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Effects of Liming and Nitrogen Fertilization on Emissions of CO_2 and N_2O From a Temporate Forest

R. BRUMME

Institute of Soil Science and Forest Nutrition, Göttingen, Germany

F. BEESE

GSF-Institute of Soil Ecology, Neuherberg, Germany

Fluxes of N_2O and CO_2 were measured simultaneously in control, *N*-fertilized, and limed plots in a 145-year-old beech stand in the Solling area in Germany using an automated chamber method. The *N*-fertilized plot received annually 140 kg of nitrogen as NH_4SO_4 since 1982; the limed plot was treated with 30 t/ha of dolomitic limestone once in 1982. On all plots, fluxes of N_2O and CO_2 underwent strong diel, daily, and seasonal variations. For the control plot, N_2O and CO_2 effluxes averaged about 1 mg $N/m^2/d$ and 2.9 g $CO_2/m^2/d$ from October until May and 3 mg $N/m^2/d$ and 4.5 g $CO_2/m^2/d$ from May until September. Annual fluxes for N_2O and CO_2 were 5.6 kg N/ha/yr and to 3.2 t C/ha/yr, respectively. Nitrogen saturation of the system due to high rates of N deposition for several decades may be responsible for the high N_2O losses. Liming drastically reduced the N_2O emission to 1.5 kg N/ha/yr and increased the CO_2 emission to 4.1 t C/ha/yr, 5 years after the treatment. Changes in N_2O/N_2 ratios are assumed to be the reason for the lower N_2O emission. Fertilization increased the N_2O emission, resulting in 7.8 kg N/ha/yr, whereas the effect on CO_2 emission with 3.8 t C/ha/yr was less pronounced.

1. INTRODUCTION

Temperate forest ecosystems have been characterized until recently as nitrogen deficient. A review of the literature shows that under these conditions the average rates of N_2O release are in the range of 1 kg N_2O–N/ha/yr [*Bowden*, 1986]. In vast areas of industrialized countries, however, high atmospheric deposition rates of nitric and sulfuric acid have changed the nitrogen and acid status of forest soils [*Ulrich*, 1983]. In many ecosystems, N saturation has been reached [*Beese and Matzner*, 1986; *Melillo et al.*, 1989; *Aber et al.*, 1989; *Skeffington*, 1990], whereby nitrogen deposition exceeds the demand of forests by a factor of 2–8 [*Beese and Matzner*, 1986]. Under these conditions, increased leaching of nitrate to the groundwater and gaseous N losses, such as nitrous oxide (N_2O) and nitrogen (N_2), may occur. In acid forest soils, nitrous oxide is the main gaseous product of denitrification; however, it is also released as a by-product of the ammonium oxidation (nitrification) [*Firestone and Davidson*, 1989]. Increased emissions of N_2O and CO_2 from soils to the atmosphere are important because these gases change the chemical and physical properties of the atmosphere. Nitrous oxide enhances the catalytic reduction of ozone [*Crutzen*, 1974; *McElroy et al.*, 1977], and both contribute to the greenhouse effect by absorbing infrared radiation in the troposphere. Nitrous oxide is of concern because its ability to warm the atmosphere is 180 times greater than that of CO_2 [*Lashof and Ahuja*, 1990], and it is increasing at 0.2–0.3 %/yr [*Rasmussen and Khalil*, 1986; *Brasseur and Hitchman*, 1988].

To prevent further acidification of forest soils and to improve the Ca^{++} and Mg^{++} supply of the trees, increasing areas of forests with acid soils in Germany have been limed in the last decade. Changes in acidity caused by liming may have an impact on nitrogen cycling and litter decomposition, both of which influence CO_2 and N_2O fluxes. Therefore management-induced changes in forest ecosystems may create side effects, that may influence biosphere-atmospheric interaction. Specifically, increased rates of CO_2 production may result from accelerated decomposition of soil organic matter stored in the forest floor, and increased emission of N_2O may result from higher rates of nitrification and higher NO_3 concentrations in the soil solution.

In this study we attempted to answer the following questions: (1) What are the rates of CO_2 and N_2O emissions from acid forest soils under conditions of high nitrogen input? (2) Do additional NH_4 inputs change CO_2 and N_2O emission rates? (3) Does liming change the CO_2 and N_2O emission rates?

