Biosphere - Atmosphere exchange of NH_3, NO and N_2O in an agroecosystem.

A. Gut, A. Blatter, M. Fahrni, A. Neftel and T. Staffelbach
Swiss Federal Research Station for Agroecology and Agriculture (FAL), Institute of Environmental Protection and Agriculture (IUL) CH - 3003 Bern, Switzerland
EMail: Andreas.Gut@iul.admin.ch

Abstract

In spring 1996 we started a four year research project to quantify the bi-directional exchange of nitrogen-containing trace gases between an agricultural ecosystem and the atmosphere. In the first year we intended to identify the most important processes that govern the exchange of nitrogen-containing trace gases between a field planted with winter wheat and the atmosphere during a vegetation period.

NH_3 profiles within the canopy were monitored with a continuous four channel analyser. We employed semipermeable membrane tubes to monitor vertical NO concentration profiles in the open pore space of the soil. N_2O profiles were taken periodically.

Preliminary results show that the NH_3 exchange is dominated by the upper part of the canopy. During daylight hours the highest concentration of NH_3 was found on the flag leaf/ear level. NO concentration in the soil was strongly enhanced in spring after the first rainfall following fertiliser application and was highest in the upper soil layer. N_2O concentration generally increased with soil depth. High concentrations were measured in spring, decreasing during plant growth. During fructification of wheat the N_2O concentration in deeper layers was elevated again.

1 Introduction

Emission of nitrogen containing trace gases account for up to 30% of the total nitrogen turnover of about 220 kT per year in Swiss agriculture (BUWAL [1]). Quantitatively the nitrogen loss in agriculture is dominated by NH_3, where the largest part is directly emitted during a few days after manure application on the field (e.g. Buijsman [2]). Another important loss is NH_3 emission from the soil/plant system. The concept of a compensation concentration has been

commonly used in this context (Schjoerring [3], Sutton [4]). At the compensation concentration production and uptake of a gas are equal. A compensation concentration exists for gases, which are consumed and produced in the same system, e.g. soil or plant canopy. The compensation concentration of NH_3 in the soil is generally low (Neftel [5]). Thus the soil is usually a sink for NH_3. NH_3 exchange is mainly influenced by the compensation concentration in the stomata of the plant. It is adjusted by pH and NH_3/NH_4^+ concentration in the apoplast.

NO and N_2O concentrations in the soil are determined by microbial activity. NO flux between soil and atmosphere occurs in both directions because atmospheric NO concentration above and below the compensation concentration of the soil can be found. In contrary the N_2O flux is usually directed from the soil into the atmosphere because the atmospheric N_2O concentration is below the compensation concentration of the soil (Conrad [6]).

The nitrogen cycle in the soil/atmosphere system involves fluxes of gases that are relevant from local to global scale:
The NH_3 compensation concentration of agricultural plants is often higher than the ambient concentration (Schjoerring [3]). As consequence agricultural fields take up almost no NH_3 from the atmosphere. NH_3 is therefore deposited over areas with semi-natural and natural vegetation with typically lower compensation concentrations (Sutton [4]). These types of vegetation will become overfertilised due to this input.

Nitrogen oxides are among the most important precursor substances for the production of tropospheric ozone on a regional to hemispheric scale (Crutzen [7]). Soils are important sources for NO (e.g. Johansson [8]).

N_2O emissions are relevant in the context of climate change and ozone destruction in the stratosphere on a global scale (Crutzen [9]).

The research project BAT (= Biosphere Atmosphere Transfer) is a four year research program which aims to quantitatively describe of the surface flux budget of nitrogen-containing trace gases on a regional scale. In this work we report results from the first year of measurements made during the growing season of 1996 in a field planted with winter wheat. We monitored the NH_3 concentration profiles in the plant canopy and vertical profiles of the NO concentration in the soil to identify the processes that drive the exchange of these gases with the atmosphere. N_2O concentration profiles of the soil were measured periodically.

