

Greenhouse gas emissions (N_2O , CO_2 and CH_4) from three forest soils near Vienna (Austria) with different water and nitrogen regimes

M. Hahn, K. Gartner and S. Zechmeister-Boltenstern

Treibhausgas-Emissionen (N_2O , CO_2 und CH_4) aus Waldböden bei Wien (Österreich) mit unterschiedlicher Wasser- und N-Versorgung

1. Introduction

Emission of N_2O from the soil surface is important as a pathway for loss of fertilizer nitrogen and as a source of atmospheric pollution. Nitrous oxide (N_2O) contributes to climate change and also participates in reactions which destroy stratospheric ozone (CICERONE, 1987). Development of aerobic and anaerobic microsites within the same soil aggregate, permits both nitrification and denitrification to occur simultaneously (DAVIDSON et al., 1986).

Soil moisture, soil temperature, pH, organic matter, O_2 status, and the capacity of soils to supply nitrifiable N affect N_2O release via nitrification and denitrification. A key factor controlling denitrification in forest soils is aeration (GAMBRELL et al., 1975; SEXSTONE et al., 1985). Nitrification requires aerobic conditions, whereas denitrification is favoured by low O_2 concentrations. Soil nitrate appears to be a more important factor regulating denitrification in forests than in agricultural soils (ROBERTSON and TIEDJE,

1984; DAVIDSON and SWANK, 1987). Factors affecting carbon availability such as freezing and thawing or wetting and drying should also be important regulators of denitrification in forest soils (EDWARDS and KILLHAM, 1986; GROFFMAN and TIEDJE, 1988).

Comprehensive studies of N_2O release from forest soils are needed to better characterize the role of denitrification and nitrification in forest ecosystem nitrogen cycles and to assess their contribution to regional and global N-budgets. Since 46 % of Austria are covered with forests (UBA, 1997) and 24 % of the total forested land on Earth are occupied by temperate forest ecosystems (BOWDEN et al., 1990) there is an urgent need to gain more information about the magnitude of individual sources and sinks and the contribution of temperate forest ecosystems to the global N_2O budget. Furthermore, the source/sink strength of forest soils for this trace gas may be significantly altered by anthropogenically increased N-deposition into such natural ecosystems. Soils in ecosystems receiving high N-deposition are likely to

Zusammenfassung

Treibhausgas-Emissionen aus Buchenwaldböden wurden während der Vegetationsperiode 1997 mittels geschlossener Kammern gemessen. Klausenleopoldsdorf 1 und Schottenwald unterschieden sich signifikant in NO_3^- und NH_4^+ Gehalten, jedoch nicht bezüglich Klima und Bodentyp, während Klausenleopoldsdorf 2 durch einen hohen Bodenwassergehalt charakterisiert war.

Die N_2O Bildungsraten waren im Schottenwald am höchsten ($70,0 \mu\text{g N}_2\text{O-N m}^{-2} \text{ h}^{-1}$), was hauptsächlich auf eine höhere Nitratverfügbarkeit zurückzuführen war. Die niedrigen N_2O Emissionsraten ($15,1 \mu\text{g N}_2\text{O-N m}^{-2} \text{ h}^{-1}$) in Klausenleopoldsdorf 2 lassen, bedingt durch den hohen Bodenwassergehalt, auf eine vollständige Denitrifikation bis zum N_2 schließen.

Klausenleopoldsdorf 1 und Schottenwald zeigten gleich hohe CO_2 Bildungsraten und fungierten als Nettosenke für Methan. Auf der Nassfläche Klausenleopoldsdorf 2 wurde Methan gebildet ($30,7 \mu\text{g CH}_4 \text{ m}^{-2} \text{ h}^{-1}$) und die CO_2 Emissionen waren geringer, was auf die vorwiegend anaeroben Bodenbedingungen zurückzuführen sein kann.

Schlagnworte: Denitrifikation, Nitrifikation, Distickstoffoxid, Methan, Kohlendioxid.

Summary

Greenhouse gas production from forest soils was measured during the 1997 vegetation period using closed chambers. Klausenleopoldsdorf 1 and Schottenwald differed significantly in NO_3^- and NH_4^+ concentrations but not in climatic conditions and soil type, while Klausenleopoldsdorf 2 was characterized by high soil water content.

N_2O emission rates were highest at Schottenwald ($70.0 \mu\text{g N}_2\text{O-N m}^{-2} \text{h}^{-1}$), which was mainly due to higher NO_3^- availability. The lower N_2O emissions at Klausenleopoldsdorf 2 ($15.1 \mu\text{g N}_2\text{O-N m}^{-2} \text{h}^{-1}$) indicated that denitrification proceeded entirely to N_2 , conditioned by high soil water content. Klausenleopoldsdorf 1 and Schottenwald showed similar CO_2 emissions and functioned as a sink for methane. At Klausenleopoldsdorf 2 methane was produced ($30.7 \mu\text{g CH}_4 \text{m}^{-2} \text{h}^{-1}$) and CO_2 emissions were low, which can be attributed to anaerobic soil conditions.

Key words: denitrification, nitrification, nitrous oxide, methane, carbon dioxide.

exhibit higher N_2O emission rates and lower CH_4 uptake compared to soils receiving lower N-deposition (BUTTERBACH-BAHL et al., 1998). Nevertheless, source/sink strengths of soils for most of the biogenic trace gases are still uncertain (BOUWMAN, 1990). This makes it crucial to understand the mechanisms and principles governing the direction and magnitude of the fluxes. While there is increasing information about the contribution of agriculture to global N_2O and CH_4 budgets, such information from forest ecosystems is scarce (BUTTERBACH-BAHL et al., 1998).

The purpose of our work was to answer the following questions: Are there differences in the magnitude and the seasonal patterns of trace gas emission rates (N_2O , CO_2 and CH_4) between the measuring plots? What is the cause of different emission rates on similar plots? What is the impact of soil properties such as soil moisture, temperature and mineral nitrogen concentrations on trace gas emission rates? The sampling plots Klausenleopoldsdorf 1 and Schottenwald differed significantly in N and C regimes and in the age of the forest stand but not in soil type. Klausenleopoldsdorf 2 was significantly different in water conditions, N regime and soil type compared to the other two investigated sampling plots. How important is the anthropogenic influence via atmospheric N-deposition?

2. Materials and methods

2.1 Study area

Two experimental plots were located in Klausenleopoldsdorf (abbreviation: KL), approximately 40 km west of Vienna, on a NNE-facing slope. The plot KL1 was located

on a moderately well-drained upslope supporting a 55-year-old stand dominated by *Fagus sylvatica* over dystic cambisol. Wetland KL2 was located at a seepage line in slope position with wet to moist conditions. The soil type was stagnic alisol (FAO, 1989).