2. STUDY AREA AND TREATMENTS

Measurements of N_2O and CO_2 released from three plots (20 m × 20 m) of a 145-year-old beech stand in the Solling area in Germany were made with three replicates on each plot in the years 1987 and 1988. The limed plot was treated with 30 t/ha of fine ground dolomitic limestone in 1982. The fertilized plot has been fertilized annually with 140 kg of nitrogen in the form of solid ammonium sulfate since 1982. A third plot was taken as control. All plots receive 35 kg N/ha/yr by atmospheric deposition [*Matzner*, 1989]. The soil is an acidic cambisol derived from loess over weathered triassic sandstone. Some properties of the soil and the humus layer (5–6 cm thick) of the three plots are summarized in Table 1.

3. METHODS

We developed an automatic chamber method for daily monitoring of N_2O and CO_2 fluxes from the soil to the

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TABLE 1. Soil Properties of the Fertilized, Limed, and Control Plots

	Soil, 0–5 cm					Humus Layer					
						O_L		O_f		O_H	
	*p*H $CaCl_2$ 0.01 M	CEC $\mu mol_c/g$	Ca + Mg % of CEC	Organic C %	C/N	Organic C %	C/N	Organic C %	C/N	Organic C %	C/N
Control	3.0	129	4.5	5.7	19	51	29	48	22	31	19
Fertilized	3.2	125	3.4	4.1	16	49	26	48	22	31	19
Limed	3.4	128	21.2	5.5	19	49	29	34	21	31	19

atmosphere [*Loftfield et al.*, 1992] to provide reliable rates where high temporal variability of biotic processes occur.

Gas fluxes from the soil into the atmosphere were measured automatically using Plexiglas chambers connected to a gas chromatograph (GC) equipped with an electron capture detector and a personal computer (Figure 1). The chambers consisted of two compartments and were sealed with a movable lid (Figure 2). They were only worked into the litter deep enough to achieve horizontal placement required for the 2-cm water trap to seal the lid. Gas samples were taken from the inner chamber 0, 30, and 60 min after closure. The outer chamber acts as a buffer zone preventing air penetration from the outside. Each compartment had a volume of 50 L and a surface area of 0.25 m^2. The personal computer registered opening and closing of the chambers and controlled the extraction of the gas samples, carried out the analysis of samples taken for N_2O and CO_2, the integration of the chromatograms, and the storage of the data. For calibration of the GC a gas standard was analyzed every 3–6 measurements. Each of the nine chambers was closed 5 times each day which resulted in 45 flux calculations in total. Soil temperatures were measured in 5- and 10-cm depth. Air, soil temperatures, and light intensities were recorded as hourly averages. Soil water tension (10-cm depth) were

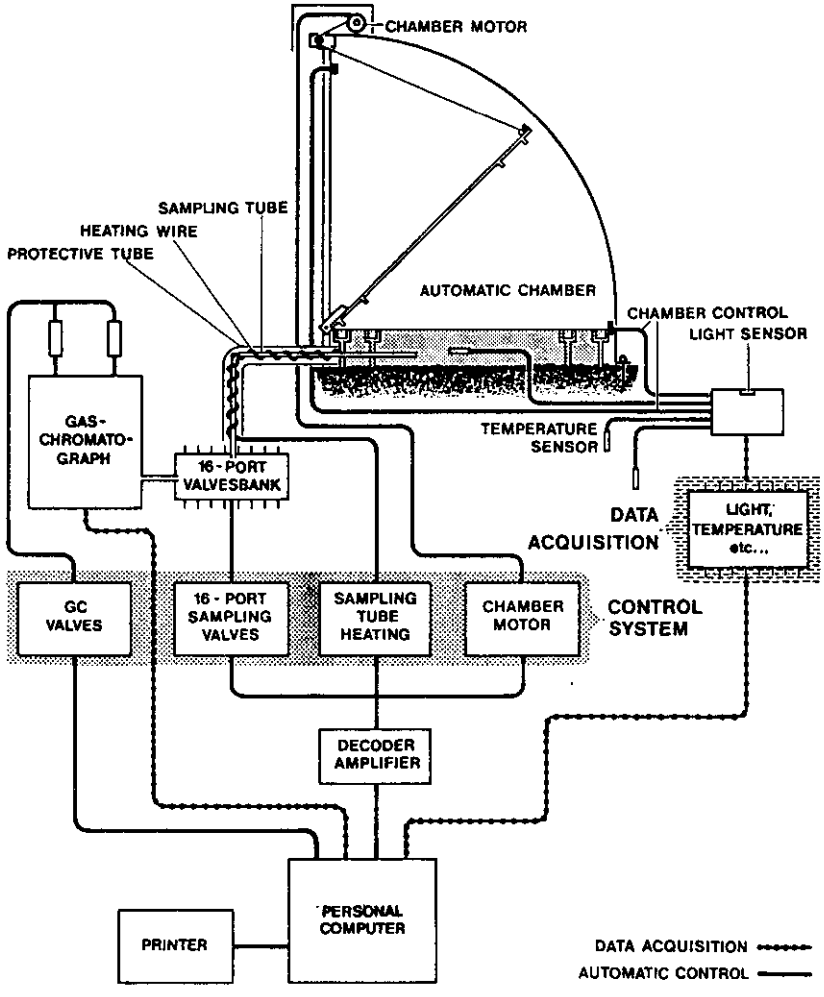

Fig. 1. Schematic diagram of the personal computer, supported control, and data acquisition.

