

## Fluxes of CO<sub>2</sub>, CH<sub>4</sub> and N<sub>2</sub>O from a temperate forest soil: the effects of leaves and humus layers

By Y. DONG<sup>1,2</sup>, D. SCHARFFE<sup>1</sup>, J. M. LOBERT<sup>3</sup>, P. J. CRUTZEN<sup>1</sup> and E. SANHUEZA<sup>1,4,\*</sup>, <sup>1</sup>MPI für Chemie, Luftchemie, Postfach 3060, 55020 Mainz, Germany; <sup>2</sup>Chinese Academy of Science, Inst. of Geography, Beijing, China; <sup>3</sup>UCSD/SIO, Center for Clouds, Chemistry and Climate, 9500 Gilman Drive #0239, La Jolla, CA 92093, USA; <sup>4</sup>IVIC, Atmos. Chem. Lab., Apartado 21827, Caracas 1020, Venezuela

(Manuscript received 2 October 1996; in final form 4 February 1998)

### ABSTRACT

Fluxes of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O from forest soils were measured with an enclosed chamber technique between October 1990 and December 1991 in a deciduous forest near Darmstadt, Germany. Flux measurements were made before and after the removal of leaves and humus layer from the forest floor, and gas fluxes from the leaves and humus alone were also measured as well as depth profiles of CH<sub>4</sub>, N<sub>2</sub>O, and soil moisture. Except for N<sub>2</sub>O, large seasonal variations were observed with generally higher gas fluxes during the summer. CO<sub>2</sub> and CH<sub>4</sub> fluxes were significantly dependent on changes in ambient temperature, whereas N<sub>2</sub>O fluxes were more affected by soil moisture. A good correlation between CO<sub>2</sub> production and CH<sub>4</sub> uptake was observed, but no relationship was found between N<sub>2</sub>O emissions and either CO<sub>2</sub> or CH<sub>4</sub> fluxes. Depth profiles of the CH<sub>4</sub> mixing ratio in soil air consistently showed an exponential decrease with depth, whereas N<sub>2</sub>O profiles were highly variable and appeared to be related to changes in soil moisture. The manipulated soil experiments indicate that the leaves and the humus layers contribute significantly to the soil-atmosphere exchange of trace gases.

### 1. Introduction

Human activities have caused significant changes in the atmospheric concentrations of several greenhouse gases, and it is believed that these changes are leading to climate warming (Houghton et al., 1996). Carbon dioxide (CO<sub>2</sub>), methane (CH<sub>4</sub>), and nitrous oxide (N<sub>2</sub>O) are currently increasing in the atmosphere at rates of 1000 ppb yr<sup>-1</sup>, 7 ppb yr<sup>-1</sup>, and 0.7 ppb yr<sup>-1</sup> (ppb = parts per billion, 10<sup>-9</sup> moles per mole), respectively (Prather et al., 1995; Sanhueza et al., 1995; Schimel et al., 1995). They were responsible for an estimated 56%, 15% and 7% of the anthro-

pogenically driven radiative forcing during the period 1958–1989 (Hansen and Lacis, 1990). Besides affecting climate, N<sub>2</sub>O and CH<sub>4</sub> are also involved in the chemistry of the stratosphere. N<sub>2</sub>O is mainly transported to the stratosphere where part of it is oxidized to nitric oxide (NO), which acts as a catalyst in ozone chemistry (Crutzen, 1970).

The soil-vegetation system was reported to affect the atmospheric budget of CO<sub>2</sub> (Schimel et al., 1995), CH<sub>4</sub>, and N<sub>2</sub>O (Prather et al., 1995; Sanhueza et al., 1995). Here we report measured fluxes of these gases from the soil of a temperate, deciduous forest and the effects of the leaves and the humus material on these fluxes. For a discussion of CO fluxes from the same study, see Sanhueza et al. (1998).

\* Corresponding author.

## 2. Experimental

Fluxes of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O from a forest soil were measured near Darmstadt, Germany (49.86°N, 8.65°E) between October 1990 and December 1991 in a deciduous forest consisting of mainly beech and oak trees with a density of ~600 trees per hectare and a litterfall production of ~570 g dry matter m<sup>-2</sup> yr<sup>-1</sup>. The forest has not been disturbed for more than 80 years and its soil is a cambisol. Above the 4–5 cm O-horizon, in this paper called “humus layer”, there is a 1–2 cm thick layer of fresh, i.e. non- or partially decomposed leaves. The mineral soil (A horizon) consists of sand between the 5–100 cm layer. The bulk soil density for the upper part of the soil (0 to 15 cm) range between 1.3 and 1.5 g cm<sup>-3</sup>. The carbon, hydrogen, and nitrogen content and the pH (in 0.01 M CaCl<sub>2</sub> solution) of leaves and various soil layers are given in Table 1.

Flux measurements were made from undisturbed soil, after the removal of leaves (soil without leaves) and after the removal of both leaves and humus layer from the forest floor (mineral soil), as well as from the leaves and humus separately. The chambers for our enclosed chamber technique were made of acrylic material and consisted of two parts, a box (without top and bottom) of ~0.45 m<sup>2</sup> surface area and a volume of ~70 l, which was inserted directly into the forest floor, and a cover, which was placed on top (sealed with a PVC gasket) and removed between experiments. A typical experiment lasted for about 30 min, during which CO<sub>2</sub> was continuously monitored with a non-dispersive infrared analyzer (Leybold Heraeus). Gas samples were supplied to the analyzer by a teflon-covered membrane pump (ASF,

