

METHANE OXIDATION IN SOIL PROFILES OF DUTCH AND FINNISH CONIFEROUS FORESTS WITH DIFFERENT SOIL TEXTURE AND ATMOSPHERIC NITROGEN DEPOSITION

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Summary—We studied methane oxidation capacity in soil profiles of Dutch and Finnish coniferous forests. The Finnish sites ($n = 9$) had nitrogen depositions from 3 to 36 kg N ha⁻¹ a⁻¹. The deposition of N on the Dutch sites ($n = 13$) was higher ranging from 50 to 92 kg N ha⁻¹ a⁻¹. The Dutch sites had also limed counterparts. Methane oxidation rates were determined by incubating soil samples in the laboratory at +15°C with 10 μl CH₄ l⁻¹ (10 ppmv CH₄). In general, CH₄ oxidation rates were highest in the uppermost mineral layers. The average CH₄ oxidation rate in the Finnish mineral soils was three times higher than that in the Dutch soils. The litter layers did not oxidize CH₄. In the Netherlands all organic horizons had a negligible capacity to oxidize CH₄. However, some Finnish organic horizons showed high CH₄ oxidation capacity. In the Netherlands, in contrast to Finland, there were some soil profiles lacking CH₄ oxidation. Higher contents of nitrate and ammonium, as well as greater production of nitrous oxide (N₂O) and lower production of carbon dioxide in the Dutch than in the Finnish forest soils reflected the high N deposition rate in the Netherlands. Not only the N deposition, but also the highly sorted soil texture (fine sand) with low amounts of both coarse and fine particles is an important reason for the low CH₄ oxidation in the Dutch soils. The proportions of fine and coarse particles, both well represented in moraine soils typical in northern Europe, correlated positively with the CH₄ oxidation. Fine particles provide a good surface for microbial growth. Coarse particles, on the other hand, enhance diffusion of CH₄ and oxygen into the soil. Methane oxidation in the Dutch mineral soils was slightly enhanced by liming. © 1997 Elsevier Science Ltd

INTRODUCTION

Methane is an important greenhouse gas currently contributing approximately 20% to global warming (Bouwman, 1990). Its concentration in the troposphere has increased for several decades by up to 1% a⁻¹ (Blake and Rowland, 1988), although there has been a decline in the rate of increase during the last decade (Dlugokency *et al.*, 1994). Methane is produced in anaerobic microbial processes in wetlands, rice paddies, ruminants and landfills. Aerated soils, such as forest soils, contain bacteria able to oxidize atmospheric CH₄. Aerated soils, thus, counteract the increase in atmospheric CH₄ concentration and global warming by annually consuming 5–60 Tg of atmospheric CH₄ (Born *et al.*, 1990;

Dörr *et al.*, 1993), which is 1–10% of the total global CH₄ emission.

Methanotrophs in forest soil profiles generally have their highest activity in the uppermost mineral layers (Crill, 1991; Adamsen and King, 1993; Koschorreck and Conrad, 1993). Gas diffusion into the soil is an important factor (Born *et al.*, 1990; Striegl, 1993) for the location of CH₄ oxidation activity in soil profiles. Therefore, CH₄ consumption can decrease with increasing soil moisture as a result of reduced CH₄ diffusion into the soil (Adamsen and King, 1993; Koschorreck and Conrad, 1993; Castro *et al.*, 1994).

Ammonium inhibits the activity of CH₄-oxidizing bacteria (Nesbit and Breitenbeck, 1992; Adamsen and King, 1993; Schnell and King, 1994) and CH₄ uptake into soil (Stuedler *et al.*, 1989; Schnell and King, 1994; Sitaula *et al.*, 1995). Inhibition in CH₄ uptake is detected soon after N fertilization (Stuedler *et al.*, 1989) and inhibition can persist for a long time (Nesbit and Breitenbeck, 1992; Schnell

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and King, 1994). It has been suggested that in addition to ammonium, nitrite generated from ammonium oxidation participates in the inhibition of CH₄ oxidation (Schnell and King, 1994; Dunfield and Knowles, 1995).

Little is known about how the combination of soil characteristics and environmental conditions affect CH₄ oxidation in forest soils in different geographical regions. We studied the CH₄ oxidation capacity of temperate coniferous soils in northwest Europe (the Netherlands) and boreal coniferous forest soils in northern Europe (Finland). These forest soils have very different textures and amounts of N deposition.

