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Ecosystem responses to reduced nitrogen and sulphur inputs into two coniferous forest stands in the Netherlands

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Abstract

Atmospheric inputs of nitrogen and sulphur were reduced to pre-industrial levels in a nitrogen and sulphur saturated Scots pine (*Pinus sylvestris*) and Douglas fir (*Pseudotsuga menziesii*) stand in the Netherlands. Starting in 1989, throughfall water was intercepted by means of a roof and replaced by simulated, clean throughfall water. Underneath the roof two plots were designed to receive either clean water (roof clean) or ambient throughfall (roof control). Outside the roof a second control plot was established (ambient control). Until 1992 a significant roof effect was found owing to differences in water application, but automation of the watering regime significantly reduced this problem.

Throughfall chemistry showed a dominance of ammonium to nitrate, whereas the reverse was observed in the soil solution. In the roof clean plots a quick response of soil solution chemistry was observed. The sulphur and nitrogen concentrations in the upper soil layers strongly decreased, as did the fluxes of these elements through the soil profile. As a result, leaching of base cations and ratios of ammonium to various cations decreased.

Decomposition studies in the Scots pine stand showed a positive effect of nitrogen deposition on the decomposition rate in the roof control plot compared with the roof clean plot, whereas in the Douglas fir stand no differences between these plots were found.

A reduction of atmospheric nitrogen and sulphur deposition in the Scots pine stand increased the species diversity of microarthropods, owing to a decreased dominance of some species at a constant species number.

In the Scots pine stand fine root biomass and the number of root tips increased as nitrogen deposition decreased, indicating an increased nutrient uptake capacity. As a result potassium and magnesium concentrations and their ratios to nitrogen in the needles increased. After 4 years of treatment, nitrogen concentrations in the needles of the roof clean plot remained high, but were significantly lower than in the needles of the control plots. In the fourth year of the experiment nitrogen concentrations in the older needles of the clean plot became lower than in the current needles, which is typical for a nitrogen limited forest ecosystem. This is in agreement with the nitrogen flux via litterfall, which was lower onto the roof clean plot than onto the control plots. Until now, no significant

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changes in nutrient concentrations in the needles of the Douglas fir stand have been observed.

Keywords: Decomposition; NIICCE model; NITREX project; Nitrogen removal; Nutrient flux; Soil fauna; Throughfall

1. Introduction

Until the 1950s nitrogen was the growth-limiting nutrient in most forest ecosystems of the Northern Hemisphere and these ecosystems had very tight nitrogen cycles. Water leaving such systems contained little nitrogen. However, during the past few decades the nitrogen cycle has become disrupted in many European forest ecosystems. Atmospheric nitrogen input to these systems has increased to levels that exceed vegetation demand (less than 10 kg N ha^{-1} $year^{-1}$) (Encke, 1986) and nitrogen has appeared in the leachate: the ecosystems have become 'nitrogen saturated' (Aber et al., 1989).

Excess nitrogen availability strongly influences organisms and processes within the forest ecosystem (Fog, 1988). High nitrogen input has a negative effect on soil fauna involved in decomposition processes and affects nutrient recycling within the ecosystem (Verhoef and Brussaard, 1990; Verhoef and Meintser, 1991). However, because of increased nitrogen input enhanced litter quality may stimulate decomposition over the long term (Tietema, 1993). Nitrification of deposited ammonium with subsequent nitrate leaching may lead to acidification, increased weathering and leaching of base cations (Van Breemen et al., 1982; Roelofs et al., 1988). Increased leaching of cations, excessive nitrogen uptake and reduced cation uptake due to ammonium antagonism cause nutritional imbalances within the trees. This is reflected in reduced tree growth and impaired tree vitality (Roelofs et al., 1985; Boxman and Roelofs, 1988; Flaig and Mohr, 1991).

Here we assess the reversibility of nitrogen-saturation in two forest ecosystems following experimental removal of nitrogen inputs. The following questions are central to this study. (1) Is nitrogen saturation reversible, and when and how does recovery proceed? (2) Does a reduction of the nitrogen input lead to recovery of tree health and changes in understory vegetation and how much time is needed for such a recovery? (3) Does reduction of the nitrogen input influence the density and diversity of soil animals, thus influencing decomposition rates?

The data were used to develop, calibrate and validate the process-oriented deterministic model NIICCE (NItrogen Isotopes and Carbon Cycling in coniferous Ecosystems) (Van Dam and Van Breemen, 1994). The model aims to predict input–output budgets of nitrogen for coniferous ecosystems depending on site characteristics and environmental conditions such as atmospheric deposition. This study was carried out within the framework of the NITREX program (Dise and Wright, 1992).

2. Materials and methods

In 1989 two research sites were established in which ambient throughfall water was intercepted by means of a roof and replaced by demineralised water to which all nutrients were added in the same amount as present in the throughfall, except for nitrogen and sulphur. The site at Ysselsteyn, southeastern Netherlands, contains Scots pine (*Pinus sylvestris* L.) of vitality class 3, indicating a needle loss of 26–60% (Smits, 1992). The second site at Speuld, central Netherlands, has a Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco) stand of vitality class 1, indicating a needle loss of 0–10%. Underneath the roofs, two plots (10 m × 10 m) were set up to receive either clean water (roof clean plot) or ambient throughfall (roof control plot). Outside each of the roofs a second control plot was established (ambient control plot).

A detailed description of the sites and of the methods is given by Dise and Wright (1992), Van Dijk et al. (1992a,b), and Boxman et al. (1994b).

2.1. Watering regime

Starting in May 1989, the throughfall water collected from the roof, stored in two 2000-l tanks, and then sprayed in weekly doses onto the roof control plot. The same volume of simulated, clean rainwater was administered to the roof clean plot. This method proved to be unsatisfactory since water collected for a whole week (sometimes as much as 30 mm) was sprayed onto the plot within 1 h. At Ysselsteyn the watering regime was automated to an almost real time watering in the beginning of 1992, and at Speuld in the summer of 1992. By means of sensors in the storage containers the watering regime was adjusted such that spraying was carried out following each 2 mm of precipitation.

2.2. Data derivation for the NIICCE model

The NIICCE model (Van Dam and Van Breemen, 1994) simulates turnover of nitrogen and carbon isotopes in coniferous ecosystems. In this study the hydrological part of the model was used to calculate element fluxes from simulated moisture fluxes and observed concentrations of major constituents in soil solution. Boundary conditions for heat and moisture transport required by NIICCE are hourly values for air temperature, global radiation, relative humidity, precipitation and wind speed. Except for precipitation, these data were derived from measurements at Wageningen, about midway between the Speuld (50 km) and Ysselsteyn (80 km) sites. Precipitation data came from measurements made 1–4 week intervals in forest clearings close to the experimental sites. These precipitation data were assigned daily frequency distributions equal to the precipitation measured at official meteorological stations (Elspeet and Ysselsteyn) situated a few kilometres from the experimental sites (Royal Dutch Meteorological Institute, 1987–1992). Daily precipitation was transformed to hourly values, using a mean precipitation intensity for the Netherlands, varying from 0.57 mm h^{-1} in February to 1.89 mm h^{-1} in August. From these intensities and the observed amount of rainfall the duration of rainfall was calculated. The rainfall intensities were adjusted to match the observed amount of rainfall. The number of precipitation events during a day was randomly varied between 1 and 3, to retain the stochastic character of rainfall events. Distribution of global radiation over 1 day was assumed to be equal to the distribution for cloudless conditions. Temperature fluctuation during the day was described by sinusoidal functions, with maximum temperature occurring at 14:00 h and minimum temperature half an hour after sunrise. Hourly relative humidity was calculated from hourly temperatures and the daily mean value of the absolute air humidity.

2.3. Soil moisture sampling

Ceramic lysimeter cups (Soil Moisture Corp., type 655X1 B1M3 high flow) were installed in the mineral soil of each plot at depths of 10 cm (eight replicates), 25 cm (four replicates), 45 cm (four replicates) and 90 cm (four replicates). Just below the organic layer, ceramic plates (Keramische Masse P80, thickness 3 mm, diameter 135 mm, Staatliche Porzellan-Manufaktur, Berlin) (four replicates) were installed. Soil moisture was collected once a month (fortnightly since April 1992) by applying underpressure to the cups. Samples were analysed separately according to Van Dijk and Roelofs (1988).

2.4. Calculation of element fluxes

Element fluxes in the soil were calculated from daily nutrient concentrations and simulated moisture fluxes as determined with the NIICCE model. The daily concentrations were obtained by assuming a linear change in concentrations between two sampling dates.

2.5. Soil temperature

At both locations soil temperatures were measured every hour at the soil surface (shaded) and at the boundary of the ectorganic and the mineral soil using thermistors in combination with a data logger.