type 7005) and circulated back to the chamber at a flow rate of 1 litre min<sup>-1</sup>. For the determination of CH<sub>4</sub> and N<sub>2</sub>O, air samples were taken from the chamber every 10 minutes with 1.1 litre, evacuated, electropolished, stainless steel canisters. These samples were analyzed within 24 hours following the experiment by gas chromatography. For methane, a Shimadzu Mini II gas chromatograph (GC) equipped with a molecular sieve 13 × column (60–80 mesh, 3.15 mm OD and 3 m length) and a flame-ionization detector was used; the carrier gas was nitrogen, with a flow rate of 30 ml min<sup>-1</sup>, the injection sample loop was ~4 ml, the oven and detector temperatures were 120°C and 150°C, respectively. The analysis of nitrous oxide was carried out with a Dani GC equipped with a Porapak N column (80–100 mesh, 3.15 mm OD and 3 m length) and a <sup>63</sup>Ni electron-capture detector; the carrier gas was a mixture of argon (95%) and methane (5%) with a flow rate of 28 ml min<sup>-1</sup>, the injection sample loop was ~5 ml; oven and detector temperatures were 70°C and 350°C, respectively. The instruments were calibrated with a two point calibration method using gravimetric gas standards (Deuste Steininger).

The sequence of measurements made during each one-day campaign in the field was as follows: First, the lower part of chamber one was pressed about 5 cm deep into the forest floor; after 60 min, it was closed and flux measurements from native, undisturbed soil were performed. After the experiment, this chamber was opened, the leaves were removed from the forest floor and were placed inside a second chamber with a closed bottom to obtain flux measurements from the leaves alone. Chamber one was closed again and flux measurements from the soil without leaves were performed.

Table 1. *Elemental composition and pH (in CaCl<sub>2</sub> solution) of leaves and various soil layers*

Layer	C-content (%)	H-content (%)	N-content (%)	pH	N
leaves	41.4	4.99	1.39		4
1 to 3 cm	13.9	1.58	0.69	3.22	4
2 to 5 cm	6.0	0.77	0.33	3.05	2
5 to 10 cm	3.4	0.41	0.15	3.26	2
10 to 20 cm	1.8	0.29	0.07	3.50	2
20 to 30 cm	0.6	0.14	0.04	3.86	2

N = number of samples.

Finally, chamber one was opened again and the humus material was removed and placed in chamber number two for flux measurements from humus alone. Chamber one was closed and flux measurements from the mineral soil were obtained. In many experiments, particularly in the case of CO<sub>2</sub>, we observed a nonlinear change in gas mixing ratios during the experiments, hence, fluxes were calculated from the first few data points of a particular run only (linear portion of the curve). For CH<sub>4</sub> and N<sub>2</sub>O, we used all available data to calculate fluxes.

Air temperature outside and inside the chamber was monitored continuously, minimum temperatures between 0 and 5°C were recorded during December and February and maximum temperatures of ~25°C during the summer months. The precipitation record for the investigation period is given in Fig. 1. Soil moisture was measured between June and December, 1991.

Depth profiles of CH<sub>4</sub> and N<sub>2</sub>O mixing ratios in soil air were measured between 5 June and 30 October, 1991. The samples were collected using 3.15 mm OD stainless steel probes (1.2 mm ID) equipped with a septum and gas-tight syringes. Probes were inserted at depths of 5, 10, 20, 30, and 50 cm into the soil below the humus layer and, after four hours, 10 ml samples were taken simultaneously from all depths. The analyses of CH<sub>4</sub> and N<sub>2</sub>O from these samples were completed within 6 hours after each experiment.

### 3. Results and discussion

Monthly means of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O fluxes from undisturbed soil, soil without leaves, and

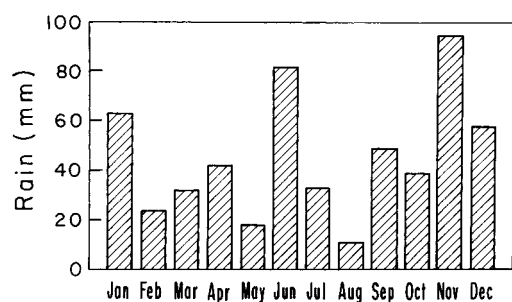


Fig. 1. Precipitation record for Darmstadt, Germany during 1991.

mineral soil are presented in Table 2. No measurements were made during the month of January because the forest soil was frozen or covered with snow. Also, several 24-h experiments did not reveal any significant diurnal cycle and the results were combined with those from the regular dataset. Table 3 lists the annual or quasi-annual means for all categories. Manipulation of the soil always produces certain artifacts (e.g., aeration, decompaction) and, as expected, the combined fluxes obtained from separated layers do not necessarily represent the fluxes from undisturbed soil. This fact can actually be used to gain considerable insight into the processes involved in soil/air exchange.

#### 3.1. Carbon dioxide

Soil organic matter and decomposing litter globally represent a reservoir of about 1580 Pg C (Pg C = 10<sup>15</sup> g carbon; Schimel et al., 1995). The emission of CO<sub>2</sub> to the atmosphere due to soil respiration has been estimated at 68 to 76 Pg C yr<sup>-1</sup>, which represents about 30% of all CO<sub>2</sub> entering the atmosphere (Raich and Schlesinger, 1992; Raich and Potter, 1995).

The annual cycle of CO<sub>2</sub> production recorded at the Darmstadt forest indicates that the variations observed in undisturbed and disturbed soils are similar, with higher releases during the summer and very low emissions during the winter (Fig. 2a). As expected from the observed seasonal variation, a linear correlation ( $R = 0.38$ ;  $n = 47$ ;  $\alpha < 0.01$ ) between CO<sub>2</sub> soil emissions and environmental temperatures (measured inside the chamber) was observed, with an estimated  $Q_{10}$  of ~1.8 ( $Q_{10}$ : increase of the flux due to a temperature increase of 10°C). This value is lower than the global, median value of about 2.4 found when soil temperatures were used in the evaluation (Raich and Schlesinger, 1992), but is in good agreement with the value of 1.5 derived by Raich and Potter (1995), who used air temperatures in their evaluation. In general,  $Q_{10}$  values calculated from air temperatures are lower than those based on soil temperatures at sites with air temperature fluctuating more than soil temperature (Kicklighter et al., 1994; Raich and Potter, 1995).