MATERIALS AND METHODS

Forest sites and soil sampling

Soil samples were taken in late autumn 1992 from six Finnish and seven Dutch coniferous forests (Table 1). The forests in western Finland had N deposition rates varying from 5 to 36 kg N ha⁻¹ a⁻¹. The sites in western Finland with high N deposition were close to fur farms. There NH₄⁺ was the main component in the N deposition (Table 1). A control site in eastern Finland (Ilomantsi) had a nitrogen deposition rate of 3 kg N ha⁻¹ a⁻¹. In the Netherlands the N deposition rates were high, from 50 to 92 kg N ha⁻¹ a⁻¹. In six of the seven Dutch forests there also were limed counterparts (Table 1).

Soil profiles were divided into litter layers (L), organic horizons (O) and uppermost mineral soils. The soils were homogenized and sieved before the experiments (mesh size for organic horizons 1 cm, and for upper mineral soils 4 mm). The Dutch soils

were transported to Finland, where they were stored at 4°C the same as the Finnish samples.

Soil physical and chemical analyses

Soil particle size analyses were done by standard sieving and sedimentation methods in the Finnish Forest Research Institute. Particles were divided into eight size classes expressed as percentages of the total weight. The size classes were ≤0.002 mm, 0.002–0.006 mm, 0.006–0.02 mm, 0.02–0.06 mm, 0.06–0.125, 0.125–0.5 mm, 0.5–2 mm and ≥2 mm.

Soil pH was determined in soil–water suspensions (1:2 v/v). Nitrate and ammonium were extracted with 2 M KCl for 1 h on a rotary shaker (100 rev min⁻¹) at room temperature. Extracts were filtered (S and S 589³ blue ribbon) and stored frozen. Content of nitrate in extracts was measured by AKEA autoanalyzer (Datex Instrumentarium). Ammonium was analyzed by a colorimetric technique according to Fawcett and Scott (1960).

Phosphorus, potassium, calcium, magnesium, iron, manganese, zinc and copper were extracted with 1 M ammonium acetate at pH 4.65 and aluminum with 1 M KCl. Concentrations of these elements were determined by an inductively-coupled plasma emission spectrophotometer ICP-AES (ARL). Contents of total carbon and total nitrogen were measured with an automatic carbon–hydrogen–nitrogen analyzer (CHN-600, Leco).

Soil incubation experiments to measure CH₄ oxidation and production of CO₂ and N₂O

The capacity of the soil samples to oxidize CH₄ was determined in triplicate 600 ml flasks with 60 ml of soil (natural moisture) and 10 μl CH₄ l⁻¹ in gas phase (21% O₂). The flasks were sealed with

Table 1. Study areas

| Site | Tree stand | | Nitrogen deposition rates | | Liming | |
|------------------------------|------------------|-----------------|---|--|---------|-------------------------------|
| | Dominant species | Age (y) | Nitrate (kg N ha ⁻¹ a ⁻¹) | Ammonium (kg N ha ⁻¹ a ⁻¹) | Year | Rate (t ha ⁻¹) |
| The Netherlands ^a | | | | | | |
| 1 | Douglas fir | 30 | 19 | 63 | | |
| 2 | Scots pine | 38 | 19 | 35 | 1985 | 3 |
| 3 | Picea Abies | 51 | 15 | 35 | 1955 | 4 |
| 4 | Picea Abies | 48 | 15 | 35 | 1986 | 3 |
| 5 | Scots pine | 30 | 19 | 63 | 1963–68 | 4 |
| 6 | Corsican pine | ND ^d | 15 | 77 | 1970 | 5.5 |
| 7 | Corsican pine | 43 | 15 | 77 | 1988 | 3 |
| Finland ^b | | | | | | |
| 1 ^c | Scots pine | 35 | 1.3 | 1.4 | | |
| 2 | Scots pine | 40 | 1.0 | 7.0 | | |
| 3a | Scots pine | 41 | 2.0 | 26 | | |
| 3b | Scots pine | 41 | ND | ND | | |
| 4a | Scots pine | 31 | 3.2 | 33 | | |
| 4b | Scots pine | 31 | 2.6 | 22 | | |
| 5a | Scots pine | 23 | 2.0 | 18 | | |
| 5b | Scots pine | 23 | ND | ND | | |
| 6 | Scots pine | 35 | 1.5 | 3.0 | | |

^aDutch forest sites also had unlimed counterparts.