2.6. Decomposition

At Ysselsteyn, needle material from the L-layer (Aoo) and F-layer (Ao) was collected outside the plots in April 1991 and air-dried at room temperature for 3–5 days. Material of the L-layer was hand-sorted for intact needles only, material of the F-layer was sieved to 2–5 mm length. About 7 g (air-dried) needle material of the L-layer and 36 g (50% wet) of the F-layer were placed in 10 cm×10 cm nylon bags with a mesh size of 4 mm for the L-layer and 2 mm for the F-layer. The weight and thickness of the litterbags containing L- and F-material was in agreement with the original weight and thickness of the organic layer as determined by Van Dijk et al. (1992a).

Litterbags with L- and F-material were placed in the field as stratified litterbag sets. The litterbags in each plot were randomly distributed, side by side in rows with 10 cm spacing, in an area of 1.5 m×2 m. The litterbag sets were sequentially removed at random after 2, 4, 6, 8, 10 and 12 months. Every litterbag set had 12 replicates. Eight replicates were used for extraction of soil fauna.

After collection, the litterbags were immediately placed in sealed plastic jars for transportation. After faunal extraction, roots and soil were removed and litterbags were dried in an oven at 50°C for 2–3 days, and the weight loss of the material in the litterbags was calculated.

At the Speuld site, litter-bag material came litter falling on the roof during a 2-week period in November 1991. The litter was air-dried and cones and twigs removed. Aliquots of 5 g (70°C dry weight) were put into polyethylene bags with a mesh size of 0.6 mm. Litterbags measuring 10 cm×10 cm were randomly distributed on the forest floor in each plot over a 2 m×3 m area. Starting in May 1992, five randomly chosen replicate litterbags were removed from the plots after 0, 1, 2, 4, 6, 9, 12 and 15 months. The bags were dried for 48 h at 70°C and the dry weight of the remaining needles was determined.

2.7. Soil fauna

By means of Tullgren-type equipment (Van Straalen and Rijninks, 1982) soil fauna from the Ysselsteyn plots was extracted. All taxa of soil fauna were collected, but only the microarthropod orders Collembola and Oribatei were identified to species. For each litterbag, the numbers of the soil fauna were counted. Counts were expressed as averages for the eight litterbag sets. Taxa of Collembola and Oribatei were combined and diversity was calculated for each plot for each sampling date using the Shannon–Weaver index:

$$H = -\sum p_i \log p_i$$

where p_i is the observed frequency of species i.

Weight loss and diversity were both subjected

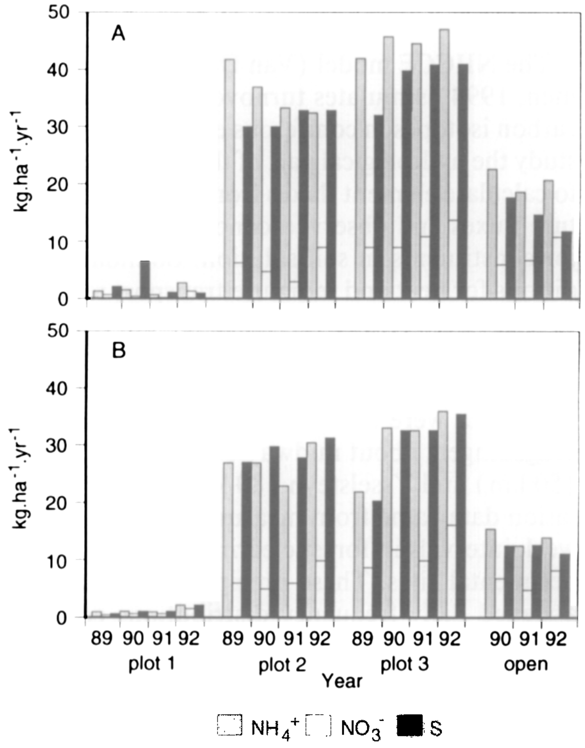


Fig. 1. Deposition fluxes of nitrogen (NH_4^+ and NO_3^-) and sulphur (total S) to the open field and plots at Ysselsteyn (A) and Speuld (B). Plot 1, roof clean; plot 2, roof control; plot 3, ambient control; open, open field precipitation.

Table 1
Watering regime at both locations (mm year^{-1})

	Ysselsteyn			Speuld		
	Roof clean	Roof control	Ambient control	Roof clean	Roof control	Ambient control
1989	497	528	447	441	440	440
1990	477	463	486	402	406	505
1991	371	376	508	358	341	467
1992	673	586	663	472	477	621
Total	2018	1953	2104	1673	1664	2033

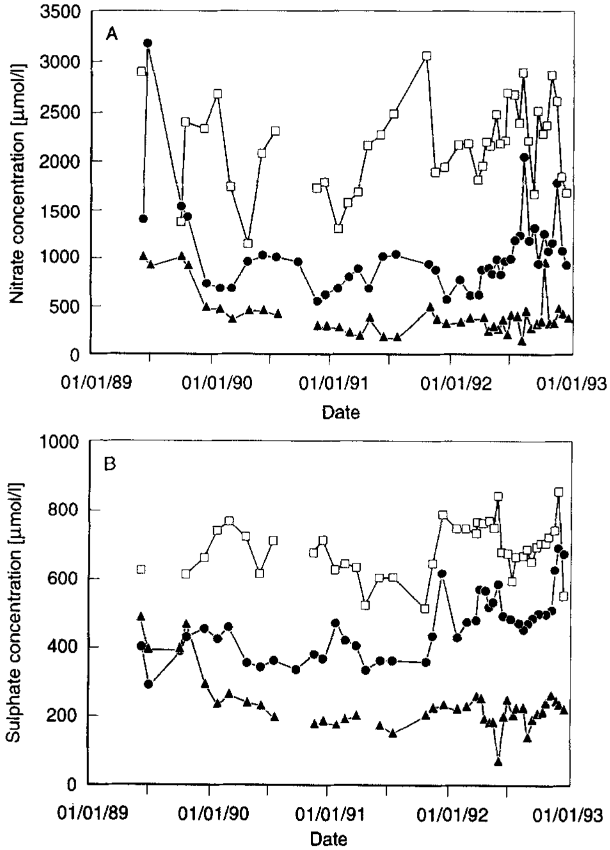


Fig. 2. Nitrate (A) and sulphate (B) concentrations in the soil solution at 90 cm depth at Ysselsteyn during 4 years of treatment. ▲, roof clean plot; ●, roof control plot; □, ambient control plot.

to a two-way analysis of variance with independent variables plot and time. Transformations were applied to the data to achieve homogeneous variance. Arcsin$\sqrt{}$ transformation was used for percentages (Sokal and Rohlf, 1981). Pearson correlation coefficients were calculated among weight loss and diversity. Calculations were done with the MGLH (Multivariate General Linear Hypothesis) module of the program SYSTAT 5.0.

2.8. Roots

In September 1992 the roots were sampled at Ysselsteyn. Nine samples per plot were taken on a 3×3 grid. Soil cores of 25 cm depth were taken with a tube 5 cm in diameter and divided into four layers: humus (H, 2–0 cm) and mineral soil (M, 0–4, 4–13 and 13–18 cm). The morphology of the fine roots (root diameter less than 1 mm) was determined according to the standard NITREX protocol (Clemensson-Lindell, 1994). Vitality class 1 indicates vital fine-roots, vitality class 2 indicates non-vital fine-roots and vitality class 3 indicates dead roots.

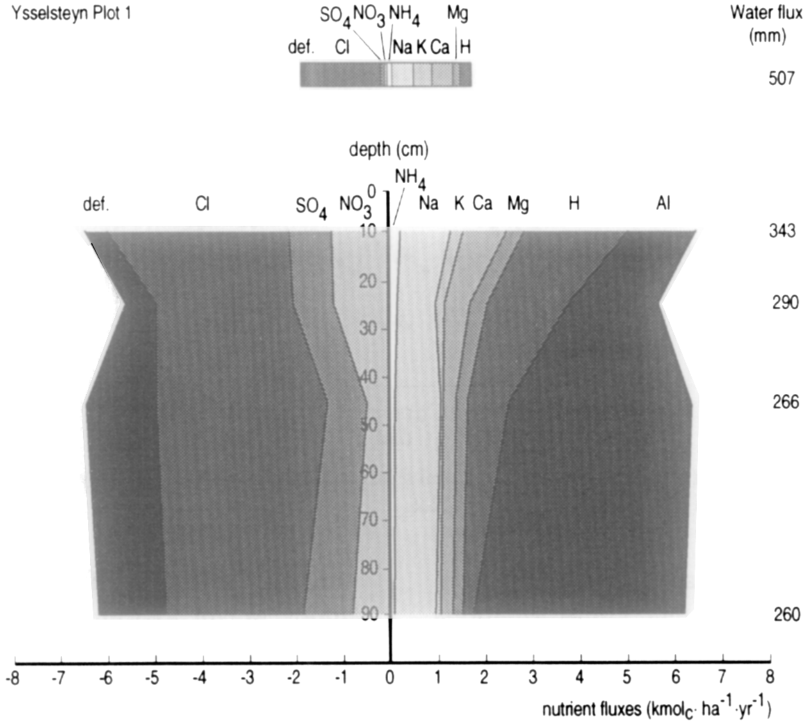
Water extractable nutrients in each layer were determined according to Van Dijk and Roelofs (1988). Chemical analyses of the fine roots were performed on the bulked roots of vitality classes 1 and 2 for each soil core. Attached soil particles were removed from the fine roots with a soft paintbrush and the roots rinsed twice with tap water and once with distilled water. After grinding in liquid nitrogen the roots were dried for 48 h at 70°C. Chemical analyses were performed according to Van Dijk and Roelofs (1988).