2 Experimental set-up and Methods

2.1 Experimental set-up

The site (46°59'42''N; 7°11'02''E) was located 20 km north-west of Bern, in the Seeland, a flat rural area on the Swiss plateau with small agricultural plots

with different intensively managed crops. The elevation of the site is 436 m above sea level. The experiment took place in a field planted with winter wheat (*Triticum aestivum, „Arina Tamaro"*) and lasted from April 11 until July 29, 1996. The field was fertilised with 40 kg ha^{-1} NH$_4$NO$_3$-N on April 17 and harvested on August 1. Some properties of the soil are listed in Table 1.

soil type	bulk density	porosity	particle distribution	pH
Mollic Gleysol	1.1 g cm^{-3}	0.6 cm^3 cm^{-3}	9% sand 53% loam 38% clay	8.0

Table 1: Overview of the soil properties at the site

Vertical NH$_3$ profiles in the plant canopy and NO profiles in the soil were measured continuously during the experiment. Figure 1 gives an overview of the experimental set-up.
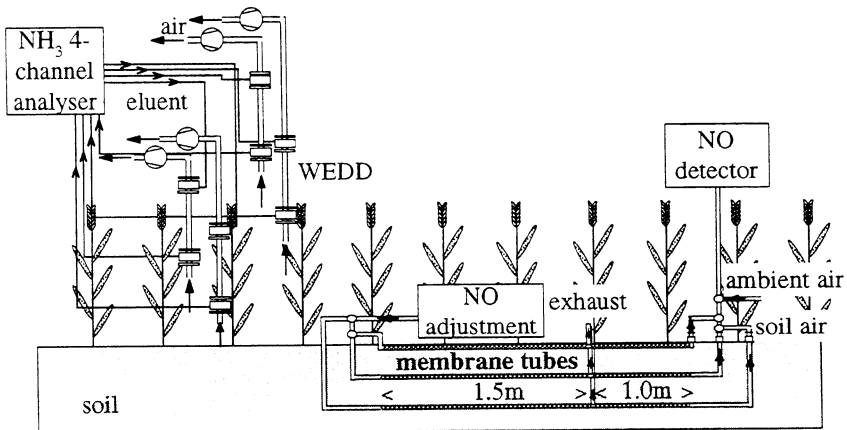

Figure 1: experimental set-up of the NH$_3$ and NO measurement. NH$_3$ was collected with wet effluent diffusion denuders (WEDD) and NO with hydrophobic, gas permeable membrane tubes

2.2 NH$_3$ measurement

NH$_3$ measurements were performed in and above the plant canopy. NH$_3$ was collected with a miniaturised wet effluent diffusion denuder (WEDD) described in Neftel [5]. Ammonium was analysed with a continuous four channel system. The ammonium concentration in the liquid is determined by its reaction with orto-phthalaldehyde in presence of sodium sulphite. Fluorescence

of the product is detected with a photomultiplier. The time resolution of the detector is 3 min and the detection limit 0.1 μg m^{-3}.

2.3 NO measurement

NO was measured at 2, 10, 25, periodically at 50 and 70 cm below the soil surface and 5 cm above the soil surface. NO in the interstitial air of the soil was collected with the hydrophobic, gas permeable polypropylene membrane tube Accurel® V8/2 PP (AKZO NOBEL, D-Wuppertal) (Gut [10]). The collection is based on diffusion, driven by the concentration gradient between the open pore space of the soil and the air flowing through the membrane tube. To prevent a concentration depletion of the soil gas the NO concentration of the air stream going into the membrane tube is adjusted to the measured concentration. The collection efficiency of the membrane tube is 100%. The concentration at four different levels of the soil/atmosphere profile was measured sequentially by switching the air flow every 15 min with valves. A profile was completed in 1 hour. The NO chemiluminescence detector is a CLD 780 TR (ECO PHYSICS, CH-Dürnten) with a detection limit of 0.01 ppbV at an integration time of 60 s.

2.4 N_2O measurement

The N_2O profiles in the soil have been measured fifteen times during the field experiment. The collection of N_2O in the interstitial air of the soil was made by connecting Tedlar bags with the membrane tubes and pushing ambient air through the tubes into the bags. The flow through the membrane tube and the length of the tube have been chosen to get a sample efficiency of 100% (Gut [10]). The samples were analysed with a tuneable diode laser absorption system (Aerodyne Research, Billerica, MA, USA) with a precision of 1% of the measured concentration.