^bSite "a" is closer to a fur farm than the site "b".

^cControl site in eastern Finland.

^dND = Not determined.

rubber septa and incubated at 15°C for 4 d. Before CH₄ addition the flasks were flushed with ambient air. At the beginning of the experiment each flask had an overpressure of 13 kPa. Gas samples (15 ml) for analyses of CH₄, CO₂ and N₂O were taken through the septa by a gas-tight syringe (Plastipak) equipped with a three-way stopcock and a needle (26G). Gas samples were taken immediately after addition of 10 µl CH₄ l⁻¹ and every 24 h during the 4-d incubation.

Gas concentrations were determined with two gas chromatographs running parallel (HP 5890 Series 2). The first GC was equipped with flame ionization (FI) and thermal conductivity (TC) detectors for CH₄ and CO₂, respectively, and the second with an electron capture (EC) detector for N₂O. The detector temperatures were 200, 200 and 350°C and the oven temperatures were 35, 35 and 55°C for FID, TCD and ECD, respectively. Both gas chromatographs had a HayeSep Q 80/100 mesh column (length 2 m) and a Porapak S 80/100 mesh column (length 2 m) in series. The gas samples were injected into the GCs by two 0.5 ml loops fitted to an automatic 10-port valve (Valco). The GC for N₂O also had an automatic six-port valve (Valco) to bypass O₂ from the ECD.

Rate of gas consumption or production was calculated from the decrease or increase in gas concentration in the gas phase. Methane oxidation generally follows first-order reaction kinetics (Crill, 1991; Adamsen and King, 1993). Because many of our soils oxidized methane according to zero-order reaction kinetics, i.e. the oxidation rate was independent of the CH₄ concentration, we determined the CH₄ oxidation rates for all samples from the change in CH₄ concentration during the first 24 h. Production rates of CO₂ and N₂O were determined from the linear increase in gas concentrations during the whole incubation (100 h).

Statistical analyses

The results were analyzed by one-way analysis of variance, correlation analysis and principal component analysis (SPSS/PC+ Inc., Chicago, U.S.A.).

RESULTS

Soil physical and chemical characteristics

As a result of the glacial period moraine soils are typical in northern latitudes. Therefore, Finnish soils have both fine and coarse particles. In contrast, highly sorted Dutch soils virtually lack the finest (<0.06 mm) and the coarsest (>0.5 mm) particles, about 80% of the particles belonging to the size class from 0.125 to 0.5 mm (fine sand) (Fig. 1).

Litter layers of the unlimed soils had higher pH in the Netherlands than in Finland. However, pH in O horizons and upper mineral soils was lower in the Netherlands than in Finland (Table 2). In the

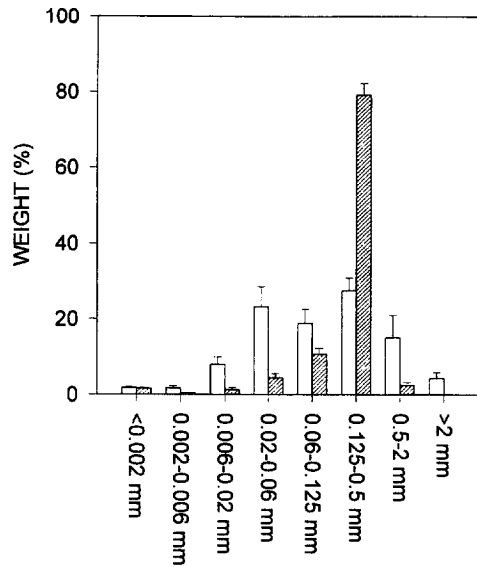


Fig. 1. Particle size fractions of the mineral soils in the Netherlands (hatched columns) and in Finland (unhatched columns). The particle size analysis was done by standard sieving and sedimentation methods in the Finnish Forest Research Institute. The columns show the mean weight percentages of the various fractions, SE of means are shown by bars.

Netherlands liming had increased pH in O horizons and in mineral soils (Table 2).

As a result of greater ammonium ($P = 0.002$) and nitrate ($P = 0.000$) depositions the Dutch soils had higher contents of nitrate and ammonium than the Finnish soils. In the Netherlands, liming has increased nitrate contents and decreased ammonium contents in the O horizons (Table 2). In Finland, the forest soils near the fur farms had more nitrate and ammonium than the soils which were a greater distance from the farms (results not shown).