2.9. Needles

Until 1993 nitrogen was measured using the open sulphuric acid/peroxide destruction method (Van Dijk and Roelofs, 1988). Since 1993, however, nitrogen has been measured with a NA-1500 C,N,S analyser (Carlo Erba Instruments, Milan, Italy). A comparison between both methods revealed that the nitrogen values obtained with the C,N,S analyser were a factor 1.10–1.14 higher.

2.10. Statistics

If not otherwise stated, the statistical analyses were performed with the general linear model (GLM) procedure available in the software package of the Statistical Analysis System Institute Inc. (SAS Institute Inc., 1988), after testing whether the data were normally distributed



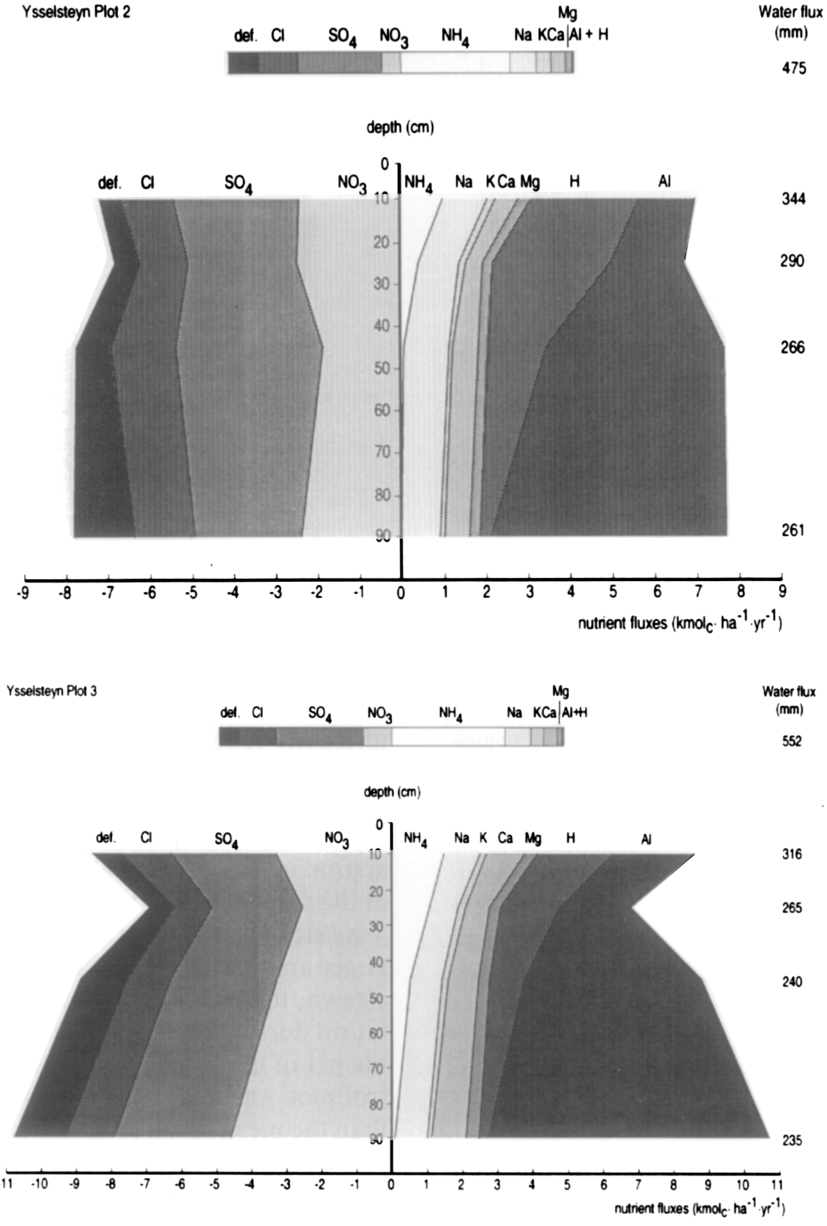


Fig. 3. Nutrient fluxes in the soil at four depths at Ysselsteyn: average values of 1990, 1991 and 1992. For comparison, nutrient fluxes in throughfall water are presented as horizontal bars above each diagram. The water flux at each soil depth is also given. Plot 1, roof clean; plot 2, roof control; plot 3, ambient control.

(univariate procedure). If not, logarithmic transformations were performed to make the variance independent of the mean (Sokal and Rohlf, 1981). The influence of the experimental factors on soil and tree parameters was analysed with ANOVA models of variance, including first-order interactions. Duncan's multiple-range test was performed on all main-effect means.

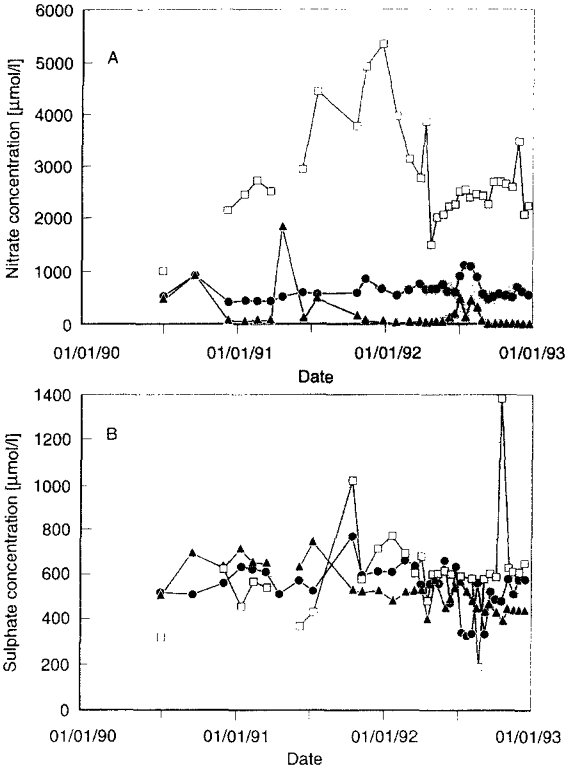
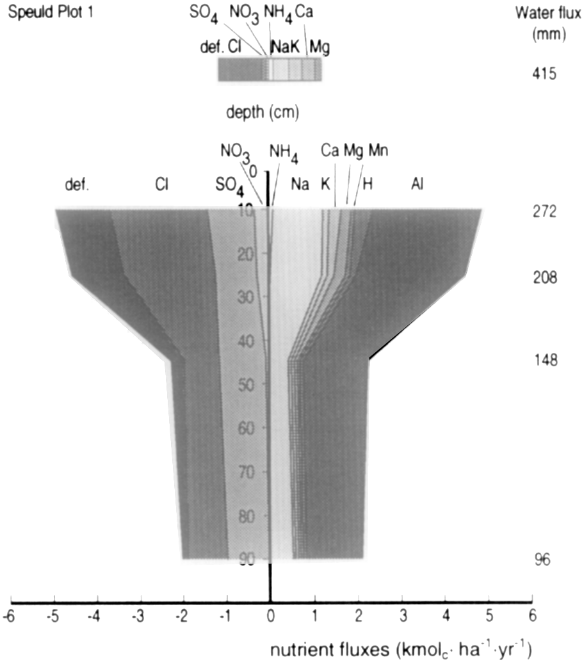


Fig. 4. Nitrate (A) and sulphate (B) concentrations in the soil solution at 90 cm depth at Speuld during 4 years of treatment. ▲, roof clean plot; ●, roof control plot; □, ambient control plot.

