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**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

# Nitric oxide and nitrous oxide emission from Hungarian forest soils; link with atmospheric N-deposition

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Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

EGU

## Abstract

Studies of forest nitrogen (N) budgets generally measure inputs to the atmosphere in wet and dry precipitation and outputs via hydrologic export. Although denitrification has been shown to be important in many wetland ecosystems, emission of nitrogen oxides from forest soils is an important, and often overlooked, component of an ecosystem nitrogen budget. During one year (2002–2003), emissions of nitric oxide (NO) and nitrous oxide (N<sub>2</sub>O) were measured from Sessile oak and Norway spruce forest soils in northeast Hungary. Accumulation in small static chambers followed by gas chromatography-mass spectrometry detection was used for the estimation of N<sub>2</sub>O emission flux. Because there are rapid chemical reactions of NO and ozone, small dynamic chambers were used for in situ NO flux measurements. Average soil emissions of NO were 1.2 and 2.1  $\mu\text{g N m}^{-2} \text{h}^{-1}$ , and for N<sub>2</sub>O were 15 and 20  $\mu\text{g N m}^{-2} \text{h}^{-1}$ , for spruce and oak soils, respectively. The previously determined nitrogen balance between the atmosphere and the forest ecosystem was re-calculated using these soil emission figures. The total (dry + wet) atmospheric N-deposition to the soil was 1.42 and 1.59  $\text{g N m}^{-2} \text{yr}^{-1}$  for spruce and oak, respectively, while the soil emissions are 0.14 and 0.20  $\text{g N m}^{-2} \text{yr}^{-1}$ . Thus, about 10–13% of N compounds deposited to the soil, mostly as NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup> and HNO<sub>3</sub>/NO<sub>3</sub><sup>-</sup>, are transformed in the soil and emitted back to the atmosphere, mostly as a greenhouse gas (N<sub>2</sub>O).

## 1. Introduction

Forest ecosystem N budgets have received considerable attention in recent years, in part due to increasing concern over increased N delivery to receiving waters from N saturated ecosystems (Aber et al., 1998; Dise et al., 1998; Pregitzer et al., 2004). In order to create predictive models of ecosystem and stream response to elevated N deposition, all pathways of N input and loss must be known (Baron et al., 1994). Nitric oxide (NO) and nitrous oxide (N<sub>2</sub>O) are known as intermediate products of nitrification and

**BGD**

2, 703–723, 2005

## NO and NO<sub>2</sub> emission from Hungarian forest soils

L. Horváth et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

EGU

denitrification processes in soils, and forest soil may be an important source for these compounds. Recent research has shown that the emission of NO and N<sub>2</sub>O depends, among other things, on forest type, soil characteristics, and on atmospheric deposition of N to the forest ecosystem (Groffman and Tiedje, 1989; Henrich and Haselwandter, 1997; Vermes and Myrold, 1992). In a cross-site study of 15 European forests sites (Pilegaard and NOFRETETE Team, 2004, 2005<sup>1</sup>), the ratio of nitrogen oxide emission to N-deposition ranged from very little up to 50%. Although denitrification has shown to be insignificant in many forest ecosystems (e.g. Gundersen, 1991; Myrold et al., 1989), it is clear that in some forests, soils may contribute to the atmospheric N-budget at both a local and a global scale. As a large part of these N emissions may occur in the form of N<sub>2</sub>O, such emissions have significant implications for global warming.

Field-based measurements of nitrogen oxide emissions from forested soils are limited. The aim of this work was to extend the forest soil nitrogen oxide emission network to Eastern Europe and provide data for its special climate, N deposition history, and vegetation. This paper summarises the Hungarian contribution to the NOFRETETE objectives based on a one year long field study of nitrogen oxide emission from Sessile oak (*Quercus petraea*) and Norway spruce (*Picea abies* (L.) Karst.) forest soils in Hungary. A second objective was to compare the emission of nitrogen oxides from soils to the atmospheric N deposition fluxes, thus placing emissions into the context of the ecosystem N budget.

## 2. Measurements

### 2.1. Field sites

Both sites used in this study lie in the forested area of Mátra Mountains, NE Hungary, and are located in Ecology Research Stations of Forest Research Institute. Co-

<sup>1</sup>Pilegaard, K. and the NOFRETETE Team: Nitrogen load and forest type determine soil emission of nitrogen oxides (NO and N<sub>2</sub>O), Biogeosciences Discuss., submitted, 2005.