The Finnish soils had more soluble phosphorus, potassium and iron than the Dutch soils (Table 2). In the Netherlands calcium content was slightly higher in the limed O horizons and mineral soils than in their unlimed counterparts (Table 2).

Methane oxidation and production of CO₂ and N₂O

Mineral soils consumed methane generally according to first-order reaction kinetics. However, some soils with low CH₄ oxidation capacity, oxidized methane according to zero-order reaction kinetics. There were more soil samples with zero-order reaction kinetics in the Netherlands than in Finland.

Generally, little net methane consumption occurred in the L layers and O horizons (Table 3). In fact, methane production was occasionally observed in these soil layers. However, in Finland some O horizons showed high methane oxidation capacity (Table 3). Greatest methane oxidation took place in the uppermost mineral soils if the oxi-

Table 2. Some physical, chemical and microbiological characteristics of the Dutch and Finnish forest soils

| Soil layer | The Netherlands | | <i>P</i> ^a | Finland | |
|---|--------------------------|--------------|-----------------------|---------------|-----------------------|
| | Unlimed | Limed | | | <i>P</i> ^b |
| Litter layer^c | | | | | |
| CO ₂ -prod. (μg cm ⁻³ h ⁻¹) | 4.02 (0.28) ^d | 5.41 (0.58) | * | 5.86 (0.60) | * |
| N ₂ O-prod. (ng cm ⁻³ h ⁻¹) | 0.25 (0.05) | 0.67 (0.32) | | 0.56 (0.20) | |
| pH (H ₂ O) | 5.31 (0.08) | 5.22 (0.13) | | 4.65 (0.07) | *** |
| Water content (%) | 69 (2.69) | 67 (3.46) | | 68 (1.82) | |
| Nitrate (μg cm ⁻³ soil) | 5.37 (2.32) | 4.05 (2.03) | | 4.01 (0.82) | |
| Ammonium (μg cm ⁻³ soil) | 25 (5.59) | 29 (7.97) | | 9.17 (1.87) | ** |
| Phosphorus (μg cm ⁻³ soil) | 8.08 (2.33) | 9.52 (2.90) | | 20 (1.86) | ** |
| Potassium (μg cm ⁻³ soil) | 26 (2.66) | 29 (7.40) | | 47 (2.76) | *** |
| Iron (μg cm ⁻³ soil) | 0.10 (0.05) | 0.90 (0.04) | | 0.17 (0.07) | |
| Calcium (mg cm ⁻³ soil) | 0.15 (0.07) | 0.20 (0.08) | | 0.15 (0.02) | |
| Magnesium (μg cm ⁻³ soil) | 40 (24) | 30 (11) | | 28 (2.86) | |
| Organic horizon^c | | | | | |
| CO ₂ -prod. (μg cm ⁻³ h ⁻¹) | 0.96 (0.05) | 1.11 (0.16) | | 2.81 (0.28) | *** |
| N ₂ O-prod. (ng cm ⁻³ h ⁻¹) | 0.08 (0.01) | 0.12 (0.03) | | 0.02 (0.01) | ** |
| pH (H ₂ O) | 3.59 (0.06) | 4.35 (0.14) | *** | 4.03 (0.05) | *** |
| Water content (%) | 65 (0.93) | 65 (1.58) | | 74 (1.38) | *** |
| Nitrate (μg cm ⁻³ soil) | 6.02 (0.77) | 18.8 (1.04) | *** | 1.00 (0.28) | *** |
| Ammonium (μg cm ⁻³ soil) | 17 (2.76) | 1.97 (0.29) | ** | 8.50 (1.63) | ** |
| Phosphorus (μg cm ⁻³ soil) | 15 (7.13) | 12 (0.09) | | 20 (3.63) | |
| Potassium (μg cm ⁻³ soil) | 43 (19) | 26 (3.72) | | 54 (8.42) | |
| Iron (μg cm ⁻³ soil) | 0.82 (0.26) | 0.45 (0.16) | | 1.02 (0.18) | |
| Calcium (mg cm ⁻³ soil) | 0.15 (0.05) | 0.75 (0.18) | ** | 0.17 (0.02) | |
| Magnesium (μg cm ⁻³ soil) | 26 (5.90) | 92 (61) | | 29 (3.52) | |
| Mineral soil (0–5 cm)^c | | | | | |
| CO ₂ -prod. (μg cm ⁻³ h ⁻¹) | 0.35 (0.02) | 0.37 (0.03) | | 1.24 (0.11) | *** |
| N ₂ O-prod. (ng cm ⁻³ h ⁻¹) | 0.01 (0.003) | 0.003 (0.00) | * | 0.001 (0.001) | ** |
| pH (H ₂ O) | 3.49 (0.04) | 3.78 (0.07) | *** | 4.26 (0.05) | *** |
| Water content (%) | 14 (1.16) | 15 (2.18) | | 25 (1.71) | *** |
| Nitrate (μg cm ⁻³ soil) | 2.34 (0.27) | 2.53 (0.62) | | 0.49 (0.13) | *** |
| Ammonium (μg cm ⁻³ soil) | 4.44 (0.67) | 4.72 (1.53) | | 3.30 (0.97) | |
| Phosphorus (μg cm ⁻³ soil) | 1.51 (0.16) | 1.50 (0.16) | | 5.79 (1.11) | ** |
| Potassium (μg cm ⁻³ soil) | 12 (1.14) | 11 (0.94) | | 18 (1.88) | * |
| Iron (μg cm ⁻³ soil) | 16 (5.84) | 21 (13) | | 225 (81) | * |
| Calcium (mg cm ⁻³ soil) | 0.05 (0.02) | 0.14 (0.03) | * | 0.06 (0.01) | |
| Magnesium (μg cm ⁻³ soil) | 13 (6.29) | 26 (10) | | 10 (1.18) | |