On the other hand, no significant changes in CO<sub>2</sub> fluxes were observed with changes of soil moisture (Fig. 3), which was measured to be

Table 2. Monthly mean fluxes ( $\pm$  standard deviation and number of measurements in parenthesis) from undisturbed and disturbed soils; positive values indicate emission to the atmosphere, negative values an uptake from the atmosphere

Month	Undisturbed soil	Soil without leaves	Mineral soil
<i>Carbon dioxide fluxes</i> (ng CO <sub>2</sub> m <sup>-2</sup> s <sup>-1</sup> × 10 <sup>-5</sup> )			
February	0.30 (1)	0.17 (1)	0.16 (1)
March	0.80 ± 0.29 (6)	0.32 ± 0.08 (3)	0.35 ± 0.07 (2)
April	0.67 (1)	0.7 (1)	0.77 (1)
May	0.83 ± 0.03 (3)	0.75 ± 0.13 (10) <sup>a</sup>	
June	1.50 ± 0.54 (4)	1.20 ± 0.42 (4)	0.90 ± 0.14 (4)
July	1.29 ± 0.12 (6)	1.23 ± 0.17 (6)	1.15 ± 0.21 (5)
August	1.16 ± 0.23 (10)	1.11 ± 0.28 (5)	1.11 ± 0.24 (5)
September	1.10 ± 0.46 (4)	0.95 ± 0.29 (4)	0.85 ± 0.22 (4)
October	1.11 ± 0.49 (7)	0.97 ± 0.29 (7)	1.00 ± 0.09 (4)
November	0.92 ± 0.09 (3)	0.86 ± 0.12 (3)	0.62 (1)
December	0.49 ± 0.31 (3)	0.26 ± 0.17 (2)	0.28 ± 0.21 (2)
<i>Methane fluxes</i> (ng CH <sub>4</sub> m <sup>-2</sup> s <sup>-1</sup> )			
February	-6.00 (1)	-5.15 (1)	-9.44 (1)
March	-12.2 ± 2.1 (6)	-4.3 ± 19.7 (3)	-12.5 ± 2.9 (3)
April	-19.7 (1)	-22.4 (1)	-22.6 (1)
May	-28.1 ± 9.1 (3)	-20.3 ± 8.8 (8) <sup>a</sup>	
June	-22.6 ± 6.0 (4)	-23.7 ± 6.2 (4)	-24.2 ± 11.4 (4)
July	-26.2 ± 4.3 (6)	-29.6 ± 4.3 (6)	-37.9 ± 4.3 (5)
August	-31.5 ± 3.0 (10)	-38.3 ± 2.2 (2)	-46.6 ± 0.09 (2)
September	-22.8 ± 2.8 (4)	-26.7 ± 5.3 (4)	-29.9 ± 3.9 (4)
October	-21.4 ± 7.6 (5)	-24.6 ± 8.1 (5)	-26.7 ± 8.4 (5)
November	-26.1 (1)	-27.6 (1)	-29.8 (1)
December	-17.7 ± 8.0 (2)	-17.6 ± 7.0 (3)	-12.3 ± 8.9 (2)
<i>Nitrous oxide fluxes</i> (ng N <sub>2</sub> O m <sup>-2</sup> s <sup>-1</sup> )			
February	-0.25 (1)	0.98 (1)	2.97 (1)
March	3.7 ± 0.39 (6)	0.91 ± 0.0 (2)	3.57 ± 0.27 (2)
April	0.12 (1)		0.41 (1)
May	0.71 ± 0.23 (2)	0.34 ± 0.23 (10) <sup>a</sup>	
June	2.17 ± 2.64 (4)	2.12 ± 1.86 (4)	0.63 ± 1.23 (4)
July	0.43 ± 0.62 (6)	0.59 ± 0.22 (6)	0.72 ± 0.59 (5)
August	0.51 ± 0.37 (10)	0.00 ± 0.29 (2)	0.02 (1)
September	1.45 ± 0.87 (4)	1.46 ± 0.80 (4)	0.13 ± 0.51 (4)
October	2.27 ± 1.83 (5)	1.45 ± 0.82 (5)	1.11 ± 0.27 (5)
November	4.09 (1)	2.96 (1)	
December	2.24 ± 0.04 (2)	1.29 ± 0.31 (3)	0.18 ± 0.26 (2)

<sup>a</sup>In most cases no measurements under undisturbed conditions were made.

between 7 and 22% of weight (18–56% WFPS). No reduction in CO<sub>2</sub> emissions was observed at high soil moisture, suggesting that CO<sub>2</sub> production was not inhibited by reduced aerobic respiration.

The combined emission rate of leaves, humus,

and mineral soil is higher than the flux observed from the undisturbed forest floor alone (Table 3), indicating the above mentioned experimental disturbance of the soil. Removal of leaves and humus causes individual layers and the underlying soil to

Table 3. Weighted annual fluxes ( $\text{ng m}^{-2} \text{s}^{-1}$ ) for undisturbed soil, soil without leaves and mineral soil, derived from monthly means of Table 2

Gas	Undisturbed soil	Soil without leaves	Mineral soil	Leaves only <sup>a</sup>	Humus only <sup>b</sup>
$10^{-5} \text{ CO}_2$	$0.88 \pm 0.35$	$0.73 \pm 0.37$	$0.68 \pm 0.34$	$0.24 \pm 0.18$	$0.27 \pm 0.12$
CH <sub>4</sub>	$-20.3 \pm 7.4$	$-20.9 \pm 9.7$	$-23.8 \pm 11.0$	$-0.86 \pm 3.3$	$-0.94 \pm 1.8$
N <sub>2</sub> O	$1.60 \pm 1.40$	$1.50 \pm 1.30$	$0.80 \pm 1.50$	$0.9 \pm 1.6$	$0.59 \pm 0.74$

<sup>a</sup> Average for all individual measurements; no seasonal variation was observed with leaves only.