## NO and NO<sub>2</sub> emission from Hungarian forest soils

L. Horváth et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

ordinates of the 8 ha Norway spruce stand (Nyirjes) are:  $\lambda=19^{\circ}57'05''$ ,  $\phi=47^{\circ}53'38''$ ,  $h=560$  m. The age of the stand is 38–40 years, and leaf area index measured in 1993 is 3.3. Co-ordinates of the 20 ha Sessile oak stand (Tetves-rét) are:  $\lambda=19^{\circ}58'01''$ ,  $\phi=47^{\circ}51'56''$ ,  $h=660$  m. The age of this stand is 71–76 years, and leaf area index was estimated as 4.0. The distance between the two stands is 5 km. Mean annual precipitation at both sites is 780 mm, and mean annual temperature is 5.7°C. The main characteristics of the stand and soil can be seen in Table 1.

## 2.2. N<sub>2</sub>O soil emission flux measurements

Nitrous oxide fluxes were determined by small static (closed) chambers following methods of Christensen et al. (1990). A total of 8–8 parallel chambers were used at both sites. Chambers were permanently placed approximately 3–4 m apart along a transect. The area and volume of the chambers are 80 cm<sup>2</sup> and 400 cm<sup>3</sup>, respectively. After closure, gas samples were taken at  $t=0$ , 10, 20 and 30 min with a syringe. A total of 6 ml of sample was injected into a 5.6 ml evacuated tube. Accumulation of N<sub>2</sub>O was measured by a gas chromatography-mass spectrometry system. Emission fluxes were calculated from the accumulation of nitrous oxide gas in the chamber. The calibration standard gas was 313 ppb N<sub>2</sub>O in N<sub>2</sub>, provided by NOAA, Environmental Monitoring and Diagnostics Laboratory. We sampled weekly except when soils were covered by snow. Pilot measurements for the spruce stand were taken June–August, 2002, and regular measurements were taken October 2002–October 2003. We calculated the non-systematic error (coefficient of variation) of sampling and analysis with repeated sampling performed on one day at the two sites. The time difference between repeated samplings was 45 min. Assuming that the emission flux from the soil was stable during this time, our bulk (sampling and analysis) error is approximately 10%. Repeated GC-MS analyses from samples of 3 October 2002 from the oak site resulted in 52.1 and 51.4  $\mu\text{g N m}^{-2} \text{ h}^{-1}$  mean calculated flux. During a test run by sampling every minute, the coefficient of variation determined from the de-trended line was 11%.

There is evidence for a systematic underestimation of soil gas flux using closed

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

---

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**L. Horváth et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

EGU

chambers. For carbon dioxide soil efflux, Rayment (2000) pointed out one bias caused by the fact that the effective volume of a chamber is larger than the physical chamber volume since it also includes the volume of air-filled spaces in the soil. This underestimation increases with a decrease in the height of the chamber. In our case (h=5 cm), using Rayment's estimates for an equivalent depth of air in soil, soil fluxes are underestimated by 24%. Conen and Smith (2000) predicted that N<sub>2</sub>O concentration increases in the soil air immediately after closure of the chamber due to high gas concentrations in the upper part of the soil profile. Christensen et al. (1996) compared 10 different closed, dynamic chambers and micro-meteorological methods and found a good agreement between results. However, they also reported that the magnitude of underestimation is 20% using small chambers.

### 2.3. NO soil emission flux measurements

For the estimation of soil NO flux, small dynamic chambers were used with the same volume described above. A HORIBA gas monitor was used to detect the concentration of NO leaving the chambers at both the spruce and oak sites. The ozone mixing ratio was simultaneously monitored. The calibration standard gas was 8 ppm NO/N<sub>2</sub>. A Thermo Electron gas calibration system was used for multi-point calibration in the mixing ratio range of 0–10 ppb. The coefficient of variation determined for a calibration gas of 8 ppb was 0.9%. For soil samples with a mixing ratio of 1 and 10 ppb, the measured coefficients of variation were 5.3% and 1.5%, respectively. Since the average mixing ratio found during the flux measurements lies between these two figures, our estimated coefficient of variation ranges between 1 and 5%.

