DEPOSITION OF NITROGEN-CONTAINING COMPOUNDS TO AN EXTENSIVELY MANAGED GRASSLAND IN CENTRAL SWITZERLAND

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Abstract
During four intensive observation periods in 1992 and 1993, dry deposition of nitrogen dioxide (NO\(_2\)) and ammonia (NH\(_3\)), and wet deposition of nitrogen (N) were determined. The measurements were carried out in a small, extensively managed litter meadow surrounded by intensively managed agricultural land. Dry deposition of NH\(_3\) was estimated by the gradient method, whereas eddy correlation was used for NO\(_2\). Rates of dry deposition of total nitrate (= nitric acid (HNO\(_3\)) + nitrate (NO\(_3^-\))), total nitrite (= nitrous acid (HONO) + nitrite (NO\(_2^-\))), and aerosol-bound ammonium (NH\(_4^+\)) were estimated using deposition velocities from the literature and measured concentrations. Both wet N deposition and the vertical NH\(_3\) gradient were measured on a weekly basis during one year.

Dry deposition was between 15 and 25 kg N ha\(^{-1}\) y\(^{-1}\), and net wet deposition was about 9.0 kg N ha\(^{-1}\) y\(^{-1}\). Daily average NO\(_2\) deposition velocity varied from 0.11 to 0.24 cm s\(^{-1}\). Deposition velocity of NH\(_3\), was between 0.13 and 1.4 cm s\(^{-1}\), and a compensation point between 3 and 6 ppbV NH\(_3\) (ppb = 10\(^{-9}\)) was found. Between 60 and 70% of dry deposition originated from NH\(_3\) emitted by farms in the neighbourhood. It is concluded that total N deposition is exceeding the critical load for litter meadows, is highly correlated to local NH\(_3\) emissions, and that NH\(_3\) is of utmost importance with respect to possible strategies to reduce N deposition in rural regions.

Keywords: Dry nitrogen deposition, wet nitrogen deposition.

INTRODUCTION

The increase in the emission of the primary NO and NH\(_3\) due to anthropogenic causes strongly increased total N deposition, which may severely influence the N status of natural ecosystems. Accumulation of inorganic N leads to soil acidification (van Breemen et al., 1982; Stumm et al., 1982; Fabian, 1987) followed by an increase in NO\(_3^-\) leaching into the groundwater and acidification of surface waters as well as a washout of plant nutrients, such as potassium, calcium and magnesium (Ulrich & Pankrath, 1983; Fabian, 1987; Bobbink et al., 1992). Furthermore, N acts as fertilizer and may damage plants and decrease biodiversity (Ellenberg, 1985, 1989; Mohr, 1993a,b, 1994; Bobbink et al., 1988).

Before strategies to reduce N loading of ecosystems can be developed, it is necessary to know the amount and composition of N inputs. Within the Cooperative Program for the Monitoring and Evaluation of the Long Range Transmission of Air Pollutants in Europe (EMEP), the concept of critical loads has been introduced. Nilsson and Grennfelt (1988) defined the critical load as 'a quantitative estimate of an exposure to one or more pollutants below which significant harmful effects on specified sensitive elements of the environment do not occur according to our present knowledge'. The critical load has been set at 5–10 kg N ha\(^{-1}\) y\(^{-1}\) for moors, at about 15 kg N ha\(^{-1}\) y\(^{-1}\) for unmanaged grasslands and at 20–30 kg N ha\(^{-1}\) y\(^{-1}\) for forests (Grennfelt & Thörnénöf, 1992). In recent years, extensive efforts have been made to produce regional maps of the critical load of N as a function of vegetation type and soil characteristics. To calculate the exceedance of critical loads, accurate information concerning the amount and speciation of N deposition is now required.

Numerous results from flux measurements of gaseous N species are reported in the literature (Erisman et al., 1989; Hanson et al., 1989; Stocker et al., 1993; Meixner et al., 1985). However, in regions with complex topography suitable field sites with sufficient fetch hardly exist. In Switzerland, semi-natural ecosystems are typically located in hilly areas not suitable for intensive farming, or they are small protected areas surrounded by intensively managed agricultural land. In spite of these difficulties, it is necessary to obtain reliable estimates of the yearly N deposition under such conditions. In this paper, we report results from measurements of the amount and speciation of total N deposition to an extensively managed litter meadow located in an area surrounded by intensive agriculture.
INSTRUMENTATION

Experimental site

The experimental site was located in the lower Reuss valley near the village of Merenschwand (20 km SW of Zurich, 385 m a.s.l.). The litter meadow (Junco-Molinion typicum; Klötzli, 1969) was a small protected area of about 60 m x 350 m with some scattered Alder Buckthorn (Frangula alnus, 25% of area). The grass and bushes were harvested once in the fall. The long axis of the field was oriented NW–SE (Fig. 1). To the NE, the site was separated from adjacent fields by a hedge of 5-6 m height and 5 m width. To the SW, the litter meadow was separated from neighbouring fields by a drainage canal of about 1 m depth and 2 m width. Prevailing winds were from NW (up-valley winds) and SE (down-valley winds), i.e. parallel to the Reuss valley (Hutter & Bürki, 1991), and in the ideal direction relative to the measuring site. Daily mean speed of wind from the main directions and from NE were generally below 2.5 m s⁻¹, and from all other directions below 1.0 m s⁻¹. Best fetch of over 200 m existed for the wind sector 285°–345° (NW). Due to heterogeneous properties of soil, vegetation, and land management, the measuring site represented a typical non-ideal terrain, according to the description by Hicks and McMillan (1988).

From June 1992 to May 1993, vertical profiles of NH₃ concentration, wet deposition, and meteorological parameters (solar radiation, air temperature and humidity, wind speed and direction, air pressure) were recorded continuously. The profile of NH₃ concentration and wet deposition were determined on a weekly

Table 1. Summary of the instruments used during the four intensive observation periods (IOP), measuring heights in m, and characteristics of the vegetation cover