The third experimental plot at Schottenwald (abbreviation: SW), approximately 10 km west of Vienna, was located on an upslope of SE-exposure supporting a 135-year-old *Fagus sylvatica* stand and being occupied by a dense cover of the spring geophyte *Allium ursinum* (L.). The soil of the experimental plot was classified as dystic cambisol (FAO, 1989). The plot itself was described in more detail by MEGER (1997), HAHN (1999) and JANDL et al. (1997). Further site characteristics are described in Table 1.

2.2 Experimental setup

Within each of the three experimental sites, four grids of 40 x 20 m were marked. A chamber for in situ measurements of gas production was placed in the centre of each plot and remained installed the entire sampling period. In 4-m-distance from each chamber corner, a wooden picket defined the sampling plot. Soil samples were taken between the sticks every two weeks.

The chambers were adapted hotbeds made of rigid polyethylene sheets forming a trap for gases emitted from the soil surface. Each chamber covered an area of 60 x 100 cm. Chambers were closed with a rigid polyethylene sheet on whose underside a compressible rubber seal was fixed and tightened by means of tape. Repeated measurements could therefore be conducted on the same location without soil disturbance.

Table 1: Site characteristics and soil parameters of the three investigated measuring plots
 Tabelle 1: Standortcharakterisierung und Bodenparameter der drei untersuchten Standorte

sampling site	upslope KL1	wetland KL2	upslope SW
soil type*	dystric cambisol	stagnic alisol	dystric cambisol
exposition, elevation	NNE, 510 m a. sl.	NNE, 510 m, at a seepage line below the ridge top	SE, 370 m
plant community	<i>Asperulo odoratae-Fagetum</i>	<i>Cardamino-Chrysosplenietum alternifolii</i>	<i>Aro maculati-Fagetum</i>
age of <i>Fagus</i> -trees (a)	55	(55)	135
water conditions	moderately well-drained	wet to moist	fresh
atm. N-deposition (kg N ha ⁻¹ a ⁻¹)	10.0	10.0	35.0
pH (in CaCl ₂) [∇]	5.0	5.2	4.3
N _{tot} (mg g ⁻¹) [∇]	4.8	n.d.	2.4
C _{org} (mg g ⁻¹) [∇]	74.5	n.d.	37.7
CEC (mmolc kg ⁻¹) [∇]	400.0	n.d.	76.9

[∇] chemical parameters analyzed from 0-5 cm soil depth; n.d. not determined

* according to FAO (1989)

Large chambers and short sampling periods reduced variability and avoided bias due to the buildup of high gas concentrations, which inhibits linear emission rates.

Samples were taken biweekly from April through November 1997 on each plot. Two replicate gas samples (30 ml) were taken from the headspace immediately after sealing; subsequent samples were taken after 2 and 4 h using a 50-ml syringe. The gas samples were injected into evacuated headspace vials fitted with rubber stoppers and aluminium caps. The headspace vials were stored at 4° C under water. Gas samples were analyzed within 7 days after sampling.

2.3 Soil samples

For N-mineralization 8 undisturbed soil cores were taken from each plot using metal cylinders (5 cm length, 6 cm diameter). The samples were split into two sets. The first set was cooled and taken to the laboratory immediately for analysis of nitrate and ammonium concentrations and gravimetric soil water content. The second set was incubated in polyethylene bags in the field. Samples were retrieved after 14 days, sieved (mesh size 5 mm) and stored in plastic bags at + 4° C. The N-mineralization rate was estimated as the difference between the initial and the final NO₃⁻ and NH₄⁺ concentrations after 14 days incubation. Ammonium and nitrate were extracted with 0.1 M potassium chloride solution and analyzed colorimetrically (KANDELER, 1996). Results are expressed as µg NO₃⁻-N g dw⁻¹ and µg NH₄⁺-N g dw⁻¹. Extractable glucose equivalents were

determined after hot water extraction by colorimetric determination of reducing sugars (ÖHLINGER, 1996b).

Soil water content of the sieved soil samples (0–5 cm depth) was determined gravimetrically as described by ÖHLINGER (1996a). In addition to gravimetric measurements the soil water content was measured biweekly by means of time domain reflectometry (TDR). This system determines the propagation velocity of an electromagnetic wave, which depends on the water content of the soil (TOPP et al., 1980). The TDR measurements were conducted in 0–15 cm and 0–30 cm soil depth and at each corner of the sampling plot in order to test the spatial variation of the soil water content.

Precipitation was measured at Alland (330 m a. sl.), which was 5 km away from our sampling plots in Klausenleopoldsdorf (Hydrographischer Dienst NÖ), and at Mariabrunn (226 m a. sl.), which was 3 km away from the sampling plot Schottenwald (Zentralanstalt für Meteorologie und Geodynamik). Soil temperature was measured in 0, 3 and 10 cm soil depth.

2.4 Gas samples

CO₂ and CH₄ were analyzed by a gas chromatograph (Hewlett-Packard 5890 II series) equipped with a thermal conductivity detector (TCD) and a flame ionization detector (FID).

To separate the gaseous components, two columns connected in series were used – Poraplot (Q 25-m-long,

0.53 mm diameter) and Carboplot (25-m-long, 0.53 mm diameter). Helium was used as a carrier (flow rate 10 ml min⁻¹). The detector, injector and oven temperatures of the TCD were set at 150, 60 and 80° C, respectively. Detector, injector and oven temperatures of the FID were 200, 60 and 80° C. The CO₂ standard was 10,000 ppm CO₂ and the CH₄ standard was 1 ppm CH₄.

Air samples were analyzed for N₂O using a GC (Hewlett-Packard 5890 II series) equipped with a ⁶³Ni-electron-capture-detector (ECD) and an autoinjector. The detector, injector and oven temperatures were 330, 50 and 30° C, respectively. The flow rate of the carrier gas (N₂ in ECD-quality) was 30 ml min⁻¹. Calibration was performed using a 5 ppm N₂O standard gas (Linde Gas). Results are reported as µg N₂O-N m⁻² h⁻¹, mg CO₂-C m⁻² h⁻¹ and µg CH₄ m⁻² h⁻¹.

