Dynamics of N_2O in Soils: Experiments and Modelling

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Abstract

Probably more than 60% of the globally emitted nitrous oxide (N_2O) originates from soils. Recently, N_2O fluxes from soils have been monitored in several studies. Regression relationships between N_2O fluxes and variables like average soil moisture content, nitrogen content and temperature did in general not explain more than 50% of the flux variance. This study aims to contribute to an improved quantitative insight into the relation between underground processes and N_2O fluxes.

At grassland soils in the Wageningen Rhizolab, we monitored N_2O fluxes as well as underground profiles of several variables. No relation was found between underground N_2O profiles and the surface fluxes. A simulation model was adapted to simulate the relevant processes. The simulated O_2 and CO_2 profiles were satisfactory, indicating that the (bulk) respiration rates used in the model were realistic. The N_2O profiles, however, were less well simulated. A possible explanation is that the model does not account for heterogeneously distributed N_2O production at microsites in the soil.

Introduction

The atmospheric concentration of N_2O, involved in the enhanced greenhouse effect and the depletion of the stratospheric ozone layer, is currently about 0.31parts per million by volume (ppmv) at an annual increase of about 0.25% [1]. At least 60% of the global gross N_2O emission is attributed to soils [2]. However, the relation between the underlying processes, denitrification, nitrification and transport, and N_2O fluxes from soils is badly quantified. Attempts to obtain simple, robust empirical relationships between field-scale variables and the fluxes were rarely succesful. In general, such empirical relationships explained at most 50% of the flux variance [3].

To improve the limited explanatory power of regression models, supplementary quantitative process-based research is desirable. Simulation models to relate N_2O fluxes to the underlying processes under wet field conditions have been published. The model of Li *et al.* [4] neglected transport of gases in the soil, which might explain that the simulated emission peaks typically occurred earlier than the measured peaks [5]. The model of Grant *et al.* [6,7] explained fluxes during the melting of a frozen snow-covered soil. As far as we know, Li *et al.* [4,5] and Grant *et al.* [6,7] did not use concentration profiles measured underground to test the models.

The aim of this paper is to contribute to the quantitative knowledge on processes underlying N_2O fluxes from grassland soils by combining experiments under controlled or intensively monitored 'realistic field' conditions and process-based modelling.

Materials and Methods

The experiment in the Wageningen Rhizolab

In the period May 1993-April 1995 we conducted an experiment in the Wageningen Rhizolab. This 'field laboratory' [8] contains 16 compartments (length × width × height: 1.25 × 1.25 × 2.00 m) used to mimick agricultural plots. In the facility the relations between underground processes and processes like plant growth or gaseous emissions are studied, under relatively controlled and well monitored conditions. Our experiment was conducted in four of the compartments. Technical aspects on the experiment are described more extensively in [9]. The rectangular compartments were filled with a fine sandy soil that was sown to perennial ryegrass (*Lolium perenne* L.). We controlled the groundwater tables, and the applications of nitrate fertiliser and irrigation water. The grass was harvested 5 times during 1993 and 8 times during 1994 at intervals of 3 to 5 weeks. Here we report results obtained for the compartment with the average groundwater table at 0.5 m depth, to which nitrate fertiliser was applied at the start of the growing seasons and directly after harvests within the growing season. $Ca(NO_3)_2$ was applied at a rate corresponding with 100 kg N/ha per application. Within the soil various sensors, samplers and (horizontal) root observation tubes were installed. The sensors monitored volumetric moisture content, temperature, electric conductivity and soil water potential. Samples of the acqueous and the gaseous phase in the soil were taken. Above-ground observations included the monitoring of N_2O fluxes and the determinations of the yield and composition of the harvested grass.

Two methods were used to monitor the N_2O fluxes. During 1993 and a part of 1994 we used small vented closed flux chambers (cylindrical-shaped; inner diameter 20 cm, height 15 cm; [10]). These were installed at two fixed spots on the soil surfaces of each compartment during the measurements. To get a better average flux estimate, the sample area was increased by using large box-shaped chambers (height: 0.5 m) covering the plots in the other observations during 1994 and those in 1995.