<table>
<thead>
<tr>
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<td>Eddy correlation</td>
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<td>Gradient method</td>
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<td>0.90 / 2.60</td>
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<td>no</td>
<td>0.86 / 2.50</td>
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<td>WEDD</td>
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<td>no</td>
<td>0.86 / 2.50</td>
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<td>0.90 / 2.60</td>
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<td>Gradient method</td>
<td>Passive samplers</td>
<td>During one year</td>
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<td>&lt;0.05 m</td>
<td>0.25 m</td>
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<tr>
<td></td>
<td>Bushes (25% cover)</td>
<td>0.50 m</td>
<td></td>
<td></td>
<td>&lt;0.05 m</td>
<td>0.25 m</td>
</tr>
</tbody>
</table>

no = not measured
Deposition of $N$-containing compounds

The reaction of luminol and NO$_2$ is known to be interfered with O$_3$ and PAN. According to Drummond et al. (1989b) this interference can be described by the equation:

$$c_k = c_1 - \alpha_0 - \frac{c_1 - c_0}{c_1} \alpha_p + \alpha_p \alpha_{PAN} \tag{1}$$

where $c_k$ = corrected NO$_2$ concentration, $c_1$ = linearly corrected LMA-3 signal, $\alpha_0 = 0.01$ ppbv NO$_2$ (ppbv O$_3$)$^{-1}$, $c_1 = 15$ ppbv NO$_2$ and $\alpha_p = 0.25$ ppbv NO$_2$ (ppbv PAN)$^{-1}$. However, Gehrig and Baumann (1992) reported a PAN interference of up to $\alpha_p = 0.5$ ppbv NO$_2$ (ppbv PAN)$^{-1}$. Our own determination of the parameters $c_1$ and $\alpha_0$ yielded a O$_3$ interference of $\alpha_0 = 0.008$ ppbv NO$_2$ (ppbv O$_3$)$^{-1}$ and $c_1 = 6.2$ ppbv. Some luminol batches purchased in 1991 (batch numbers less than 920300) showed an interference of up to $\alpha_0 = 0.06$ ppbv NO$_2$ (ppbv O$_3$)$^{-1}$.

Determination of NH$_3$, HNO$_3$ and HONO

NH$_3$, HONO and HNO$_3$ were sampled with the newly developed wet effluent diffusion denuder (WEDD) technique (Vecera & Dasgupta, 1991). NO$_2$ and NO$_3$ were analysed by ion chromatography (IC) (Genfa & Dasgupta, 1989). The response time for the system was 5 min. The detection limit of the wet denuder for HNO$_3$ and HONO was 0.05 ppbv. The precision of the signal was 5% at 1 ppbv in ambient air. NH$_4^+$ was analysed as the fluorescence signal produced by reaction with o-phthalaldehyde in the presence of sodium sulphite (Genfa & Dasgupta, 1989). With a 1-min averaging period the detection limit of the continuous-flow instrument (FIA) was 0.1 ppbv. A two-channel instrument was used for gradient measurements.

Determination of aerosol-bound NH$_4^+$, NO$_3^-$ and NO$_2^-$

In order to measure the aerosol-bound quantity of NH$_4^+$, NO$_3^-$ and NO$_2^-$ the mist chamber system (MCS) developed by Simon and Dasgupta (1995) was used (see also Blatter et al., 1993) with a collection efficiency for NaNO$_3$ aerosols of 100%, and of 98% for Na$_2$SO$_4$ aerosols using 600 mg min$^{-1}$ of steam (aerosol concentration $2.7 \times 10^5$ g m$^{-3}$, mean diameter $2.5 \times 10^{-7}$ m). The calculated efficiency based on Henry’s law was more than 99% for gaseous HNO$_3$ and more than 95% for NH$_3$. This system may be used to measure total nitrate (HNO$_3$ + NO$_3^-$) and total nitrite (HONO + NO$_2^-$). The response time was 5 min with a detection limit of 2 nmol m$^{-3}$. A single measuring system was used in combination of both the WEDD and the MCS collection systems.

During IOP-2 and IOP-4 the two systems were fixed to two separate platforms mounted to opposite sides of a 6-m long ladder. A computer-controlled motor mounted to the top of the ladder was used to move the two platforms up and down in opposite direction. This allowed the platform to be switched or to be placed at identical height. NH$_3$ concentrations measured at identical levels mostly did not differ by more than 5%. Larger deviations were observed during rapid
fluctuations in concentration. The heights at which measurements were made are listed in Table 1.

**NH₃ measurements with passive samplers**
The passive NH₃ samplers described by Blatter et al. (1992) were used for NH₃ concentration determinations on a weekly basis. Laboratory tests yielded a standard deviation for the slope of the calibration curve of 2.5%. In field conditions, the passive samplers showed deviations of less than 5% in comparison with the WEDD. Three to five boxes, each containing four samplers, were mounted at levels between 0.5 and 5.5 m above ground. The precision (2σ) was better than 7%. Outliers (1 value out of 4) were rejected.

**Meteorological measurements**
Self-constructed 6-cup-anemometers described by Rosset (1990) were used for the measurement of horizontal wind speed. The detection limit was about 0.15 m s⁻¹, and stable rotation could be obtained above 0.2 m s⁻¹. Four to six anemometers were installed at the heights given in Table 1. Dry bulb temperature was measured with active semi-conductors (Analog Devices AD590 JF). Deviations between sensors were smaller than ± 0.05°C. A sonic anemometer of type Solent (former Gill, Great Britain) was used for the measurement of three-dimensional wind speed fluctuations (Eugster, 1994). The coordinate system of the wind vector measurements was rotated to be in line with the mean streamlines of the averaging period of 30 min (Zeman & Jensen, 1987). The sampling rate was approximately 21 Hz.

**Bowen-ratio measurements**
Description of the technical details of the Bowen ratio measuring system Campell was given by Rosset (1990). Due to the use of water for the determination of the wet bulb temperature, measurements below the freezing point were not possible. Global radiation was measured at 1 m above ground, and three soil heat flux plates were installed at 1.5 cm below the soil surface.

**Measurements of wet deposition of nitrogen**
Rain was sampled on a weekly basis with a wet/dry precipitation collector (Model 301 Aerochem Metrics, Florida). Concentrations of NH₄⁺ and NO₃⁻ were determined by ion chromatography. Measurements were repeated when the balance between anions and cations differed by more than 10% of the sum of ion equivalents. When the second measurement still yielded an imbalance of more than 10%, the data from this sample were rejected. This occurred occasionally with very small rain water samples.