Ten minute concentration averages were recorded. Flow rate of the air through the dynamic chamber was 0.327 L min<sup>-1</sup>. The flux was calculated according to Meixner (1997) as follows, taking into account the rapid chemical reaction of nitric oxide and ozone:



The photolysis rate of NO<sub>2</sub> inside the dark chambers was estimated to be zero. Under steady state conditions the mass balance equation for NO can be written as:

$$F_f + F_m + F_{bl} + F_{gp} = 0, \quad (1)$$

where  $F_f$  is the soil flux,  $F_m$  is the difference between fluxes entering and leaving the chamber,  $F_{bl}$  is the term for the wall effect, and  $F_{gp}$  is the loss of NO due to the chemical reaction with ozone. According to our measurements  $F_{bl}$  is negligible because of the relatively short residence time (~80 s) of the gas mixture in the chamber. The soil flux (in units of  $\mu\text{g N m}^{-2} \text{h}^{-1}$ ) can be calculated by the following equation where the two terms on the right represent the input and output fluxes from the chamber and the effect of the chemical reaction, respectively:

$$F_f = \{[\text{NO}]_{\text{out}} - [\text{NO}]_{\text{in}}\} f_1 f_2 * Q/A + \{k[\text{NO}]_{\text{out}}[\text{O}_3]_{\text{out}}\} f_1 * V/A, \quad (2)$$

where  $Q$  is the flow rate ( $0.327 * 10^{-3} \text{ m}^3 \text{ min}^{-1}$ ),  $A$  is the surface of the soil covered by the chamber [ $8 * 10^{-3} \text{ m}^2$ ],  $[\text{NO}]_{\text{out}}$  and  $[\text{NO}]_{\text{in}}$  are the NO mixing ratios leaving and entering the chamber [ppb],  $f_1 = 14/V_t$ ,  $V_t$  is the molar volume at the given temperature  $t$  [°C] ( $V_t = 0.0795 * t + 22.41$ ),  $f_2 = 60 [\text{min h}^{-1}]$  is the conversion term from minutes into hours,  $V$  is the volume of the chamber, including tubing ( $4.44 * 10^{-4} \text{ m}^3$ ),  $k$  is the reaction rate constant calculated as  $k = 1.8 * 10^2 \exp(-1400/T)$  [ppb<sup>-1</sup> h<sup>-1</sup>] (Seinfeld and Pandis, 1998), where  $T$  is the air temperature [K], and  $[\text{O}_3]_{\text{out}}$  is the mixing ratio of ozone (ppb) leaving the chamber. Because the latter term was not recorded, soil flux was calculated first using  $[\text{O}_3]_{\text{out}}$  assuming that:  $[\text{O}_3]_{\text{in}} \approx [\text{O}_3]_{\text{out}}$ . This approximation is valid, if ozone is in excess of NO. In our case, the average mixing ratios of NO and ozone were 1.43 and 47.1 ppb, respectively, showing order of magnitude differences in the mixing ratios. However, in some cases the mixing ratio of NO was high and  $[\text{O}_3]_{\text{in}} > [\text{O}_3]_{\text{out}}$ . For this reason  $[\text{O}_3]_{\text{out}}$  was estimated from the following equation:

$$[\text{O}_3]_{\text{out}} = [\text{O}_3]_{\text{in}} - \{[\text{NO}]_{\text{out}} - [\text{NO}]_{\text{in}}\} * F_{gp} / F_m = \{k[\text{NO}]_{\text{out}}[\text{O}_3]_{\text{out}}\} * V / Q f_2, \quad (3)$$

which takes into account NO loss via reaction with ozone through the ratio of  $F_{gp} / F_m$ . From this equation the exact calculation of  $[\text{O}_3]_{\text{out}}$  is impossible, but through

**NO and NO<sub>2</sub> emission from Hungarian forest soils**

L. Horváth et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

---

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**L. Horváth et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

iteration we are able to estimate the mixing ratio of ozone in the output air. First,  $[O_3]_{out}$  was calculated assuming  $[O_3]_{in} \approx [O_3]_{out}$  for the calculation of  $F_{gp}$ . Then  $F_{gp}$  was recalculated with the inferred  $[O_3]_{out}$ . This process was repeated 5 times until there was no further change in the estimate of  $[O_3]_{out}$ . The use of the correction for  $[O_3]_{out}$  gives a lower value for the mean estimated soil fluxes by 10%. The difference is much lower when the mixing ratio of NO is low. The variation in diurnal flux rate was checked by longer (approximately 24 h) measurements. The signal was relatively stable for 20 h, but a small decrease was observed at night.

At the spruce site, NO flux was measured once or twice a month. Flux rates were determined 4 times at the oak site, and the magnitude of yearly mean flux was estimated using the ratio of fluxes in spruce and oak forest during the same period (in May, June, and September, 2003). The ratio of observed fluxes during these measurements was 1.744. This factor was used to generate monthly values for NO flux in oak. This is a rough estimate, but the magnitude of NO flux (especially for the comparison to N<sub>2</sub>O) can be estimated this way.

Physical and chemical characteristics of the soil were measured in parallel with soil emission flux measurements. Soil temperature and moisture at three depths (5, 10 and 20 cm) has been recorded in the spruce stand since 1999 and since November, 2002 in the oak stand. Soil moisture data are available only at 20 cm at the spruce stand.