<sup>b</sup> Humus was measured between June and November, 1991, only.

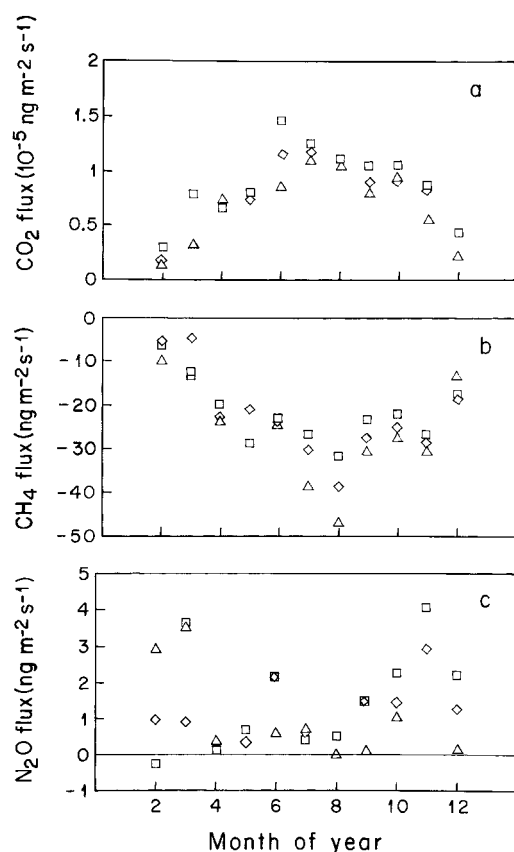


Fig. 2. Seasonal variation of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O fluxes from undisturbed soil (squares), soil without leaves (diamonds), and mineral soil (triangles). Standard deviations and number of measurements are given in Table 2.

be better aerated, resulting in greater respiration rates. After removal of leaves or leaves and humus from the forest floor, a reduction of CO<sub>2</sub> emissions from the remaining soil was observed in most

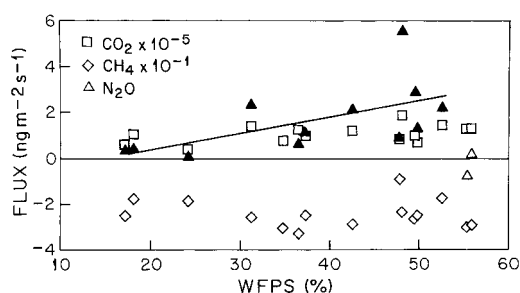


Fig. 3. Variations of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O fluxes with changes in soil WFPS. The line corresponds to a linear regression ( $R = 59$ ;  $n = 12$ ,  $\alpha < 0.01$ ) between N<sub>2</sub>O emissions and WFPS below 55%. WFPS was calculated after Saxton et al. (1986).

cases, with a larger effect at lower temperatures (Fig. 4). Also, larger differences between undisturbed and disturbed conditions were observed during rainy periods (e.g., months of June, September, November), suggesting that enhanced moisture promotes CO<sub>2</sub> production from the litter material. On average, the data presented in Fig. 4 show reductions in CO<sub>2</sub> emissions relative to those from undisturbed soils of ~12% and ~16% after the removal of leaves and leaves and humus, respectively. Considering the enhanced aeration effect, the emissions measured from disturbed soils represent the upper limits for their contribution to the emissions from undisturbed soil. On average, leaves could produce as much as 22% of the flux observed in undisturbed soil, while humus alone could contribute up to 25% (Fig. 4).

Annual emissions of CO<sub>2</sub> from our experiments with undisturbed forest soil were estimated at  $2800 \text{ g CO}_2 \text{ m}^{-2} \text{ yr}^{-1}$ . Annual CO<sub>2</sub> fluxes from temperate, broad-leaved, and mixed forest soils reported by Raich and Schlesinger (1992) show a

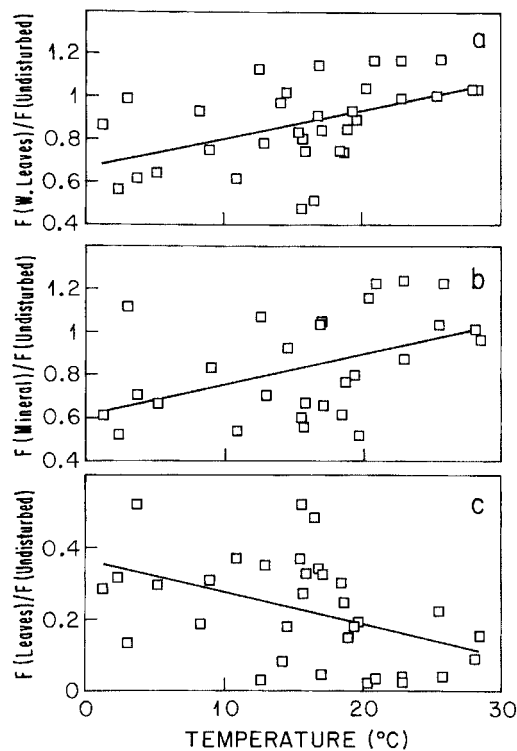


Fig. 4. Temperature dependence of (a) the ratio between the  $\text{CO}_2$  emission from the soil without leaves to the emission from undisturbed soil ( $R = 0.50$ ;  $n = 33$ ;  $\alpha < 0.01$ ), (b) the ratio between mineral soil and undisturbed soil ( $R = 0.45$ ;  $n = 29$ ; 95%  $\alpha < 0.05$ ), and (c) the ratio between leaves only and undisturbed soil ( $R = 0.45$ ,  $n = 33$ ;  $\alpha < 0.01$ ).

large range from  $1100$  to  $5130 \text{ g m}^{-2} \text{ yr}^{-1}$  with a mean rate of  $2370 \pm 190 \text{ g m}^{-2} \text{ yr}^{-1}$ , in rather good agreement with the present estimates.