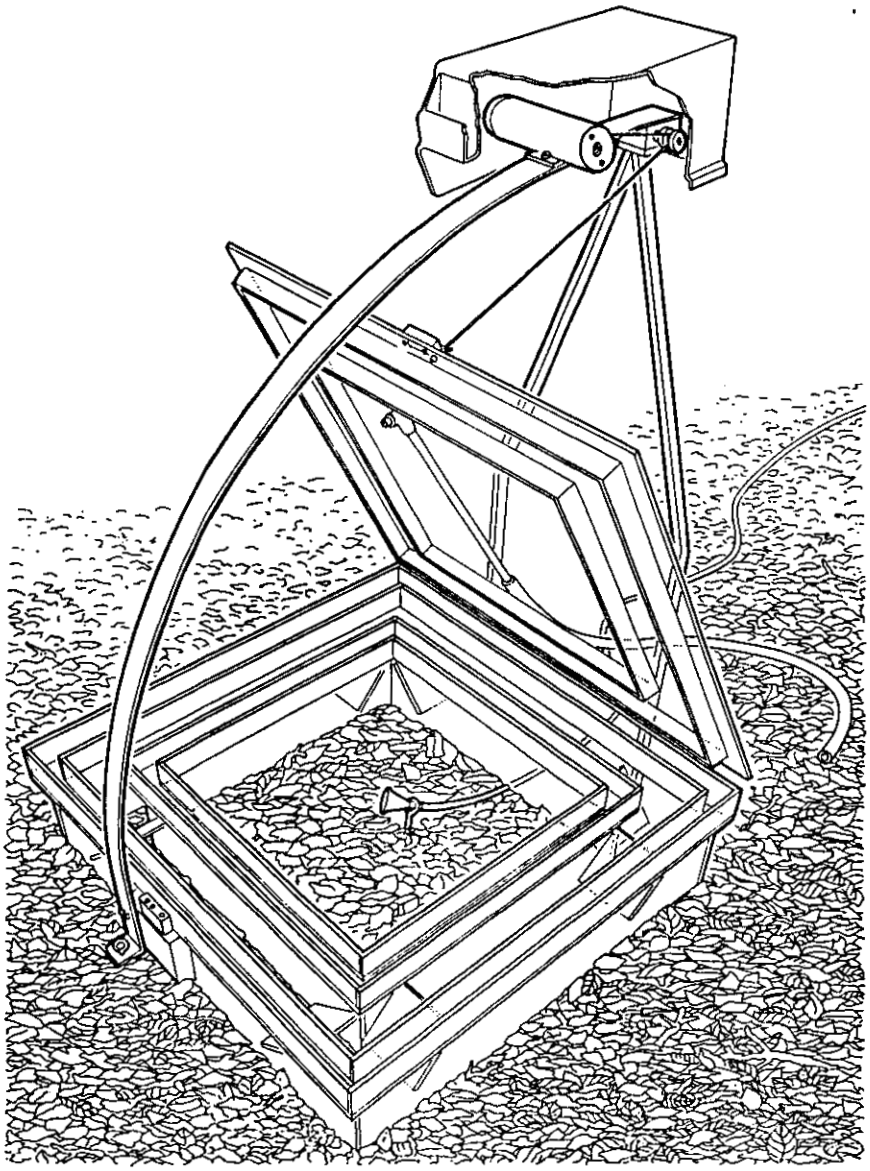

Fig. 2. Lockable double chamber for collecting gaseous losses from soils.

measured once a week and soil water solutions were collected over a month by suction cups in 10-cm depth and were analyzed for nitrate [*Meiwes et al.*, 1984].

4. RESULTS

4.1. *Temporal Variations*

Nitrous oxide and CO_2 emission showed strong diel and daily variations due to changing environmental conditions (Figure 3). The amplitude of diel fluxes is not constant. As an estimate based on a 20-day plot in June 1988, one can assume that the highest values are 10 to 220% higher for CO_2 and 10 to 470% higher for N_2O than the lowest rate measured each day.