#### 2.4. Atmospheric N deposition

For the estimation of total atmospheric N-input to the canopy, dry deposition was inferred as  $DD = C v$ , where  $C$  is the daily mean concentration measured in 2001–2003,  $v$  is the mean dry deposition velocity in the different seasons, determined during earlier field trials. This estimate considered the net dry flux of ammonia (NH<sub>3</sub>) and nitrogen dioxide (NO<sub>2</sub>) gases and nitric acid (HNO<sub>3</sub>) vapour as well as the dry flux of ammonium (NH<sub>4</sub><sup>+</sup>) and nitrate (NO<sub>3</sub><sup>-</sup>) ions in aerosol particles.

Dry deposition velocity of NO<sub>2</sub> gas was measured for three years (1996 and 1998) by the gradient method described in Horváth et al. (1997, 1998). Dry flux was calcu-



---

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**L. Horváth et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

EGU

lated as  $F = -K_H dC/dz$ , where  $K_H$  is the turbulent diffusion coefficient for the sensible heat flux,  $dC/dz$  is the concentration gradient. Concentration gradient was determined by concentration measurements at different heights (28, 23, 18 m) above the 16 m height canopy by a HORIBA APNA-350E NO<sub>x</sub> gas monitor. Concentration averages of 5 minute were logged at each level. Diffusion coefficients of gases were calculated according to the Monin-Obukhov's semi-empirical similarity theory (Weidinger et al., 2000) for stable and unstable stratification for the layer between 28 and 18 m.

Concentration of ammonia, nitric acid and ammonium and nitrate particles were determined on the basis of 24-h sampling according to the EMEP (1996) filter pack method. Ammonia gas and ammonium particles were measured as ammonium in the solution of sampling filters by indophenol-blue spectrophotometry. Nitric acid and nitrate particles were analysed by ion-chromatography. Dry deposition velocity was determined during an intensive campaign in 1998 (Horváth et al., 2001), using the gradient technique. Concentration gradient was determined by two filter pack samplers settled at the height of 28 and 18 m. Dry flux ( $DD$ ) was calculated by the Monin-Obukhov's similarity theory mentioned above. Mean dry deposition velocity was calculated as  $v = DD/C$ , where  $C$  is the concentration at the upper height.

Wet, throughfall and stemflow deposition of nitrate and ammonium were also determined during 2001–2003. Wet deposition ( $WD$ ) of nitrogen was determined as  $WD = C^* p$ , where  $C$  is the concentration of ammonium or nitrate measured in precipitation,  $p$  is the precipitation volume. Daily precipitation samples were taken by a wet-only collector out of the forest canopy. Ammonium and nitrate ions were determined by spectrophotometry and ion-chromatography, respectively.

Rain and snow samples were collected for throughfall by 5 collectors (0.2 m<sup>2</sup> cross-section area) randomly placed under the canopy in each stand, and for stemflow by flexible tubes attached to the trunk of 10 select trees in each stand. Details of sampling and measurement are described in Kovács and Horváth (2004) and Horváth (2004). At the spruce site, ozone concentrations are regularly recorded, and daily 24-h concentrations of N and S compounds are measured. There are various meteorological

and forestry measurements at the two sites (wind, air temperature, humidity, litter fall, forest growth, etc.).

The relation of wet (*WD*), dry (*DD*), throughfall (*TF*), stemflow (*SF*) depositions can be expressed by the equation:

$$5 \quad WD + DD = TF + SF + NCE, \quad (4)$$

where *NCE* denotes net canopy uptake, or the amount of deposited N taken up by the plant through stomata (Lovett and Lindberg 1993, Ferm and Hultberg, 1999).

### 3. Results

#### 3.1. N<sub>2</sub>O soil flux

10 Soil N<sub>2</sub>O flux measured between October 2002 and October 2003 ranged from 0 to 70 μg N m<sup>-2</sup> h<sup>-1</sup>, depending on the season, with averages of 15 and 20 μg N m<sup>-2</sup> h<sup>-1</sup> for spruce and oak, respectively (Fig. 1). Flux was not measured from the beginning of January until the end of March, 2003 due to snow. Yearly averages were calculated from the monthly means of 4–5 measurements per month assuming zero flux for the  
15 winter period with snow cover (Table 2). During this period soil temperature was close to 0°C. There were weak positive correlations between N<sub>2</sub>O flux and soil temperature in both stands for all depths where soil temperature was recorded (Table 3). There were weak negative correlations between N<sub>2</sub>O flux and soil moisture, most significantly in the oak stand.

