

Determination of Soil-Related Trace Gas Fluxes during the Cultivation of Renewable Raw Materials

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The emission of climate- and environmentally relevant trace gases affects the environmental balance of renewable raw materials. Depending on the intensity and the kind of nitrogen fertilizing, additional nitrous oxide or ammonia is emitted, and methane degradation in the soil is influenced. These trace gas fluxes are measured using the flux chamber method in connection with gas chromatography (GC) and Fourier-transform infrared spectroscopy (FT-IR). The GC technology provides the advantage of higher accuracy for some selected trace gases. The FT-IR measurements show that the release of ammonia caused by fertilizing with calcium ammonium nitrate is below the detection limit. The nitrous oxide emission factor measured at a loamy-sandy location is based on GC results. The average quantity of all fertilizer-induced N₂O-N emissions only amounts to approximately 0.2 % of the annual quantity of N-fertilizer applied. In addition, the GC measurements show that the degradation of atmospheric methane in the soil depends on the soil temperature. The mean annual value of methane degradation at the examined location amounts to 0.7 kg CH₄ ha⁻¹ a⁻¹.

Keywords

Emission rates, ammonia, nitrous oxide, methane, FT-IR spectroscopy, gas chromatography

Nitrous Oxide in the Environmental Balance

The publication of the ecological balance of rapeseed oil and rapeseed oil methyl-ester led to an intensive discussion about the emission of nitrous oxide (N₂O) and the quantities of nitrogen fertilizer that are really necessary for the cultivation of renewable raw materials [1]. Subsequently, several authors [such as 2-6] addressed this problem more intensively. The UBA (Federal Environment Office) study [1] reached the somewhat controversial result that, with regard to the impact on the climate, rapeseed oil as a diesel substitute does not provide substantial advantages as compared with diesel fuel. The environmental effects of the fertilizer-induced N₂O emissions are an important buttress of the argumentation chain. In the newer UBA study on the assessment of the use of rapeseed oil/RME in comparison with diesel fuel [7], N₂O formation is also a dominant negative factor in the environmental balance. In the ecological balance [1], a fertilizer-induced N₂O-N emission of 2% to 3% of the of the annual quantity of N fertilizer applied is assumed to be the probable range. Since nitrous oxide is a

very efficient greenhouse gas, this N₂O release may already be sufficient to cancel out the CO₂ credit in the greenhouse balance (climatic balance) achievable with bio fuel, depending on the cultivation intensity. With regard to the greenhouse effect, only additional CO₂ credits due to the comprehensive utilization of rape give biofuel an advantage over diesel fuel. In addition, nitrous oxide contributes to the depletion of ozone in the stratosphere and thus exerts a negative influence on the environmental balance of biofuels. In the more current evaluation [7], the conversion factors recommended by the IPCC [8] are employed. According to this assessment, the N₂O-N emission value amounts to 1.25% in temperate latitudes and is supposed to vary between 0.25% and 2.25% depending on the soil- and climatic conditions. Since N₂O release is dependent upon many factors, significant deviations may occur depending on the location.

Different land utilization alters the climate-relevant fluxes of the trace gases N₂O and CH₄ [2]. In reference [2], soil emissions at four locations (poplar plantations, rape field, oak forest, and permanent fallow land) were examined over a

two-year vegetation period using the GC flux chamber method and weekly sampling. Great differences in the amount and the dynamics of N₂O emissions were found. Moreover, it was shown that the intensity of utilization cannot be employed as an indication of the emission level. On poplar plantations, there were only low N₂O emissions in a range between 0.2 and 0.6 kg ha⁻¹ a⁻¹. Fallow land, rape field, and oak forest showed a higher level ranging between 1.0 and 2.5 kg ha⁻¹ a⁻¹. The soil type also exerts a dominant influence on the emission of nitrous oxide. If crop rotation was the same, an average of 1.5% of the applied nitrogen fertilizer was released as N₂O from sandy loam as compared with only 0.8% from loamy silt [4].