2.5 Statistical analysis

SPSS 7.0 for Windows was used for statistical analysis of significant differences in trace gas fluxes between the three measuring plots after testing the distribution of data sets (Kolmogoroff-Smirnoff) using a non-parametric statistical test (Mann-Whitney U-test). Correlation coefficients between averaged trace gas emission rates and temperature, gravimetric soil water content, TDR measurements and NO₃⁻ and NH₄⁺ concentrations were computed using Spearman correlation analysis.

3. Results

The seasonal patterns of N₂O, CH₄ and CO₂ flux rates during April and November 1997 are shown in Fig. 1 a–c;

Fig. 1 d presents results from TDR and air temperature measurements.

3.1 Nitrous oxide (N₂O)

Over the entire observation period all sampling plots were net sources of N₂O. During the 1997 vegetation period all sampling plots showed similar seasonal patterns of N₂O emission rates, with increasing N₂O production in spring and maximum rates in mid-summer (Fig. 1a). Correlation analysis showed a high correlation ($p < 0.01$) of N₂O emissions of the two upslope plots KL1 and SW ($r = 0.919$) and of upslope KL1 and wetland KL2 ($r = 0.794$).

The seasonal mean of N₂O emission rates at upslope SW (mean ± S.E.: 70.0 ± 16.7 µg N₂O-N m⁻² h⁻¹) was significantly higher ($p < 0.05$) than that at upslope KL1 (range: 27.3 ± 5.5 µg N₂O-N m⁻² h⁻¹). These pronounced differences in N₂O flux rates between the two upslope plots were observed most of the year (Fig. 1a). While at upslope SW maximum emissions of N₂O reached 248.6 µg N₂O-N m⁻² h⁻¹ in mid-July, at upslope KL1 maximum N₂O emissions were only 76.5 µg N₂O-N m⁻² h⁻¹. The lowest seasonal N₂O emission rates (Table 2) were observed at wetland KL2 (15.09 ± 1.8 µg N₂O-N m⁻² h⁻¹). Mean N₂O emission rates of upslope KL1 and wetland KL2 differed only in June and July (Fig. 1a).

Upslope SW showed a positive correlation between N₂O emission rates and gravimetric soil water content ($r = 0.745$; $p < 0.01$). A negative correlation was found between N₂O emission rates and NO₃⁻ concentrations at upslope KL1 and SW. Results of cross-correlation analysis revealed lag1 between N₂O and NO₃⁻.

Table 2: Mean seasonal emission rates of N₂O, CH₄ and CO₂ as well as NO₃⁻ and NH₄⁺ concentrations in soil, gravimetric soil water content and air temperature (mean ± S.E.) of the sampling plots

Tabelle 2: Mittlere saisonale Emissionsraten von N₂O, CH₄ und CO₂ sowie von NO₃⁻ und NH₄⁺ Konzentrationen im Boden, gravimetrisch bestimmter Bodenwassergehalt und Lufttemperatur (Mittelwert ± Standardfehler) der untersuchten Standorte

sampling plot	upslope KL1	wetland KL2	upslope SW
µg N ₂ O-N m ⁻² h ⁻¹	27.3 ± 5.5 a*	15.09 ± 1.8 a	70.0 ± 16.7 b
µg CH ₄ m ⁻² h ⁻¹	-20.2 ± 3.7 a	30.7 ± 11.0 c	-11.5 ± 1.7 b
mg CO ₂ -C m ⁻² h ⁻¹	52.1 ± 8.8 b	24.0 ± 2.5 a	56.8 ± 8.8 b
NO ₃ ⁻ (µg N g ⁻¹ dw)	0.33 ± 0.05 b	0.21 ± 0.04 a	1.04 ± 0.16 c
% nitrate from N _{min}	17.2 ± 2.6 b	10.2 ± 1.7 a	51.4 ± 4.0 c
NH ₄ ⁺ (µg N g ⁻¹ dw)	1.7 ± 0.2 b	2.1 ± 0.2 b	0.9 ± 0.1 a
soil water content (%)	31.4 ± 1.4 a	39.3 ± 0.9 b	29.7 ± 1.0 a
air temperature (°C)	15.0 ± 1.6 a	15.0 ± 1.6 a	16.0 ± 1.4 a
soil temperature (°C)	11.4 ± 1.3 a	11.4 ± 1.3 a	12.6 ± 1.1 a

* Different letters represent significantly different means between the sampling plots (Mann-Whitney U-test).

