Biogeochemistry **38:** 255–280, 1997.

The effects of ammonium sulphate deposition and root sinks on soil solution chemistry in coniferous forest soils

M. CARNOL[1,2], P. INESON[2], J.M. ANDERSON[3], F. BEESE[4],
M.P. BERG[5], T. BOLGER[6], M.-M. COÛTEAUX[7], P. CUDLIN[8],
S. DOLAN[6], M. RAUBUCH[4] & H.A. VERHOEF[5]
[1]*University of Liège, Dept. Botany B22, Microbial Ecology, 4000 Liège, Belgium;* [2]*Institute of Terrestrial Ecology, Merlewood Research Station, Grange-over-Sands, Cumbria LA11 6JU, U.K.;* [3]*Dept. of Biological Sciences, Hatherly Laboratories, University of Exeter, Prince of Wales Road, Exeter EX4 4PS, U.K.;* [4]*Institute of Soil Science and Forest Nutrition, University Göttingen, Büsgenweg 2, 37077 Göttingen, Germany;* [5]*Dept. of Ecology and Ecotoxicology, Section Soil Ecology, Vrije Universiteit Amsterdam, De Boelelaan 1087, 1081 HV Amsterdam, The Netherlands;* [6]*Dept. of Zoology, University College, Dublin 4, Ireland;* [7]*CNRS, Route de Mende, BP 5051, 34033 Montpellier, Cedex, France;* [8]*Institute of Landscape Ecology, Academy of Sciences of the Czech Republic, Na sádkách 7, 37007 Ceské Budejovice, Czech Republic*

Accepted 21 January 1997

Key words: aluminium, forest soils, lysimeters, nitrification, nitrogen, roots

Abstract. The effects of enhanced $(NH_4)_2SO_4$ deposition on soil solution cation and anion concentrations and annual ionic fluxes were followed using a standardised experimental protocol in six European coniferous forests with contrasting soil types, pollution inputs and climate. Native soil cores containing a ceramic suction cup were installed in the field, roofed and watered every two weeks with local throughfall or local throughfall with added $(NH_4)_2SO_4$ at 75 kg NH_4^+-N ha^{-1} a^{-1}. Living root systems were established in half of the lysimeters. Untreated throughfall NH_4^+-N deposition at the sites ranged from 3.7 to 29 kg ha^{-1} a^{-1}. Soil leachates were collected at two weekly intervals over 12 months and analysed for volume, and concentrations of major anions and cations. Increases in soil solution NO_3^- concentrations in response to N additions were observed after 4–9 months at three sites, whilst one sandy soil with high C:N ratio failed to nitrify under any of the treatments. Changes in NO_3^- concentrations in soil solution controlled soil solution cation concentrations in the five nitrifying soils, with Al^{3+} being the dominant cation in the more acid soils with low base saturation. The acidification responses of the soils to the $(NH_4)_2SO_4$ additions were primarily related to the ability of the soils to nitrify the added NH_4^+. pH and soil texture seemed important in controlling NH_4^+ leaching in response to the treatments, with two less acidic, clay/clay loam sites showing almost total retention of added NH_4^+, whilst nearly 75% of the added N was leached as NH_4^+ at the acid sandy soils. The presence of living roots significantly reduced soil solution NO_3^- and associated cation concentrations at two of the six sites. The very different responses of the six soils to increased $(NH_4)_2SO_4$ deposition emphasise that the establishment of N critical loads for forest soils need to allow for differences in N storage capacity and nitrification potential.

Introduction

With the exception of ecosystems with high N fixation, N has been a major limiting factor in the productivity of forest ecosystems in Europe. However, due to changing agricultural practices and increased usage of fossil fuels over recent decades, this situation has changed substantially; the N load to large areas of forests in Europe now exceeds the annual growth increment of trees, indicating that excess soil N accumulation may be taking place (Agren & Bosatta 1988). In contrast to the decreasing inputs of S to forest ecosystems, N deposition does not seem to be declining (Grennfelt & Hultberg 1986) and this elevated N deposition has been related to the forest dieback observed in Europe (Nihlgard 1985). High NH_4^+ deposition may enhance nitrification, leading to soil acidification, NO_3^- and cation leaching, Al^{3+} mobilisation and root damage (e.g. Gundersen & Rasmussen 1990). Nutrient imbalances due to an increased NH_4^+/cation ratio have also been related to forest decline (Roelofs et al. 1985).