2.5 Auxiliary measurements

Most of the commonly measured meteorological parameters were measured with a Campbell Bowen Ratio system. Global radiation was measured at 1.5 m above the soil surface. Wetness grids were installed at different heights in and above the canopy. Wetness grids detect condensed water, e.g. dew or rain, on surfaces by measuring the resistance between two conductors. If the surface of the grids is dry, the resistance is high and if the surface is wet the resistance is low. The parameters measured continuously in the soil were: Temperatures at nine depths between 0 and 50 cm and soil moisture at 2, 4, 10 and 25 cm.

NO$_3^-$ and NH$_4^+$ content of the soil was measured three times and organic carbon and total nitrogen twice during the campaign. C_{org} and N_{tot} were extracted from the soil by burning the samples at 950°C in an oxygen stream. CO_2 was measured with infrared spectroscopy and the products of N_{tot} in a

thermal conductivity cell. C_{org} is carbonate corrected. NH_4^+ was extracted from fresh soil with a 2 M KCl solution and NO_3^- with water. Both solutions were centrifuged and filtrated. NH_4^+ was measured with flow injection-fluorescence analysis and NO_3^- with ion chromatography. The results are shown in Table 2.

	C_{org}/N_{tot}		NO_3^- [ppm]			NH_4^+ [ppm]		
depth	25.3.	27.7.	25.3.	22.5.	17.7.	25.3.	22.5.	17.7.
0-10 cm	8.6	8.7	10.4	9.2	2.54	0.23	0.51	0.97
20-30 cm	10.2	8.8	5.5	1.0	2.0	0.14	0.36	0.91
68-72 cm	-	10.3	-	-	-	-	-	-

Table 2: NO_3^- and NH_4^+ concentration and C_{org}/N_{tot} ratio in 0-10, 20-30 and 68-72 cm soil depth together with the date where soil samples were taken

3 Results

3.1 NH₃ concentration profile in the canopy

The NH_3 concentrations within the canopy at 0.1, 0.5, 0.9 m (region of flag leaf and ear) above ground and the concentration above the canopy (1.2 m) are shown in Figure 2 together with the surface wetness on the soil surface and above the canopy, measured with wetness grids, and the global radiation. Two days at the beginning of June are presented, which show typical diurnal variations of the NH_3 concentrations in and above the canopy.
At the beginning of June 3 during night the soil surface is wet from the nocturnal dew and begins to dry at 9:00 the next morning. At 1.3 m above the soil surface or 0.1 m above the plant canopy the wetness grid detects humidity from 1:30 until 8:00. During this period with dew on the surfaces the NH_3 concentration in the canopy is below the concentration above the canopy and the lowest concentration is near the soil surface. When the surfaces are drying the ammonia in the liquid evaporates. The drying process near the soil takes place from 9:00 till 15:00. The concentration 0.1 m above the soil surface increases slowly. At 1.3 m above the soil surface the drying is fast and leads to peaks in the NH_3 concentrations at 0.5, 0.9 and 1.2 m.
Between the evaporation peak on June 3 and sunset on June 4 the NH_3 profile increases from the soil surface (0.1 m) to a maximum at the top of the plant canopy (0.9 m) followed by a decrease above the canopy (1.2 m). Only for two hours before midnight of June 4 the NH_3 concentration above the canopy is equal to the concentration at 0.9 m. During the night from June 3 to June 4 the surfaces in the upper canopy region remain dry. Therefore, there is no deposition and the evaporation peak does not occur in the morning of June 4. Towards midday a rise of the NH_3 concentration at all heights is measured. Between 17:00 and 21:00 the concentration at 0.5, 0.9 and 1.2 m rises sharply and then declines after 21:00 when dew formation begins.