^aStatistical significance (*P*) of the difference between unlimed and limed forest soils in the Netherlands was tested by analysis of variance, * = *P* < 0.05, ** = *P* < 0.01, *** = *P* < 0.001.

^bStatistical significance (*P*) of the difference between the unlimed Dutch and Finnish forest soils was tested by analysis of variance, * = *P* < 0.05, ** = *P* < 0.01, *** = *P* < 0.001.

^cThe average bulk density in L layer, O horizon and mineral soil was 0.04 g⁻¹ dw cm⁻³, 0.11 g⁻¹ dw cm⁻³ and 0.75 g⁻¹ dw cm⁻³, respectively.

^dSE of the mean in parentheses (*n* = 5–9).

ation rates were expressed per soil volume (CO₂ production) were most active in the L layers (Table 3), which compensates for the very different bulk densities of various soil layers. In contrast to CH₄ oxidation, N₂O production and respiration (Table 2). The average CH₄ oxidation rate in the Finnish mineral soils was three times higher than in the

Table 3. Methane oxidation capacity in the Dutch and Finnish forest soils

| Soil layer | The Netherlands | | <i>P</i> ^a | Finland | |
|--|---------------------------------------|--------------|-----------------------|--------------|-----------------------|
| | Unlimed | Limed | | | <i>P</i> ^b |
| Litter layer | | | | | |
| ng CH ₄ g ⁻¹ soil h ⁻¹ | 0.07 ^c (0.12) ^d | 0.22 (0.06) | | 0.33 (0.09) | |
| ng CH ₄ cm ⁻³ soil h ⁻¹ | 0.002 (0.003) | 0.01 (0.02) | | 0.01 (0.00) | |
| Organic horizon | | | | | |
| ng CH ₄ g ⁻¹ soil h ⁻¹ | -0.05 (0.03) | -0.04 (0.00) | | -4.60 (1.88) | |
| ng CH ₄ cm ⁻³ soil h ⁻¹ | -0.01 (0.00) | -0.00 (0.00) | | -0.47 (0.19) | |
| Mineral soil (0–5 cm) | | | | | |
| ng CH ₄ g ⁻¹ soil h ⁻¹ | -0.44 (0.24) | -1.21 (0.75) | | -1.43 (0.23) | ** |
| ng CH ₄ cm ⁻³ soil h ⁻¹ | -0.30 (0.09) | -0.78 (0.44) | | -1.02 (0.16) | *** |

^aStatistical significance (*P*) of the difference between unlimed and limed forest soils in the Netherlands was tested by analysis of variance, * = *P* < 0.05, ** = *P* < 0.01, *** = *P* < 0.001.

^bStatistical significance (*P*) of the difference between the unlimed Dutch and Finnish forest soils was tested by analysis of variance, * = *P* < 0.05, ** = *P* < 0.01, *** = *P* < 0.001.