### 3.2 Methane

The consumption of methane due to oxidation in aerated soils has been reported to be significant in the global budget of methane (Reeburgh et al., 1994), with current sink estimates ranging from  $15$  to  $45 \text{ Tg yr}^{-1}$  ( $\text{Tg} = 10^{12} \text{ g}$ ; Prather et al., 1995; Sanhueza et al., 1995). Measurements of methane fluxes in temperate forests are very sparse and the estimated contribution of these ecosystems to the global methane uptake by soils is very uncertain. A compilation of measurements made to date is given by Castro et al. (1995).

The annual cycle of  $\text{CH}_4$  fluxes observed at the Darmstadt forest site shows a strong seasonality with 4–5 times higher consumption rates during the summer compared to the winter (Fig. 2b). This seasonality is likely due to changes in temperature and soil moisture during the year, as the highest consumption was observed in August, when high temperature and low rainfall coincided in this region. These results differ from the weaker seasonal variations ( $\sim 50\%$ ) found at other forest sites in Germany (Born et al., 1990; Dörr et al., 1993; Koschorreck and Conrad, 1993).

In general,  $\text{CH}_4$  uptake rates were slightly higher after the leaves and the humus layer were removed from the undisturbed forest floor (Fig. 2b), with an annual, mean increase of 17% for the mineral soil compared to the undisturbed soil (Table 3). Results of flux measurements made with leaves and humus alone indicate that neither emit nor consume large amounts of  $\text{CH}_4$  (Table 3), hence, the increased  $\text{CH}_4$  consumption in disturbed soils should be mainly due to a higher permeation rate, which promotes  $\text{CH}_4$  oxidation through an enhanced contact between atmospheric  $\text{CH}_4$  and  $\text{O}_2$  and the biologically active soil layer. This is in accordance with Crill (1991) and Koschorreck and Conrad (1993) who found that the main  $\text{CH}_4$ -oxidizing activity was located in a zone at the top of the mineral layer and that the organic layer (here referred to as humus layer) was not very active.

The annually averaged  $\text{CH}_4$  consumption rate from undisturbed soil was  $-20.3 \pm 7.4 \text{ ng m}^{-2} \text{ s}^{-1}$ , which is consistent with a range of  $-4$  to  $-40 \text{ ng m}^{-2} \text{ s}^{-1}$  reported for other temperate forests in Germany (Dörr et al., 1993; Koschorreck and Conrad, 1993) and in the United States (Keller et al., 1983; Crill, 1991; Bowden et al., 1993; Peterjohn et al., 1994; Castro et al., 1993 and 1995; Yavitt et al., 1993).

Relations between methane fluxes and air temperatures are shown in Fig. 5. In all instances, a negative correlation was found, indicating higher uptake rates with increasing temperature. A  $Q_{10}$  of about 1.3 was determined for the undisturbed soil, and slightly higher values for those experiments in which the litter was removed. Castro et al. (1995) found an important dependence of  $\text{CH}_4$  fluxes on soil temperatures between  $-5$  and  $10^\circ\text{C}$ , whereas methane consumption was independent of soil temperatures between  $10$  and  $20^\circ\text{C}$ .

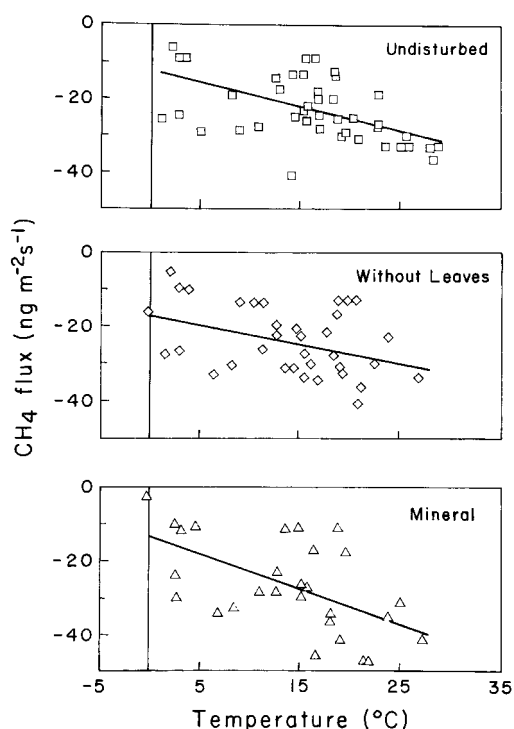


Fig. 5. Relationship between CH<sub>4</sub> fluxes and air temperature. Undisturbed soil:  $R = 0.50$ ,  $n = 43$ ,  $\alpha < 0.01$ ; soil without leaves:  $R = 0.39$ ,  $n = 38$ ,  $\alpha < 0.05$ ; mineral soil:  $R = 0.56$ ,  $n = 28$ ,  $\alpha < 0.01$ .

Born et al. (1990) found that the permeability of methane in the soil is the most influential parameter on CH<sub>4</sub> fluxes. Supporting this idea, Koschorreck and Conrad (1993) and Castro et al. (1995) found that at high WFPS (i.e., >60%) soil moisture exerts strong control on CH<sub>4</sub> uptake. We did not find any significant variation in methane uptake with changes in soil moisture (Fig. 3), because soil moisture was always below 60% WFPS.