Nitrous oxide emission also underwent a strong seasonal variation. Two main periods of N_2O emission can be distinguished on the control and fertilized plot (Figure 4). On the control plot, low rates (about 1 mg $N/m^2/d$) occurred from

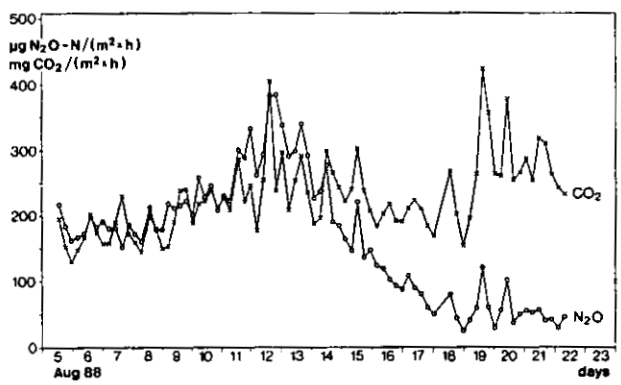

Fig. 3. Diel and daily changes of N_2O and CO_2 emission rates for August 1988 in chamber 6 of the control plot.

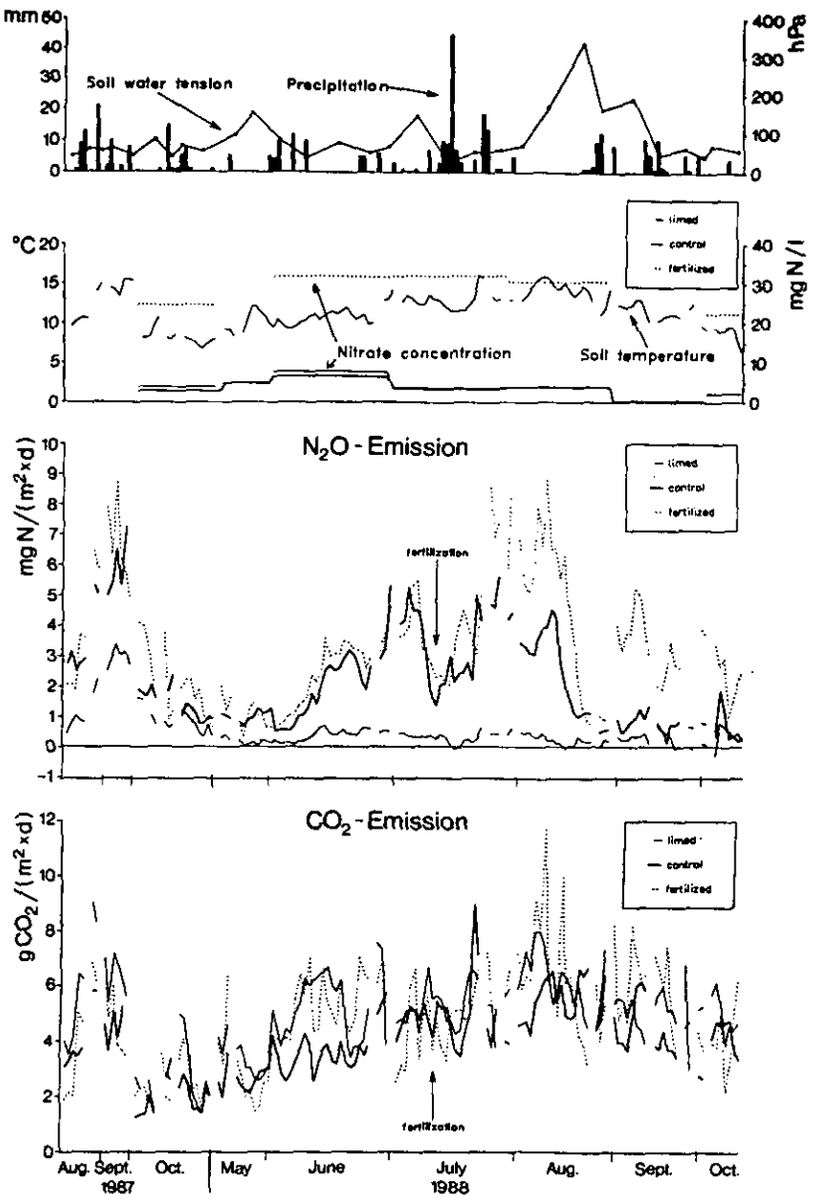

Fig. 4. Daily mean values of CO_2 and N_2O emission of three chambers and monthly nitrate concentration of soil solution (10-cm depth) of three plots, soil water tension (10-cm depth), soil temperature (5-cm depth), and precipitation (interrupted monitoring is indicated by breaks).