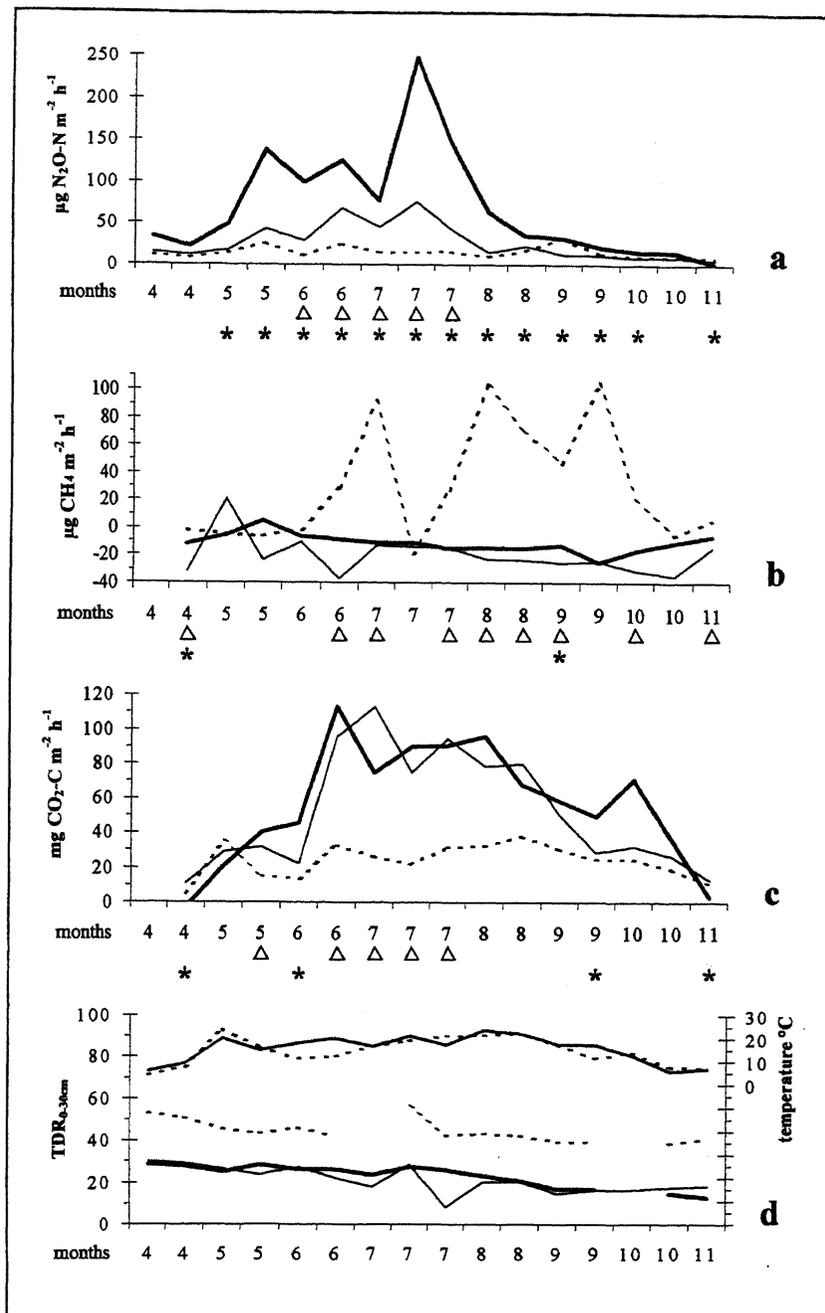


Figure 1: Seasonal patterns of trace gas emissions for the measuring period April to November 1997 at three different forest stands near Vienna: a – Seasonal pattern of N_2O emission rates. b – Seasonal course of methane production at wetland KL2 and methane oxidation at the two upslope plots SW and KL1. c – Net- CO_2 -release from soil. d – Results of TDR measurements in 0–30 cm depth ($\text{TDR}_{0-30\text{cm}}$), averaged air temperature (continuous line) and soil temperature in 3 cm depth (broken line). Slender line: upslope KL1; broken line: wetland KL2; bold line: upslope SW

Δ results of mean difference test (U-test; $p < 0.05$): significantly different means between upslope KL1 and wetland KL2

* significantly different means between the two upslope plots KL1 and SW at the 0.05 level

Abbildung 1: Saisonaler Verlauf der Spurengas-Emissionen von April bis November 1997 an drei unterschiedlichen Waldstandorten nahe Wien: a – Saisonaler Verlauf der N_2O Emissionsraten. b – Methanbildung auf der Nassfläche KL2 und Methanabbau an den beiden Hangstandorten SW und KL1. c – Netto CO_2 -Bildung. d – Ergebnisse der TDR-Messung in 0–30 cm Tiefe ($\text{TDR}_{0-30\text{cm}}$), gemittelte Lufttemperatur (durchgehende Linie) sowie Bodentemperatur in 3 cm Tiefe (unterbrochene Linie). Dünne Linie: Hang KL1; gestrichelte Linie: Nassfläche KL2; dicke Linie: Hang SW

Δ Ergebnisse des Mittelwertvergleiches (U-Test; $p < 0,05$): signifikanter Mittelwertunterschied zwischen Hang KL1 und Nassfläche KL2

* signifikant unterschiedliche Mittelwerte zwischen den beiden Hangstandorten KL1 und SW ($p < 0,05$)

3.2 Methane (CH₄)

Measured net CH₄ fluxes are the balance of methane production and consumption. While at wetland KL2 a net CH₄ production was detected in most cases, the soil of the two upslope plots KL1 and SW functioned as a net sink of CH₄ during the entire sampling period (Fig. 1b). Seasonal mean oxidation rates of atmospheric methane at upslope KL1 were in the range of $-20.2 \pm 3.7 \mu\text{g CH}_4 \text{ m}^{-2} \text{ h}^{-1}$ (mean \pm S.E.; see Table 2). CH₄ consumption at upslope SW was in the range of $-11.5 \pm 1.7 \mu\text{g CH}_4 \text{ m}^{-2} \text{ h}^{-1}$. The mean CH₄ production rate obtained at wetland KL2 was $30.7 \pm 11.0 \mu\text{g CH}_4 \text{ m}^{-2} \text{ h}^{-1}$, which was significantly higher than CH₄ oxidation at the two upslope plots KL1 and SW ($p < 0.001$). Maximum CH₄ emission rates at KL 2 were obtained in summer ($105.5 \mu\text{g CH}_4 \text{ m}^{-2} \text{ h}^{-1}$). Low CH₄ production occurred in July during a period of heavy rainfalls. Correlation analysis (Spearman) showed a negative correlation between net CH₄ emission rate and gravimetric soil water content ($r = -0.679$, $p < 0.01$) at wetland KL2.

3.3 Carbon dioxide (CO₂)

All sampling plots showed net CO₂ production over the entire sampling period (Fig. 1c) regardless of the presence of photosynthetically active undergrowth. A comparison of CO₂ emission rates of the different measuring plots revealed similar seasonal trends. A correlation ($p < 0.01$) between the plots KL1 – KL2 ($r = 0.739$) and KL1 – SW ($r = 0.875$) could be shown.

Net CO₂ emission rates were highest at the two upslope plots KL1 and SW. Seasonal mean CO₂ emission rates ranged from $52.1 \pm 8.8 \text{ mg CO}_2\text{-C m}^{-2} \text{ h}^{-1}$ (mean \pm S.E.) at upslope KL1 to $56.8 \pm 8.8 \text{ mg CO}_2\text{-C m}^{-2} \text{ h}^{-1}$ at upslope SW, there was no significant difference in means. Maximum CO₂ emissions were measured in June at upslope SW ($113.0 \text{ mg CO}_2\text{-C m}^{-2} \text{ h}^{-1}$). At wetland KL2, the seasonal mean CO₂ emission rate was in the range of $24.0 \pm 2.5 \text{ mg CO}_2\text{-C m}^{-2} \text{ h}^{-1}$, which is 2.3 magnitudes lower than the mean values of the two upslope plots (Table 2).

CO₂ and N₂O emission rates were positively correlated at all sampling plots. A correlation between CO₂ and temperature was also found at all plots.

3.4 Impact of soil and climate parameters on trace gas emission rates

While positive values of N-mineralization indicate ammonification and nitrification (NO₃⁻/NH₄⁺ release), negative values indicate net immobilization of mineral nitrogen in soil. At upslope KL1 positive values were measured over the entire sampling period, while wetland KL2 and upslope SW showed both positive and negative values. With regard to N-mineralization, no significant statistical differences between the three investigated plots could be demonstrated. In early May N-immobilization occurred at all sampling plots; at this time air temperature and mineral nitrogen concentrations were elevated.