The evaluation of agricultural methods and technology, as well as the assessment of the consequences of a transition to new technical solutions and methods in the wake of alternative or future sources of income for farmers have become part of research and development in agricultural engineering with regard to growing standards for the environmentally friendly supply of agricultural products. Since raw materials from biological production are gaining in importance as an additional source of income for farmers, climatically and environmentally relevant consequences must be examined in addition to economic aspects. Therefore, quantitative information regarding the impact on soil, water, and air are necessary for the ecological assessment of techniques. Credits and additional emissions of carbon dioxide (CO₂), N₂O, and other gases - especially ammonia (NH₃) and methane (CH₄) - are integrated into the environmental balance of renewable raw materials. With the studies presented here, more detailed knowledge of the influence of the process design (fertilizing and plant species) on soil-related gas fluxes during the supply of renewable raw materials is to be gained.

Soil-Related Gas Fluxes

Biological activity in the soil leads to gas fluxes between the atmosphere and the soil. Many organisms need oxygen. De-

pending on the soil conditions and the bacteria population, some are able to use hydrogen (H_2), carbon monoxide (CO), or CH_4 as an energy source. For this reason, CH_4 can be released from soils or be consumed there. CH_4 emissions resulting from the anaerobic transformation of organic substance are mainly found in hydromorphic soils (moor locations) [9]. Sulphur-containing gases of biogenic origin (H_2S , C_2S , COS, CH_3SH , CH_3SCH_3 , etc.) are also formed under anaerobic conditions [10]. Due to the low flow rates, soil emissions of gases containing sulphur do not affect the environmental balance of renewable raw materials [11]. Carbon dioxide (CO_2) and the oxidation states of nitrogen (N_2O , NO, and NO_2) are emitted as gaseous metabolic products. Even though soil emissions of nitrogen monoxide (NO) can be significantly higher than N_2O emissions [12, 13], NO is of minor climatic relevance due to the short retention time in the atmosphere. NO and the oxidation product nitrogen dioxide (NO_2) must be taken into account in the environmental balance because they influence the tropospheric ozone content [14]. All in all, however, current knowledge about biogenous NO emissions are not yet sufficient for the integration of NO soil emissions into the environmental balance of renewable raw materials. For this reason, biogenous NO emissions are not evaluated in the UBA study [1], and in the study [7], NO_x field emissions are considered equal to zero.

Fertilizing as a form of nutrient supply influences biological activity in the soil. Since nitrate- [15] and ammonium ions [16, 17] may inhibit methanogenesis, nitrogen fertilizer should restrict methane formation. Methanotrophic bacteria oxidize methane and reduce or prevent methane emissions, especially in wetland areas, where more methane is produced [18, 19]. The oxidation of atmospheric methane is not yet understood in all details. At present, the molecular-biological processes during oxidation are still the object of research [20]. The known methanotrophic bacteria cannot oxidize methane in the atmospheric concentration range. An unknown subgroup of methanotrophic bacteria, which shows a specifically high methane affinity, is assumed to be the cause of atmospheric methane degradation [20]. Methane degradation in the soil leads to a concentration gradient. Since methane transport in the soil is mainly controlled by diffusion, it is considered certain that soil properties (such as pore volume, pore continuity, water content, and air capacity) determine the diffusion resistance in the soil and, hence, the degradation rates. On areas used for agri-

culture, different degradation rates must therefore be expected depending on soil type, soil cultivation, plant cover, plant species, and fertilizing. Furthermore, the soil is a source and an absorber of other hydrocarbons. Since these substance fluxes are far below the methane level [21], they have no influence on the greenhouse gas balance of renewable raw materials. Therefore, they will not be discussed here.