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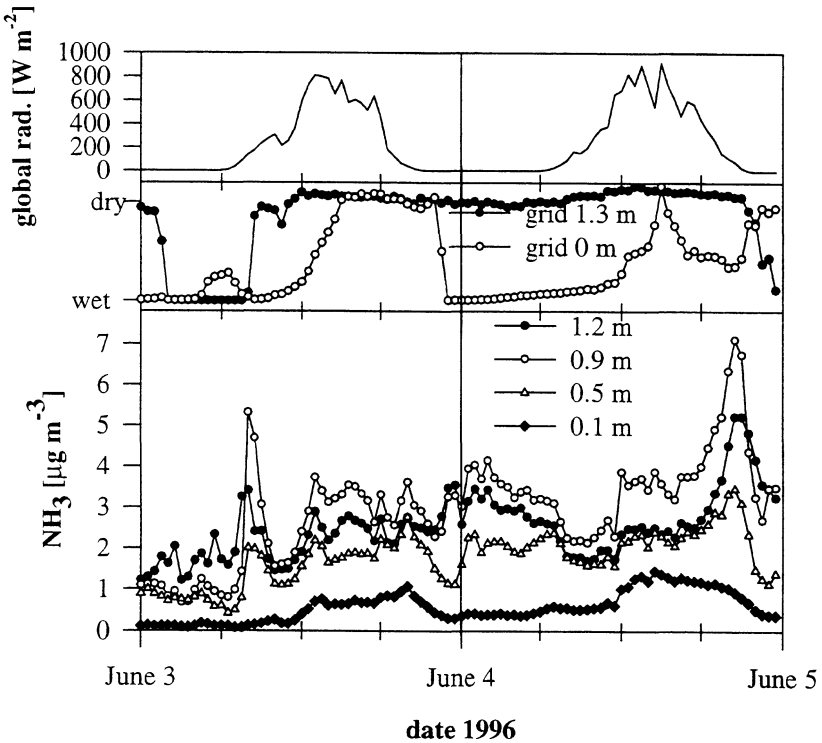

Figure 2: NH$_3$ concentration in the canopy (0.1, 0.5, 0.9 m) and above the canopy (1.2 m); surface wetness (wetness grids); global radiation; half hour means

3.2 NO concentration profile in the soil

Of the five soil depths where NO was measured (2, 10, 25, 50 and 70 cm) the highest concentration of the profile was found either at 2 or 25 cm soil depth. Figure 3 shows the seasonal variability of the NO concentration at 2 and 25 cm soil depth during the campaign together with the plant height, the growth stages of the plants and special activities.

The most pronounced variation in the NO concentration at 2 cm soil depth takes place after fertilisation of the field. On April 17 the field was fertilised with 40 kg ha^{-1} NH$_4$NO$_3$-N. Six days later 5 mm rain fell on the dry soil surface and dissolved and transported the mineral fertiliser into the soil, which resulted in an immediate increase in the NO concentration in the top soil layer by 1 - 2 orders of magnitude. The effect of the fertiliser on the NO concentration in the soil lasted about 20 days. After the fertiliser event until fructification low NO concentrations were measured and the concentration decreased with increasing

soil depth. From the beginning of fructification until the end of measurements the concentration at 25 cm rose continuously.
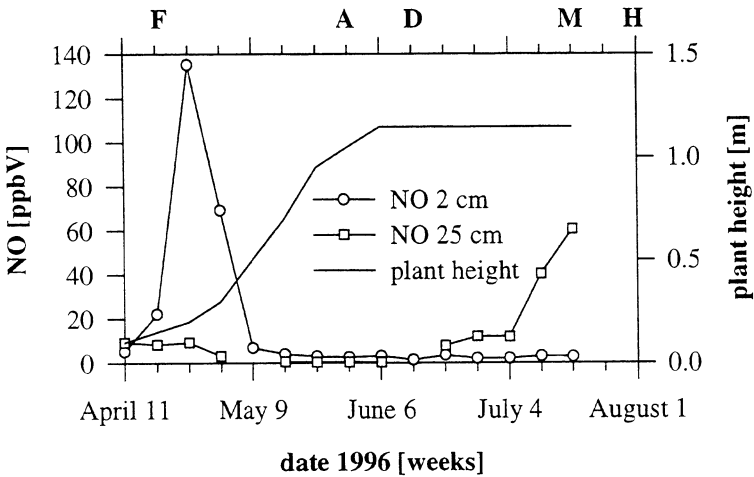

Figure 3: NO concentration in 2 cm and 25 cm soil depth during the whole measuring campaign together with the height of the wheat; weekly means; F: fertilisation; A: anthesis; D: fructification; M: ear maturity; H: harvest

Four typical days are presented in Figure 4 to explain qualitatively the daily variations at the beginning of the measurements, after fertiliser application, during plant growth and during fructification. The NO concentration is presented together with the temperature. Four days without precipitation have been chosen.