Under the warm and moist conditions in spring and summer, nitrogen mineralization was elevated and nitrogen availability consequently increased. With regard to extractable NO₃⁻, highest concentrations were detected at upslope SW (51.4 % nitrate of mineral nitrogen; Fig. 2). Mean NO₃⁻ concentrations differed significantly between the two upslope plots SW and KL1 ($p < 0.001$) and between upslope KL1 and wetland KL2 ($p < 0.01$) (Table 2). NH₄⁺ concentration was significantly lower ($p < 0.05$) at upslope SW than at upslope KL1 and wetland KL2 (Table 2). The highest NH₄⁺ concentrations were detected at wetland KL2.

No correlation between N-mineralization and N₂O emission rates was found for any of the sampling plots. N₂O emissions and NO₃⁻ concentrations correlated negatively ($p < 0.05$) at upslope KL1 ($r = -0.635$) and at upslope SW ($r = -0.538$). N₂O and NH₄⁺ were not correlated at these plots. At upslope SW a negative correlation between N₂O emission rates and total mineral nitrogen ($r = -0.574$; $p < 0.05$) was found.

Since mean temperatures of the three sampling plots did not differ significantly and high correlations between the plots were observed, the air and soil temperatures were averaged. The highest air temperature was recorded in August (23.8° C); the mean air temperature was 15.5° C and mean soil temperature was 12.0° C. Soil temperature was on average 3.5° C lower than air temperature (Fig. 1d).

Gravimetric soil water content showed similar seasonal patterns at upland KL1 and at wetland KL2. Although the latter site showed significantly higher ($p < 0.001$) values ($39.3 \pm 0.9 \%$) than the two upslope plots KL1 and SW (31.4 % and 29.7 %, respectively). KL1 and SW did not differ significantly in mean water content (Table 2). These two plots exhibited a negative correlation ($p < 0.05$)

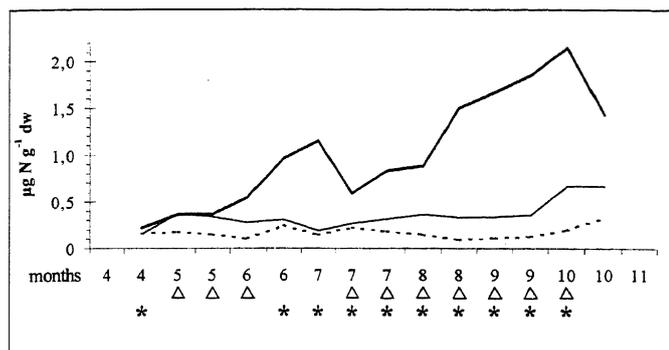


Figure 2: Concentrations of extractable nitrate (in $\mu\text{g N g}^{-1} \text{ dw}$) from three forest soils during the 1997 vegetation period. Slender line: upslope KL1; broken line: wetland KL2; bold line: upslope SW

Δ Results of mean difference test (U-test; $p < 0.05$): significantly different means between upslope KL1 and wetland KL2

* significantly different means between the two upslope plots KL1 and SW at the 0.05 level

Abbildung 2: Nitratkonzentrationen (in $\mu\text{g N g}^{-1} \text{ TG}$) von drei Waldböden während der Vegetationsperiode 1997. Dünne Linie: Hang KL1; gestrichelte Linie: Nassfläche KL2; dicke Linie: Hang SW

Δ Ergebnisse des Mittelwertvergleichs (U-Test; $p < 0,05$): signifikanter Mittelwertsunterschied zwischen Hang KL1 und Nassfläche KL2

* signifikant unterschiedliche Mittelwerte zwischen den beiden Hangstandorten KL1 und SW ($p < 0,05$)

between water content and NO_3^- concentrations ($r = -0.640$ at KL1; $r = -0.534$ at SW), while a positive correlation was detected at wetland KL2 ($r = 0.640$; $p < 0.05$).

Precipitation during the sampling period (April to November) was 734 mm at Klausenleopoldsdorf sampling plots (KL1 and KL2) and 683 mm in Schottenwald (SW). Seasonal means did not differ significantly. Heavy rainfalls early in July coincided with highest N_2O emission rates.

Seasonal means (\pm S.E.) of TDR measurements in 0–30 cm depth were 21.1 ± 1.5 vol% at upslope KL1, 44.6 ± 1.4 vol% at wetland KL2 and 22.8 ± 1.4 vol% at upslope SW (Table 2). The values at wetland KL2 were highest and differed significantly from the other sites. Differences in seasonal means were significant between the plots KL1 – KL2 and SW – KL2 at the level $p < 0.001$. No significant differences were detected between the two upslope plots KL1 and SW. Gravimetric soil water content and TDR measurements correlated closely at upslope KL1 and at wetland KL2 (KL1: $r = 0.864$ and KL2: $r = 0.813$; $p < 0.01$ for $\text{TDR}_{15\text{cm}}$), but not at upslope SW. TDR measurements and NO_3^- concentrations correlated negatively at upslope

SW ($r = -0.841$ for $\text{TDR}_{15\text{cm}}$ and $r = -0.868$ for $\text{TDR}_{30\text{cm}}$; $p < 0.01$). Correlations of TDR measurements with trace gas emissions or soil parameters were not found.

Extractable glucose equivalents indicate the C-availability for microorganisms in soil. Wetland KL2 showed the lowest amount of available glucose ($6.3 \text{ g gluc g}^{-1} \text{ dw}$), while values at upslope SW were twice as high ($12.5 \text{ g gluc g}^{-1} \text{ dw}$). At upslope KL1 $9.6 \text{ g gluc g}^{-1} \text{ dw}$ were measured. Mean values differed significantly between the three plots. Extractable glucose equivalents correlated closely with air temperature at upslope KL1 ($r = 0.842$) and at wetland KL2 ($r = 0.806$; $p < 0.01$). No correlations between glucose equivalents and trace gas emission rates, soil water content or mineral nitrogen concentration were detected.

4. Discussion

Our results indicate different seasonal patterns and magnitudes of N_2O emission rates between the investigated plots. But what was the cause of the temporarily increased N_2O emission rates? How did seasonal variations in temperature, soil moisture, mineral nitrogen and carbon availability influence trace gas emission rates? Why did higher soil moisture at wetland KL2 not cause elevated N_2O emission rates via denitrification? Did the higher nitrogen loads via atmospheric deposition at upslope SW provoke higher emission rates or were they caused by a higher nitrification activity?

4.1 Nitrous oxide (N_2O)

The two upslope plots KL1 and SW showed similar seasonal patterns in N_2O emission rates ($r = 0.919$; $p < 0.01$) but significantly different mean emissions ($p < 0.05$) (Fig. 1a and Table 2). Emission rates were by far the highest at upslope SW, whereas the lowest and steadiest rates were measured at wetland KL2.