NH_3 losses during and after manure spreading cover a wide range depending on the spreading and other influencing factors [22]. Contact time with the atmosphere, the pH value, and the weather conditions are decisive for the volatilization of NH_3 . Few reliable data are available concerning NH_3 emissions from soils, i.e. after the working in or seeping in of ammonium-containing fertilizers and due to the ammonification of protein-containing organic substance (green manuring). The good water solubility, the fast ion exchange, and the high adsorbability of NH_3 at virtually all surfaces strongly inhibit NH_3 soil emissions. NH_3 stimulates nitrous oxide formation through the nitrification- and denitrification processes in the soil. For this reason, higher N_2O emissions are observed as a result of the application of ammonium-containing nitrate fertilizers than as the result of pure nitrate fertilizing [14].

Trial Plots and Gas Measurement Technology

Since the beginning of 1997, accompanying gas flux measurements have been taken on a trial field of the ATB [23], where the environmentally sound cultivation of renewable raw materials is studied under practical conditions. The distribution of the clay content (6.2%), the humus content (1.55%), and the pH value (5.74) over the trial area shows great uniformity [23, 24]. The field is divided up into 40 plots having $624 m^2$ each. Four plots each with one plant species are grouped into four fertilization levels (A - NPK fertilization with $150 kg N ha^{-1} a^{-1}$; B - wood ashes and $75 kg N ha^{-1} a^{-1}$; C - straw ashes and $75 kg N ha^{-1} a^{-1}$; D - reference field without fertilization). The gas samples are taken at fixed measuring points with different fertilization levels and plant species. For this purpose, flux chambers are put on the sealing rings embedded in the soil for a period of ca. 100 minutes (figure 1). The gas fluxes result from the concentration differences in the chamber between the beginning and the end of the measuring period. The increase in con-

centration in the gas flux chamber is determined by the properties of the flux chamber (volume-area relation), while the gas flux rate at the soil surface depends on the gradient of the gas concentration in the soil (or on the diffusion resistance). Studies on the concentration increase in the gas flux chambers on the measuring areas of the experimental field showed that the increase in concentration can be assumed to exhibit linear approximation even if the collection periods amount to 120 minutes [6]. In 1997 and 1998, weekly sampling and subsequent FT-IR gas analysis were carried out [6]. The sample volume corresponded to approximately 4% of the flux chamber volume. Since the beginning of 1999, the gas samples have been evaluated four times per week using an automated GC [25, 26]. The gas sample taken from the flux chamber during GC measurements is smaller than 0.2% of the chamber volume. Due to the measuring principle (examined, evacuated gas cylinder, automatic density check of the filled sample bottles before the GC measurement), only correct samples are used for further evaluation.

With the chosen measuring arrangement (gas cuvette with an optical measuring length of 20 m and a measuring volume of $5 dm^3$, nitrogen-cooled MCT detectors, 40-fold accumulation, gas-sampling bag with a volume of approximately $7 dm^3$, flux chamber volume $190 dm^3$ with a volume - measuring surface relation of $0.647 m$), the FT-IR gas analysis led to variation coefficients of 7% during the analysis of N_2O and CH_4 in atmospheric concentra-