The first day is April 16 at the beginning of the measurements. The soil was dry from a long drought period. The highest NO concentration in the soil profile is measured most of the time at 25 cm soil depth. The NO concentration at 2 cm soil depth follows the temperature in the corresponding depth. At 8:00 an elevated NO concentration of 8.9 ppbV at 5 cm above the soil surface is measured due to morning traffic emissions and a low stable boundary layer. At the same time the NO concentration at 2 cm soil depth rises and reaches a maximum concentration of 7.5 ppbV at 9:00.

On April 25, shortly after fertilisation of the field, the highest NO concentrations, up to 230 ppbV, were measured at 2 cm soil depth and the lowest at 25 cm soil depth. The concentrations at the three soil depths vary with the temperature at 2 cm depth. The variation is decreasing with increasing soil depth. At 21:00 when a nocturnal boundary layer begins to develop a pronounced peak concentration of 206 ppbV is measured at 2 cm depth. Due to elevated NO concentration in the soil also the NO concentration at 5 cm above

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the soil surface is clearly elevated compared to all the other cases presented in Figure 4.
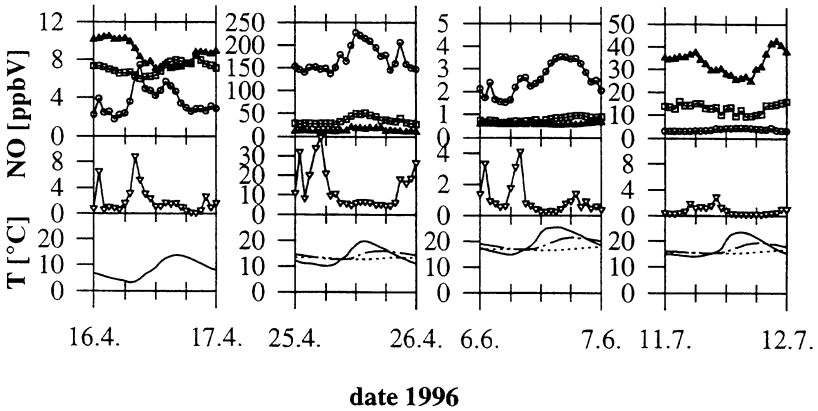

date 1996

Figure 4: NO concentration at 2 cm (circles), 10 cm (squares) and 25 cm soil depth (triangles pointed upwards); NO concentration 5 cm above the soil surface (triangles pointed downwards); temperature at 2cm (solid line), 10 cm (line-dot-line) and 25 cm soil depth (dotted line); NO concentrations are 15 min means, measured within the same hour; temperature in hourly means

The concentration variations at 2, 10 and 25 cm soil depths on June 6, during plant growth, follow the temperature variations at the corresponding depths. At 2 cm soil depth the NO concentration rises between 8:00 and 9:00 together with the concentration 5 cm above the soil surface due to morning traffic emissions and low stable boundary layer.

On July 11, during fructification, the highest concentration is measured at 25 cm and the lowest concentration at 2 cm soil depth. The pattern of the daily NO concentration variations on July 11 is similar to the pattern of April 16. The gradient of the NO concentration is more pronounced and the relative variation at 25cm soil depth is higher than at 10 cm and 2 cm.

3.3 N_2O concentration profile in the soil

The N_2O profiles in the soil measured during the campaign are shown in Figure 5 together with the atmospheric concentration, the plant height, the growth stages of the plants and special activities.