An exponential decrease in CH<sub>4</sub> mixing ratios with depth was observed in our experiments (Fig. 6), which is expected to occur in soils where CH<sub>4</sub> consuming microorganisms are horizontally homogeneously distributed (Born et al., 1990). The largest decrease in mixing ratios (>50%) was observed within the first 10 cm, whereas mixing ratios leveled off below 20 cm; Koschorreck and Conrad (1993) also found that no CH<sub>4</sub>-oxidation occurs in soils below about 20 cm depth.

A positive correlation ( $R = 0.49$ ;  $n = 43$ ;

$\alpha < 0.01$ ) was observed between CH<sub>4</sub> uptake and CO<sub>2</sub> production. This relationship should be mainly driven by temperature, but also indicates that the flux-controlling processes are affected in a similar way by other environmental properties such as soil moisture, amount of leaf litter, etc. This is an important point to take into consideration in the development of regional models, and in the interpretation and extrapolation of results obtained in the field.

### 3.3. Nitrous Oxide

Nitrous oxide is emitted from many different sources, most of which are associated with large uncertainties, hence, its atmospheric budget is still not well constrained (Prather et al., 1995; Sanhueza et al., 1995; Bouwman et al., 1995). The principal source of N<sub>2</sub>O is microbial nitrification and denitrification in soils and oceans. Estimates of the contribution of temperate forests to the global N<sub>2</sub>O budget range from 0.05 to 2 Tg N per year (Prather et al., 1995); a most probable value of 0.5 Tg N yr<sup>-1</sup> was recently used by Bouwman et al. (1995).

Monthly means of measured N<sub>2</sub>O fluxes in undisturbed and disturbed soils are summarized in Table 2, Fig. 2c. Fluxes from all experiments show a large scatter with a possible, weak seasonal trend towards higher fluxes in the winter months. This increase in winter times may be due to a combination of several factors: (i) enhanced anaerobic N<sub>2</sub>O production due to higher soil moisture during the fall/winter months, (ii) enhanced mineralization of leaf material during the fall, which will enhance the emission of N<sub>2</sub>O (Schmidt et al., 1988), and (iii) increased anaerobic production of N<sub>2</sub>O because of a thicker leaf layer with reduced O<sub>2</sub> exchange. The effect that appears to be strongest is the increased production due to enhanced soil moisture, which is particularly obvious during the rainy months of June and November (Figs. 1, 2c).

An increase of emissions was observed between 18% and 51% WFPS, with a decrease above ~55% WFPS (Fig. 3). An increase in N<sub>2</sub>O production by denitrification is expected with increasing anaerobic conditions caused by a higher WFPS (Williams et al., 1992). The decrease in N<sub>2</sub>O emissions above 55% WFPS is probably based on an inhibition of the transport of N<sub>2</sub>O

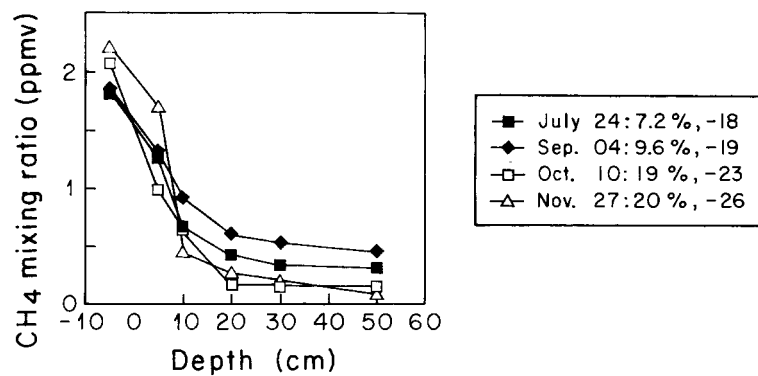


Fig. 6. Depth profiles of  $\text{CH}_4$  mixing ratios in soil air. Dates, gravimetric soil moisture in the 0 to 7 cm layer, and measured fluxes ( $\text{ng m}^{-2} \text{s}^{-1}$ ) are given in the legend. Data at negative depth represent ambient air levels.

from the soil to the atmosphere. Hence,  $\text{N}_2\text{O}$  remains in the soil for further breakdown to  $\text{N}_2$ .

For comparison, we compiled  $\text{N}_2\text{O}$  fluxes from other publications in Table 4. The mean, annual emission of  $\text{N}_2\text{O}$  at the Darmstadt forest site was  $1.6 \text{ ng m}^{-2} \text{ s}^{-1}$ . Our results are in the lower part of the range reported by Schmidt et al. (1988) for measurements made at six different forest sites in the vicinity of Mainz, Germany, but are much lower than the rates found in the Solling area, Germany (Brumme and Beese, 1992). They are also lower than the fluxes measured by Goodroad and Keeney (1984) in deciduous and pine forests in Wisconsin, but similar to those from a mixed hardwood forest in Massachusetts (Keller et al., 1983) and significantly higher than the emission

from pine and hardwood forests in Massachusetts (Bowden et al., 1990).