3. Results

3.1. Throughfall fluxes to the forest floor of the plots

The interception of throughfall in the roof clean plot reduced the deposition of nitrogen and sulphur to 1-2 kg N ha^{-1} year^{-1} at both locations (Fig. 1). For 4 successive years the deposition onto the ambient control plot was 45-60 kg N ha^{-1} year^{-1} and 30 kg S ha^{-1} year^{-1}, respectively. The water additions to the roofed plots were less than the ambient controls (Table 1), particularly in 1991 when there were many heavy short rain events. In the course of 1992 the watering regime was automated and the gutter system was adjusted so that this problem was significantly reduced. The throughfall flux of ammonium and nitrate to the roof control plot were somewhat lower as to the ambient control plot. The throughfall fluxes of the other nutrients were approximately equal in all plots (Fig. 2). The pH of the open field and throughfall precipitation at Ysselsteyn was higher than in Speuld, owing to a larger amount of dissolved ammonium. At Ysselsteyn, the mean pH of the open field precipitation during the experimental period was 5.61, the pH of the throughfall onto the ambient control plot was 5.88, which was somewhat lower than the mean throughfall pH of the roof control plot (6.29). The mean pH of the rainwater to the roof clean plot was 5.36. In Speuld these pH values were 5.16, 5.47, 6.00 and 5.83, respectively.



3.2. Nutrient fluxes in the soil

At Ysselsteyn, the reduced input of nitrogen and sulphur to the roof clean plot resulted very quickly in reduced ammonium, nitrate and sulphate concentrations in the soil solution (Fig. 2). Over the period 1990-1992 the inorganic nitrogen flux in the soil was reduced by more than 50%

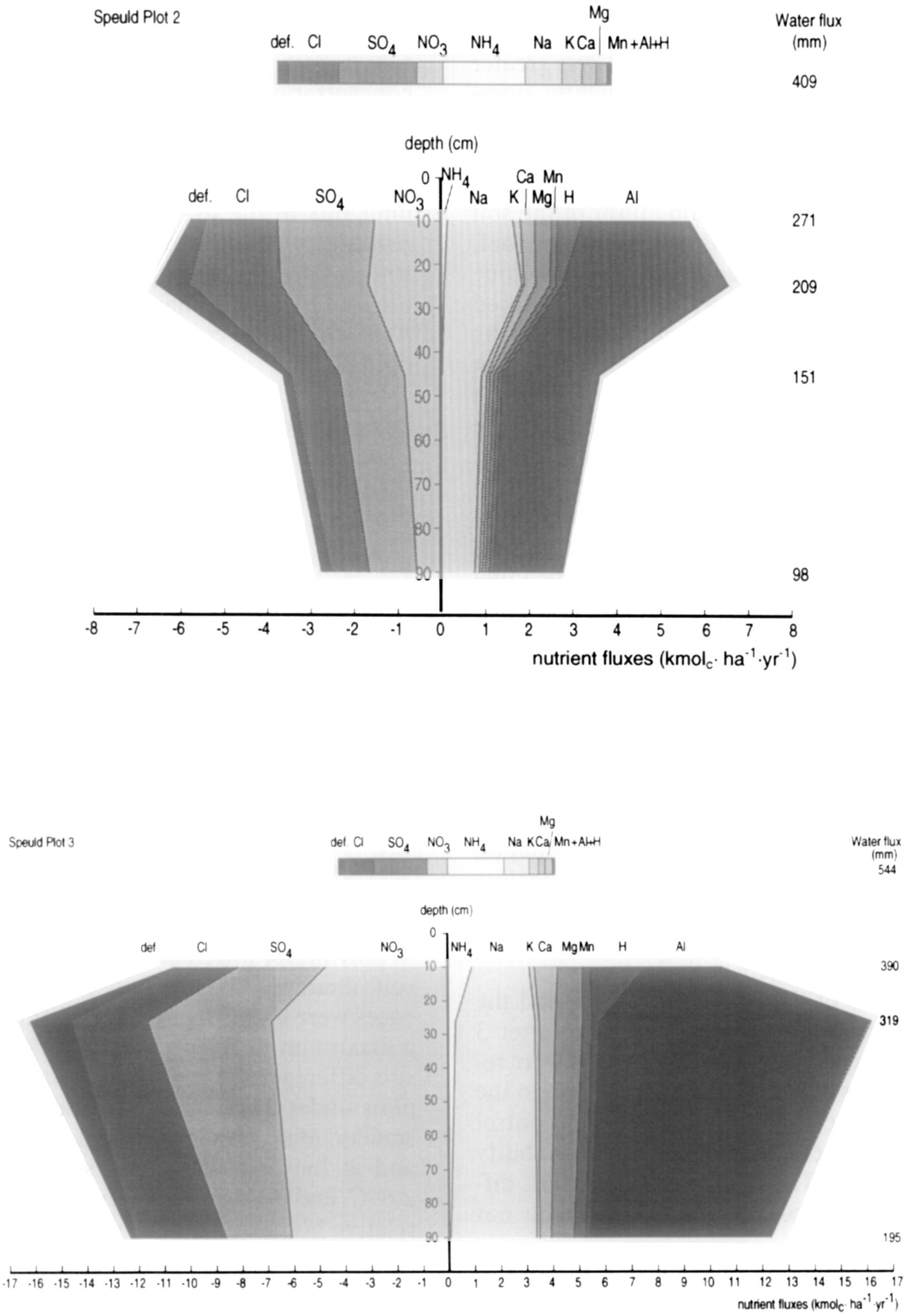


Fig. 5. Nutrient fluxes in the soil at four depths at Speuld: average values of 1991 and 1992. For comparison, nutrient fluxes in throughfall water are presented as horizontal bars above each diagram. The water flux at each soil depth is also given. Plot 1, roof clean; plot 2, roof control; plot 3, ambient control.

compared with the flux in the roof control plot (Fig. 3). In the upper layer of the mineral soil the reduction in ammonium (80%) was greater than that of nitrate (45%). Consequently, dominance of nitrate over ammonium was more pronounced in the roof clean plot than in the roof control plot. At a depth of 45 cm in the mineral soil there was almost no ammonium in the soil solution in either of the plots underneath the roof, and no ammonium leached from the rooting zone (at 90 cm). The total sulphur flux was reduced by 70%. At a depth of 10 cm, most cations showed higher fluxes in the roof clean plot than in the roof control plot. At 90 cm, however, this situation was reversed. At all depths the chloride flux was two to three times higher in the roof clean plot than in the roof control plot, due to a technical error in the spraying equipment. Nitrogen fluxes were 30–50% lower in the roof control plot than in the ambient control plot, depending on the soil depth. In both there was very little ammonium at 90 cm. Sulphur fluxes were also similar in both plots. The fluxes of most cations were lower in the roof control plot than in the ambient control plot.

In all plots at Ysselsteyn some ion fluxes showed similar depth-dependent trends: the fluxes of ammonium, potassium and hydrogen ions decreased with increasing soil depth, whereas the aluminium flux and the negative charge deficit (probably caused by organic anions) increased; the nitrate flux first decreased and than increased again with depth.

Soil extracts of the organic top layer and the upper mineral soil, made at Ysselsteyn after 3 years of treatment, confirmed that there were reduced amounts of ammonium and nitrate in the roof clean plot compared with the roof control plot, although there was high spatial variability within the plots. There were no significant differences between both control plots (data not shown).

At Speuld, reduction of the nitrogen input to the roof clean plot resulted in a strong decrease in the nitrogen concentration in the soil solution (Fig. 4(A)) and the nitrogen flux in the soil was reduced by more than 80% compared with the roof control plot (Fig. 5). Below 45 cm there was no nitrate remaining. Even at 10 cm depth, little, if any, ammonium was present in the soil solution in both plots underneath the roof. Sulphate concentrations in the roof clean plot were reduced less drastically. At greater depths especially, differences between the roofed plots were small (Fig. 4(B)). Depending on depth, the sulphur flux in the soil of the roof clean plot was reduced by 20–60% compared with the roof control plot. Cation fluxes were generally smaller in the roof clean plot than in the roof control plot. At 90 cm, however, ion fluxes were very small in both plots.

In the control plot outside the roof (the ambient control plot), ion concentrations in the soil solution and fluxes in the soil were much larger than in the other plots, particularly at greater depth.

3.3. Temperature, humidity and light

Owing to the presence of the roofs soil temperatures were somewhat higher in the roofed plots. At Speuld these differences were smaller owing to a dense canopy structure.

In Ysselsteyn the roof resulted in an average daily temperature difference at the soil surface of about 0.1 °C. The maximum differences occurred at very warm or very cold spells of the day with an absolute maximum difference at the soil surface of 3.7 °C (Fig. 6). Differences between the plots decreased rapidly with depth, resulting in only minor differences at the organic-mineral soil interface. Average daily temperature differences were lower than 0.06 °C at this depth with a maximum difference of 1.2 °C. At the Speuld site differences in soil temperature between the plots under the roofs and outside the roof were smaller. Maximum differences at the soil surface and at the organic-mineral soil interface were 2.6 °C and 0.8 °C, respectively. Differences in relative humidity were mostly within 2%, while light intensity was reduced by at most 15% (Van Dijk et al., 1992b).