Impact of soil properties and climatic conditions:

Temperature was probably not the main reason for the higher N_2O emission rates at upslope SW, since temperature and N_2O emission rates did not correlate at any of the plots and no significant differences in temperature between the sampling plots were detected.

Gravimetric soil water content was significantly higher at wetland KL2. The reduction of N_2O to N_2 increases with increasing soil water content. The low N_2O emissions rates

at wetland KL2 could therefore be explained by the fact that denitrification processes proceeded completely to N_2 due to the anaerobic soil conditions here. Nitrification at wetland KL2 was inhibited by the anaerobic soil conditions, and consequently NO_3^- availability was low and NH_4^+ concentration high. Nitrate might have been a limiting factor for denitrification processes at KL2. Our hypothesis is that the significantly lower amount of available carbon at KL2 was a limiting factor for microbial activity. This interpretation is supported by results of CO_2 measurements at wetland KL2.

At the two upslope plots (KL1 and SW) NO_3^- concentrations were negatively correlated with gravimetric soil water content: precipitation events led to nitrate leaching here. At upslope SW, the soil water content affected N_2O emission rates positively. Soil conditions permitted both denitrification and aerobic nitrification, resulting in high emission rates at SW.

Gravimetric soil water content and results of TDR measurements correlated closely at KL1 and KL2 but not at SW probably because gravimetric water content was determined in 0–5 cm depth and TDR measurements in 0–15 and 0–30 cm depth. Layering of sediments and abundant fine material at SW causes temporarily waterlogged conditions, as evident from manganese concretions (JANDL et al., 1997), and causes varied water conditions in different soil horizons. Upslope SW is drained by lateral water flow.

In autumn, N_2O emission rates at upslope KL1 and SW were as low as in spring, while NO_3^- concentrations were elevated. These low rates might be due to the dry soil conditions in autumn 1997. Wetland KL2 showed relatively high rates, especially in September (maximum $32.5 \mu g N_2O-N m^{-2} h^{-1}$). We assume that a decreasing soil water content in autumn could have created aerobic soil conditions favourable for nitrification. Heavy rainfalls in early July coincided with the highest N_2O emission rates. Frequent precipitation could have had a positive effect on the nitrate-reductase-activity (J. M. ANDERSON, pers. comm.). However, only at upslope KL1 precipitation and N_2O emissions did correlate positively.

Mineralization rates were low at all sampling sites, perhaps due to the short incubation period or to rapid nitrification and immobilization. Favourable temperature and moisture conditions in spring and summer promoted the decay and mineralization of organic matter and, consequently, the high availability of mineral nitrogen (Fig. 2). At upslope KL1, ammonification and nitrification occurred

over the entire measuring period, while upslope SW showed N -release in early spring and in summer and N -immobilization in May and in September/October. At wetland KL2, N -immobilization and N -release varied considerably. In early May, increasing temperatures boosted microbial activity; N -immobilization occurred at all sampling plots.

In forest ecosystems, high nitrification rates are expected when nitrogen availability in the soil is high (MEGER, 1997). The highest N_2O emission rates were measured at upslope SW plot, where by far the highest soil nitrate and the lowest soil ammonium concentrations were measured. Extractable NO_3^- was 51.4 % of total mineral nitrogen at upslope SW, while at upslope KL1 and at wetland KL2 nitrate amounted to only 17.2 % and 10.2 %, respectively. The $NH_4^+ : NO_3^-$ ratio was 0.9 at SW, 5.2 at KL1 and 10.0 at KL2. This indicates high nitrification rates at upslope SW.

The upslope plots KL1 and SW exhibited a negative correlation between N_2O emission rates and NO_3^- concentrations. This fact could be explained by a retarded N_2O production compared to NO_3^- release via mineralization. Such a negative correlation may also be the effect of the low N_2O emission rates in autumn combined with high NO_3^- availability.

Simultaneously with high NO_3^- concentrations, N_2O emission rates at upslope SW were highest in June/July as a consequence of high nitrification rates. During this period, JANDL et al. (1997) observed a sharp decline of soil solution pH here, which was partially due to H^+ ions emitted during nitrification processes. Soil pH itself is an important additional modulator of N trace gas production and emission (BUTTERBACH-BAHL et al., 1997). At low soil pH, N_2O release increases compared to N_2 release because N_2O -reductase is sensitive to low pH. In acid soils (pH < 4.5) with large amounts of organic matter, heterotrophic organisms take over the transformation of NH_4^+ to NO_3^- (FOCHT and VERSTRAETE, 1977). In the slightly acidic soil of upslope SW (pH 4.3) heterotrophic nitrifiers could have played a considerable role.

The highest NH_4^+ concentrations and the lowest NO_3^- concentrations in soils were observed at wetland KL2. Nitrification rates here were probably low as a result of oxygen limitations and measured N_2O production mainly due to the denitrification process (HAHN, 1999). Maximum N_2O emitted at wetland KL2 was approximately $32.5 \mu g N_2O-N m^{-2} h^{-1}$. N_2O emission via denitrification might be determined by the reduced nitrate supply during nitrification.

Impact of the vegetation and microbial community:

N-availability and consequently the magnitude of N_2O emission rates may be altered by the vegetation, by the age of a forest stand, soil conditions, and climatic parameters (MEGER, 1997). Factors such as stand age and species characteristics (e.g. litter quality) may also affect N-availability (SCOTT and BINKLEY, 1997). Since the N-demands, N-storage pools and shoot : root ratios of adult and young trees are different (GESSLER et al., 1998), tree age might influence absolute rates of nitrogen uptake per root. At upslope SW the investigated *Fagus sylvatica* stand is 135 years old (JANDL et al., 1997), while beech trees at upslope KL1 are 55 years old. The $NH_4^+ : NO_3^-$ ratio differed significantly between these plots ($NH_4^+ : NO_3^-$ ratio: 5.2 at KL1 and 0.9 at SW). This could be a consequence of the different N-demand of the vegetation depending on tree age and therefore different rates of mineral nitrogen uptake.

Soil parameters may have influenced microbial community diversity and activity at the three plots. For example, microbial biomass, C_{org} and N_{tot} content in the soil were significantly higher at upslope KL1 versus upslope SW (Table 1), while extractable glucose equivalents at KL1 were significantly lower. The cation exchange capacity at KL1 was 400 mmolc kg^{-1} . Nutrient conditions at KL1 might have been favourable for microorganisms so that microbial biomass here was higher than at SW. KL1 and SW showed similar CO_2 emissions, but N_2O emission rates were elevated at the former, probably due to a higher NO_3^- availability.