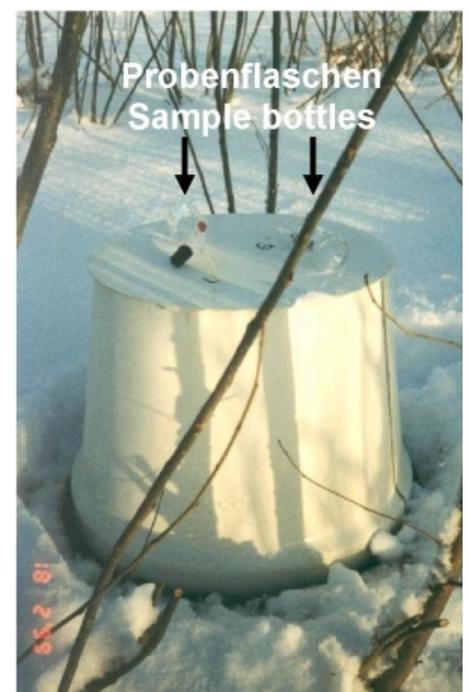


Figure 1: Gas flux chamber with two sample bottles on top

tion ranges and of about 20% during NH_3 measurement at very low concentrations (<0.5 ppm). Hence, the standard deviation during the establishment of the mass-related flux of all three gases lies at about $20 \mu\text{g m}^{-2} \text{h}^{-1}$ ($1.8 \text{ kg ha}^{-1} \text{a}^{-1}$). The greater insecurity in the measurement of the NH_3 concentration results from the strong adsorbability of NH_3 . The FT-IR detection limit for NH_3 , N_2O , and CH_4 is better than 10^{-7} (0.1 ppm) and can be improved through further accumulation. The systematic measurement error caused by calibration is partly compensated for by the difference measurements during gas flux analysis.

The GC technology applied here enables the measurement error during the flux measurement of the atmospheric trace gases N_2O and CH_4 to be reduced significantly. The instrument employed is an automated GC with FID- and ECD detectors (up to 64 sample bottles, automatic calibration at the beginning of each series of measurements, measuring process controlled by a PC, automatic storage of the measurement results in tabular form for further evaluation). The used sample volume is 3 cm^3 , the sample bottles can hold 100 cm^3 , and the volume of the gas flux chambers amounts to 64 dm^3 with a volume - measuring surface relation of 0.315 m. The three gases CH_4 (FID), N_2O , and CO_2 (ECD) are measured using the GC. In order to suppress the influence of a zero drift due to room temperature fluctuations, the sample bottles (initial and final concentration) which belong to one gas flux chamber are always measured one after another. In addition, the concentrations of the calibration gases are measured at the end of a series of measurements. This allows a zero drift to be established and corrected, if necessary. The detection limit is better than 10^{-8} , and the variation coefficient amounts to 1.2% for N_2O and 2.1% for CH_4 at natural air concentrations. The automated GC measuring instruments have a standard deviation of $0.6 \mu\text{g m}^{-2} \text{h}^{-1}$ ($50 \text{ g ha}^{-1} \text{a}^{-1}$) for CH_4 and $0.17 \mu\text{g m}^{-2} \text{h}^{-1}$ ($15 \text{ g ha}^{-1} \text{a}^{-1}$) for N_2O when examining the soil-related fluxes.

Emissions of Ammonia, Nitrous Oxide, and Methane

Ammonia

Even though nitrochalk was applied, i.e. a fertilizer which contains ammonium, no fertilizer-induced NH_3 emissions could be detected using the FT-IR measuring technology, in contrast to N_2O measurements. During the entire two year measurement period, there were only single peaks on

different plots, which are attributed to the depositing of animal excrement. Since nitrochalk consists of lime (CaCO_3) and ammonium nitrate (NH_4NO_3), the same quantity of both ammonium nitrogen and nitrate nitrogen are applied during fertilizing. In connection with the FT-IR- NH_3 standard deviation of $20 \mu\text{g m}^{-2} \text{h}^{-1}$, this would allow the conclusion to be drawn that fertilizer-induced NH_3 emissions on the plots with the highest fertilization level should be able to be observed as of approximately $2 \text{ kg NH}_3 \text{ ha}^{-1} \text{a}^{-1}$. The NH_3 detection limit of the flux measurements is better than $5 \mu\text{g m}^{-2} \text{h}^{-1}$. Therefore, it is likely that these emissions are significantly lower.