3.4. Decomposition

After 1 year no significant weight loss was measured in the fragmentation layer of all three

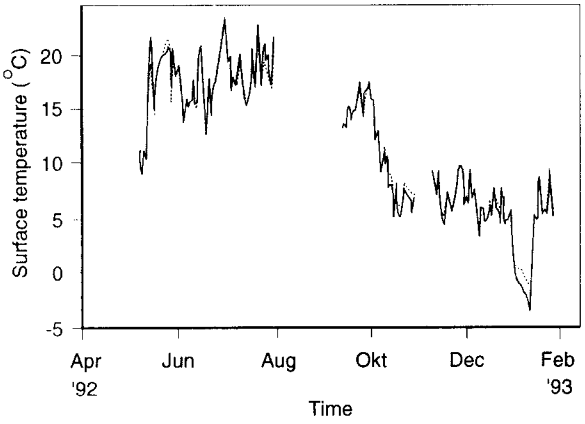


Fig. 6. Soil temperature at the soil surface below (dotted line) and outside (solid line) the roof at Ysselsteyn.

plots at Ysselsteyn (Fig. 7). The L-layer material underneath the roof decomposed at a similar rate in both roofed plots, but at the end of the year, average (±SD) weight loss was less in the roof clean plot than in the roof control plot (32.25±6.70% vs. 40.65±8.02%). In the ambient control plot the litter decomposition rate showed a different pattern and was slower (33.47±6.30%) compared with the roof control plot. A two-way analysis of variance for litter material of all three plots (Table 2) showed significant interactions of the factors plot and time. This indicates not only that the weight loss differed significantly between the plots, but also that the increment per interval differed for each of the three plots. Comparing two plots (the roof clean plot versus the roof control, and the roof control plot versus the ambient control plot) with a two-way analysis of variance the same results were obtained (all $P \leqslant 0.001$). In Speuld no significant differences in decomposition rates between the roof clean plot and the roof control plot were found. A two-way analysis of variance showed no significant differences between both roofed plots during 15 months. During the first 9 months a significant ($P \leqslant 0.05$) roof effect on the decomposition was found. The decomposition rate in the ambient control plot was higher than in both roofed plots.

3.5. Soil fauna

In total 50 species of the two microarthropod groups were found; 17 species of Collembola and 33 species of Oribatei. The number of species during the year were similar in all three plots (Table 3), except for September when considerably fewer species were found underneath the roof, probably because of a very dry period. *Isotoma notabilis* (Schäffer, 1896) was the dominant collembolan species and *Oribatula tibialis* (Nicolet, 1833) the dominant oribatid species.

The diversity of Collembola and Oribatei combined (Table 3) was low in the roof control plot in the winter period (November to March). In the roof clean plot, the diversity index increased in the winter period owing to a decrease of the number of individuals of several dominant species. This resulted in a constant diversity in the roof clean plot throughout the year. The highest diversity indices were found in the ambient control plot with 1.028 decits and 1.012 decits in March and May, respectively. A two-way analysis of variance for all three plots showed significant interactions of the factors plot and time ($P \leqslant 0.001$). The same results were obtained when two plots (the roof clean plot versus the roof control, and the roof control plot versus the ambient control plot) were tested (all $P \leqslant 0.001$).

In order to obtain information about the relationship between decomposition and soil fauna diversity, Pearson correlation coefficients were calculated. The different treatments underneath the roof showed no interaction; only the diversity of the soil fauna of the ambient control plot showed a significant positive correlation with decomposition ($P \leqslant 0.05$; $r = +0.83$).

3.6. Fine roots

At Ysselsteyn, the fine root biomass and the number of root tips showed very similar distribution patterns (Fig. 8). In all soil layers more fine roots and root tips were found in the roof clean plot. The amount of roots and root tips of vitality class 1 was three to four times higher in the roof clean plot than both control plots. In all

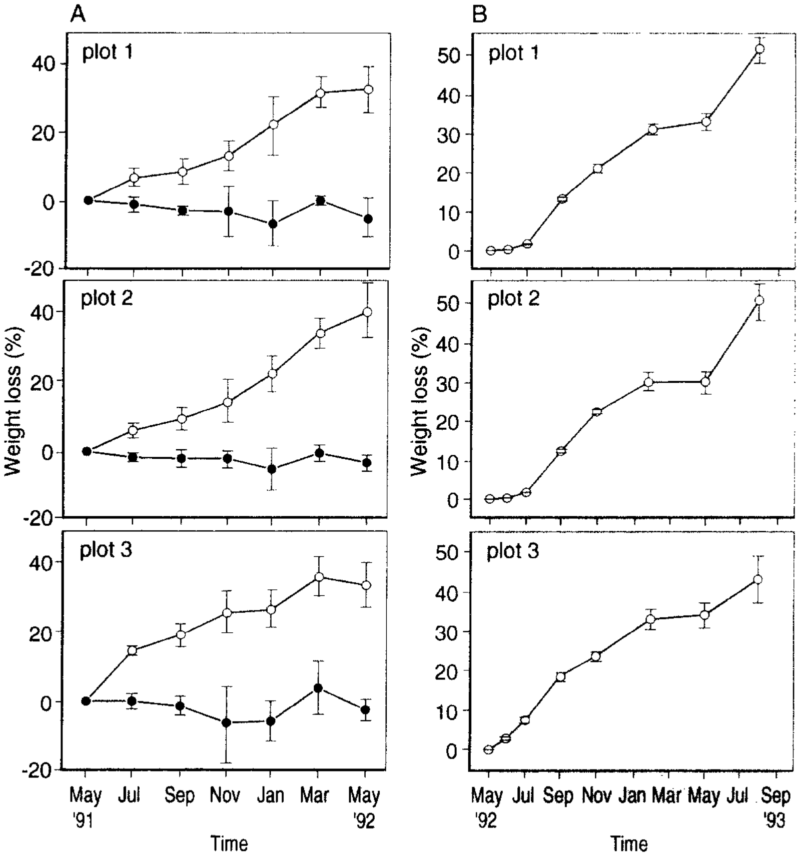


Fig. 7. Loss of dry weight of needles from the L-layer (•) and the F-layer (•) at: (A) Ysselsteyn; (B) Speuld. Bars indicate the standard deviations. Plot 1, roof clean; plot 2, roof control; plot 3, ambient control.

Table 2
Two-way analysis of variance for percent dry weight loss of L-needles in litterbags at Ysselsteyn

Source of variation	d.f.	MS[a]	*F*-ratio	Sign
Main effects				
Plot	2	832.7	27.6	0.000***
Time	5	4067.2	134.8	0.000***
Two-way interaction				
Plot × time	10	156.1	5.2	0.000***
Error	194	30.2		
Total	211	139.4		

[a] Mean squares ($n=12$).

plots most roots were of vitality class 2. The NH_4^+/NO_3^- ratio in the aqueous soil extracts was inversely correlated with the root biomass ($P\leqslant0.1$, Fig. 9) or the number of root tips ($P\leqslant0.1$, data not shown). The fine roots in the different plots did not differ in nutrient content (mean concentrations: N, 1.67%; K, 0.10%; Mg, 0.02%; P, 0.11%; Al, 0.46%; Fe, 0.21%). Only calcium was significantly higher in the roots of

Table 3
Number of species and diversity (Shannon–Weaver index; H) for Collembola and Oribatei combined, in three different treatments at Ysselsteyn ($n=8$)

	Roof clean		Roof control		Ambient control	
	Number	H	Number	H	Number	H
July 1991	32	0.879	27	0.943	33	0.669
September 1991	20	0.692	24	0.690	31	0.510
November 1991	32	0.930	33	0.517	33	0.990
January 1992	32	0.837	29	0.509	30	0.760
March 1992	38	0.754	33	0.447	32	1.028
May 1992	31	0.823	41	0.736	30	1.012