#### 20 3.2. NO soil flux

NO flux was small compared to fluxes of N<sub>2</sub>O (Fig. 2). Significant soil NO flux was observed only during the summer. Average NO fluxes were 5.0 and 6.4 μg N m<sup>-2</sup> h<sup>-1</sup> for spruce and oak, respectively. Correlation between emission rates of the two gases was calculated for the two sites when emission data were available (n=20; Table 3).

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

### 3.3. N-deposition

The total atmospheric N-deposition to the canopy, calculated as the sum of wet + dry deposition, was estimated as  $1.7 \text{ g N m}^{-2} \text{ yr}^{-1}$  (Horváth, 2004). Deposition to the soil was estimated from throughfall (*TF*) and stemflow (*SF*) measurements, and should equal N deposition to the canopy minus net canopy exchange (*NCE*). *NCE* was calculated to be 16% of total N deposition to the spruce forest canopy, and 6.5% for the oak forest. Though litter fall deposition is much higher than other deposition forms, it represents the internal circulation of N-compounds inside the canopy (Table 4). Emission of NO and N<sub>2</sub>O together returns 10–13% of the atmospheric deposition to the soil (*TF+SF*).

## 4. Discussion

Our results can be compared to those collected at 15 European forest sites as part of the EU-funded project NOFRETETE. The NOFRETETE locations represent different forest types (coniferous vs. deciduous) and span the typical nitrogen deposition rates found across Europe. In that survey (Pilegaard et al., 2005<sup>1</sup>), NO emissions were much greater (ca. 8 times) from coniferous forests than from deciduous forests, and N<sub>2</sub>O emission rates were slightly lower (ca. 2/3) for coniferous forests. While the total (NO + N<sub>2</sub>O) emissions are much higher for European coniferous forests, in the current study there were no significant differences between the spruce and oak sites for flux rates of either NO or N<sub>2</sub>O, although the oak site had slightly higher mean flux rates. At our Hungarian coniferous site the total (NO+N<sub>2</sub>O) mean soil emission is the half of the European average, and is roughly two times higher at our deciduous site compared to other European sites. This is a consequence of the depressed NO flux from our spruce soil and the enhanced N<sub>2</sub>O flux observed from the oak forest soil. Because NO is mainly derived from nitrification and N<sub>2</sub>O is mainly derived from denitrification, well aerated soils should favor the production of NO whereas the presence of anaerobic

**BGD**

2, 703–723, 2005

## NO and NO<sub>2</sub> emission from Hungarian forest soils

L. Horváth et al.

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**EGU**

microsites, due to thick litter layers or compaction, should favor N<sub>2</sub>O production. The lack of significant differences in flux rates from our two sites in this study might suggest that our soils are quite similar with respect to moisture regimes. It is also quite possible that the differences between conifer and deciduous sites observed in the larger survey are due to factors other than tree species, i.e. the primary factors that caused forest stand differences in the first place, such as soil depth or temperature regime. Only further studies with paired conifer/deciduous stands can answer this question.

In our study, all gas flux rates were positively correlated with soil temperature and oak emission rates were negatively correlated with soil moisture. The positive correlation with soil temperature is not surprising, as many indices of soil microbial activity, such as respiration, are positively related to temperature (Chen et al., 2000; Curiel Yuste et al., 2004; Franzluebbers et al., 2002; Sulzman et al., 2005). The negative correlation with soil moisture was surprising, as soil microbial processes are usually positively correlated with soil water, or other factors that control the supply of readily mineralizable substrates (Qi and Xu, 2001; Franzluebbers et al., 2002). Only when soil moisture becomes too high does microbial respiration tend to decrease (Chen et al., 2000). This negative correlation suggests that our oak soils were wet enough that N<sub>2</sub>O production was decreasing, and perhaps N<sub>2</sub> emissions from denitrification were increasing.

The estimate of canopy uptake by foliage in these two stands was 16% and 6.5% of total inorganic N deposition for the spruce and oak sites, respectively. This is quite low compared to results found for a variety of forests in the United States measured as part of the Integrated Forest Study (Lovett and Lindberg, 1993), where net canopy exchange was estimated to be about 40% of inorganic N deposition. These authors found a good correlation between *NCE* and total deposition, indicating that as deposition increases, N consumption in the canopy increases. There are many possible explanations for this discrepancy, including variations in methodology for measuring total N inputs. Alternatively, climatic differences and histories of N deposition to the sites may cause real differences in *NCE* among sites. The Hungarian forests in the current study have received chronic elevated N deposition for a longer period of time than

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**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**L. Horváth et al.