Nitrous Oxide

The first successful qualitative measurements of fertilizer-induced N_2O emissions

were already taken in the trial years 1997 and 1998 [6]. Only the weekly sampling and the measuring accuracy, which was insufficient for difference measurements in the atmospheric concentration range, prevented more precise quantitative evaluation. The transition to GC measurement technology and the quadrupling of the measurement frequency allowed the course of the emissions to be registered more precisely. Significantly increased nitrous oxide emissions occur as of the day after fertilizer application and can be detected for about three months afterwards (figure 2). The accumulation of emissions makes the differences between the individual plant plots visible (figure 3). Rye, triticale, and orchard grass show similar courses and annual sums. The accumulated average values of fertilized and unfertilized plant plots add

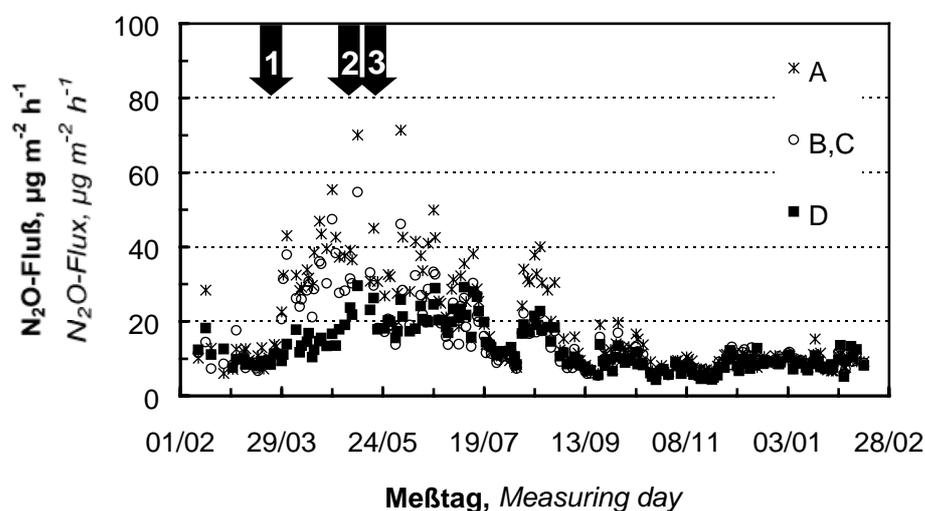


Figure 2: Mean values of N_2O soil emission rates from plots A ($150 \text{ kg N ha}^{-1} \text{a}^{-1}$), B, C ($75 \text{ kg N ha}^{-1} \text{a}^{-1}$), and D (not fertilized) over the course of 1999/2000. The dates of fertilizer application are indicated by arrows:

- 1 - Fertilization of A, B and C with 50 kg N ha^{-1} each on March 26, 1999
- 2 - Fertilization of A with 50 kg N ha^{-1} and of B and C with 25 kg N ha^{-1} each on May 7, 1999
- 3 - Fertilization of A with 50 kg N ha^{-1} on May 18, 1999

