellebrand & Scholz 1998). The weekly gas sampling and the lower FT-IR accuracy hampered a quantitative determination of the N₂O emission factor. Due to GC analysis, a significant improvement could be achieved. Increased emissions of N₂O were detectable from the first day after fertilising over a three-month time period (Fig. 3).

The variation between the different plants can be recognised by the accumulation of emissions (Fig. 4). Rye, triticale, and orchard grass gave similar values. The accumulated means of fertilised and non-fertilised plant plots result in an annual emission of 170 mg N₂O m⁻² (1.1 kg N₂O-N ha⁻¹) for rye and orchard grass. In the case of triticale plots, the annual emission is 155 mg N₂O m⁻² (1.0 kg N₂O-N ha⁻¹). Plots with poplars (120 mg N₂O m⁻²; 0.8 kg N₂O-N ha⁻¹) and with willows (90 mg N₂O m⁻²; 0.6 kg N₂O-N ha⁻¹) had lower emissions.

Fertiliser-induced N₂O emissions were observed in all fertilised plots. The lowest factor of 0.1% was found in willow plots, whereas rye and triticale reached values above 0.3%. It is not clear, if these differences between plant plots are really caused by the type of plants or, if these variations correspond to the natural stochastic variability of N₂O soil emissions. The results found here are supported by other studies (Teepe 1999). Teepe recorded lower emissions for plots with short rotation woods than for cultivated fields.

The N₂O emission factor was calculated by taking the accumulated difference between the mean values of all equally fertilised plots and the non-fertilised plots related to the level of fertilisation (Fig. 5). At both levels, 75 kg N ha⁻¹ a⁻¹ and 150 kg N ha⁻¹ a⁻¹, the emission factor is near 0.2%. This supports Bowman's linear relation for the loamy sandy soil of the experimental field. Of course, the results measured here are at the lower end of the range of N₂O emission factor, which is recommended by IPCC (1995&1996) for the fertilisation-based N₂O inventories. The emissions found here do not essentially influence the environmental balance of biofuels and other non-food crops. Measurements carried out over several years are necessary to support the findings.



Conclusions

Fertiliser induced emissions of nitrous oxide depend on the type and level of fertilisation. The N₂O emissions from plots with cereals and grass are nearly twice as high compared to plots with short rotation woods. Fertilisation of loamy sandy soils leads to low N₂O-N emissions of about 0.2% of the applied nitrogen. These low emissions of nitrous oxide do not essentially influence the environmental balance of the cultivation of biofuels and allow the fertilised production of non-food crops.

Fig. 5 Accumulated fertiliser-induced soil emission of N₂O-N as a ratio to the total annual fertilisation level in the course of 1999/2000 (Fertiliser-related mean values of all plots A, as well as mean values of all plots B and C as difference to the mean values of all plots D).

A: Fertilisation level of 150 kg N ha⁻¹ a⁻¹;

B, C: Fertilisation level of 75 kg N ha⁻¹ a⁻¹

D: No fertilisation

Dates of fertilising are indicated by arrows (see Fig. 3).

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