---

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

have the US forests, and thus might be experiencing greater N saturation. Similarly, it is quite likely that chemical species of N in deposition, particularly in dry deposition, may be significantly different, and react differently on leaf surfaces. It is also possible that *NCE* is variable from year to year. An earlier (1996–1998) study of atmospheric fluxes in these same forests (Horváth, 2004) measured higher N in wet deposition but lower N in throughfall and produced a higher estimate of *NCE*, more similar to estimates in the Lovett and Lindberg (1993) survey. Throughfall deposition has been shown to be quite variable both spatially and temporally, and only long-term studies can address the variability of true *NCE*.

Comparing N deposition numbers and gas efflux values, we found that factoring in NO and N<sub>2</sub>O emissions reduces the estimate of net N deposition to the canopy from 1.7 g N m<sup>-2</sup> yr<sup>-1</sup> to 1.5–1.6 g N m<sup>-2</sup> yr<sup>-1</sup>, and thus does not affect this estimate significantly. We did not measure N<sub>2</sub> emission from the soil, which can be significant especially at high moisture levels, and thus our estimates of N flux from the soil is an underestimate. Still, fluxes of NO and N<sub>2</sub>O were found to be a measurable portion of N deposition to soil, which was estimated as 1.42 and 1.59 g N m<sup>-2</sup> yr<sup>-1</sup> for spruce and oak, respectively, and thus these soil gas emissions are around 10–13% of the total atmospheric N deposition to the soil. It is also important to note that while deposition to the soil is mostly as NH<sub>3</sub>/NH<sub>4</sub><sup>+</sup> and HNO<sub>3</sub>/NO<sub>3</sub><sup>-</sup>, these compounds are transformed in the soil and can be emitted as a greenhouse gas (N<sub>2</sub>O) back into the atmosphere. Thus, forest soils play a role in the transformation of deposited N compounds from regional or continental pollution sources into a greenhouse gas that acts at a global scale.

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**BGD**

2, 703–723, 2005

---

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

EGU

## References

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**BGD**

2, 703–723, 2005

---

## **NO and NO<sub>2</sub> emission from Hungarian forest soils**

L. Horváth et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**EGU**

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**BGD**

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---

## **NO and NO<sub>2</sub> emission from Hungarian forest soils**

L. Horváth et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**EGU**

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**BGD**

2, 703–723, 2005

---

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

---

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

EGU



**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

**Table 1.** Stand and soil characteristics of the Norway spruce and Sessile oak stands used in this study (surveyed in 2000).

			spruce	oak
Stand density	trees ha <sup>-1</sup>		1188	776
Stand height	m		20.1	20.5
Mean breast diameter	cm		20.1	22
Rooting depth	m		>1	>1
Soil type			Eutric Leptosol	Mollic Leptosol
Organic layer type			moder/mull	mull
Organic layer C stocks	humus layer	g C kg <sup>-1</sup>	236	144
Organic layer N stocks	humus layer	g N kg <sup>-1</sup>	12	9.3
Organic layer, C:N ratio	humus layer		19.7	15.4
Organic layer, pH	humus layer		4.5	5.7
Mineral soil, clay:silt:sand	0–10 cm	%	15:29:56	17:38:45
Mineral soil, clay:silt:sand	10–20 cm	%	18:31:51	20:42:38
Mineral soil, clay:silt:sand	20–40 cm	%	21:26:53	25:38:37
Mineral soil, C stocks	0–5, 5–10, 10–20, 20–40 cm	g C kg <sup>-1</sup>	55.2, 29.8, 15.1, 9.6	65.2, 36.5, 27.7, 22.7
Mineral soil, N stocks	0–5, 5–10, 10–20, 20–40 cm	g N kg <sup>-1</sup>	4.3, 2.5, 1.2, 0.8	4.8, 3.1, 2.2, 2.0
Mineral soil, C:N ratio	0–5, 5–10, 10–20, 20–40 cm		12.9, 11.9, 12.2, 12.0	13.5, 11.9, 12.4, 11.5
Mineral soil, pH	0–5, 5–10 cm		3.6, 3.9	4.7, 4.3