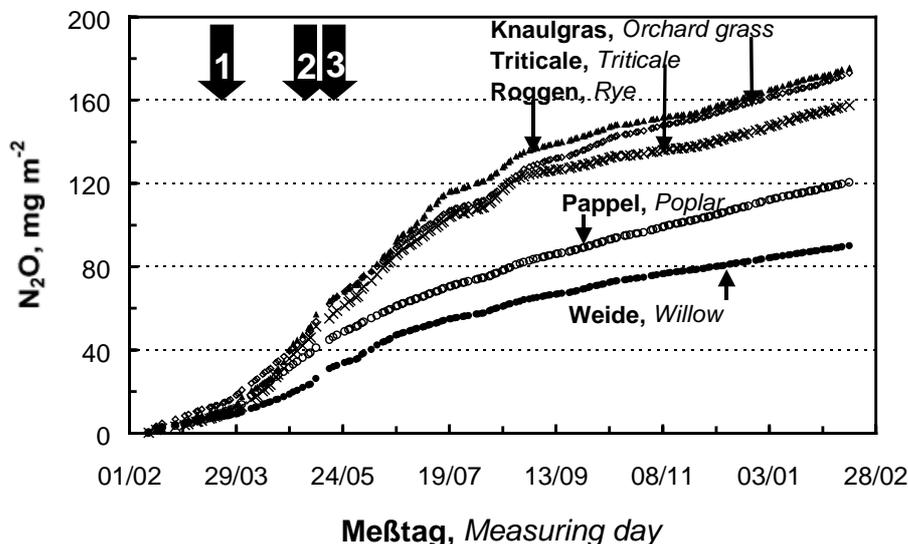


Figure 3: Mean values (A, B, C, D) of the accumulated $\text{N}_2\text{O-N}$ soil emissions on plant plots over the course of 1999/2000.

The dates of fertilizer application are indicated by arrows (see Fig. 2)

up to an annual emission of 170 mg N₂O m⁻² (1.1 kg N₂O-N ha⁻¹) on rye- and orchard grass plots and 155 mg N₂O m⁻² (1.0 kg N₂O-N ha⁻¹) on triticale plots. Annual emissions from poplar plots (120 mg N₂O m⁻²; 0.8 kg N₂O-N ha⁻¹) and willow plots (90 mg N₂O m⁻²; 0.6 kg N₂O-N ha⁻¹) are significantly lower.

Fertilizer-induced nitrous oxide emissions occur on all fertilized plots. Plots of willows (low emission level) and orchard grass (higher emission level) remain below 0.1% N₂O-N emissions in relation to the annual quantity of N applied (figure 4). Triticale and rye, however, exhibit stronger dynamics with N emission factors of more than 0.3%. Figure 4 shows seemingly decreasing emission rates on willow plots during the period before the first application of fertilizer. The same applies to the course of the values from rye plots as of June 1999. The reason for this development on these plant plots is the greater increase in accumulated N₂O emissions from the unfertilized reference plots during these periods. The difference between fertilized (A, B, C) and unfertilized (D) plots developed during the accumulation period of the individual plant species. All in all, it is still an open question whether these courses are really caused by the plants or rather by a stochastic local and time-related variation. The insights gained here are corroborated by studies from other authors. When wood plantations and field areas were compared, lower emissions were measured on areas used for the cultivation of short-rotation wood [2]. If the accumulated difference between the mean N₂O emissions of all equally fertilized plots are considered in relation to the quantity of N fertilizer applied (figure 5), it becomes clear that the fertilizer-induced N₂O emissions amount to 0.2% both at a fertilization level of 75 kg N ha⁻¹ a⁻¹ and at a level of 150 kg N ha⁻¹ a⁻¹. The results gained here at a loamy-sandy location thus also confirm the linear approach as an appropriate approximation for the measurement of N₂O emissions. However, the values measured here are at the lower end of the percentage range recommended by the IPCC [7] for the measurement of fertilizer-induced N₂O emissions. Therefore, the nitrous oxide emissions at the examined location impair the environmental balance of renewable raw materials only to a small extent. More measurements over several years are necessary to substantiate this value further.

Methane

In the measuring period as of 1997, virtually only „negative CH₄ soil emissions“, i.e. degradation of atmospheric CH₄ in the