**METHODS**

**Gradient method**
Under steady-state conditions the vertical flux $F_c$ of a chemically inert substance is independent of height:

$$ F_c = -K \frac{\Delta c}{\Delta z} $$

where $K$ = turbulent mass transfer coefficient, $c$ = concentration and $z$ = height along the vertical axis. The coefficient $K$ must be calculated from micrometeorological measurements and the gradient is determined by at least two concentration measurements at different heights. Using the aerodynamic method (Businger et al., 1971; Monteith & Unsworth, 1990; Zeller et al., 1990), $K$ is calculated from the friction velocity $u^*$, the Karman constant $k = 0.4$, the non-dimensional vertical gradient of mass $\phi$ and the zero plane displacement $d$ by

$$ K = u^* k(z - d) \phi^{-1} $$

$\phi$ is a function of the Monin–Obukhov-Length $L = \frac{x^2}{g}$ (Monin & Obukhov, 1954) and can be parametrized (Panofsky & Dutton, 1984) according to

$$ \phi = \begin{cases} 
1 + \frac{5.5x^2}{L} & \text{stable, } L \geq 0 \\
1 - \frac{16x^2}{L} & \text{unstable, } L < 0 
\end{cases} $$

where $\theta$ is the turbulent potential scaling temperature, $g$ the acceleration due to gravity, and $\theta$ the average potential temperature. With the aerodynamic gradient method, $u^*$ and $\theta$ were determined from measured profiles of horizontal wind speed and temperature.

Often, $d$ is assumed to be $0.67 \times h$ ($h$ = vegetation height). The relationship between $d$ and $h$ was tested by calculating $d$, $u^*$, and $\theta$ from vertical profiles of wind speed and temperature by numerical iteration (Hesterberg, 1994; method proposed by Marquardt, 1963). The average of these calculations confirmed the relationship, but $d$ tended to decrease with increasing friction velocity $u^*$. By using $d = (\alpha u^* + h)$, $\alpha$ ranged between 2.4 and 4.0 s.

Alternatively, $K$ could be determined directly from $\theta$ by measuring the sensible heat flux $H$ with a sonic anemometer and a fast-response temperature sensor (Businger et al., 1971; Zeller et al., 1990), or with the Bowen-ratio measuring system. Applying the Bowen-ratio method, $K$ is calculated from the energy balance above the surface from net radiation, soil heat flux and profiles of temperature and humidity (Stull, 1988; Monteith & Unsworth, 1990).

Eddy correlation and aerodynamic method yielded similar $K$ values within a range of 20%. The average $K$ from the aerodynamic method and from eddy correlation measurements was routinely used. In comparison to those two methods a larger scatter was observed in the Bowen-ratio data (Buwal, 1994a), even after testing the quality of the data according to Ohmura (1982). We therefore used the $K$ values from the Bowen-ratio measuring system only in cases where no data was available from the other two methods.

**NO₂ flux measurements by eddy correlation**
The turbulent flux of a gas, such as NO₂, in any direction can be expressed by the covariance $\text{cov} < u_c >$ of
the time series of the wind speed along the direction of \( u \) and the concentration \( c \). Thus, the vertical turbulent flux of the gas is

\[
F_c = \text{cov}(w, c) = \overline{w'c'}
\]  

(5)

where \( w \) is the vertical wind speed and \( w' \) and \( c' \) are the momentaneous deviations from the averages \( \overline{w} \) and \( \overline{c} \). Due to the alignment with the mean streamlines (Zeman & Jensen, 1987) the mean vertical wind speed \( w \) is always zero, such that eqn (5) is valid. The time-series of concentration was detrended by linear regression, then shifted by 1–2 s relative to the wind speed time series to correct for the lagtime caused by the gas inlet system of the LMA-3. This lagtime was determined from the clear peak of the cross-correlation of \( w \) and \( c \).

The LMA-3 is not an ideal instrument for measuring NO\textsubscript{2} fluctuations at high frequencies. To estimate the error of the eddy correlation flux measurement obtained from the LMA-3, Eugster and Senn (1995) developed a cospectral correction model which was used to correct the measured fluxes. The underestimation of the NO\textsubscript{2} flux was in the range of 11–22 % on average. The correction model used the measured cospectrum by Kaimal et al. (1972). A similar model using a different approach can also be found in Moore (1986). The reported NO\textsubscript{2} fluxes in this work were corrected for these damping losses. For the determination of the mean NO\textsubscript{2} deposition, the data from the unusable wind sectors (64°–138°) were not used. Furthermore, data were rejected in case of an observed positive momentum flux, \( \overline{u'w'} > 0 \), where \( u' \) is the momentaneous deviation from the average \( \overline{u} \) (Eugster et al., 1993). The distance between the points of wind speed measurement and the inlet system of the LMA-3 was 8 cm, which was small enough for eddy correlation measurements at 2 m above ground as suggested by Dyer et al. (1982).

The reproducibility of the LMA-3 was tested during IOP-4 with two instruments working in parallel. Linear regression analysis of the two concentration measurements yielded a slope of 0.979 ± 0.006 and an intercept of (0.29 ± 0.04) ppbv NO\textsubscript{2} \( (r^2 = 0.96) \). Regression analysis of NO\textsubscript{2} fluxes yielded a slope of (1.058 ± 0.007) and an intercept of (0.89 ± 0.10) \times 10^{-3} \text{ppbv m s}^{-1} \( (r^2 = 0.96) \). Thus the eddy correlation technique with use of a LMA-3 detector is capable of determining NO\textsubscript{2} fluxes with a reproducibility of 6 %.

**Resistance analogy**

A commonly used approach in describing fluxes is the resistance analogy. This approach assumes that the flux is proportional to \( c \). The proportionality coefficient \( v_d \) is generally known as the deposition velocity:

\[
F_c = -v_d \cdot c
\]  

(6)

Both \( v_d \) and \( c \) are referenced to a fixed height above ground. \( v_d \) can be expressed as the reciprocal value of a series of resistances:

\[
v_d = \frac{1}{R_a + R_b + R_c}
\]  

(7)

where \( R_a \) = aerodynamic resistance, \( R_b \) = boundary layer resistance, and \( R_c \) = canopy resistance. \( R_a \) and \( R_b \) can be calculated from friction velocity and wind speed (Monteith & Unsworth, 1990). \( R_b \) and \( R_c \) depend on the nature of the gas, and on surface characteristics.

For some gases, such as NH\textsubscript{3} or NO\textsubscript{2}, deposition may occur depending on the condition of the vegetation/soil system which has a finite partial pressure. Formally this can be accounted for in eqn (6) by a compensation point \( c_0 \) and introducing \( v_d^* \) as a deposition velocity:

\[
F_c = -v_d^* \cdot (c - c_0)
\]  

(8)

Net emission occurs when the concentration drops below \( c_0 \), otherwise net deposition takes place.