The N_2O profiles in the second half of March increase from the deepest measured soil layer at 70 cm towards the soil surface. The concentration at 70 cm soil depth is about 6 ppmV and the concentration at 10 cm is between 400 and 900 ppbV, close to the concentration in the atmosphere. Fertiliser application did not substantially stimulate N_2O production.
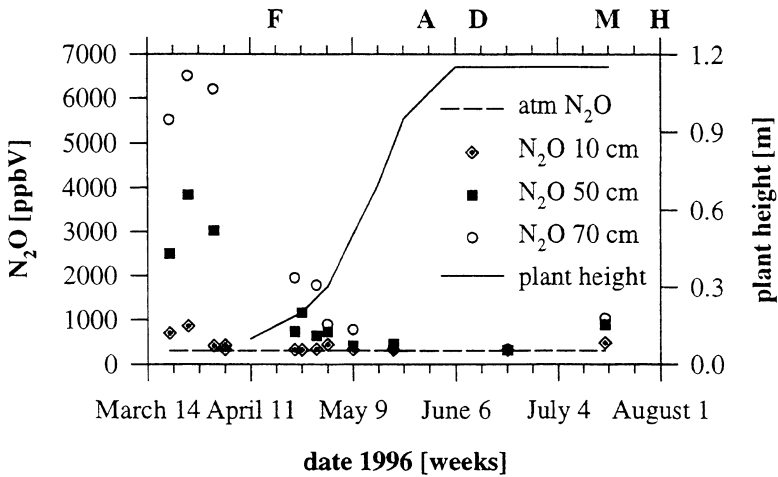
Figure 5: N_2O concentrations in the soil and the atmosphere („atm N_2O")
together with the height of the wheat; N_2O concentration symbols represent one
measurement; F: fertilisation; A: anthesis; D: fructification; M: ear maturity; H:
harvest

With the beginning of plant growth the N_2O concentrations, at all soil depths
measured, decline and are close to the atmospheric concentration from May 2.
The N_2O profile measured in July, close to maturity, slightly inreases again
from 70 cm to the soil surface.

4 Discussion

4.1 NH₃ exchange

During the night when the surface of the plant and the soil is covered with dew
ammonia is deposited on wet surfaces. Drying of the surfaces leads to an
evaporation of NH_3 in the morning, where most of the deposited ammonia is
emitted again. This evaporation peak was observed during days where dew
covered the plant surfaces during the night and evaporated in the morning.
        The causes for the evening peak of June 4, which was observed on
several days cannot be definitely explained. One explanation is a change of the
pH and the NH_3/NH_4^+ concentration in the apoplast of the plants. Another is a
decrease of the eddy-diffusivity in the atmosphere towards the evening together
with strong emission from the plants.
If the surfaces remain dry during day or night ammonia is emitted into the
atmosphere from the flag leaf/ear region of the plants. The highest NH_3
concentration in the canopy profile was always measured on the flag leaf, ear

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level from full development of the flag leaf around May 23 until fruit maturity at the end of July. Harper et al. [11] found NH_3 emissions from a field planted with wheat (*Triticum aestivum L.*) during fertiliser application and from the appearance of the flag leaf until leaf senescence. The ammonium/ammonia concentration and pH in the apoplast as well as the stomata opening of the flag leaf and/or ear plays therefore a key role in the ammonia exchange between wheat and the atmosphere. NH_3 deposition on surfaces covered with dew is of minor importance for the exchange budget because the deposited NH_3 is mostly reemitted during evaporation.

4.2 NO and N_2O exchange

The seasonal NO concentration variation at 2cm soil depth is mainly caused by fertiliser application. After the long drought period the stimulation of the NO production in this layer indicates a combined water and N-limitation of the production. A similar effect was observed by several authors (e.g. Johansson [8]).

The soil layer at 25 cm depth is in the region of the plough depth, where decaying straw was incorporated into the soil. An active microbial population can be expected in this layer. Before fertilization the NO concentration at 25 cm depth exceeds the concentration at 2 cm by a factor up to 5 and declines after April 25 when plants are growing in height. The concentration begins to raise at the beginning of fructification and is up to 10 times higher than the concentration at 2 cm when the ear is mature.

The cause of the concentration decrease and increase at 25 cm depth cannot be definitively explained. Our first explanation is a high nitrogen demand of the plants during growth and a smaller nitrogen demand when full length is reached. This trend was found in the measurement of the NO_3^- concentration of the soil at 25 cm depth (see Table 2) but not at 2 cm. Harper [11] found in a field planted with wheat (*Triticum aestivum, L.*) that the soil N availability rate, defined as mineralization rate minus fertiliser uptake rate minus total plant uptake rate, decreased when plants were growing in height. Therefore, we assume that the availability of NO_3^- and to some extent NH_4^+ limits the microbial NO production.