^cOxidation of methane has a minus sign, production is positive.

^dSE of the mean in parentheses (*n* = 5–9).

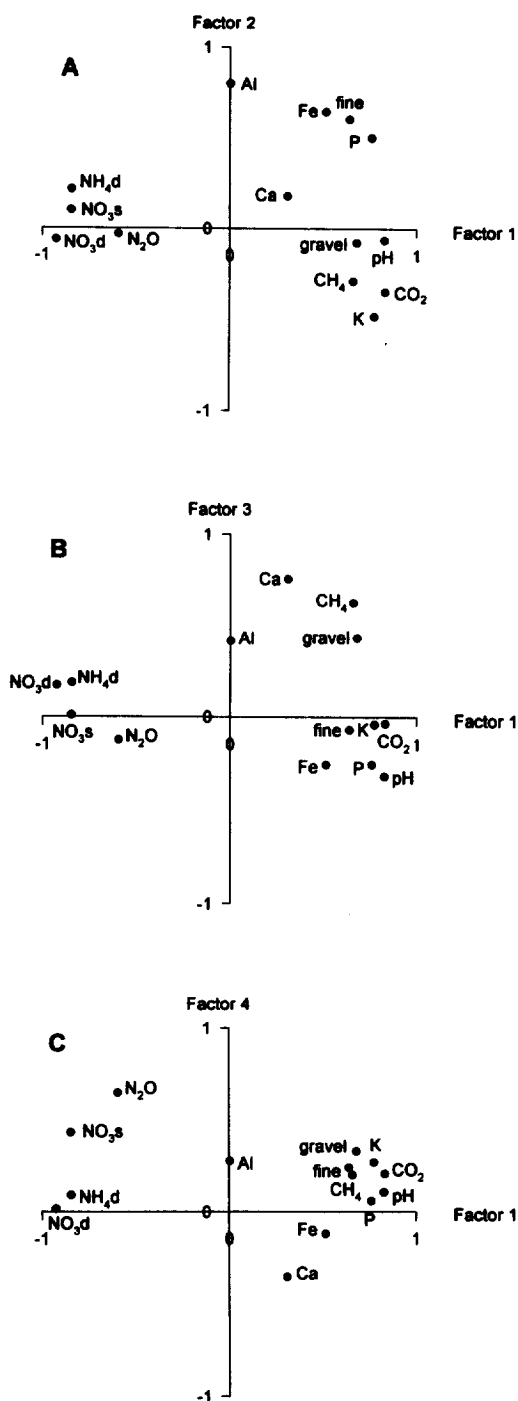


Fig. 2. Interactions between methane oxidation, soil characteristics and atmospheric nitrogen load in unlimed mineral soils are shown by the principal component analysis with the first and the second factors (A), the first and the third factors (B) and the first and the fourth factors (C). In principal component analysis the first two factors explained 65% of the variance (A). If also the third and the fourth factors were included, 77 and 85% of the variance was explained. In the analysis CH_4 oxidation has positive values. "Fine" means particles < 0.06 mm and "gravel" particles > 2 mm. NH_4d and NO_3d are ammonium and nitrate depositions, respectively. NO_3s means concentration of nitrate in soil.

Dutch soils (Table 3). Almost half of the Dutch forest sites had negligible capacity to oxidize methane. In Finland the mineral soils of the sites located 150–300 m from the fur farms consumed slightly more methane (mean $1.20 \text{ ng CH}_4 \text{ cm}^{-3} \text{ h}^{-1}$) than the mineral soils near (ca. 50 m) the fur farms (mean $0.59 \text{ ng CH}_4 \text{ cm}^{-3} \text{ h}^{-1}$) (the difference was not statistically significant, $P = 0.38$). General microbial activity shown as CO_2 production was lower in the Dutch than in the Finnish soils (Table 2). In contrast to the CO_2 production the N_2O production was higher in the O horizons and mineral soils in the Netherlands than in Finland (Table 2). Liming slightly increased the CO_2 production in the L layers and decreased the N_2O production in the mineral soils in the Netherlands (Table 2).

Environmental factors associated with methane oxidation

The results of the principal component analysis (PCA) suggest that CH_4 oxidation in mineral soils had negative interactions with nitrogen deposition and content of nitrate in soil [Fig. 2(a–c)], whereas it had positive associations with content of potassium [Fig. 2(a,c)] and calcium [Fig. 2(b)]. Methane oxidation also had positive interaction with soil pH [Fig. 2(a,c)]. In fact, liming slightly increased pH (Table 2) and CH_4 oxidation rate (Table 3) in the mineral soils of the Dutch forests.