**RESULTS**

**Nitrogen dioxide flux**

Concentrations and fluxes of NO\textsubscript{2} were corrected for the \( O_3 \) and PAN interference of the LMA-3 according to eqn (1) (Hesterberg, 1994). \( O_3 \) concentrations were measured on site, and PAN concentrations observed by Wunderli and Gehrig (1991) near the city of Zurich were used. \( O_3 \) and PAN fluxes were estimated from concentration and deposition velocity (eqn (6)). \( O_3 \) deposition velocity was assumed to be equal to the observed NO\textsubscript{2} deposition velocity. PAN deposition velocity was chosen at half the observed NO\textsubscript{2} deposition velocity (Erisman et al., 1994). On average the calculated \( O_3 \) and PAN interferences to the NO\textsubscript{2} deposition were quite small with -2 % in IOP-1, 1 % in IOP-2, -1 % in IOP-3, and 1 % in IOP-4 with extreme values between -12 and +50 %.

In the presence of \( O_3 \), NO emitted from soils is rapidly oxidized to NO\textsubscript{2}, producing an upward NO\textsubscript{2} flux. Consequently, NO\textsubscript{2} fluxes are decreasing with height. At ambient \( O_3 \) levels, the time constant for the oxidation of NO to NO\textsubscript{2} is in the same order of magnitude as the time necessary for transporting emitted NO to the measuring height by turbulent eddies. Hence, measured fluxes of NO\textsubscript{2} reflect approximations of net NO\textsubscript{2} (= NO + NO\textsubscript{2}) fluxes. Assuming a constant rate of NO emission throughout one IOP and a constant NO oxidation efficiency, an indirect approach was used to estimate the order of magnitude of the NO emission rate. The slopes of the linear regression analysis according to eqn (8) reflect mean deposition velocities, the intercepts correspond to NO\textsubscript{2} fluxes. Denitrification and nitrification processes in soils lead to negligible NO\textsubscript{2} emission rates in comparison to NO emission rates (Galbally, 1989). Hence, the estimated NO\textsubscript{2} emission corresponds to the release of NO, when an oxidation efficiency of 100 % is assumed. This approach yielded mean NO emission rates of 1.6, 0.7 and 0.7 kg NO-N ha\textsuperscript{-1} y\textsuperscript{-1} during IOP-1, 2 and 4, respectively (Table 2 and
Table 2. Number of observations (A), percentages of accepted data (pc), concentration of NO₂, the linear regression analysis using eqn (8), dry deposition of NO₂, (positive sign corresponds to emission), and deposition velocity of NO₂ (eqn (6)) for different IOPs (± standard deviation on concentrations, depositions and deposition velocities, ± standard error on slopes and intercepts)

<table>
<thead>
<tr>
<th>IOP</th>
<th>A</th>
<th>pc</th>
<th>Conc (ppbv)</th>
<th>Linear regression analysis</th>
<th>Deposition (kg N ha⁻¹ y⁻¹)</th>
<th>Vd (cm s⁻¹)</th>
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<tr>
<td></td>
<td></td>
<td></td>
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<td>Slope v_d</td>
<td>r²</td>
<td>Intercept  v_d</td>
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<tr>
<td></td>
<td></td>
<td></td>
<td>(kg N ha⁻¹ y⁻¹)</td>
<td>(cm s⁻¹)</td>
<td></td>
<td>(kg N ha⁻¹ y⁻¹)</td>
</tr>
<tr>
<td>IOP-1</td>
<td>514</td>
<td>74.3</td>
<td>7.4 ± 1.2</td>
<td>1.55 ± 0.23</td>
<td>-0.245 ± 0.016</td>
<td>0.39</td>
</tr>
<tr>
<td>IOP-2</td>
<td>831</td>
<td>64.3</td>
<td>5.2 ± 1.6</td>
<td>0.68 ± 0.16</td>
<td>-0.206 ± 0.013</td>
<td>0.31</td>
</tr>
<tr>
<td>IOP-3</td>
<td>1137</td>
<td>63.2</td>
<td>19.9 ± 2.4</td>
<td>-0.11 ± 0.17</td>
<td>-0.101 ± 0.004</td>
<td>0.46</td>
</tr>
<tr>
<td>IOP-4</td>
<td>1081</td>
<td>69.8</td>
<td>4.9 ± 1.6</td>
<td>0.67 ± 0.15</td>
<td>-0.238 ± 0.015</td>
<td>0.26</td>
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</table>

*Average of mean daily variation.

Fig. 2. Linear regression analysis of the relationship between measured NO₂ flux (neg. = deposition) and NO₂ concentration using eqn (8) (— regression line; — 95% confidence intervals on the slopes).

Fig. 3. Average diurnal variations in concentration, flux (neg. = deposition) and deposition velocity of NO₂.

No significant NO flux could be observed in winter (IOP-3). An annual total of 0.7 kg NO-N ha⁻¹ y⁻¹ lies in the range of NO emission rates of 0.1–4 kg N ha⁻¹ y⁻¹ given in, the literature for non-cultivated, unfertilized soils (Betlach & Tiedje, 1982; Johansson & Galbally, 1984; Anderson & Levine, 1987; Shepherd et al., 1991). NO₂ fluxes reported in this study were corrected for this effect. The largest correction had to be applied during summer (IOP-1) when about 50% of the NO₂ deposition was compensated for by oxidized NO leaving the soil. No corrections were applied during winter (IOP-3) when the intercept was not significantly different from zero.

In the presence of active vegetation (IOP-1, 2, and 4), the diurnal pattern of NO₂ flux reflected the influence of stomatal uptake (Fig. 3). Largest deposition rates between 3 and 6 kg N ha⁻¹ y⁻¹ were observed during the late morning hours. Stomatal closure could have been responsible for the decline to about 2 kg N ha⁻¹ y⁻¹ around noon. The average deposition was between 1.8 and 3.1 kg N ha⁻¹ y⁻¹ during these IOP's (Table 2). During wintertime, only a small diurnal variation proportional to the NO₂ concentration was observed and the mean flux of 3.7 kg N ha⁻¹ y⁻¹ was much higher than during the campaigns in spring and summer, due to higher NO₂ concentrations of 20 ppbv with daily means of up to 25 ppbv during stable, foggy weather situations.

The mean diurnal variation of the measured deposition velocity during the growing season was controlled by stomatal activity. Minimum values of around 0.1 cm
Taking into account errors associated with concentrations and fluxes.

During winter (IOP-3) $V_d$ was nearly constant throughout the day and the daily mean of 0.11 cm s$^{-1}$ was about the half of the averages of 0.19–0.24 cm s$^{-1}$ observed during the summer IOP's.