Prior to fructification the daily concentration variation at all soil depths can mostly be explained with variations in soil temperature except during fertiliser application and precipitation. The ambient NO concentration influences the concentration at 2 cm depth during morning traffic. After fertiliser application the NO concentration down to 10 cm soil depth was influenced by the concentration in the top soil layer as the correlation with temperature at 2 cm shows. At the beginning of fructification the relative NO concentration variation at 2, 10 and 25 cm increases with depth in contradiction to the relative temperature variations. We assign this NO variation to a day/night plant activity cycle in N-demand.

Figure 3 and 4 demonstrate that large differences in the dynamic of the NO concentration in the soil over short vertical distances exist without influencing each other, except after fertiliser application. This can be explained by the relaxation depth of NO. The relaxation depth represents the depth over which a change in the prevailing conditions is equilibrated. Galbally [12] found NO relaxation depths in the range of some mm to cm, due to large consuption rates and small diffusional transport. We conclude that only the concentration in the top soil layer is of importance for the NO flux into the atmosphere.

N_2O has been mainly produced at deeper soil levels. This has also been postulated in the works of Wienhold [13] and Burton [14]. The reason for the high N_2O concentrations at 70 cm depth could be a layer, at 58 - 68 cm depth, just above the layer where N_2O was measured, with a very high content of organic carbon (40%). The C_{org}/N_{tot} ratio at 70 cm, measured once at the end of the campaign, was 10.3 (see Table2). It reflects a high content of organic carbon and a total nitrogen content, which is comparable to the upper soil layers.
The seasonal variation of the N_2O concentration at 70 cm can not be compared with the NO_3^- and NH_4^+ concentration of the soil because at 70 cm depth it was not measured. Nevertheless the variation is similar to the variation of the NO concentration at 25 cm. The hypothesis of N-limitation, due to plant demand during growth, presented for the explanation of the NO concentration at 25 cm, can be taken into account to explain the N_2O concentration at 70 cm. The lack of a substantial response to fertiliser application indicates that the conditions in the upper soil layer did not favour N_2O production and that the nitrogen of the fertiliser did not penetrate to depths where the main N_2O production occurred.
        If the relaxation depth of N_2O is calculated with the approach of Galbally [12] and N_2O uptake rates determined in the laboratory by Bandibas [15] from 18 different soils we get depths greater than 1 m. We therefore assume that the N_2O production at around 70 cm depth drives the N_2O flux from the soil into the atmosphere.

5 Conclusion

The most important parameters for the NH_3 exchange are the ambient air concentration and the compensation concentration of the plant canopy, measured at the level of flag leaf/ear. pH and NH_4^+ concentration in the apoplast and stomata opening of the flag leaf and/or the ear determine the compensation concentration. Further experiments have to be done to determine the compensation concentration of the flag leaf and the ear to determine the contribution to the flux. Whether the evening peak of the ammonia concentration is caused by a drop in eddy diffusivity, change of plant activity or both has to be investigated by further experiments. The flux above the plant canopy has to be measured to quantify the observed processes.

The NO exchange is controlled by the top soil layer. The most important factor for the variability is fertiliser application. Temperature controls the daily variation of the concentration in the upper soil layer. To quantify the observed processes flux measurements with chambers in addition to measurement of vertical profiles of NO and biological inert ^{222}Rn in the soil are planned. The processes in deeper layers are of minor importance because changes in the soil concentration are equilibrated within short distances. To understand the processes at the plough depth measurements of the nitrogen content of the soil in high time resolution would be needed.

First estimates lead to the conclusion that N_2O flux is determined by production processes at soil depths around 70 cm. Flux measurements with static chambers and eddy correlation have to be done to confirm this hypothesis. The highest N_2O concentration gradient in the soil profile has been measured in spring and an elevated gradient at maturity of the ear. First considerations lead to the conclusion that N_2O production is N-limited. N_2O profiles in the soil have to be measured in higher time resolution to identify the processes influencing N_2O concentration in the soil adequately. Soil profiles of nitrogen will have to be determined to obtain a better understanding of the limitation of N_2O production.

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