The concept of a 'critical load', 'the maximum deposition of a given compound which will not cause long-term harmful effects on ecosystem structure and function according to present knowledge' (Hornung & Skeffington 1993; Nilsson & Grennfelt 1988), has been developed as a tool in European pollution control policy (De Vriess et al. 1992). This approach takes into account the ecological effects of pollutants (Skeffington 1993), hence the importance of improving our understanding of processes, e.g. soil processes in relation to forest vitality, on a wider scale. The responses of similar forest ecosystems to increased NH_4^+ deposition can be very different (Feger 1992) and our understanding of the underlying processes are poor. Methodological differences frequently impede comparisons of studies across sites. Additionally, studies using lysimeter or incubation approaches usually exclude living tree roots, and may effectively simulate N-saturated systems (sensu Aber et al. 1989), where 'availability of inorganic N is in excess of total combined plant and microbial nutritional demand'. However, soil type, pollution inputs and climate may be important factors in determining the effect of N deposition on a particular ecosystem and data on these interactions are needed in order to understand regulating processes and to reach generally applicable conclusions.

In this study we examined the effects of enhanced $(NH_4)_2SO_4$ deposition on soil solution chemistry in the presence of living tree roots by following an identical experimental protocol in six European coniferous forest soils situated across a pollution gradient. This study therefore represents a basis for comparing the effects of increased N deposition on sites with different soil types and pollution histories, concepts central to the setting of critical N loads at regional or ecosystem scales.

Methods

Site description and experimental design

Six forest sites, located in The Netherlands (Wekerom), Germany (Solling), United Kingdom (Haldon and Grizedale), France (Fontainebleau) and Ireland (Clonegal) were selected across a European pollution transect of N and S (Figure 1). Five of these sites were used in previous CORE studies (Core et al. 1992). However, the site in Ireland (Kilkenny) was substituted because of altered management, and Clonegal was chosen for its similarity to the Kilkenny site. Main site characteristics are given in Table 1, and detailed site characteristics can be found in Raubuch (1992) and Coûteaux & Sallih (1994). Two sites were established on acid, sandy soils in Pinus sylvestris stands (Fontainebleau and Wekerom) with low and high N deposition. Two Picea abies plots (Haldon, Clonegal) were on less acidified soils ($pH > 4.2$) with past N deposition of 9.6 and 17.3 kg ha^{-1} a^{-1}. Grizedale and Solling were relatively acid soils with past N depositions of 13.3 and 37.1 kg ha^{-1} a^{-1}.

Soil cores were taken at each site using Plexiglas cylinders (14.4 cm inner diam $*$ 24 cm deep) fitted onto a metal corer, which were forced into the ground, using the CORE protocol described by Raubuch (1992). After checking that the soil cores were undamaged, the bases were trimmed, a 5 cm depth of acid washed sand was placed at the bottom of each core and a cap was fitted and sealed with silicone adhesive. A ceramic cup (P80, Fa. KPM Berlin) was inserted diagonally into the sand and connected to a 2-liter glass bottle to which a suction of 0.040–0.065 MPa was applied (Figure 2). The whole experimental unit (soil core and ceramic cup) is referred to as a lysimeter.

Twenty-eight lysimeters were installed in a randomised block design at each of the six sites, using soil cores derived from that site. The experimental design and protocols were identical at each site, but the approach differed from that of CORE et al. (1992), since lysimeters were not transplanted between sites and because lysimeters were roofed to enable control of throughfall inputs (Figure 2). Each site used therefore ‘its own’ soil only. The following four treatments were applied with 7 replicate lysimeters per treatment: control (C), $(NH_4)_2SO_4$ additions (+N), with roots (+R), with roots and $(NH_4)_2SO_4$ additions (+R+N).

One half of the lysimeters contained living roots, which was achieved by inserting 3 to 4 small living tree rooting systems into each lysimeter, after Anderson et al. (1990). This process involved deciding the approximate position of the lysimeter, and then carefully ’teasing’ out small (< 4 mm diameter), live mycorrhizal root branches from the forest floor. These root

Table 1. Characteristics of the CORE sites. Soil pH and C:N given for 0–5 cm of the mineral soil horizons. Buffer ranges given according to Ulrich (1981): cation exchange (pH 4.2–5.0), aluminium (pH 2.8–4.2). CEC (total cation exchange capacity) and texture after Coûteaux and Sallih (1994). Past N input: (NH_4^+-N + NO_3^--N) March 1989–90, from Raubuch (1992). ∗Data from similar nearby site; nd: not determined.