**NH$_3$ deposition**

The successful use of the gradient technique requires stationary conditions, i.e. the turbulent surface layer above the ground must be in equilibrium with the ground. Hence, the data must be evaluated according to the following criteria. (a) Wind direction must be in the appropriate sector where fetch requirements are fulfilled. (b) Concentration differences must be larger than the detection limit (0.2 ppbv for NH$_3$). (c) Wind speed must be larger than 0.3 m s$^{-1}$ in order to get a reliable measurement of eddy diffusivities $K$. (d) No large variations in NH$_3$ concentration due to nearby emissions should occur, i.e. 30-min averages should not change by more than 20 %. (e) The NH$_3$ compensation point $c_0$ of our experimental field is likely to be lower than 10 ppbv so therefore, emission should not have occurred during periods with a concentration above 10 ppbv NH$_3$.

In summary, NH$_3$ flux data were rejected when one of the following criteria was met:

1. wind direction in bad sector (0°–285° or 345°–360°)
2. $Ac < 0.2$ ppbv between the two heights
3. mean wind speed at 2 m smaller than 0.3 m s$^{-1}$
4. $Ac/c > 20$ % for 30-min averages
5. NH$_3$ emission occurs above 10 ppbv NH$_3$

Table 3 lists the parameters of the analysis from which fluxes were calculated from mean concentrations as function of the criterion passed. Only 5–10 % of the data passed all criteria. Up to 95 % of all data were rejected applying criteria (a) and (d), but large changes in NH$_3$ fluxes only occurred during IOP-1. Based on linear regression analysis and by accounting for errors of fluxes and concentrations (Mandel, 1964) the arithmetic average NH$_3$ flux varied only between 8.2 and 8.9 kg N ha$^{-1}$ y$^{-1}$ when a fraction of 0.02 of the variance of concentration determination to the variance of the flux determination was used. Because of the relatively large uncertainty of $F_c$ compared to the uncertainty of the concentration, simple linear regression analysis was applicable.

Figure 4 shows a plot of the data with the regression lines from the final calculation. The compensation point was determined to be 5.6 ± 6.9 in IOP-1, 2.9 ± 4.2 in IOP-2, and 4.8 ± 2.7 ppbv NH$_3$ in IOP-4 (+ standard error). Unfortunately, the uncertainty of these values was of similar magnitude as the mean value, and in some cases the sign of $c_0$ changed depending on the criterion passed. Only for IOP-4, was the standard error of $c_0$ smaller than $c_0$ itself, indicating clearly the existence of a compensation point. Over unfertilized grassland $c_0$ is reported to be not significantly different from zero (Andersen et al., 1993; Sutton & Fowler, 1993; Sutton

Table 3. Number of observations (A), percentages of accepted data (pc), concentration of NH$_3$, the linear regression analysis using eqn (8) (pos. = emission), and concentration-weighted NH$_3$ flux (pos. = emission) depending on quality criteria used during each IOPs (± standard deviation on concentrations, ± standard error on slopes and intercepts)

<table>
<thead>
<tr>
<th>IOP</th>
<th>Criterion</th>
<th>A</th>
<th>pc</th>
<th>Conc (ppbv)</th>
<th>Intercept $v_d^*$, $c_0$</th>
<th>Linear regression analysis</th>
<th>Flux (kg N ha$^{-1}$ y$^{-1}$)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td>Slope $v_d^*$</td>
<td>$r^2$</td>
<td></td>
</tr>
<tr>
<td>IOP-1 None 242</td>
<td>100.0 ± 9.5</td>
<td>10.0 ± 9.5</td>
<td>2.7 ± 1.2</td>
<td>-0.22 ± 0.05</td>
<td>0.07</td>
<td>-0.86</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a  41</td>
<td>16.9</td>
<td>9.6 ± 6.6</td>
<td>-4.4 ± 1.6</td>
<td>-0.20 ± 0.08</td>
<td>0.12</td>
<td>1.26</td>
</tr>
<tr>
<td></td>
<td>b  37</td>
<td>15.3</td>
<td>10.4 ± 6.5</td>
<td>-5.4 ± 1.9</td>
<td>-0.23 ± 0.09</td>
<td>0.16</td>
<td>1.58</td>
</tr>
<tr>
<td></td>
<td>c  35</td>
<td>14.5</td>
<td>9.6 ± 5.4</td>
<td>-6.9 ± 2.1</td>
<td>-0.33 ± 0.11</td>
<td>0.21</td>
<td>1.44</td>
</tr>
<tr>
<td></td>
<td>d  15</td>
<td>6.2</td>
<td>13.5 ± 5.6</td>
<td>-1.5 ± 1.7</td>
<td>-0.12 ± 0.07</td>
<td>0.20</td>
<td>0.50</td>
</tr>
<tr>
<td></td>
<td>e  13</td>
<td>5.4</td>
<td>13.3 ± 6.0</td>
<td>-1.3 ± 1.5</td>
<td>-0.13 ± 0.06</td>
<td>0.30</td>
<td>-0.86</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>9.3 ± 4.5$^a$</td>
<td>-1.3$^b$</td>
<td>-0.13$^b$</td>
<td>-0.85$^b$</td>
<td></td>
</tr>
<tr>
<td>IOP-2 None 402</td>
<td>100.0</td>
<td>13 ± 12</td>
<td>2.0 ± 2.8</td>
<td>-0.17 ± 0.10</td>
<td>0.008</td>
<td>-5.74</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a  63</td>
<td>15.7</td>
<td>9.0 ± 6.9</td>
<td>-2.1 ± 5.4</td>
<td>-0.79 ± 0.27</td>
<td>0.12</td>
<td>-15.40</td>
</tr>
<tr>
<td></td>
<td>b  51</td>
<td>12.7</td>
<td>10.3 ± 7.0</td>
<td>-1.0 ± 7.2</td>
<td>-0.77 ± 0.33</td>
<td>0.10</td>
<td>-16.09</td>
</tr>
<tr>
<td></td>
<td>c  45</td>
<td>11.2</td>
<td>10.5 ± 7.4</td>
<td>-0.8 ± 8.0</td>
<td>-0.79 ± 0.35</td>
<td>0.10</td>
<td>-16.55</td>
</tr>
<tr>
<td></td>
<td>d  32</td>
<td>8.0</td>
<td>9.3 ± 5.9</td>
<td>-1.3 ± 7.2</td>
<td>-0.50 ± 0.38</td>
<td>0.05</td>
<td>-12.31</td>
</tr>
<tr>
<td></td>
<td>e  31</td>
<td>7.7</td>
<td>8.8 ± 5.3</td>
<td>-5.2 ± 7.2</td>
<td>-1.01 ± 0.39</td>
<td>0.17</td>
<td>-17.02</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>12.4 ± 2.2$^a$</td>
<td>-10.8$^b$</td>
<td>-1.3$^b$</td>
<td>-19.3$^b$</td>
<td></td>
</tr>
<tr>
<td>IOP-4 None 305</td>
<td>100.0</td>
<td>12 ± 12</td>
<td>-5.5 ± 2.1</td>
<td>-0.58 ± 0.08</td>
<td>0.23</td>
<td>-6.58</td>
<td></td>
</tr>
<tr>
<td></td>
<td>a  42</td>
<td>13.8</td>
<td>9.3 ± 6.3</td>
<td>-2.3 ± 1.9</td>
<td>-0.40 ± 0.10</td>
<td>0.30</td>
<td>-6.03</td>
</tr>
<tr>
<td></td>
<td>b  36</td>
<td>11.8</td>
<td>9.9 ± 6.4</td>
<td>-1.8 ± 2.3</td>
<td>-0.38 ± 0.11</td>
<td>0.26</td>
<td>-6.26</td>
</tr>
<tr>
<td></td>
<td>c  26</td>
<td>8.5</td>
<td>8.7 ± 6.6</td>
<td>-1.5 ± 2.6</td>
<td>-0.39 ± 0.14</td>
<td>0.25</td>
<td>-6.59</td>
</tr>
<tr>
<td></td>
<td>d  16</td>
<td>5.2</td>
<td>8.9 ± 6.2</td>
<td>-4.5 ± 2.3</td>
<td>-0.53 ± 0.12</td>
<td>0.56</td>
<td>-6.54</td>
</tr>
<tr>
<td></td>
<td>e  16</td>
<td>5.2</td>
<td>8.9 ± 6.2</td>
<td>-4.5 ± 2.3</td>
<td>-0.53 ± 0.12</td>
<td>0.56</td>
<td>-6.54</td>
</tr>
</tbody>
</table>