In about 70% of our experiments,  $\text{N}_2\text{O}$  emissions decreased when leaves or leaves and humus were removed, but this was not consistently so. On the one hand, most of this observed reduction in emission can probably be explained by the fact that both leaf and humus layers alone produce significant amounts of  $\text{N}_2\text{O}$ . Removing the leaves from the undisturbed soil does not show a large effect on the total flux (1.6 to 1.5). Removal of the humus layer has the largest impact and can mostly be accounted for by the emissions of the humus layer alone ( $0.8 + 0.6 = 1.4 \approx 1.5$ ). However, the observed mean emission of the leaves layer alone is much more than the reduction it causes when

Table 4. Summary of published  $\text{N}_2\text{O}$  emission rates from temperate forests

Site	Type of forest	Flux ( $\text{ng N}_2\text{O m}^{-2} \text{ s}^{-1}$ )	Ref.
Massachusetts, USA	mixed hardwood	1.5	Keller et al. (1983)
Wisconsin, USA	deciduous	4.1	Goodroad and Keeney (1984)
	pine	24.2	
Mainz, Germany	deciduous	1.7–7.0	Schmidt et al. (1988)
Massachusetts, USA	hardwood	0.2	Bowden et al. (1990)
	pine	0.1	
Solling, Germany	beech	36–100	Brumme and Beese (1992)
Villigen and Schluchsee, Germany	spruce	1.3–13.1	Papen et al. (1994)
Darmstadt, Germany	beech and oak	1.6	present study



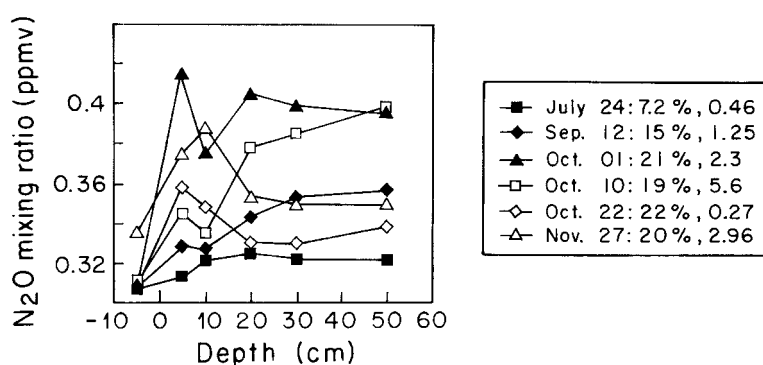


Fig. 7. Depth profiles of N<sub>2</sub>O mixing ratios in soil air. Dates, gravimetric soil moisture in the 0 to 7 cm layer, and measured fluxes (ng m<sup>-2</sup> s<sup>-1</sup>) are given in the legend. Data at negative depth indicate ambient air levels.

removed from the undisturbed soil. This is somewhat peculiar and might be an artifact caused by humus particles that were removed along with the leaves. On the other hand, enhanced O<sub>2</sub> levels at the mineral soil, caused by the removal of leaf and humus layers, should also decrease anaerobic N<sub>2</sub>O production and its emission. But the missing diffusion barrier should increase transport of mineral-soil N<sub>2</sub>O to the atmosphere. Hence, it is difficult to easily quantify the effects of the leaves and humus layers on N<sub>2</sub>O emissions. However, it is apparent that these layers seem to significantly affect N<sub>2</sub>O fluxes, with the mineral soil and humus layer being most important in the overall emission.

Depth profiles and the top-layer soil moisture are given in Fig. 7. Generally, N<sub>2</sub>O mixing ratios increase with depth and appear to be somewhat regulated by soil moisture, with higher N<sub>2</sub>O mixing ratios at higher water contents, which would support the arguments outlined above. Locally saturated soils or horizontal transport might also explain some of the results that deviate from general patterns.

#### 4. Conclusions

The annually weighted, average flux rates for CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O from the undisturbed forest soil are 8.8 × 10<sup>4</sup> ng m<sup>-2</sup> s<sup>-1</sup>, -20.3 ng m<sup>-2</sup> s<sup>-1</sup>, and 1.6 ng m<sup>-2</sup> s<sup>-1</sup>, respectively, which generally agree with reports from similar sites. Temperature exerts a controlling influence on CO<sub>2</sub> emission and CH<sub>4</sub> uptake, whereas N<sub>2</sub>O fluxes seem to be mostly affected by the WFPS. In general, the results indicate that fresh and partly decomposed leaves and humus affect the soil-atmosphere exchange of the investigated gases. It was estimated that ~20% of the emitted CO<sub>2</sub> and up to 50% of the emitted N<sub>2</sub>O is produced by leaves and humus. The consumption of methane occurs mainly in the mineral soil and the humus layer acts mainly as a barrier against diffusion. Changes in thickness, accumulation rates, removal, or degradation of leaves and humus layer are likely to affect the fluxes of CO<sub>2</sub>, CH<sub>4</sub>, and N<sub>2</sub>O between soil and atmosphere.

#### REFERENCES

- Born, M., Dörr, H. and Levin, I. 1990. Methane consumption in aerated soils of the temperate zone. *Tellus* **42B**, 2–8.
- Bouwman, A. F., Van der Hoek, K. W. and Olivier, J. G. J. 1995. Uncertainties in the global source distribution of nitrous oxide. *J. Geophys. Res.* **100**, 2785–2800.
- Bowden, R. D., Steudler, P. A., Melillo, J. M. and Aber, J. D. 1990. Annual nitrous oxide fluxes from temperate forest soils in the northeastern United States. *J. Geophys. Res.* **95**, 13997–14005.
- Bowden, R. D., Castro, M. S., Melillo, J. M., Steudler, P. A. and Aber, J. D. 1993. Fluxes of greenhouse gases between soils and the atmosphere in a temperate forest following a simulated hurricane blowdown. *Biogeochem.* **21**, 61–71.
- Brumme, R. and Beese, F. 1992. Effects of liming and nitrogen fertilization on emissions of CO<sub>2</sub> and N<sub>2</sub>O from a temperate forest. *J. Geophys. Res.* **97**, 12,851–12,858.
- Castro, M. S., Steudler, P. A., Melillo, J. M., Aber, J. D.