In contrast to the CH_4 oxidation, the production of N_2O was favored by nitrogen deposition and high content of nitrate in soil [Fig. 2(a–c)]. Similar to CH_4 oxidation, the production of CO_2 in the mineral soils had negative interactions with nitrogen deposition and content of nitrate in soil and positive interactions with soil pH and content of potassium [Fig. 2(a–c)] and phosphorus [Fig. 2(b,c)].

Methane oxidation in the mineral soils had a strong positive interaction with the coarse soil particles [Fig. 2(a–c)]. Methane oxidation [Fig. 2(c)] and production of CO_2 [Fig. 2(b,c)] in mineral soils also had positive association to the fine particles as had contents of phosphorous [Fig. 2(a–c)] and potassium [Fig. 2(b,c)].

DISCUSSION

In most of our mineral soils methane oxidation was a first-order process as can be expected at concentrations below saturation (Crill, 1991; Adamsen and King, 1993), but there also were soils oxidizing methane according to zero-order reaction kinetics. Similarly, Bender and Conrad (1993) noted that oxidation of methane in soils was not always a pure first-order reaction. Oxidation of CH_4 followed first-order reaction kinetics until the concentration of CH_4 lowered to from 0.2 to $0.3 \mu\text{l CH}_4 \text{ l}^{-1}$. Below this threshold mixing ratio methane was oxi-

dized slowly, and the process did not follow first-order reaction kinetics.

In general, CH₄ oxidation rates were highest in the uppermost mineral layers. The litter layers did not oxidize CH₄. Many L layers, as well as some O horizons, were slight sources of methane. These results support some earlier findings that the uppermost layers of forest soil can act as a net source of methane (Yavitt *et al.*, 1990; Adamsen and King, 1993). In contrast to the CH₄ oxidation, the production of N₂O and CO₂ was greatest in the L layers. This agrees with the results of Martikainen *et al.* (1993), who measured the highest N₂O production in oxic conditions from the L layer in the soil profile of a forest having high N deposition.

In Finland some O horizons showed high CH₄ oxidation, but in the Netherlands all O horizons had negligible capacity to oxidize CH₄. The low CH₄ oxidation activity in the O horizons of the Dutch coniferous forests may reflect the high N deposition, which has increased content of mineral nitrogen (ammonium and nitrate) in soil. It is well documented that ammonium (Nesbit and Breitenbeck, 1992; Adamsen and King, 1993) and nitrite (produced in ammonium oxidation) may inhibit the activity of CH₄ oxidizing bacteria in soil (Schnell and King, 1994; Dunfield and Knowles, 1995). The L layer receives the highest ammonium load, which could explain the lack of CH₄ oxidation in this layer (Table 3). However, L layers from sites with low N deposition in Finland also showed low CH₄ oxidation activity. Thus, N deposition alone could not explain the low CH₄ oxidation in the L layers.

The average CH₄ oxidation rate in the Finnish mineral soils was three times higher than in the Dutch mineral soils. High N deposition rate is one probable reason for the low CH₄ oxidation rates in the Dutch mineral soils. The effect of high N load could be also seen in other biological processes. The Dutch soils produced more N₂O and less CO₂ than the Finnish soils. Nitrogen load is known to enhance N₂O production (Brumme and Beese, 1992; Sitaula and Bakken, 1993) and reduce general microbial activity in forest soil determined as CO₂ production (Martikainen *et al.*, 1989, 1990; Smolander *et al.*, 1994).

One of the key questions in understanding the mechanism of soil CH₄ consumption is the identification of the soil microorganisms responsible for the oxidation. Stuedler *et al.* (1996) have suggested that soils with little nitrification have high methane consumption rates because CH₄ oxidation is dominated by methanotrophs. In contrast, soils with high nitrification rates have low CH₄ consumption because CH₄ oxidation is carried out mainly by ammonium oxidizing nitrifiers. According to this assumption there could be differences in the microbial community responsible for the CH₄ oxi-

dation in the N saturated Dutch forest soils and in the Finnish forests with low N deposition.