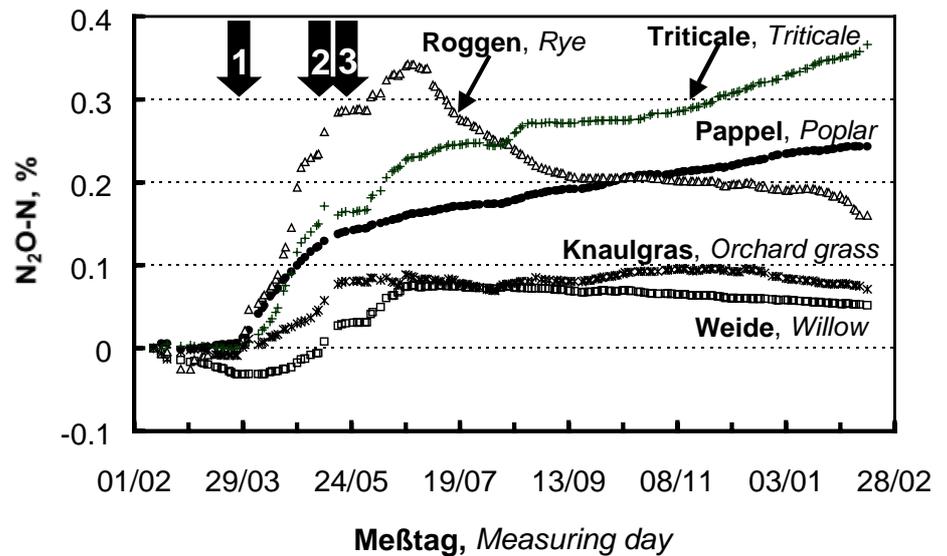


Figure 4: Accumulated fertilizer-induced soil emission ratio of N₂O-N on plant plots in relation to the total annual fertilization level over the course of 1999/2000 (weighted average of the accumulated N₂O-N emissions in relation to the application of N on plots A, B, and C; difference value as compared with the accumulated value of plot D at the individual plant locations). The dates of fertilizer application are indicated by arrows (see Fig. 2)

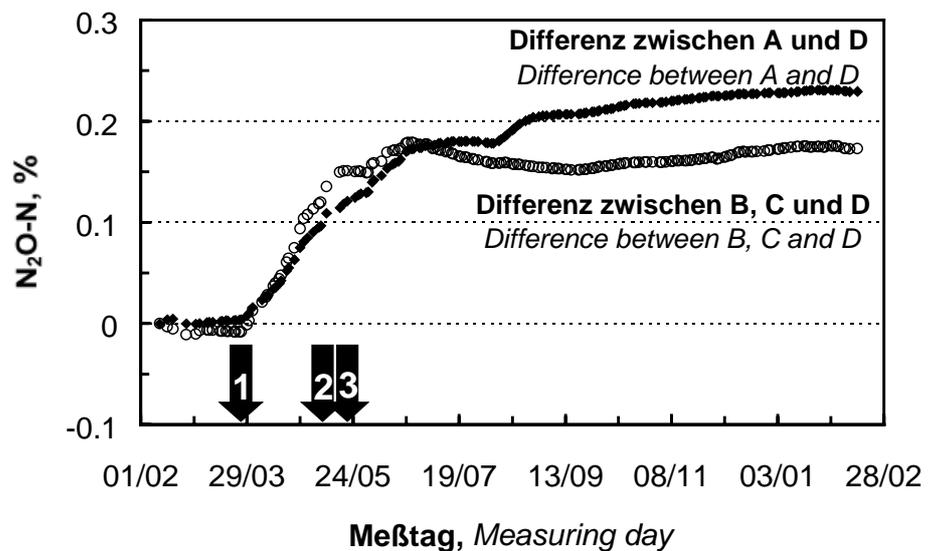


Figure 5: Accumulated fertilizer-induced soil emission ratio of N₂O-N in relation to the total annual fertilization level over the course of 1999/2000 (fertilizer-related mean values of all plots A and mean values of all plots B and C; difference value as compared with the mean value of all plots D). A: fertilization level of 150 kg N ha⁻¹ a⁻¹; B, C: fertilization level of 75 kg N ha⁻¹ a⁻¹. The dates of fertilizer application are indicated by arrows (see Fig. 2)

soil, have been measured. The gas-chromatographic analyses as of the beginning of 1999 allowed the quantitative measurement of the methane degradation rates (table 1). CH₄ degradation correlates very well with the soil temperature (figure 6). While from December until March the degradation rates diminish to values around 5 µg CH₄ m⁻² h⁻¹, measurement values between June and November generally range from 10 to 20 µg CH₄ m⁻² h⁻¹. Depending on the plant species, different fertilizer effects were established. On the willow plots, fertilizing reduced CH₄ degradation by 30%. On poplar- and triticale plots, however, there were no fertilizer-induced differences. On orchard grass-