Plant roots themselves strongly influence microbial growth and activity in the rhizosphere. Plants affect microbial activity through their uptake of water and nutrients, through the release of root exudates, and through other changes in the physical and chemical microenvironment of microbes (VAN VEEN et al., 1993).

The vegetation of upslope SW had a considerable impact on nutrient dynamics, especially on the amount of available nitrate and ammonium. Upslope SW is covered with a dense *Allium ursinum*-dominated herb layer. The mineralization of *Allium*-leaves in mid-summer resulted in a high availability of nitrate. STARK and HART (1997) indicated that NO_3^- assimilation and rapid microbial turnover will continually deplete NO_3^- pools, and minimal NO_3^- leaching will occur despite significant nitrification rates. However, results of ion exchange bags from the 1996 vegetation period (MEGER, 1997) and of tension lysimeters (JANDL et al., 1997) showed considerable NO_3^- leaching into deeper soil horizons in June/July at upslope SW. In mid-July 1997, high nitrate leaching occurred after heavy rains. In this peri-

od the highest N_2O emission rates at upslope SW coincided with a lower NO_3^- concentration in the soil and elevated NO_3^- leaching rates (data from ion exchange bags).

The influence of atmospheric N-deposition on trace gas emissions in forest ecosystems:

BUTTERBACH-BAHL et al. (1997) reported that the high N_2O emission rates of a beech forest in Germany might be due to high nitrogen inputs by atmospheric deposition. N_2O fluxes at the site receiving high atmospheric N-input by wet deposition were 1.5-5-fold higher. Results of N-fertilization experiments by PAPEN et al. (1994) indicated that elevated N_2O emission rates of a natural spruce forest stand might be the consequence of chronic N-loads. Upslope SW, located close to the city of Vienna, received an atmospheric N-input of 35 $kg N a^{-1} ha^{-1}$, while atmospheric N-deposition at upslope KL1 was in the range of 10 $kg N a^{-1} ha^{-1}$ (NEUMANN und SMIDT, 1997). We therefore suppose that the higher atmospheric N-deposition at SW could also have affected the N_2O emission rates.

4.2 Methane (CH_4)

The regulation of methane is determined both by the production in anoxic soil and by CH_4 consumption in unsaturated soil. In our study, CH_4 consumption was observed at the two upslope plots SW and KL1, while wetland KL2 was a net source of methane (Fig. 1b). KL2 differed significantly from SW and KL1 ($p < 0.001$) in gravimetric soil water content. The low to negative CH_4 emission rates at the two latter plots were due to low methane production in the anaerobic parts of the soil and increased methane oxidation in the aerobic part. At wetland KL2, high methane emissions were the result of high CH_4 production because of the waterlogged soil conditions and the low CH_4 consumption. BORKEN (1996) indicated that gravimetric soil water content, which also controls oxygen and methane diffusion in the soil, was the most important factor controlling methane oxidation. In our study, CH_4 emission rates at KL2 and gravimetric soil water content correlated negatively. This could be related to the high variability of methane emission rates here during the 1997 vegetation period. The large variability of CH_4 emission rates at KL 2 was mainly due to differences in soil moisture. In July, during a period of heavy rainfall, methane production at KL2 decreased. We assume that the decomposition of organic matter was low because of the anoxic soil conditions and

that low C-availability limited the activity of methane-producing microorganisms.

4.3 Carbon dioxide (CO₂)

Net CO₂ release from soil is a result of production and plant assimilation. The intensity of microbial activity may be concluded from CO₂ emission rates, but a differentiation of CO₂-producing processes is not possible. Mineralization of organic matter and root respiration contribute considerably to CO₂ emissions, their seasonal dynamic being controlled by temperature and soil water content. The three sampling plots showed analogous seasonal trends in CO₂ production. The low CO₂ emission rates measured at wetland KL2 resulted from inhibited microbial activity due to anaerobic soil conditions and/or significantly lower carbon availability here. CO₂ emission rates of both upslope plots, KL1 and SW, were equal and were significantly higher than at KL2. Our results confirmed that CO₂ production was positively influenced by temperature.

5. Conclusion

Since the edaphic and climatic conditions of the two upslope plots KL1 and SW were comparable, we conclude that the difference in NO₃⁻ availability was the main factor responsible for the significantly higher N₂O emission rates at SW. Nitrate concentrations at SW were significantly higher than at the other sites (51 % nitrate of total mineral nitrogen). N-availability strongly depended on microbial activity, which was influenced by carbon availability, temperature and soil water content. Variations in nitrogen availability also could be due to the vegetation. Low N₂O emission rates at wetland KL2 might have been due to the anaerobic soil conditions, so that denitrification proceeded completely to N₂. Furthermore, nitrification might have been inhibited by anaerobic soil conditions, so that low nitrate availability was a limiting factor for denitrification.

Regarding net CH₄ fluxes, the two upslope plots KL1 and SW functioned as methane sinks, while at wetland KL2 methane was produced. High gravimetric soil water content and the resulting anaerobic soil conditions seemed to be the main reason for CH₄ production at wetland KL2.

CO₂ production showed analogous seasonal patterns at KL1 and SW. Low CO₂ production at wetland KL2 resulted from inhibited microbial activity due to anaerobic soil

conditions. Temperature apparently had a considerable impact on CO₂ production.

To obtain more precise estimations of seasonal N₂O emission rates from different natural ecosystems further studies are required. BUTTERBACH-BAHL et al. (1997) suggested the development of process-based models and their validation with data from continuous measurements of trace gas fluxes and environmental parameters over entire years. Based on our present experience, this approach would also offer insight into the trace gas emission rates of large areas and over longer periods, where continuous measurement is not practicable.

Acknowledgements

Special thanks are given to F. Ruhm, E. Leitgeb, R. Albert and B. Schraufstädter.