September/October to May, and elevated mean rates (approximately 3.5 mg $N/m^2/d$) occurred during the vegetation period. This seasonal fluctuation was not observed on the limed plot. There was only one short period with increased N_2O emission of 3.2 mg $N/m^2/d$ at the end of 1987. In 1988 the rates remained below 0.8 mg with an average of 0.5 mg $N/m^2/d$.

Soil respiration rates followed a similar but less-pronounced seasonal pattern, especially on the control plot. Fluctuations due to temperature changing are more pronounced for CO_2, resulting in peaks with a duration of several days. Despite these different patterns of CO_2 and N_2O there are periods with high correlations between both fluxes, for example, in September 1987 and directly after fertilization in 1988.

During the active period of N_2O emission (May to September), precipitation events are often accompanied by lower temperatures and lower emission rates of N_2O on the control and fertilized plot in 1988 (Figure 4). After precipitation periods, often fluxes increased together with soil water tensions and even the temperatures. At the middle of August this period abruptly ended when soil water tension exceeded 200 hPa. The effects of precipitation frequency on the CO_2 emissions were less pronounced.

Fertilization with ammonium sulfate in this nitrifying acid soil [*Lang and Beese*, 1985] resulted in higher N_2O fluxes than observed on the control plot after a lag time of 14 days (Figure 4). Despite higher nitrate concentrations on the fertilized plot the emission of N_2O prior to the fertilization was only slightly higher than on the control plot. The effect

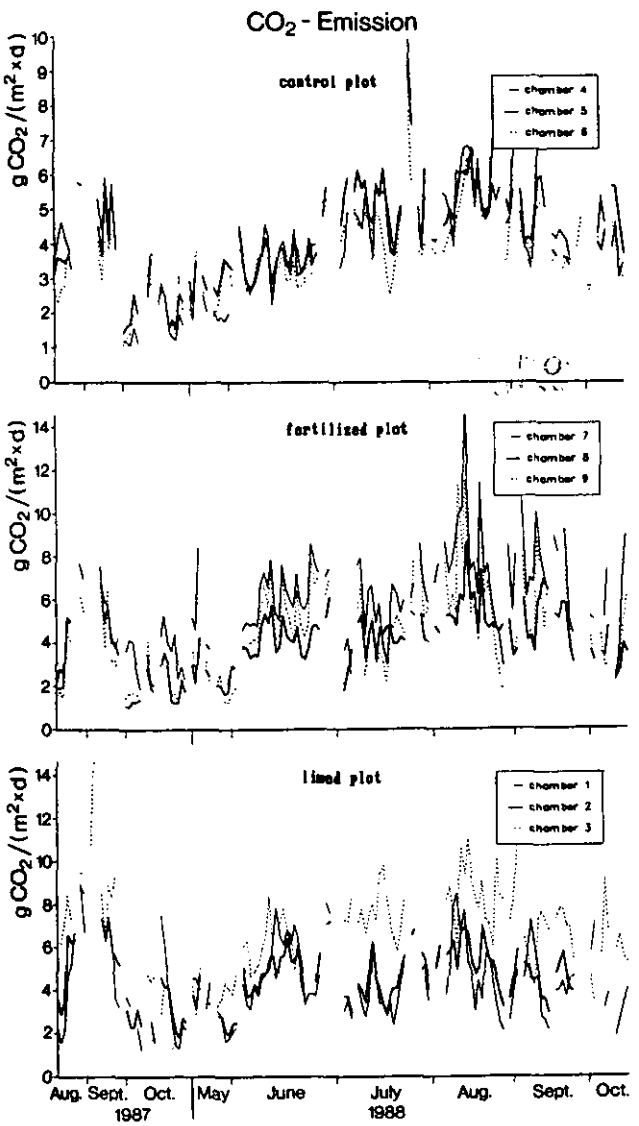

Fig. 5. Daily mean values of CO_2 emission on a control, fertilized, and limed plot of three chambers on each plot.