$^{a}$Average of mean daily variation.

$^{b}$Taking into account errors associated with concentrations and fluxes.
Fig. 4. Linear regression analysis of the relationship between NH₃ flux (neg. = deposition) and NH₃ concentration using eqn (8) (--- regression line; --- 95% confidence intervals on the slopes; □ data points used for regression analysis; ⋄ rejected data).

Fig. 5. Average diurnal variations in concentration, flux calculated by linear regression parameters (eqn (8)) (neg. = deposition), and deposition velocity Vd* of NH₃. Because of the lack of data, the diurnal pattern of Vd* is based on 3-h means.

et al., 1993) which was in disagreement with the observation made in the IOP-4. A compensation point possibly appears when a strong surface loading with NH₃ and NH₄⁺ in the presence of high concentrations in combination with low temperature and wet surfaces occurs. Parts of the already deposited NH₃ and NH₄ are re-emitted with increasing temperature, decreasing air concentration of NH₃ and drying surfaces. Probably, re-evaporation conditions in the early afternoon and large emissions from the nearby intensively managed fields increasing ambient NH₃ concentrations up to 100 ppbv (30-min average) led to the high value of c₀ in comparison to values reported in the literature. This compensation mechanism may have caused the small net deposition rates of 0.8 ± 2.0 kg N ha⁻¹ y⁻¹ (± standard error) measured during June (IOP-1). High rates of NH₃ deposition of about 17 ± 9 kg N ha⁻¹ y⁻¹ were measured during IOP-2 when vegetation was tall (up to 1.40 m) and solar radiation was lower. In the spring (IOP-4), when vegetation was short again, a lower deposition of 6.5 ± 3.0 kg N ha⁻¹ y⁻¹ was measured.

Figure 5 shows the mean diurnal pattern of NH₃ concentration, NH₃ flux calculated by linear regression and mean concentration, and deposition velocity Vd*. In IOP-1 and IOP-4 diurnal variations in NH₃ concentra-

Monitoring of NH₃ concentrations with passive samplers
Weekly average NH₃ concentrations were obtained with passive samplers (Fig. 6). Extreme values of up to 50 ppbv NH₃ were observed when manure was applied to neighbouring fields. This occurred at sowing time and after harvests when the fields were ploughed (e.g. in
Deposition of N-containing compounds

Fig. 6. Weekly variation in (a) flux (pos. = deposition) and (b) concentration of NH$_3$ measured with passive samplers.

July, late November, mid March). The annual mean concentrations at 0.5 and 5 m above ground were about 6 and 16 ppbv, respectively.

To estimate deposition, weekly average values of $K$ were calculated by linear combination of daily mean global radiation, wind speed and temperature. The parameters used were determined from measurements during the IOPs (Hesterberg, 1994). Diurnal variations in the concentration gradient and in $K$ were corrected for by multiplying deposition rates by the time-averaged factor $K/(bc/bz)/(K-ec/bz)$. This factor was 0.32 during IOP-1, 0.91 during IOP-2, and 0.45 during IOP-4. Hence, this approach resulted in an uncertainty in the estimated dry deposition rate of NH$_3$ of about 60%.

A disadvantage of the determination of NH$_3$ deposition with passive samplers is that no screening for erroneous data can be applied, e.g. for data influenced by wind from an unsuitable sector. With a fetch of only about 30 m with winds from SW or NE, the measuring height should not have exceeded 30 cm, according to the fetch criterion suggested by Businger (1986). In order to minimize this error, the NH$_3$ flux was calculated from concentrations measured at the lowest two levels between 0.5 and 1.5 m. The resulting average NH$_3$ deposition was 16 ± 9 kg N ha$^{-1}$ y$^{-1}$, with highest deposition rates of 30 kg N ha$^{-1}$ y$^{-1}$ and one emission of 1.5 kg N ha$^{-1}$ y$^{-1}$ (at 3.3 ppbv NH$_3$). By linear regression analysis of the relationship between $F_c$ and $c$, it was found that $c_0 = 4.6 ± 1.3$ ppbv NH$_3$ and $v_{a*} = 1.40 ± 0.13$ cm s$^{-1}$ ($r^2 = 0.51$, ± standard error). The value of $c_0$ was in agreement with the value observed during the IOPs, whereas $v_{a*}$ was comparatively high.

HNO$_3$ deposition
During IOP-2, an attempt was made to determine the HNO$_3$ deposition rate. The mean concentration of 0.23 (0.14 median) ppbv was very low. The precision of the system (uncertainty of 0.1 ppbv) was not sufficiently high to allow the measurement of gradients (Meixner & Wyers, 1990). An additional problem was the high ratio between NH$_3$ and HNO$_3$. The product of NH$_3$ and HNO$_3$ concentrations was lower than the product of vapour pressures of NH$_3$ and HNO$_3$ over solid ammonium nitrate (NH$_4$NO$_3$) only during daytime (Seinfeld, 1986). Hence, the gradient method yielded unrealistic emission rates.