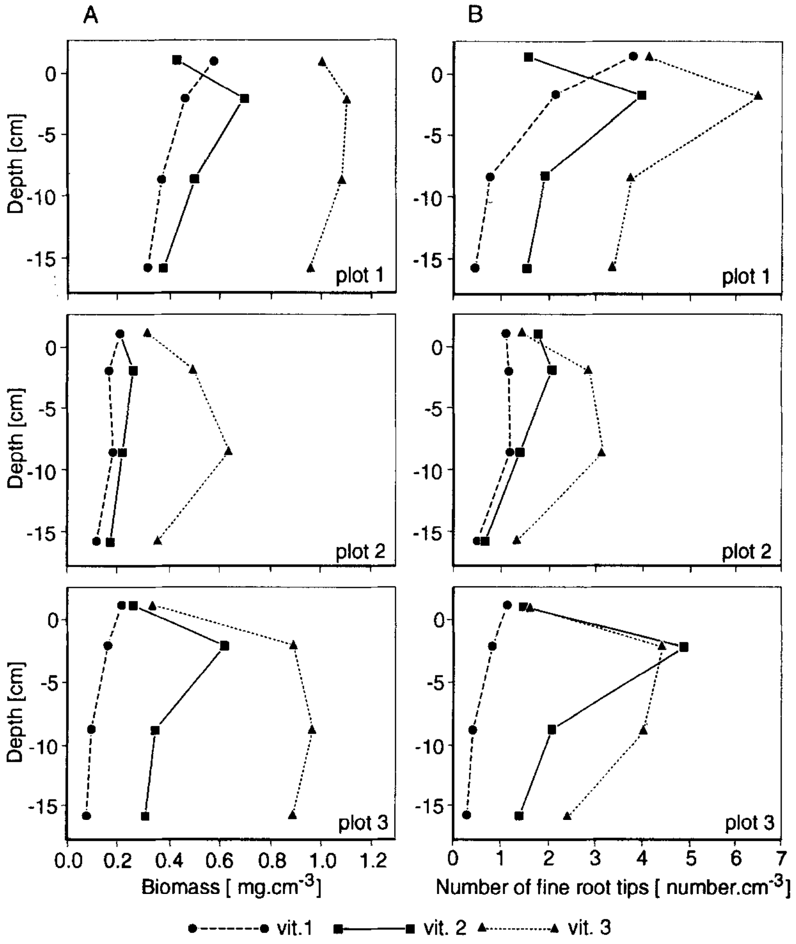


Fig. 8. Distribution of fine root biomass (A) and root tips (B) in soil cores sampled at Ysselsteyn in September 1992. Plot 1, roof clean; plot 2, roof control; plot 3, ambient control. Depth in centimetres.

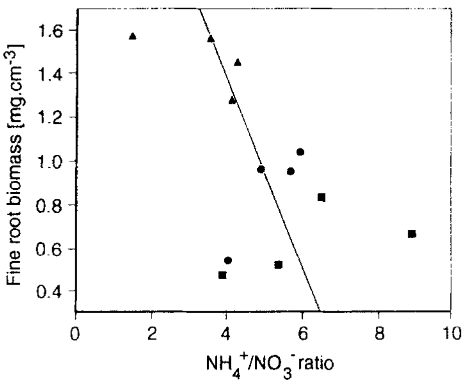


Fig. 9. Effect of varying NH_4^+/NO_3^- ratios (mol/mol) in the soil extract on the fine root biomass at Ysselsteyn. ▲, data from the roof clean plot at four depths; ■, data from the roof control plot at four depths; ●, data from the ambient control plot at four depths. $P \leq 0.1$.

the roof clean plot compared with the ambient control plot (0.15% and 0.12%, respectively).

3.7. Needles

As yet, lowering the nitrogen input in the roof clean plot at Ysselsteyn has not resulted in a significant decrease in nitrogen concentration of the 0.5-year-old needles (Table 4). In 1992, however, the N-concentration in the needles of the roof clean plot was significantly lower than in the control plots. Nitrogen concentrations in the 1.5-year-old needles were generally higher than in the 0.5-year-old needles. However, since 1992 the older needles in the roof clean plot tended to have lower N-concentrations than the current needles. In 1993, concentrations of arginine were significantly lower ($P \leq 0.05$) in the needles of the roof clean plot (81 μmol g^{-1} dry weight (DW)) than in the needles of the roof control plot (157 μmol g^{-1} DW) or the ambient control plot (128 μmol g^{-1} DW). During 4 years of reduced nitrogen and sulphur inputs the potassium and magnesium in the needles of the roof clean plot gradually increased to concentrations that were significantly higher ($P \leq 0.05$) than in the roof control plot and the ambient control plot. As a result the nutritional balance for potassium and for magnesium improved significantly.

At Speuld, lowering the nitrogen input did not significantly affect the nitrogen concentrations in the needles (Table 5). In 1993, no differences were observed between both roofed plots, but nitrogen in the needles of the ambient control plot was significantly higher. In all plots the nitrogen concentrations in the older needles were higher than in the current ones. In the potassium, magnesium and calcium concentrations no differences between the plots were induced (data not shown).

3.8. Litterfall

The amount of litterfall at Ysselsteyn was, on average, some 30% higher than at Speuld (Table 6). The amount of litterfall at Ysselsteyn was similar in all plots.

At the start of the experiment there were no differences in nutrient concentrations of the litter of the different plots at Ysselsteyn, except that magnesium was lower in both roofed plots (data not shown). During the course of the experiment, the nitrogen concentration in the litter onto the roof clean plot became significantly lower than in the two control plots. As a result the N-flux in litterfall onto the roof clean plot was on average some 10% lower. Potassium concentrations declined in the roof control plot (data not shown).

At Speuld the roof clean plot had the lowest litter inputs, which could be attributed to a blow down of a dominant tree in 1990. At the start of the experiment, calcium and phosphorus concentrations were higher in the litter onto the ambient control plot, the magnesium concentration was highest in the litter onto the roof clean plot. During the experiment, differences in magnesium and potassium between the plots disappeared, whereas calcium remained higher in the ambient control plot. The time trend for magnesium showed the same tendency in all plots: decreasing in 1990 and 1991, but increasing in 1992 (data not shown). Nitrogen remained relatively stable during the first three years, but in 1992 a tendency to lower values in the roof clean plot was observed.

Table 4
Chemical composition (% DW) and nutritional balance of the needles at Ysselsteyn. The needles were sampled in January of the reported years. Different letters in columns indicate nutrient means which are significantly different at $P \leq 0.05$ between the plots

	0.5-year-old needles				1.5-year-old needles			
	1990	1991	1992	1993	1990	1991	1992	1993
Roof clean								
%N	2.07b	2.12	2.30b	2.05	2.22b	2.35	2.26	1.91b
%K	0.51	0.54a	0.63a	0.60	0.35	0.35	0.37	0.39
%Ca	0.22a	0.20	0.19	0.24	0.36	0.35	0.25	0.26
%Mg	0.054	0.055	0.044a	0.050a	0.049	0.047	0.033	0.032
%P	0.12	0.13	0.12	0.14	0.10b	0.12	0.10	0.12b
K:N	25.1	26.3a	27.9a	29.6a				
Mg:N	2.6a	2.6a	1.9a	2.4a				
P:N	5.8a	6.0	5.1a	6.8				
Roof control								
%N	2.35b	2.47	2.55a	2.24	2.27b	2.52	2.50	2.46a
%K	0.54	0.48b	0.56b	0.52	0.37	0.36	0.35	0.40
%Ca	0.22a	0.24	0.18	0.21	0.34	0.34	0.29	0.33
%Mg	0.052	0.048	0.035b	0.037b	0.040	0.045	0.032	0.030
%P	0.12	0.13	0.12	0.14	0.11ab	0.12	0.11	0.14a
K:N	24.6	20.0b	22.1b	23.4b				
Mg:N	2.3a	2.0b	1.4b	1.6b				
P:N	5.4a	5.1	4.6b	6.3				
Ambient control								
%N	2.89a	2.32	2.64ab	2.21	2.98a	2.88	2.47	2.33a
%K	0.57	0.51ab	0.53b	0.53	0.39	0.32	0.34	0.39
%Ca	0.17b	0.18	0.19	0.20	0.26	0.30	0.24	0.27
%Mg	0.050	0.051	0.040ab	0.037b	0.043	0.050	0.036	0.034
%P	0.13	0.12	0.11	0.13	0.12a	0.12	0.10	0.13a
K:N	20.3	22.3ab	20.5b	24.9ab				
Mg:N	1.7b	2.2ab	1.5b	1.8b				
P:N	4.5b	5.3	4.3b	6.3				

3.9. Ground vegetation

Only at Ysselsteyn was an understory was present. No differences in above-ground bramble biomass (*Rubus* sp.) were found between the plots since the start of the experiment. In all plots bramble biomass decreased at the same rate (data not shown). Biomass of the ferns (*Dryopteris dilatata* (Hoffm.) A. Gray) decreased distinctly over the experimental period in all plots, but most pronounced in the roof clean plot (Table 7). No differences in nutrient composition of the above ground parts of either brambles or ferns were found between the plots (data not shown).