Simulation Modelling

We adapted the model of Leffelaar [11], that couples microbiological transformations in denitrification and aerobic respiration with the transport of gases, water and solutes. The adapted model will be described more extensively in [12]. Here we report results on the testing of the microbiological and gas transport modules of the adapted model. We made the following major changes and assumptions:

1. we assumed that the microbial biomass kg^{-1} soil was 0.1*(the value used in [11]),
2. changes to describe the linear geometry of the soil column above the groundwater table at 0.5 m depth instead of a radial geometry (height: 2.59 cm, radius: 4.9 cm),
3. we simulated for the case that no N fertilisation or irrigation had been applied for at least 2 days. The experimental data suggested that transport of water had became very slow then. Ignoring water transport, we followed the evolution in the gaseous phase from hypothetical atmospheric inital conditions throughout this phase towards a pseudo steady state (section 3.2) after 4 days,
4. zero flux boundary conditions for gas fluxes at 0.5 m depth were assumed,

5. we assumed that transport through the gaseous phase only ceased at a gas-filled porosity equal to zero.

The computer program consisted of a small CSMP III core, calling a large number of FORTRAN 77 subroutines [11]. Simulations were done on a VAX machine.

Results and Discussion

The experiment in the Wageningen Rhizolab

The N_2O fluxes for the studied compartment are plotted versus time (Fig. 1), as well as the average values at 0.1 m depth of the soil moisture contents and the concentrations of N_2O and O_2 in the gaseous phase. Fertiliser together with an amount of irrigation water was applied at the moments indicated by arrows (Fig. 1a).

A negative correlation was found between the number of days elapsed since the preceding application of fertiliser and irrigation water, and the magnitude of the N_2O fluxes (Spearman's rank correlation test, $p = 0.05$). Like several authors [3], we conducted regression analysis to establish relationships between soil variables and fluxes. Like in most other studies, the explained variances were low. For example, volumetric soil moisture content (Fig. 1b) and nitrate concentration in the soil solution at 0.1 m depth, did not explain more than 50% of the variance of ln (N_2O flux). The N_2O concentration in the gaseous phase at 0.1 m depth (Fig. 1c) at all measurement dates are above the atmospheric concentration. Like [13], we did not find a relation between this concentration and the magnitude of the N_2O fluxes: high fluxes could go with low concentrations at 0.1 m and vice versa. Furthermore, we note that the concentrations of molecular oxygen (O_2) in the gas samples (Fig. 1d) point at relatively aerobic conditions in the gaseous phase at 0.1 m depth. Also in the deeper layers, low O_2 concentrations rarely occurred (for example, O_2 concentrations below 3% were only found on 5 of the 31 days on which profiles were measured, and these observations were restricted to depths below 0.25 m). It is suggested that N_2O emission from well aerated soils starts with N_2O production at anaerobic microsites [14,15,16,17].

Simulation Modelling

In Figure 2 measured profiles of O_2, CO_2 and N_2O in the gaseous phase are shown (symbols). We assume that these profiles represent a pseudo steady state situation, i.e. that the net concentration changes as a result of the processes production, consumption and transport are relatively slow. The simulation model describing these processes was applied to simulate the profiles (Fig. 2, dashed and full lines), using measured profiles of the soil moisture content, and nitrate and water soluble carbon in the acqueous the phase as input. The simulated O_2 and CO_2 profiles represent a slowly changing situation: at all depths the relative concentration changes were $\leq 15\%$. Taking this into account and that we did not optimise parameters to obtain the result, the correspondence between the measured and simulated O_2 and CO_2 profiles (Fig. 2a) is satisfactory, indicating that the parameters for aerobic respiration in the soil are reasonable. However, the simulated N_2O profile does not match the measurements (Fig. 2b). The reason for this is N_2O production in the model only takes place if the O_2 percentage is below 1%, and that situation that did not occur in the simulation. The underlying assumption for this 1% criterion is that the soil is completely homogeneous.