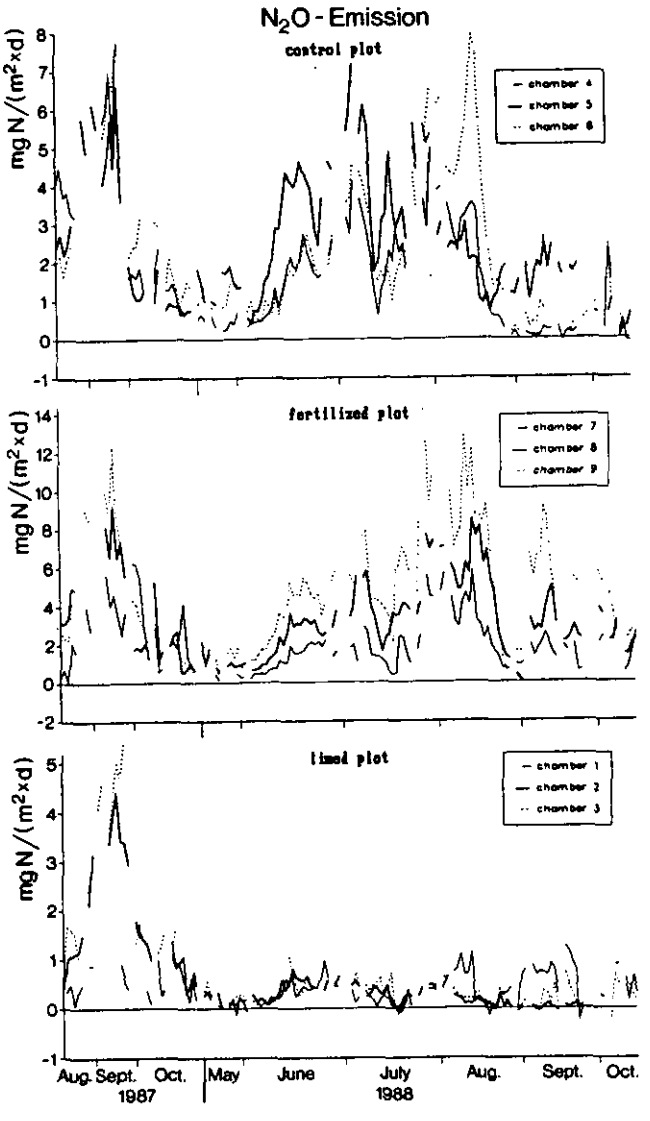

Fig. 6. Daily mean values of N_2O emission on a control, fertilized, and limed plot of three chambers on each plot.

of fertilization on the CO_2 emission was less pronounced than on N_2O, but the fluctuations were higher than on the other plots.

Temporal variation of CO_2 and N_2O emissions into the three chambers of one plot is very similar (Figures 5 and 6), despite different levels of emission rates among the chambers. The results show that the highest levels of N_2O emission rates of one chamber of a plot were not accompanied by the highest levels of CO_2.

4.2. *Annual Emission Rates of N_2O*

Monthly N_2O emissions of the acidic cambisol varied from 0.2 to 1.8 kg N/ha (Table 2). Assuming that the lowest monthly rates occurred during wintertime, emission rates of 4.6, 6.1, and 6.1 kg N/ha/yr from chambers 4, 5, and 6 (Table 3) resulted in a mean rate of 5.6 ± 0.9 kg N/ha/yr.

After fertilization with ammonium sulphate the nitrate concentration in soil solution increased 8 times compared to the control plot (Figure 4). The mean annual emission rate of N_2O was increased by 39% to 7.8 ± 2.9 kg N/ha/yr. The variation of the chambers (Table 3, Figure 6) was 2 times higher than that of the control. From the heavy N input of 140 kg N/ha/yr only 1.6% was transformed to N_2O.

Liming drastically reduced the N_2O emissions. Although approximately the same nitrate concentrations were found as in the untreated soil, the N_2O emissions dropped to about 1.5 ± 0.5 kg N/ha/yr, only 26% of the control flux rate. The variations were comparable to the fertilized plot (Table 3).

4.3. *Annual Emission Rates of CO_2*

The monthly rates of CO_2 from the acidic cambisol varied between 170 and 450 kg C/ha (Table 2). Making the same assumptions as before, an annual emission rate of 3.2 ± 0.3 t C/ha/yr was calculated. The emissions of CO_2 were increased by about 16% on the nitrogen-fertilized plot and about 31% on the limed plot. This resulted in an annual emission of 3.8 ± 1.0 and 4.1 ± 0.9 t C/ha/yr, respectively. The variation between the chambers was significantly reduced compared to N_2O (Table 3).