References

- BORKEN, W. (1996): Methanaufnahme und Kohlendioxid-freisetzung von Waldböden. Berichte des Forschungszentrums Waldökosysteme der Universität Göttingen, Reihe A, Bd. 137.
- BOUWMAN, A. F. (1990): Soils and the Greenhouse Effect. Proceedings of the International Conference on Soils and the Greenhouse Effect, organized by International Soil Reference and Information Centre. John Wiley and Sons, Chichester.
- BOWDEN, R. D., P. A. STEUDLER, J. M. MELILLO and J. D. ABER (1990): Annual nitrous oxide fluxes from temperate forest soils in the North-eastern United States. *J. Geophys. Res.* 95, 13997–14005.
- BUTTERBACH-BAHL, K., R. GASCHE, L. BREUER and H. PAPAN (1997): Fluxes of NO and N₂O from temperate forest soil: impact of forest type, N-deposition and of liming on the NO and N₂O emissions. *Nutrient Cycling in Agroecosystems* 48, 79–90.
- BUTTERBACH-BAHL, K., R. GASCHE, CH. HUBER, K. KREUTZER and H. PAPAN (1998): Impact of N-input by wet deposition on N-trace gas fluxes and CH₄-oxidation in spruce forest ecosystems of the temperate zone in Europe. *Atmospheric Environment* 32(3), 559–564.
- CICERONE, R. J. (1987): Changes in atmospheric ozone. *Science* 237, 35–42.
- DAVIDSON, E., W. T. SWANK and T. PERRY (1986): Distinguishing between nitrification and denitrification as

- sources of gaseous nitrogen production in soil. *Applied and environmental microbiology* 52(6), 1280–1286.
- DAVIDSON, E. A. and W. T. SWANK (1987): Factors limiting denitrification in soils from mature and disturbed south-eastern hardwood forests. *Forest Science* 33, 135–144.
- EDWARDS, A. C. and K. KILLHAM (1986): The effect of freeze/thaw on gaseous nitrogen loss from upland soils. *Soil Use and Management* 2, 86–91.
- FAO (1989): Unesco soil map of the world. Revised Legend. World Soil Resources Report 60, 1988, Rome. Reprinted as Technical Paper 20, ISRIC, Wageningen.
- FOCHT D. D. and W. VERSTRAETE (1977): Biochemical ecology of nitrification and denitrification. *Adv. Microb. Ecol.* 1, 135–214.
- GAMBRELL, R. P., J. W. GILLIAM and S. B. WEED (1975): Denitrification in subsoils of the North Carolina coastal plain as affected by soil drainage. *Journal of Environmental Quality* 4, 311–316.
- GESSLER, A., S. SCHNEIDER, D. VON SENGBUSCH, P. WEBER, U. HANEMANN, C. HUBER, A. ROTHE, K. KREUTZER and H. RENNENBERG (1998): Field and laboratory experiments on net uptake of nitrate and ammonium by the roots of spruce (*Picea abies*) and beech (*Fagus sylvatica*) trees. *New Phytol.* 138, 275–285.
- GROFFMAN, P. M. and J. M. TIEDJE (1988): Denitrification hysteresis during wetting and drying cycles in soil. *Soil Science Society of America Journal* 52, 1626–1629.
- HAHN, M. (1999): Spurengas-Emissionen (N₂O, CO₂ und CH₄) aus Buchenwald-Ökosystemen im Wiener Wald. Diplomarbeit an der Naturwissenschaftlichen Fakultät der Universität Wien.
- JANDL, R., H. KOPESZKI and G. GLATZEL (1997): Effect of a dense *Allium ursinum* ground cover on nutrient dynamics and mesofauna of a *Fagus sylvatica* woodland. *Plant and Soil* 0, 1–11.
- KANDELER, E. (1996): Ammonium and Nitrate. In: F. SCHINNER, R. ÖHLINGER, E. KANDELER and R. MARGESIN (eds.): *Methods of soil biology*, Springer Verlag, Berlin, 406–410.
- MEGER, S. (1997): Stickstoffkreislauf und Methanoxidation in Buchenwäldern. Diplomarbeit an der Naturwissenschaftlichen Fakultät der Universität Wien.
- NEUMANN, M. und S. SMIDT (1997): Niedrige Schadstoffeinträge in Österreichs Wälder. *Österreichische Forstzeitung* 10, 53–54.
- ÖHLINGER, R. (1996a): Methods in Soil Physics. In: F. SCHINNER, R. ÖHLINGER, E. KANDELER and R. MARGESIN (eds.): *Methods of Soil Biology*, Springer Verlag, Berlin, 396.
- ÖHLINGER, R. (1996b): CM-cellulase activity. In: F. SCHINNER, R. ÖHLINGER, E. KANDELER and R. MARGESIN (eds.): *Methods of Soil Biology*. Springer Verlag, Berlin, 190–193.
- PAPEN, H., H. HERMANN, K. BUTTERBACH-BAHL und H. RENNENBERG (1994): Emissionen von N₂O, NO und NO₂ aus Böden zweier Fichtenstandorte im Schwarzwald. Europäisches Forschungszentrum für Maßnahmen zur Luftreinhaltung (PEF) im Kernforschungszentrum Karlsruhe (Hrsg). *Forschungsbericht KfK-PEF* 127.
- ROBERTSON, G. P. and J. M. TIEDJE (1984): Denitrification and nitrous oxide production in old growth and successional Michigan forests. *Soil Science Society of America Journal* 48, 383–389.
- SCOTT, N. A. and D. BINKLEY (1997): Foliage litter quality and annual net N mineralization: comparison across North American forest sites. *Oecologia* 111, 151–159.
- SEXSTONE, A. J., T. B. PARKIN and J. M. TIEDJE (1985): Temporal response of soil denitrification rates to rainfall and irrigation. *Soil Science Society of America Journal* 49, 99–103.
- STARK, J. M. and S. C. HART (1997): High rates of nitrification and nitrate turnover in undisturbed coniferous forests. *Nature* 385, 61–64.
- TOPP G. C., J. L. DAVIES and A. P. ANNAN (1980): Electromagnetic determination of soil water content: Measurements in coaxial transmission lines. *Water Resour. Res.* 16, 574–582.
- UMWELTBUNDESAMT – UBA (1997): State of the environment in Austria. Federal Environmental Agency Austria. Federal Ministry for Environment, Youth and Family, Vienna, 46–60 and 145–146.
- VAN VEEN, J. A., P. J. KUIKMAN and E. BREMER (1993): The regulation of carbon and nitrogen turnover in the rhizosphere. In: R. GUERRERO and PEDRÓS-ALIÓ (eds.): *Trends in Microbial Ecology*. Spanish Society for Microbiology.

Address of authors

Mag. Maria Hahn, Mag. Karl Gartner, Dr. Sophie Zechmeister-Boltenstern, Forstliche Bundesversuchsanstalt, Institut für Forstökologie, Seckendorff-Gudentweg 8, A-1131 Wien.
e-mail: sophie.zechmeister@FBVA.bmlf.gv.at

Eingelangt am 16. August 1999

Angenommen am 28. Dezember 1999