As an alternative, HNO$_3$ fluxes were estimated by the inferential method using eqn (6). The canopy resistance $R_c$ for extensively managed land given by Wesely (1989) was used. HNO$_3$ deposition was estimated to be about 0.91 kg HNO$_3$-N ha$^{-1}$ y$^{-1}$. Assuming a NO$_3^-$/(HNO$_3$ + NO$_2^-$) ratio of 0.45 according to Lindberg et al. (1990) and Meixner et al. (1985), HNO$_3$ concentrations were estimated to be 1.1 ppbv HNO$_3$ using the data for total nitrate measured during the IOP-4 and IOP-3. This resulted in a deposition rate of 2.0 kg HNO$_3$-N ha$^{-1}$ y$^{-1}$. Assuming identical concentrations during IOP-1 and using the measured concentrations during IOP-2, an annual deposition of about 1.1 kg N ha$^{-1}$ y$^{-1}$ was derived.

HONO deposition
HONO concentrations measured during IOP-2 showed a peak during the early morning hours, and values were generally below 0.1 ppbv during daytime. Because of these low values, fluxes based on 30-min values showed a large scatter. Hence, HONO fluxes could only be estimated by the inferential method (eqn (6)). Calculated annual HONO deposition was 0.16 kg N ha$^{-1}$ y$^{-1}$ which was a negligible contribution to the total N input (< 1%).

Deposition of aerosol-bound NO$_3^-$, NO$_2^-$ and NH$_4^+$
Measurements with the MCS provided the sum of the gas phase and aerosol-bound portions of NH$_3$, HNO$_3$ and HONO, or the corresponding ions. Due to the lower deposition efficiency of aerosols, their gradient is small, and the precision of the MCS was too low for flux measurements. Therefore, deposition was estimated by the inferential method. The deposition velocity of aerosol-bound NO$_3^-$, NO$_2^-$ and NH$_4^+$ was assumed to be 0.12 cm s$^{-1}$ (van Aalst, 1983; Asman & Maas, 1986; Asman & Jaarsveld, 1990; Keuken et al., 1990). Calculated annual depositions were 0.3 kg NO$_3^-$-N ha$^{-1}$ y$^{-1}$, less than 0.01 kg NO$_2^-$-N ha$^{-1}$ y$^{-1}$ and 1.1 kg NH$_4^+$-N ha$^{-1}$ y$^{-1}$.

Wet deposition of nitrogen
Wet deposition was measured on a weekly basis from July 1992 to May 1993 (Fig. 7). During winter, small amounts of precipitation were observed leading to small rates of N deposition of 0.4 kg NO$_3^-$-N ha$^{-1}$ y$^{-1}$ and 1.3 kg NH$_4^+$-N ha$^{-1}$ y$^{-1}$ (January 1993). In summer, the
precipitation of up to 7 mm d⁻¹ delivered high deposition rates corresponding to 31 kg NO₃⁻·N ha⁻¹ y⁻¹ and 33 kg NH₄⁺·N ha⁻¹ y⁻¹. The annual averages were 3.4 kg NO₃⁻·N ha⁻¹ y⁻¹ and 5.6 kg NH₄⁺·N ha⁻¹ y⁻¹. These rates were in quite good agreement with those previously observed in Switzerland (Fuhrer, 1986a; Galli Purghart, 1989) and lay within the range of variation between different years.

**DISCUSSION**

**Annual input of nitrogen and speciation**

The arithmetic mean NH₃ deposition measured during three IOPs of 8.2 ± 3.4 (± standard error) kg NH₃·N ha⁻¹ y⁻¹ was corrected according to the annual concentration in order to obtain the annual deposition rate. Measurements with passive samplers yielded an annual mean NH₃ concentration at 2 m above ground of 13.1 ppbv, which was slightly higher than the mean for the three IOPs (11.2 ppbv). The correction leads to a NH₃ flux of 9.2 ± 3.8 kg NH₃·N ha⁻¹ y⁻¹ which differs from the mean flux of 16 ± 9 kg NH₃·N ha⁻¹ y⁻¹ determined with passive samplers. For further discussions, the average of both values, i.e. 13 ± 5 kg NH₃·N ha⁻¹ y⁻¹ will be used. For the calculation of the annual mean NO₂ deposition, a simple interpolation of the fluxes observed during the four IOPs was 2.8 ± 0.6 kg N ha⁻¹ y⁻¹ (± standard deviation) (Eugster, 1994). The sum of dry deposition of total nitrate, total nitrite and NH₄⁺ was 2.7 kg N ha⁻¹ y⁻¹. Rates of deposition of NO, PAN and N₂O₅ of 0.8, 0.1 and 0.2 kg N ha⁻¹ y⁻¹, respectively, were estimated from concentrations reported for rural regions and deposition velocities measured over grass (Hesterberg, 1994). They contribute only a few percent to the total input. Table 4 lists all estimated and measured annual N inputs. The sum of all fluxes was dominated by the dry and wet deposition of NH₃·N and NH₄⁺·N. followed by wet nitrate deposition and by dry NO₂ deposition (Fig. 8). Total N input was about 28 kg N ha⁻¹ y⁻¹ of which 32 % was due to wet deposition. Possible occult N deposition is not taken into account. This input could contribute significantly to the total N input in hilly and forested regions (Fabian, 1987; Dash, 1988; Grosch and Schmitt, 1988; Kroll & Winkler, 1988; Bobbink et al., 1992), but reported values for flat terrain

![Fig. 7. Weekly variation of (a) wet N deposition (—— NO₃⁻; —— NH₄⁺) and (b) precipitation.](image)

![Fig. 8. Relative contribution of different N-containing compounds to the total annual N deposition (100% = 28 kg N ha⁻¹ y⁻¹) over an extensively managed litter meadow. Possible emission is included for NO and NH₃. Occult deposition was neglected.](image)

**Table 4. Measured concentrations, estimated annual N depositions, measured deposition velocities, and fraction of different species of dry, wet and total deposition to an extensively managed litter meadow. Possible emission is included for NO and NH₃, but not for the other species (e.g. HONO, HNO₃, N₂O₅). Occult deposition was neglected.**