and rye plots, a doubling of the degradation rates was measured at the fertilization level 150 kg N ha⁻¹ a⁻¹ (table 1). Further studies lasting several years are necessary to resolve the question of whether these effects are really plant-specific and not caused by local soil variability. As an average value over all plots, a CH₄ degradation rate of 700 g CH₄ ha⁻¹ a⁻¹ was established at the examined loamy-sandy location.

Conclusions

Measurements of soil-related gas fluxes with gas flux chambers and FT-IR gas

analysis over two vegetation periods enable the conclusion to be drawn that ammonium-containing mineral fertilizer does not lead to significant NH_3 soil emissions. The N_2O soil emissions from grain- and grass plots are almost twice as high as those from plots with short rotation wood. Independent of the fertilization level, nitrogen fertilizing at loamy-sandy locations only releases approximately 0.2% of the fertilizer nitrogen as N_2O . This emission factor is at the lower end of the percentage range recommended by the IPCC [8] for the measurement of fertilizer-induced N_2O -N emissions. Therefore, the nitrous oxide emissions at the examined location impair the environmental balance of renewable raw materials only to a very small extent. A clear, fertilizer-induced inhibition of the degradation of atmospheric methane in the soil cannot be proved. The results rather speak in favour of fertilizer-stimulated methane degradation at locations used for agriculture.

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Table 1: Degradation of methane in the soil ($\text{kg ha}^{-1} \text{a}^{-1}$) on plant plots at different levels of fertilization

Plants	Level of fertilization			Mean value
	0 $\text{kg N ha}^{-1} \text{a}^{-1}$	75 $\text{kg N ha}^{-1} \text{a}^{-1}$	150 $\text{kg N ha}^{-1} \text{a}^{-1}$	
Poplar	0.25	0.29	0.50	0.35
Willow	1.00	0.57	0.51	0.69
Rye	0.46	0.63	0.96	0.68
Orchard grass	0.52	0.75	0.98	0.75
Triticale	0.89	1.02	0.91	0.94
Mean value	0.62	0.65	0.77	0.68

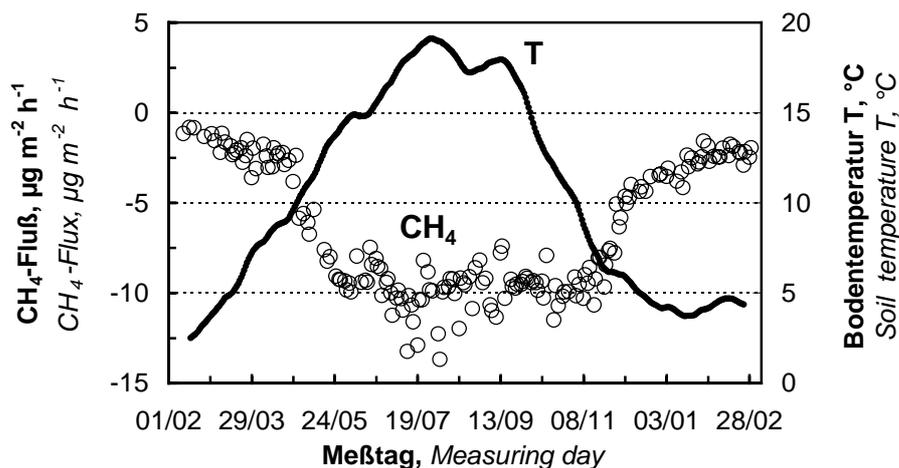


Figure 6: Soil temperature at a depth of 20 cm (sliding mean value of 30 days) and mean uptake of atmospheric methane by the soil over the course of 1999/2000

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