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

**Table 2.** Monthly and yearly means of NO and N<sub>2</sub>O fluxes.

period	flux ( $\mu\text{g N m}^{-2} \text{h}^{-1}$ )			
	spruce		oak	
	NO	N <sub>2</sub> O	NO	N <sub>2</sub> O
October, 2002	0.94	35.2	n.a.	36.0
November, 2002	1.21	18.1	n.a.	4.7
December, 2002	0.63	2.6	n.a.	2.0
January, 2003	0**	3.7	n.a.	1.1
February, 2003	0**	0**	n.a.	0**
March, 2003	0**	0**	n.a.	30.9
April, 2003	1.04	4.0	n.a.	5.7
May, 2003	1.17	11.3	3.10	16.8
June, 2003	0.99	14.9	2.13	28.6
July, 2003	5.66	22.5	n.a.	35.8
August, 2003	1.23	36.5	n.a.	48.9
September, 2003	1.59	28.2	1.31	32.8
mean	1.20	14.8	2.09*	20.3

\* Estimated from the ratio of spruce/oak fluxes from 3 months where data were available for both stations (mean of May, June, September for spruce: 1.25, for oak: 2.18, ratio=1.744)

\*\* Estimated (snow cover on soil)

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**NO and NO<sub>2</sub> emission from Hungarian forest soils**

L. Horváth et al.

**Table 3.** Correlation (*r*) of N<sub>2</sub>O flux with soil temperature and moisture and with NO flux. All relationships are significant at *P*=0.05. NS=not significant.

	NO emission	t <sub>soil</sub> 5 cm	t <sub>soil</sub> 10 cm	t <sub>soil</sub> 20 cm	w <sub>soil</sub> 5 cm	w <sub>soil</sub> 10 cm	w <sub>soil</sub> 20 cm
spruce	0.40	0.45	0.67	0.67			NS
oak		0.66	0.64	0.72	-0.63	-0.68	-0.67

Title Page

Abstract

Introduction

Conclusions

References

Tables

Figures

◀

▶

◀

▶

Back

Close

Full Screen / Esc

Print Version

Interactive Discussion

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.

**Table 4.** Atmospheric deposition and emission of nitrogen compounds at the spruce and oak stands.

Site	Spruce	Oak	Spruce	Oak	Spruce	Oak
Deposition (2001–2003)	Nitrate (NO <sub>3</sub> <sup>-</sup> )		Ammonium (NH <sub>4</sub> <sup>+</sup> )		Total N	
	(g N m <sup>-2</sup> yr <sup>-1</sup> )					
Atmospheric wet deposition to the canopy (WD)	0.25	0.25	0.29	0.29	0.54	0.54
Atmospheric dry deposition to the canopy (DD)*	0.56	0.56	0.60	0.60	1.16	1.16
Throughfall deposition** (TF)	0.49	0.31	0.85	1.01	1.34	1.32
Stemflow deposition** (SF)	0.02	0.08	0.06	0.19	0.08	0.27
Atmospheric deposition to the soil (TF+SF)	0.51	0.39	0.91	1.20	1.42	1.59
Atmospheric dry deposition to the soil (TF+SF-WD=DD-NCE)	0.26	0.14	0.62	0.91	0.88	1.05
Net canopy exchange (NCE=WD+DD-TF-SF)					0.28	0.11
Total atmospheric deposition to the canopy (WD+DD=TF+SF+NCE)					1.70	1.70
NCE as % of total N deposition					16%	6.5%
Litter fall deposition** (LF)					4.77	9.12
Total deposition to the soil (TF+SF+LF)					6.19	10.7
Emission	Nitrous oxide (N <sub>2</sub> O)		Nitric oxide (NO)		Total N	
soil emission (SE)	0.13	0.18	0.011	0.018	0.14	0.20
	(g N m <sup>-2</sup> yr <sup>-1</sup> )					
soil emission/atmospheric deposition to the soil (%) [100*SE/(TF+SF)]					10%	13%

\* estimated from Horváth (2004) following measurements between 1996–98 in spruce, assuming the same dry deposition rate for oak and spruce sites

\*\* courtesy of Miklós Manninger and Judit Sitkey, Forest Research Institute, Hungary