- and Millham, S. 1993. Exchange of  $N_2O$  and  $CH_4$  between the atmosphere and soil in spruce-fir forests in the northeastern United States. *Biogeochem.* **18**, 119–135.
- Castro, M. K., Steudler, P. A., Melillo, J. M., Aber, J. D. and Bowden, R. D. 1995. Factors controlling atmospheric methane consumption by temperate forest soils. *Global Biogeochem. Cycles* **9**, 1–10.
- Crill, P. M. 1991. Seasonal patterns of methane uptake and carbon dioxide release by a temperate woodland soil. *Global Biogeochem. Cycles* **5**, 319–334.
- Crutzen, P. J. 1970. The influence of nitrogen oxides on the atmospheric ozone content. *Quart. J. Roy. Meteorol. Soc.* **96**, 320–325.
- Dörr, H., Katruff, L. and Levin, I. 1993. Soil texture parameterization of the methane uptake in aerated soils. *Chemosphere* **26**, 697–713.
- Goodroad, L. L. and Keeney, D. R. 1984. Nitrous oxide emission from forest, marsh, and prairie ecosystems. *J. Environ. Qual.* **13**, 448–452.
- Hansen, J. E. and Lacis, A. A. 1990. Sun and dust versus greenhouse gases: an assessment of their relative roles in global climate changes. *Nature* **346**, 713–719.
- Houghton, J. T., Meira Filho, L. G., Callander, B. A., Harris, N., Kathenberg, A. and Maskell, K. (eds). 1996. *Climate change 1995: the science of climate change*. Cambridge University Press, New York.
- Keller, M., Goreau, T. J., Wofsy, S. C., Kaplan, W. A. and McElroy, M. B. 1983. Production of nitrous oxide and consumption of methane by forest soils. *Geophys. Res. Lett.* **10**, 1156–1159.
- Kicklighter, D. W., Melillo, J. M., Peterjohn, W. T., Rastetter, E. B., McGuire, A. D. and Steudler, P. A. 1994. Aspects of spatial and temporal aggregation in estimating regional carbon dioxide fluxes from temperate forest soils. *J. Geophys. Res.* **99**, 1303–1315.
- Koschorreck, M. and Conrad, R. 1993. Oxidation of atmospheric methane in soil: Measurements in the field, in soil cores and in soil samples. *Global Biogeochem. Cycles* **7**, 109–121.
- Papen, H., Hermann, H., Butterbach-Bahl, K. and Rennenberg, H. 1994. *Emissions of  $N_2O$ ,  $NO$ , and  $NO_2$  from soils of spruce stands in the black forest*. Report from the Fraunhofer Institut für Atmosphärische Umweltforschung, Garmisch Partenkirchen, Germany, October, 1994.
- Peterjohn, W. T., Melillo, J. M., Steudler, P. A., Newkirk, K. M., Bowles, F. P. and Aber, J. D. 1994. Responses of trace gas fluxes and N availability to experimentally elevated soil temperatures. *Ecol. Appl.* **4**, 617–625.
- Prather, M., Derwent, R., Ehhalt, D., Fraser, P., Sanhueza, E. and Zhou, X. 1995. Other trace gases and atmospheric chemistry. In: *Climate change 1994. Radiative forcing of climate change*. J. T. Houghton, L. G. Meira Filho, J. Bruce, H. Lee, B. A. Callander, E. Haites, N. Harris and K. Maswell, eds. Cambridge Univ. Press, Cambridge, 73–126.
- Raich, J. W. and Potter, C. S. 1995. Global patterns of carbon dioxide emissions from soils. *Global Biogeochem. Cycles* **9**, 23–36.
- Raich, J. W. and Schlesinger, W. H. 1992. The global carbon dioxide flux in soil respiration and its relationship to vegetation and climate. *Tellus* **44B**, 81–99.
- Reeburgh, W. S., Roulet, N. T. and Svensson, B. H. 1994. Terrestrial biosphere-atmosphere exchange in high latitudes. In: *Global atmospheric-biospheric chemistry*. R. G. Prinn, ed. Plenum Press, New York, 165–178.
- Sanhueza, E., Fraser, P. and Zander, R. 1995. Source gases: trends and budgets. In: *Scientific assessment of ozone depletion: 1994*. WMO/UNEP. WMO Report No.37, 2.1–2.38.
- Sanhueza, E., Dong, Y., Scharffe, D., Lobert, J. M. and Crutzen, P. J. 1998. Carbon monoxide uptake by temperate forest soils: The effects of leaves and humus layers. *Tellus* **50B**, 51–58.
- Saxton, K. E., Rawls, W. J., Romenberg, J. S. and Papendick, R. I. 1986. Estimating generalized soil-water characteristics from texture. *Soil Sci. Soc. Am. J.* **50**, 1031–1036.
- Schimmel, D., Enting, I., Heimann, M., Wigley, T., Raymond, D., Alves, D. and Siegenthaler, U. 1995. The carbon cycle. In: *Climate change 1994. Radiative forcing of climate change*. J. T. Houghton, L. G. Meira Filho, J. Bruce, H. Lee, B. A. Callander, E. Haites, N. Harris and K. Maswell, eds. Cambridge Univ. Press, Cambridge, 35–71.
- Schmidt, J., Seiler, W. and Conrad, R. 1988. Emission of nitrous oxide from temperate forest soils into the atmosphere. *J. Atmos. Chem.* **6**, 95–115.
- Williams, E. J., Hutchinson, G. L. and Fehsenfeld, F. C. 1992.  $NO_x$  and  $N_2O$  emissions from soils. *Global Biogeochem. Cycles* **6**, 351–388.
- Yavitt, J. B., Simmons, J. A. and Fahey, T. J. 1993. Methane fluxes in a northern hardwood forest ecosystem in relation to acid precipitation. *Chemosphere* **26**, 721–730.