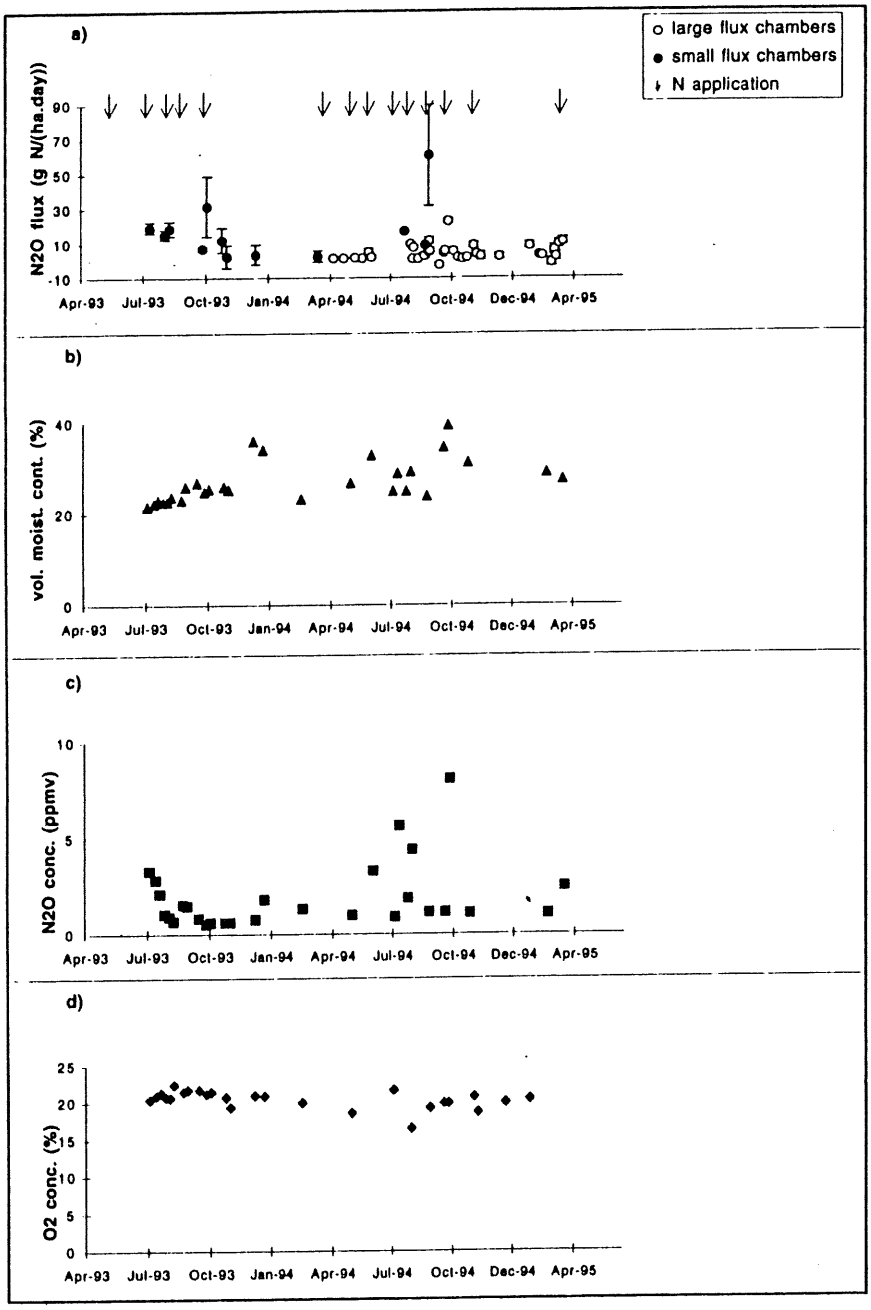

Fig. 1: Time series of: **(a)** N_2O flux measured with small or large flux chambers (g N ha^{-1} day^{-1}); flux of 190 ± 30 g N ha^{-1} day^{-1} at July 31, 1994, not shown. Average values at 0.1 m depth of: **(b)** volumetric moisture content (%; $n = 4$), **(c)** N_2O concentration (ppmv; $n = 2$), and **(d)** O_2 concentration (%; $n = 2$). Object: Rhizolab compartment with a fine sandy soil planted to *Lolium perenne* L.; groundwater table at about 0.5 m; arrows: applications of 100 kg N/ha as $Ca(NO_3)_2$ and irrigation water.