Title Page

Abstract Introduction

Conclusions References

Tables Figures

◀ ▶

◀ ▶

Back Close

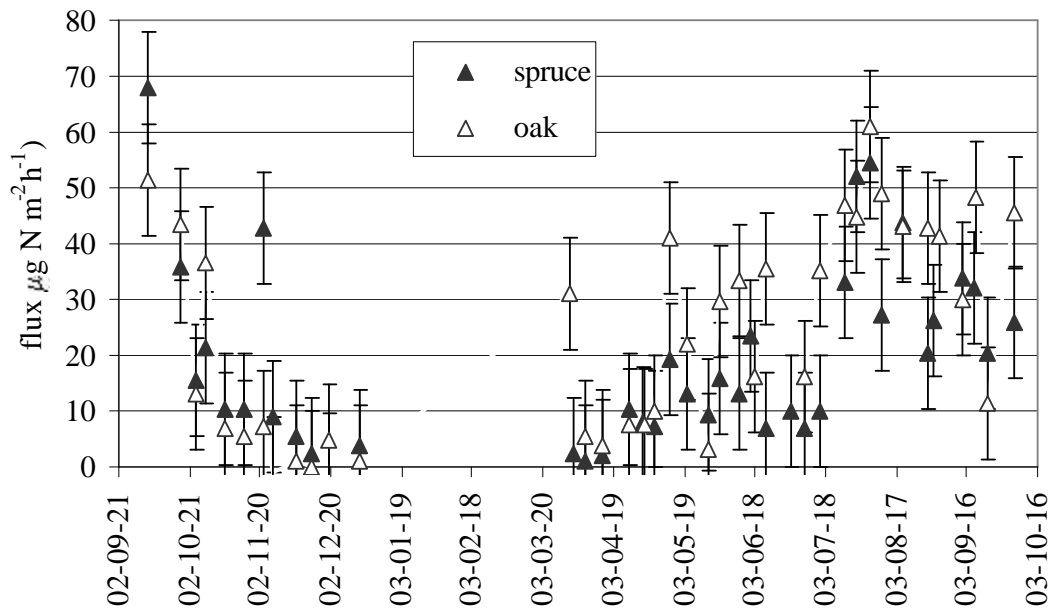
Full Screen / Esc

Print Version

Interactive Discussion

**NO and NO<sub>2</sub> emission from Hungarian forest soils**

L. Horváth et al.

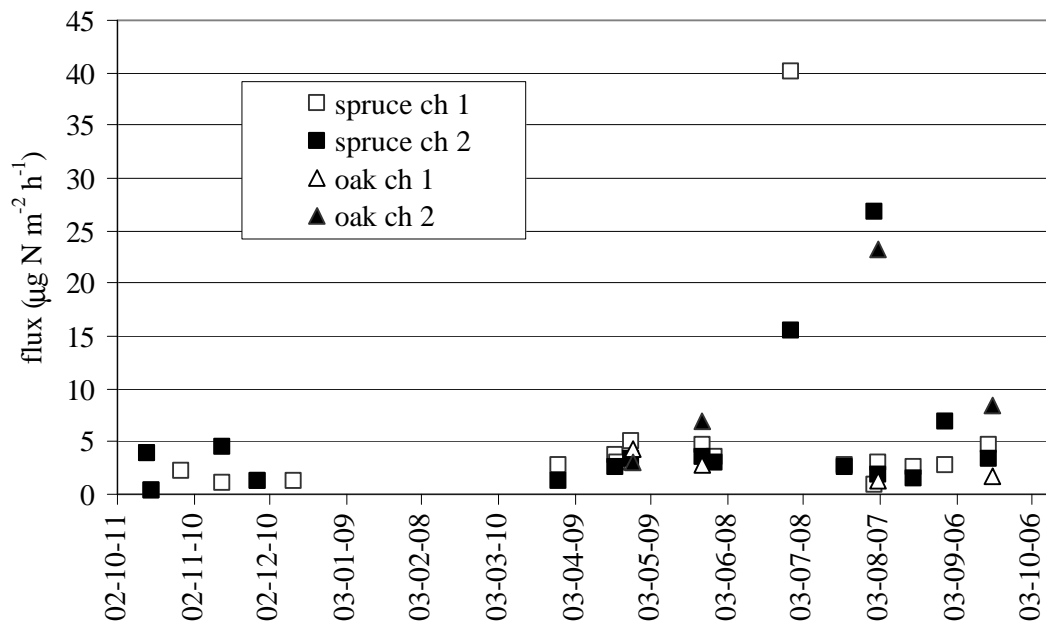


**Fig. 1.** N<sub>2</sub>O soil emission flux (mean of 8-8 chambers) error bars represent the 10% bulk error (CV) of sampling and measurement.

Title Page	
Abstract	Introduction
Conclusions	References
Tables	Figures
◀	▶
◀	▶
Back	Close
Full Screen / Esc	
Print Version	
Interactive Discussion	

**NO and NO<sub>2</sub> emission  
from Hungarian  
forest soils**

L. Horváth et al.



**Fig. 2.** NO soil emission flux measured by two chambers (ch1 and 2).

[Title Page](#)[Abstract](#)[Introduction](#)[Conclusions](#)[References](#)[Tables](#)[Figures](#)[◀](#)[▶](#)[◀](#)[▶](#)[Back](#)[Close](#)[Full Screen / Esc](#)[Print Version](#)[Interactive Discussion](#)

EGU