TABLE 2. Mean Monthly Emission Rates of CO_2 and N_2O on a Control, N Fertilized, and Limed Plot

	Control		Fertilized kg/ha/month		Limed	
	CO_2-C	N_2O-N	CO_2-C	N_2O-N	CO_2-C	N_2O-N
Aug. 1987	332	1.03	325	1.10	510	0.36
Sept. 1987	376	1.78	390	1.96	538	0.88
Oct. 1987	172	0.45	206	0.66	237	0.28
May 1988	221	0.31	254	0.35	288	0.06
June 1988	292	0.57	428	0.70	438	0.11
July 1988	423	1.09	407	1.28	444	0.09
Aug. 1988	454	0.78	535	1.45	524	0.08
Sept. 1988	358	0.24	485	1.04	419	0.08
Oct. 1988	347	0.21	356	0.77	417	0.11

5. DISCUSSION

High temporal variability of trace gas losses from soil to the atmosphere, as indicated by our data, can result in large errors in determination of fluxes with the chamber methods [*Mosier*, 1989]. Because most of the published emission rates for N_2O and many for CO_2 were determined by single flux determinations once a week. We have used a high temporal resolution to overcome hourly, daily, and seasonal variations. The amplitude of daily fluxes indicates the largest errors using single flux determination per day. As indicated by our data, the differences of the highest daily rates to the lowest were 10 to 470%. To calculate the deviations which may result from single short term flux measurements per week, we have calculated the mean hourly emission rates from 700 measurements (140 days) for each chamber and compared these data with results obtained from 30 measurements (once a week between 0630 and 1130). The results are shown in Table 4. The deviations for the CO_2 emission rates using one determination each week lie between −7% and +11% related to the automatic chambers. The mean deviation is less than 1%. The slight deviation for CO_2 resulted partly from the daytime chosen for the measurements. The deviations for the N_2O emission rates are worse. The deviations lie in the range of +3% to +50% and were on average +21%. This is due to higher rates of N_2O emission between 0630 and 1130. Liming and fertilization resulted in higher deviations compared with the control plot. In general, it was found that regular weekly measurements under the conditions studied resulted in reasonable estimates of the total emissions.

Our measurements exhibit a clear seasonal pattern for N_2O and CO_2 emission rates, upon which are superimposed daily and hourly variations (Figures 3 and 4). Soil temperature and soil water tension can explain most of the behavior of the emissions. The influence of the controlling factors is basically the same at all subplots (chamber level), indicated by the parallel pattern of emissions (Figures 5 and 6). External factors such as temperature and water probably explain only part of the temporal variation. Additional variations may also be explained by soil factors, such as organic matter or microbial processes.

The annual N_2O flux of 5.6 ± 0.9 kg N/ha released from the acidic cambisol is substantially higher than rates found in other studies. *Schmidt et al.* [1988] only found annual rates between 0.26 and 0.96 kg N/ha on six different deciduous forest stands in Germany. Similar annual rates were found for deciduous forests: 0.47–1.4 kg N/ha [*Goodroad et al.*, 1984] in Wisconsin, 0.15 kg N in New Hampshire [*Keller et al.*, 1983], 0.017 kg for a mixed hardwood stand in Massachusetts [*Bowden et al.*, 1990], and 1 kg N in Massachusetts [*Duxbury et al.*, 1982]. Higher annual rates of 2.4–3.2 kg N/ha were observed in coniferous forest ecosystems in Wisconsin [*Goodroad et al.*, 1984]; however, *Bowden et al.* [1990] showed that coniferous forest ecosystems do not necessarily emit more N_2O than deciduous forests. In a red pine plantation in Massachusetts only 0.01 kg N/ha/yr were emitted. We also detected only low N_2O emission rates in a spruce stand adjacent to the beech stand described in this paper.

The high N_2O emission rates from the Solling site may be caused by the high long term N depositions. Since the beginning of measurements at the Solling site in 1969, yearly nitrogen deposition rates have averaged 35 kg N/ha [*Matzner*, 1989]. With more or less constant leaching of 5 kg

TABLE 3. Annual Emission Rates of CO_2 and N_2O of Nine Chambers on a Control, N-Fertilized, and Limed Plot

	Limed			Control			Fertilized		
Chamber	1	2	3	4	5	6	7	8	9
				CO_2					
t C/ha/yr	3.7	3.6	5.2	3.1	3.5	3.1	4.9	3.2	3.2
cv		21.5%			8.3%			26.5%	
				N_2O					
kg N/ha/yr	1.0	1.5	2.0	4.6	6.1	6.1	4.9	7.7	10.7
cv		34.5%			16.2%			37.1%	

Coefficient of variation, cv.