In addition to high N deposition the highly sorted soil texture (fine sand) reduces CH₄ oxidation capacity in the Dutch soils. There are two probable reasons why moraine soils in northern Europe favor the activity of methane oxidizing microbes. Firstly, as a result of the coarse fractions the gas diffusivity is high allowing rapid methane and oxygen penetration into the mineral soil. Diffusion associated with soil texture (Dörr *et al.*, 1993) and moisture (Adamsen and King, 1993; Koschorreck and Conrad, 1993; Castro *et al.*, 1994) has been shown to be an important factor controlling methane uptake in soils. Secondly, methanotrophs isolated from soil require surfaces to be active (Schnell and King, 1995), and most of the methane oxidation activity has been found to be associated with the finest soil particles (<0.05 mm) (Bender and Conrad, 1994). Not only the finest particles but also the particles between 0.5 and 2 mm dia, well represented in the Finnish but not in the Dutch soils (Fig. 1), have been shown to have importance for the activity of methane-oxidizing bacteria in soil. These particles have the highest specific methane oxidation activity (activity per weight unit) (Bender and Conrad, 1994). The texture of the moraine soils may well be extremely favorable for methane oxidation by providing good gas diffusivity (gravel) and extensive surface area (fine particles) for the CH₄ oxidizing microbes. As suggested by the PCA, the activity of heterotrophic microbes in the mineral soils was also favored by the fine particles, which shows the general importance of the fine particles to the soil microbes (Harvey *et al.*, 1984; Albrechtsen, 1994).

The PCA shows that content of some nutrients (K, Ca) could have positive interactions with the CH₄ oxidation. Some of the nutrients also had associations with the fine particles. In moraine soils the fine fractions have the highest content of many elements (Lintinen, 1995). Therefore, in addition to the greater surface area of the fine fractions, their nutrients may favor the microbial activities, including methane oxidation.

The higher contents of nitrate and ammonium in the Dutch than in the Finnish forest soils reflect the high N deposition rate in the Netherlands. As a result of the high ammonium load, pH in the L layers was higher in the Netherlands than in Finland. However, pH in O horizons and upper mineral soils was lower in the Netherlands than in Finland. Content of nitrate in mineral soils correlated negatively with soil pH ($n = 16$, $P = 0.003$, $r = -0.69$), which may indicate that protons produced in nitrification contributed to the lower pH in the Dutch soils. The nitrogen-saturated Dutch forests are known to have a relatively high nitrifica-

tion activity (Van Breemen *et al.*, 1987; Tietema *et al.*, 1992).

Nitrification activity in forest soils depends on soil pH (De Boer *et al.*, 1992; Martikainen and De Boer, 1993). Limed O horizons had more nitrate and less ammonium than their unlimed counterparts, indicating that liming had increased the nitrification activity. This result agrees with earlier results that liming stimulates nitrification in forest soils (De Boer *et al.*, 1993; Priha and Smolander, 1995). In addition to nitrification, liming is known to increase microbial activity and biomass in long-term field experiments (Priha and Smolander, 1995; Smolander *et al.*, 1994). We found some increase in the CO₂ production in the limed L layers of the Dutch soil profiles. Liming had a minor effect on the N₂O production in the Dutch soils. Liming has reduced the N₂O emissions from a temperate forest soil (Brumme and Beese, 1992).

The reported effects of acidity on methane oxidation in soil are contradictory. There are observations that soil pH does not correlate with soil methane uptake (Dörr *et al.*, 1993). Artificial acidification of forest soil by sulfuric acid has even stimulated methane uptake (Sitaula *et al.*, 1995). In some studies liming had no effect on methane consumption of forest soil (Yavitt *et al.*, 1993). In a Swedish study liming increased the methane uptake of an acidic forest soil (Åsa Klemmedtsson, pers. commun.). Similarly, the limed counterparts among the studied Dutch forest sites consumed more methane than the unlimed sites. The limed soils had higher calcium content and higher pH. According to our results, calcium had a positive interaction with methane oxidation in unlimed Dutch and Finnish mineral soils.

Our results show that the northern moraine forest soils have a higher capacity to oxidize methane than the temperate, fine sandy forest soils with more sorted particle-size distribution. The low natural CH₄ oxidation capacity of temperate forest soils in northwest Europe may further be reduced by the high N deposition. Our results show that some of the coniferous forests in northwest Europe have negligible CH₄ oxidation activity in their soil profiles, probably as a result of the soil texture and high N deposition.

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