It might well be, however, that there are spots in the soil that are less accessible due to water accumulation or that respire more due to a concentration of some organic material. Such spots may be anaerobic while there is a higher O_2 percentage in the surrounding soil. This may be the case in space and time, e.g. within a few hours after irrigation, not monitored by us. That local anaerobic conditions might be crucial for N_2O production was already suggested in merely theoretical work [14,15,16,17], and supported by more recent experimental work [18].

As a straightforward way to account for this in the model, we explored the behaviour of the model assuming that a higher critical O_2 percentage (for example 10%) in the bulk part of the gaseous phase still allows N_2O production, namely in 'hot spots'. Preliminary results indicated that in this way satisfactory N_2O profiles could be obtained after parameter optimisation. However, to avoid degeneration of the explanatory model by parameter optimisation, further quantitative studies are necessary on the relation between overall aerobicity and local anaerobicity and this is the subject of ongoing research [our work, 19].

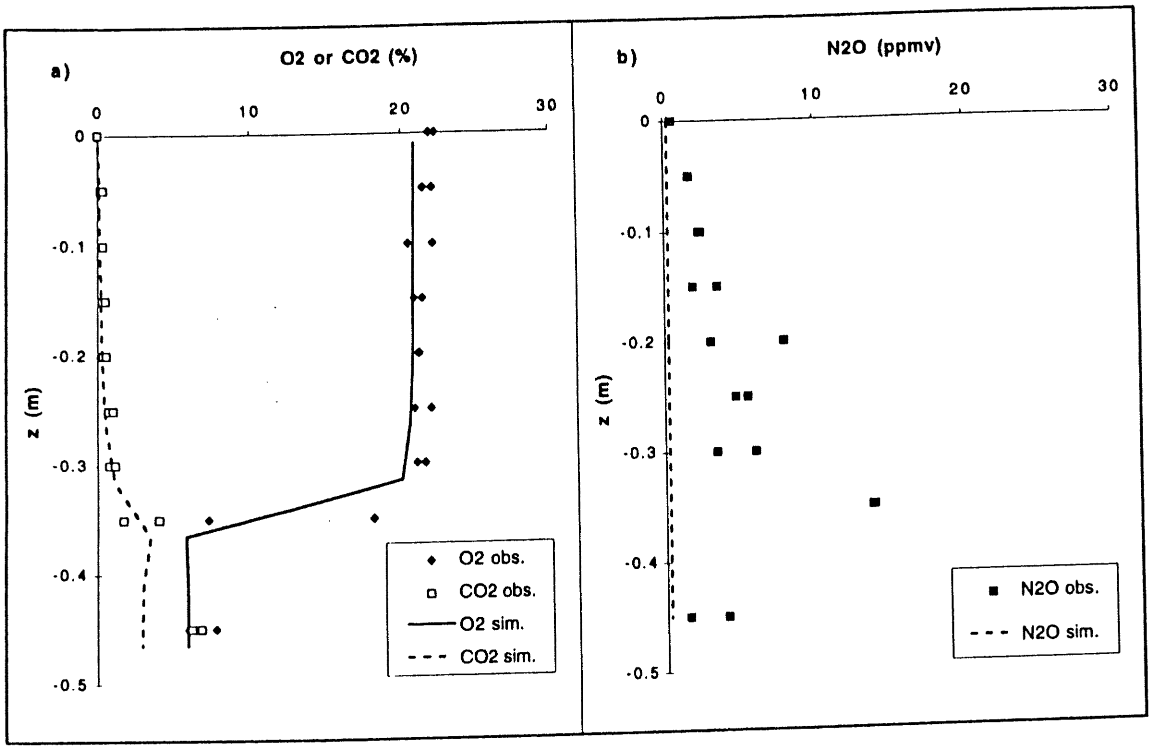

Fig. 2: Measured (symbols) and simulated (curves) profiles in the gaseous phase for the Rhizolab compartment: **(a)** O_2 and CO_2 concentration (%) versus depth z (m), **(b)** N_2O concentration (ppmv) versus z. Data for March 16, 1995.

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