TABLE 4. Mean Hourly Emission Rates of CO_2 and N_2O of Nine Chambers Calculated for the Automatic Chambers Using All Fluxes and for the Case of One Flux Determination Each Week

	Limed			Control			Fertilized		
Chamber	1	2	3	4	5	6	7	8	9
				mg $CO_2/m^2/d$					
CO_2									
automatic chamber*	172	183	266	164	173	149	227	161	172
one flux determination each week†	181	173	279	166	161	141	233	168	190
changes,‡ %	+5.2	−5.5	+4.9	+1.2	−6.9	−5.4	−1.7	+4.3	+10.5
				μg $N/m^2/h$					
N_2O									
automatic chamber*	16.7	27.3	34.4	84.7	107	93.4	76.4	133	185
one flux determination each week†	22.8	34.6	43.3	87.4	119	103	114	141	219
changes,‡ %	+36.5	+26.7	+25.9	+3.2	+11.2	+10.3	+48.6	+6.0	+18.4

*700 emission rates within 140 days.
†30 emission rates of 30 days within 140 days.
‡One flux determination each week related to automatic chambers.

N as measured separately and losses of 5.7 kg N caused by N_2O emissions, this ecosystem receives 24.3 kg N per year which may be stored in organic matter, trees, or may be lost by N_2.

After fertilization with ammonium the emission rates of N_2O increased after a lag time of 14 days, indicating that nitrification may influence the N_2O losses. Nitrification plays an important role in this acid soil. Heterotrophic microorganisms may be responsible for nitrification, as shown by *Lang and Beese* [1985]. The higher nitrate concentrations in the soil solution before fertilization seemed to be of minor importance to the N_2O emission. Only 1.6% of the fertilizer nitrogen were transformed to N_2O amounting to a total of 7.8 ± 2.9 kg N/ha/yr. This is in agreement with the results of *Bowden et al.* [1991], who found that after fertilization with 150 kg N/ha/yr as NH_4NO_3 to a mixed hardwood forest and a red pine plantation no more than 0.2% of the added N were transformed to N_2O.

Five years after liming, the N_2O emissions were reduced drastically to annual rates of about 1.5 ± 0.5 kg N/ha/yr. Nitrate concentration in the soil solution was approximately the same as found in the control (Figure 4) but the $pH(H_2O)$ in the humus layer increased from 4.5 to 6.5. These results indicate that the long-term effects of liming result not only in prevention of further acidification of soil by acidic deposition but also in a 74% reduction of N_2O emissions. An explanation for the reduced N_2O release may be found in changed N_2O/N_2 ratios of the gases emitted. Laboratory investigations have shown a shift to higher N_2 proportion after increasing pH [*Knowles*, 1981].

Estimates of global fluxes of N_2O from temperate deciduous forests covering 4.41×10^6 km^2 [*Steudler et al.*, 1989; *Bowden et al.*, 1990] amounted to 2.47 Tg N/yr. *Schmidt et al.* [1988] estimated fluxes from temperate forests of 0.7–1.5 Tg N/yr and *Bowden et al.* [1990] a flux of 0.012 Tg N/yr. The results show that areas with higher emissions exist and their contribution to global fluxes must be quantified.

Carbon dioxide release from soils is originally from two different sources, the decomposition of soil organic matter and the respiration of living roots. Both are measured together in the chamber. The annual flux from the control amounts to 3.2 ± 0.3 t C/ha. After fertilization and liming, the mean rates of CO_2 emission were increased by about 16% to an annual flux of 3.8 ± 1.0 t C/ha and about 31% to 4.1 ± 0.9 t C/ha. These data show that changes in soil chemistry combined with changes in the microbial community may result in accelerated decomposition of organic matter and a reduction of carbon pools of soils. Accelerated CO_2 emission rates on the limed plot were not accompanied by a higher leaching of nitrate (Figure 4).

From these data it is evident that changes in soil chemistry caused by atmospheric deposition (H^+ and N) or site management drastically may change the emission rates of climate relevant gases. A sensitivity analysis indicates that short time measurements as mostly carried out in the past cannot ever lead to realistic output rates. The data also show that quasi-continuous measurements may improve our knowledge to quantify the influence of the parameters governing the trace gas emission under field conditions. From the data it is obvious that our knowledge of the external and internal soil factors controlling the processes of trace gas emissions is still not good enough for the prognosis of emission rates under changing conditions. This stresses the necessity of using quasi-continuous measurements over longer time periods as a tool for parameter identification and model verification.

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F. Beese and R. Brumme, Institute of Soil Science and Forest Nutrition, Buesgenweg 2, W-3400 Göttingen, Germany.

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