<table>
<thead>
<tr>
<th>Species</th>
<th>Concentration (ppbv)</th>
<th>Dry and wet deposition (kg N ha⁻¹ y⁻¹)</th>
<th>Deposition velocity (cm s⁻¹)</th>
<th>Fraction of dry deposition (%)</th>
<th>Fraction of wet deposition (%)</th>
<th>Fraction of total deposition (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>NH₃</td>
<td>13.1</td>
<td>13</td>
<td>0.13 - 1.40</td>
<td>68.8</td>
<td>4.6</td>
<td>20.1</td>
</tr>
<tr>
<td>NO₂</td>
<td>9.4</td>
<td>2.8</td>
<td>1.0 - 1.20</td>
<td>14.8</td>
<td>20.0</td>
<td>62.2</td>
</tr>
<tr>
<td>HNO₃ + NO₅⁻</td>
<td>1.65</td>
<td>1.4</td>
<td>5.8</td>
<td>3.9</td>
<td>2.2</td>
<td>20.1</td>
</tr>
<tr>
<td>NH₄⁺</td>
<td>4.8</td>
<td>1.1</td>
<td>5.8</td>
<td>3.9</td>
<td>2.2</td>
<td>20.1</td>
</tr>
<tr>
<td>NO, N₂O₅</td>
<td>0.6</td>
<td>0.6</td>
<td>3.2</td>
<td>2.2</td>
<td>2.2</td>
<td>20.1</td>
</tr>
<tr>
<td>HONO, NO₅⁻, PAN</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>NH₄⁺ wet</td>
<td>5.6</td>
<td></td>
<td></td>
<td>62.2</td>
<td>20.1</td>
<td>20.1</td>
</tr>
<tr>
<td>NO₅⁻ wet</td>
<td>3.4</td>
<td></td>
<td></td>
<td>37.8</td>
<td>12.2</td>
<td></td>
</tr>
<tr>
<td>Total</td>
<td>27.9</td>
<td></td>
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<td></td>
</tr>
</tbody>
</table>
are low (0.1 kg NO$_3^-$-N ha$^{-1}$ y$^{-1}$ and 0.4 kg NH$_4^+$-N ha$^{-1}$ y$^{-1}$; Fuhrer, 1986b).

Our observations are in agreement with results from other studies. Stocker et al. (1993) reported a NO$_x$ (= reactive N oxides) dry deposition rate between 2.1 and 6.9 kg N ha$^{-1}$ y$^{-1}$, which is similar to our estimate of 4.4–7.3 kg N ha$^{-1}$ y$^{-1}$. The deposition velocities for NO$_2$ (between 0.11 and 0.26 cm s$^{-1}$) are similar to the observations made by Dollard et al. (1987) (0.25 cm s$^{-1}$), Erismann et al. (1989) (0.33 cm s$^{-1}$) and Lövblad et al., 1993 (0.2–0.5 cm s$^{-1}$). NH$_3$ deposition of 10 kg N ha$^{-1}$ y$^{-1}$ determined by Erismann et al. (1989) was similar to the 13 kg N ha$^{-1}$ y$^{-1}$ measured here. In the framework of the Convention on Long Range Transboundary Air Pollution coordinated by the United Nations Economic Commission for Europe, a map of the exceedance of the critical loads has been produced for Switzerland based on a 1 km x 1 km grid (Buwal, 1994b). The procedure to determine the N input followed the guidelines produced by the Coordination Centre for Effects (CCE, 1993), and parametrization of deposition was simple, without using a source–receptor model. The emission of NH$_3$ has been calculated by using widely accepted emission factors and statistical data for animal husbandry in individual communities. For the grid point near Merenschwand, the calculation differs by 24 % for NH$_3$, by -21 % for NO$_x$ and by 8 % for total N deposition (Buwal, 1994a).

Environmental consequences of large nitrogen inputs

The total N input exceeds the critical load for litter meadows and moors given by Grennfelt and Thörnölf (1992), and it is likely that the N input exceeds the critical load for most natural ecosystems located on the Swiss plateau. Consequently, changes in biodiversity, eutrophication, and deficiencies due to enhanced leaching of nutrients such as potassium, calcium and magnesium, could occur in these ecosystems, and are already detectable (Ulrich & Pankrath, 1983; Ellenberg, 1985, 1989; Fabian, 1987; Bobbink et al., 1992). For the experimental field used in the present study the critical load of nitrogen is about 15 kg N ha$^{-1}$ y$^{-1}$, and the actual load exceeds the critical load by a factor of nearly 2. Over forests, the deposition velocity for NH$_3$ is larger by a factor of 2–3 (Sutton et al., 1992; Wieters et al., 1992; Andersen & Hovmand, 1993; Duyzer et al., 1994) and by a factor of about 1.5–5 for NO$_x$ (Grosch & Schmitt, 1988; Hanson et al., 1989). Assuming the dry deposition velocity is doubled over forests as compared to our measurements and identical concentrations, a total net dry deposition of about 40 kg N ha$^{-1}$ y$^{-1}$ and a total deposition of up to 50 kg N ha$^{-1}$ y$^{-1}$ can be estimated for forests. These values reach the upper limit of the range of critical load values, and it suggests that the critical load for nitrogen is exceeded in forests as well.

For the ecosystem studied in this work, N availability was not likely to be limiting because natural mineralization of organic material in the soil may have delivered up to 60 kg N ha$^{-1}$ y$^{-1}$ (A. Grub, personal communication). During the last 15 years, no effects on vegetation from N overloading were observed (J. Fischer, personal communication). Hence, dry N deposition is not likely to have a direct effect on this meadow.

Atmospheric N input increases the N-pool in the soil. This leads to enhanced emissions of NO and nitrous oxide (N$_2$O) and increased NO$_x$ leaching into the groundwater. Emission of NO stimulates formation of photooxidants during the summer, whereas N$_2$O is a greenhouse gas and enhances O$_3$ destruction in the stratosphere (Finlayson-Pitts & Pitts, 1986). The presence of a compensation point of NH$_3$ decreases the dry deposition of NH$_3$ considerably (ApSimon et al., 1994), and, in turn, NH$_3$ is transported over larger distances and is deposited preferentially to surfaces of ecosystems with a lower compensation point. Often, such ecosystems are sensitive to additional N input (Ellenberg, 1985). With respect to possible strategies to reduce N loading of natural and semi-natural ecosystems, NH$_3$ is particularly important when these systems are surrounded by intensively managed land. In urban areas, changes in ambient concentrations are likely to lead to a lower fraction of NH$_3$ relative to the fraction of NO$_x$ (Jaeschke et al., 1992).

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