At Ysselsteyn, no carpophores of mycorrhizal fungi were found. Only carpophores of one litter-decaying species, *Hygrophoropsis aurantiacus* (Wulf. ex Fr.), were present. In 1989, 731 carpophores were observed in the roof clean plot, 360 in the roof control plot and 211 in the ambient control plot. In 1990, 23 carpophores were observed in the roof clean plot, 29 in the roof control plot and 9 in the ambient control plot. In 1991 and 1992 no carpophores could be col-

Table 5
Nitrogen concentrations (% DW) in the needles at Speuld. The needles were sampled in January in the reported years. Different letters in columns indicate nutrient means which are significantly different at $P \leq 0.05$ between the plots

	0.5-year-old needles			> 1.5-year-old needles		
	1991	1992	1993	1991	1992	1993
Roof clean						
%N	1.50	1.58	1.87b	1.80	1.74	2.06
Roof control						
%N	1.43	1.57	1.87b	1.66	1.76	2.16
Ambient control						
%N	1.55	1.56	2.06a	1.67	1.73	2.31

Table 6
Amount (kg ha^{-1} year^{-1}), nitrogen flux (kg ha^{-1} year^{-1}) and nitrogen concentration (% DW) of the litterfall at Ysselsteyn and Speuld

		Roof clean	Roof control	Ambient control
Ysselsteyn				
1989	Amount[a]	2612	3160	3141
	N-flux[a]	36	46	50
	%N	1.39	1.50	1.54
1990	Amount	3597	4038	4292
	N-flux	46	58	64
	%N	1.29b	1.46a	1.51a
1991	Amount	3643	3619	3453
	N-flux	43	51	49
	%N	1.21b	1.40a	1.47a
1992	Amount	3169	2738	3634
	N-flux	39	37	56
	%N	1.29b	1.39ab	1.54a
Speuld				
1989	Amount[a]	2708	2555	2080
	N-flux[a]	35	30	27
	%N	1.27	1.21	1.33
1990	Amount	2399	3396	3089
	N-flux	31	44	42
	%N	1.28	1.31	1.37
1991	AMount	2113	2756	2788
	N-flux	27	33	38
	%N	1.32	1.26	1.37
1992	Amount	1551	1856	2187
	N-fllux	20	24	33
	%N	1.22b	1.32ab	1.45a

[a] Expressed in kg ha^{-1} per 192 day period.

Rows with different letters indicate means which are significantly different at $P \leq 0.05$.

Table 7
Remaining percentage of aboveground biomass of ferns (*Dryopteris dilatata*) from 1990 to 1992 at Ysselsteyn (1990 = 100%)

	Roof clean	Roof control	Ambient control
1990	100 (26.4 g m^{-2})	100 (33.8 g m^{-2})	100 (20.3 g m^{-2})
1991	44	78	83
1992	37	64	84

Table 8
Collected fruitbodies of fungi in 1989 and 1992 at Speuld (number per 100 m^{-2}). Fruitbodies growing on dead wood were omitted

	Roof clean		Roof control		Ambient control	
	1989	1992	1989	1992	1989	1992
Total number of fruitbodies	135	394	85	319	97	185
Mycorrhizal species	44	323	19	281	2	128
Laccaria cf. *proxima*	2	–	–	–	–	–
Lactarius necator	–	27	–	2	–	–
Lactarius theiogalus	25	132	3	164	–	25
Russula ochroleuca	16	148	16	106	1	102
Xerocomus badius	1	6	–	6	1	1
Xerocomus chrysenteron	–	5	–	–	–	–
Xerocomus subtomentosus	–	5	–	3	–	–

lected. At Speuld carpophores of seven species of mycorrhizal fungi were found. The roof clean plot was the most productive, both in 1989 and 1992, whereas the ambient control plot was the least productive (Table 8).

4. Discussion

The atmospheric input of nitrogen and sulphur to the clean plots at both locations was reduced to approximately 1-2 kg ha^{-1} $year^{-1}$. The deposition of nitrogen onto the roof control plot was somewhat lower than onto the ambient control plot. This can be attributed to (i) water loss during heavy showers due to an insufficient transport capacity of the gutter system and (ii) loss due to denitrification and/or binding to litter debris in the storage containers. The raised pH of the water in the containers pointed in this direction (Van Dijk et al., 1992a,b). Deposition of nitrogen onto the ambient plots was high, particularly in comparison with a critical load value of 15–20 kg N ha^{-1} $year^{-1}$ (Bobbink et al., 1992).

4.1. Nutrient fluxes in the soil

The inorganic nitrogen and sulphur concentrations in the soil solution of the roof clean plot at Ysselsteyn responded very quickly to the reduction of the atmospheric nitrogen and sulphur inputs. Unfortunately, no data on soil solution chemistry during the pre-treatment period and the first weeks after the start of the treatment are available. In the deeper soil layers, where the response to the treatment was slower, both the nitrate and the sulphate concentrations in the roof clean plot diverged from those in the roof control plot. Soil extracts made before the start of the experiment revealed no significant differences between the plots in availability of ammonium, nitrate or sulphur (data not shown). The results of a similar experiment at the Solling NITREX site (Germany), also showed a rapid decrease in nitrogen and sulphur concentrations in the soil solution upon decreased deposition (Bredemeier et al., 1995). Nevertheless, the nitrogen flux out of the 10 cm soil layer in the roof clean plot at Ysselsteyn was almost ten times higher than the input flux to the forest floor via throughfall water. This nitrogen must have been produced by mineralisation of organic nitrogen. Because of this mineralisation, differences between the three plots in nitrogen flux at 10 cm depth were much smaller than differences in the nitrogen flux via throughfall water. The shift in dominance of ammonium over nitrate in throughfall water to nitrate dominance in the soil solution must be attributed to adsorption and/or nitrification of ammonium, or possibly preferential uptake of ammonium by plant roots (Arnold, 1992). The nitrogen flux was further reduced in deeper soil layers, and only 0.8 $\mathrm{kmol_c}$ (12 kg) N $\mathrm{ha^{-1}}$ $\mathrm{year^{-1}}$ left the rooting zone. As input via throughfall water amounted to 2 kg N, the net nitrogen loss from the ecosystem in the roof clean plot was approximately 10 kg N $\mathrm{ha^{-1}}$ $\mathrm{year^{-1}}$ during the last 3 years. However, unless losses by denitrification and leaching of organic nitrogen were considerable, the real net nitrogen loss must have been smaller, because of nitrogen uptake by aerial tree parts, which was not prevented by the present experimental setup (Pérez Soba, 1990; Eilers et al., 1992). Assuming uptake of nitrogen in the canopy it is unlikely there was any net loss of nitrogen in the roof clean plot. The much higher nitrogen input via throughfall water in the control plots was matched by an equally higher nitrogen efflux from the rooting zone. Apparently, reducing the nitrogen input via throughfall water in the roof clean plot at Ysselsteyn, resulted only in a reduction of the nitrogen flow through the ecosystem, but not in a reduction of the total nitrogen pool within the ecosystem. These forest ecosystems are very effective in retaining nitrogen. However, the amount of readily available inorganic nitrogen has drastically been reduced and in that respect the system has become unsaturated.

The sulphur flux in the roof clean plot at Ysselsteyn was also much higher in the soil than in throughfall water. Sulphur in the soil was released by processes such as mineralisation and desorption. Net sulphur losses from the plots were somewhat larger than net nitrogen losses and amounted to 9–15 kg S $\mathrm{ha^{-1}}$ $\mathrm{year^{-1}}$. They were rather similar in all plots in spite of the higher sulphur fluxes in the control plots.

The higher fluxes of chloride in the roof clean plot at Ysselsteyn were caused by a technical error. In May 1991, the pump that adds a concentrated salt solution to the demineralised water to change it into simulated, clean rainwater, came into operation while there was no supply of demineralised water. The concentrated salt solution dripped into the roof clean plot, resulting in a chloride addition. Potassium, magnesium and calcium were added simultaneously. These additions are not included in the throughfall fluxes shown in Fig. 2(a), as the amounts deposited are unknown and the distribution over the plot was uneven. This addition may explain why the fluxes of these cations in the upper soil layer were also higher in the roof clean plot than in the roof control plot. In deeper soil layers the fluxes of cations, particularly those of calcium and aluminium, were lowest in the roof clean plot, probably as a result of the reduced fluxes of the associated anions, nitrate and sulphate. This depression of cation leaching by the reduction in nitrogen and sulphur deposition was partly masked by the inadvertent addition of chloride. Differences in composition of the soil solution between both control plots were considerable. They were probably caused by the method of watering the trees underneath the roof until 1992 (see Van Dijk et al., 1992a,b). After December 1991, when the sprinkling system was automated, differences between the two polluted plots gradually became smaller.

Both the roof clean plot and the roof control plot at the Speuld location received more nitrogen from throughfall water than they lost by leaching from the rooting zone. The roof clean plot retained approximately 2 kg N $\mathrm{ha^{-1}}$ $\mathrm{year^{-1}}$ and the roof control plot about 30 kg N $\mathrm{ha^{-1}}$ $\mathrm{year^{-1}}$. Also other ions were leached only in very small quantities. The striking decrease in ion fluxes with increasing soil depth in both plots underneath the roof, was the result of the very low amounts of precipitation. As the trees took up most of the water in the upper soil layers, almost no water and consequently no ions leached from the rooting zone. As in Ysselsteyn, the reduced cation fluxes in the roof clean plot compared with the roof control plot were probably caused by the reduced nitrate and sulphate fluxes through the soil profile.

The much higher ion fluxes through the soil profile in the ambient control plot were the result of both a higher water flux and higher ion concentrations in the soil solution. The latter was most likely due to the method of watering the plots underneath the roof before the automation of the sprinkling system in the summer of 1992. In the ambient control plot nitrogen leaching from the rooting zone exceeded input via throughfall water by 40 kg N ha^{-1} $year^{-1}$. This is more than at Ysselsteyn in spite of the somewhat lower atmospheric nitrogen input.

4.2. Decomposition

When comparing both roofed plots a positive effect of nitrogen deposition on decomposition rate at Ysselsteyn was observed, at least in the early stage (L-layer). In contrast, no differences between plots were found at Speuld. At Ysselsteyn, there was no effect of nitrogen deposition on decomposition rate of the F-layer material. F-material did not decompose at a measurable rate in any of the three plots. Low decomposition rates may have been due to influx of material from the L-layer. The very slow rate of decomposition of F-material compared with observations of Berg et al. (1987), may indicate that the substrate quality is affected by nitrogen. Fog (1988) concluded that nitrogen is often beneficial to decomposition when it is already in surplus. Kuyper (1989) reviewed the effects of atmospheric deposition on decomposition processes in forests and concluded that in the first stage of the process nitrogen will speed up decomposition but in the second stage decomposition will be inhibited. The significantly different patterns of the decomposition process between both control plots indicate a 'roof effect', caused by the roof itself or by the watering procedure.

4.3. Soil fauna

The reduction of atmospheric nitrogen and sulphur deposition increased the species diversity of microarthropods, owing to a decreased dominance of some species (*Isotoma notabilis, Oribatula tibialis*) at a constant species number. This is apparently the first report of effects on soil fauna of reduced acidification.

The relation between decomposition of litter and soil fauna diversity was inconsistent. At the ambient control plot the soil fauna diversity was positively correlated with decomposition. This is in contrast to a 3 year study in which fauna diversity was stable while decomposition increased (Takeda, 1988). In general, acid deposition causes species numbers and species diversity of soil fauna to decrease owing to the increase of a few dominant species (Schaefer, 1990). Acid deposition may affect competition between soil animals, and thereby regulate the structure of soil animal communities (Hågvar, 1990). A reduction of atmospheric input may restore competition, decrease the dominance of species, and increase species diversity. However, not all species that disappeared may return, possibly because the substrate quality has not changed or because levels of nitrogen in the substrate are still high, so that the species composition may retain an acidophilic character. Biologically, soils seem to be able to recover from acidification but this is a slow process and the degree of recovery is probably incomplete.

4.4. Roots

The root distribution pattern found at Ysselsteyn is consistent with that at the NITREX site at Gårdsjön, Sweden (Clemensson-Lindell and Persson, 1995). Vital roots were only found in small amounts and predominantly in the clean plot. Fine roots of vitality class 2 dominated. At Gårdsjön most roots were found in the humus layer, whereas at Ysselsteyn most roots were present in the upper mineral soil layer. In Gårdsjön two to three times more roots were found (Clemensson-Lindell and Persson, 1993). Lowering the N-input seems to have stimulated root growth in the roof clean plot. The increased number of fine root tips in the roof clean plot may indicate a better nutrient uptake, as indicated by the slightly increased concentrations of potassium and magnesium in the needles.

An increased N-input often results in a deteriorating root system (Encke, 1986; Gezelius, 1986; Van Dijk et al., 1990; Boxman et al., 1991). Not only does the absolute amount of N-input seem to be important, but also the NH_4^+/NO_3^- ratio in the soil water (Boxman et al., 1991). No significant differences in nutrient concentrations of the roots between the plots were found. Enhanced uptake of nutrients may lead to enhanced transport to the above ground parts of the trees. Hydroculture experiments with *Pinus nigra* have shown that upon ammonium addition differences in nutrient status are more pronounced in the needles than in the root system (Boxman et al., 1991). When the nutrient status is compared with fine roots of *Pinus sylvestris* from clean pristine areas (Helmisaari, 1991), the nitrogen concentration is approximately four times higher while the potassium and magnesium concentrations were two to three times lower, indicating an unfavourable nutrient balance for both elements. The phosphorus and calcium concentrations were approximately equal.

4.5. Needles

Before the start of the experiment, in 1989, no significant differences were found in needle nutrient contents between the plots at either location (Van Dijk et al., 1992a,b). At Ysselsteyn the needles had high nitrogen concentrations, whereas the potassium, magnesium and phosphorus concentrations were very low (Anonymous, 1990). During the 4 years of treatment that nutrient concentrations in the needles varied considerably, probably related to meteorological conditions (Hütll, 1989; Helmisaari, 1990; Katzensteiner et al., 1992). As yet, the nitrogen concentration in the current needles of the clean plot at Ysselsteyn have not responded to a reduction in nitrogen inputs. Nitrogen concentrations above 2% are considered very high (optimal level approximately 1.4–1.8%; Anonymous, 1990). This may be caused by nitrogen uptake by the canopy (Lindberg et al., 1986; Péres Soba, 1990; Eilers et al., 1992) and by the fact that the amount of nitrogen present in the ecosystem is still high. Nevertheless, N was lower in the needles of the roof clean plot than control plots.

The significant decrease in arginine level, as a sink for excess nitrogen, corresponds to a difference in nitrogen concentration of 0.4%. Since the total nitrogen concentration in the 0.5-year-old needles was only 0.2% lower this means that the arginine pool decreased more rapidly than the total N-concentration, possibly indicating enhanced protein synthesis in the roof clean plot. Here the nitrogen concentration of the 1.5-year-old needles was lower than that of the 0.5-year-old needles, the normal situation in nitrogen-limited ecosystems. This might indicate reallocation and is consistent with the decreased nitrogen flux in litter. The potassium and magnesium concentrations increased significantly in the roof clean plot compared with the other plots. This may be related to a better root quality, a favourable NH_4^+/NO_3^- ratio and to decreased cation leaching. For calcium and phosphorus no differences between the plots were found. In fertilisation trials with K, Mg, Ca, and P to restore the nutrient balance in the soil the fastest responses were found for potassium and magnesium (Boxman et al., 1994a). This means that the nutritional balance of potassium and magnesium relative to nitrogen has improved, the former even to a level above that considered deficient (Anonymous, 1990).

Although the ecosystem at Speuld is nitrogen-saturated the firs have normal nitrogen concentrations in their needles (Anonymous, 1990). The trees are growing reasonably well (Evers and Steingröver, 1991) which suggests a dilution effect on nitrogen in the needles. The supply of the other nutrients from the soil to the trees is apparently sufficient, except for phosphorus, which is too low. When nitrogen limitation ceases the phosphorus supply often becomes limiting (Mohren et al., 1988; Houdijk and Roelofs, 1993). The nitrogen saturation effect is most pronounced in the older needles, which had higher nitrogen concentrations than the current ones. In 1993 nitrogen in the needles of the ambient control plot increased, which is consistent with the observations of E.G. Steingröver (personal communication, 1993), who also has recorded a gradual increase over the last few years. That nitrogen did not increase in the roofed control may be due to the drought in this plot, although this is not the case for potassium, magnesium and calcium uptake, which are also sensitive to drought stress (Schulze et al., 1989).

4.6. Ground vegetation

The decrease in biomass at Ysselsteyn can be attributed to the harvesting of the standing crop of the understory every autumn. However, the fern biomass in the roof clean plot decreased more than in both control plots, and this may be related to a decreased nitrogen concentration or a changed NH_4^+/NO_3^- ratio in the soil solution. This is consistent with the reverse situation: on increasing the N-input the ground vegetation is changed into a nitrophilous one, while most mycorrhizal carpophores disappear (Bobbink et al., 1992). The clean plots at both locations appear to be the most productive with respect to carpophore production, but a longer time series is needed to confirm this observation.

4.7. Ecosystem response

When the nitrogen and sulphur inputs into nitrogen and sulphur saturated forest ecosystems are reduced, the nutrient concentrations in the soil solution and the output fluxes decrease rapidly. In contrast, tree response, as measured by the nutrient concentrations in the needles, is delayed by at least several years. The ecosystems are recovering from excess nitrogen availability: in the roof clean plots the species diversity of microarthropods has increased, although the relation to decomposition is not yet clear. The nutrient balance in the needles has improved, which may be related to an enhanced root quality, a favourable NH_4^+/NO_3^- ratio in the soil solution and a decreased leaching of base cations. The nitrogen concentrations in older needles are lower than in current needles, which is normal in nitrogen-limited coniferous ecosystems. This points to reallocation of nitrogen from the older to younger needles and is consistent with decreasing arginine concentrations in the needles and a deceased nitrogen flux via the litter.

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