		Clonegal (IRL)	Fontainebleau (F)	Grizedale (UK)	Haldon (UK)	Solling (D)	Wekerom (NL)
Stand		Picea abies	Pinus sylvestris	Picea abies	Picea abies	Picea abies	Pinus sylvestris
Texture		clay/silty clay∗	loamy sand	clay	clay loam	silty clay loam	sand
Soil Type		brown soil	podsol soil	brown soil	brown soil	acid brown soil	podsol soil
CEC (cmol kg^{-1})	1–5 cm	34.2∗	nd	67.3	37.8	78.0	34.7
	5–10 cm	19.2∗	nd	99.2	17.4	26.7	8.9
Base saturation (%)	1–5 cm	23.8∗	nd	16.2	22.9	5.9	7.7
	5–10 cm	16.8∗	nd	6.0	16.3	19.7	6.3
Buffer range		cation exchange	aluminium	aluminium	cation exchange	aluminium	aluminium
pH_{H2O}		4.2∗	3.9	3.7	4.3	3.3	3.9
C:N		14.5	27.0	15.8	13.8	17.0	22.0
Past N input (kg ha^{-1} a^{-1})		9.6∗	10.5	13.3	17.3	37.1	52.1

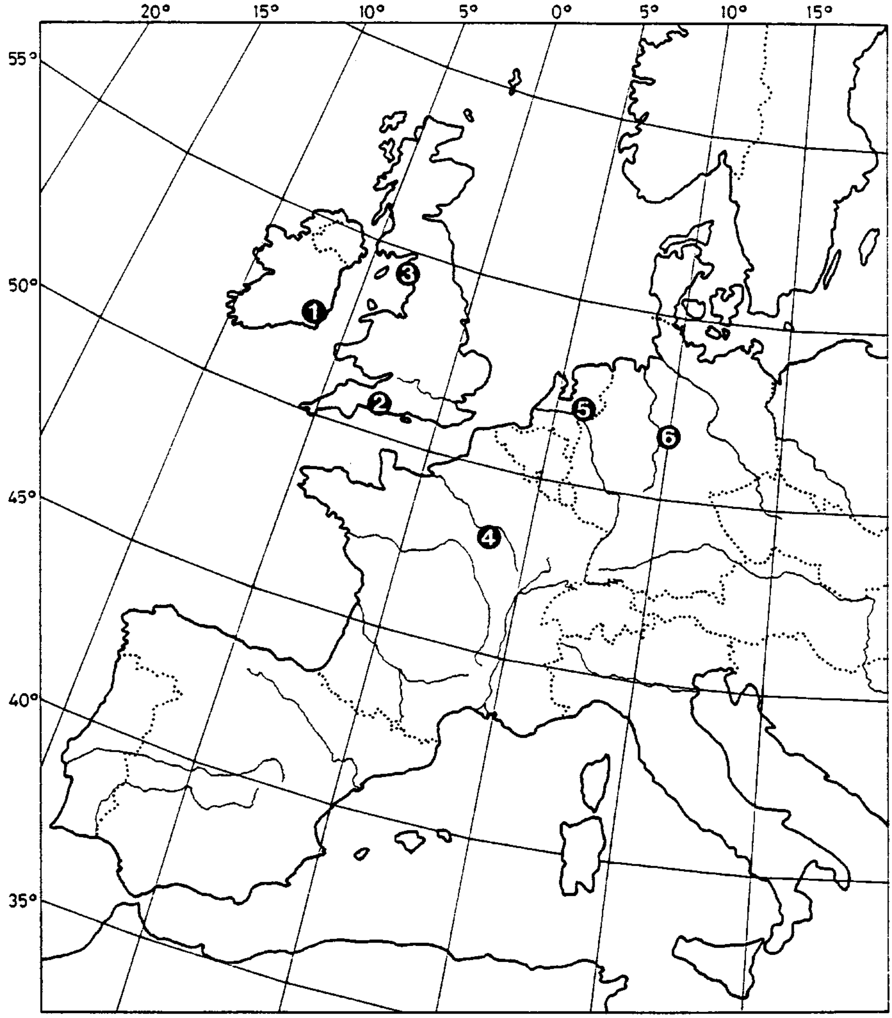

Figure 1. The CORE sites: 1 Clonegal, 2 Haldon, 3 Grizedale, 4 Fontainebleau, 5 Wekerom, 6 Solling.

systems were then temporarily protected using moist tissue whilst a hole, suitable for accommodating a lysimeter, was dug. The roots were then inserted, unbroken and undamaged, through 1–2 holes in the side of the lysimeter, into the organic horizon within each lysimeter. In order to enable root systems to be placed into the organic horizons, the top organic layer of the soil was temporarily lifted from the soil, and the roots were placed onto the remaining organic layer on the exposed organic soil surface. The top organic layer was then replaced over the intact root system and the holes in the side of

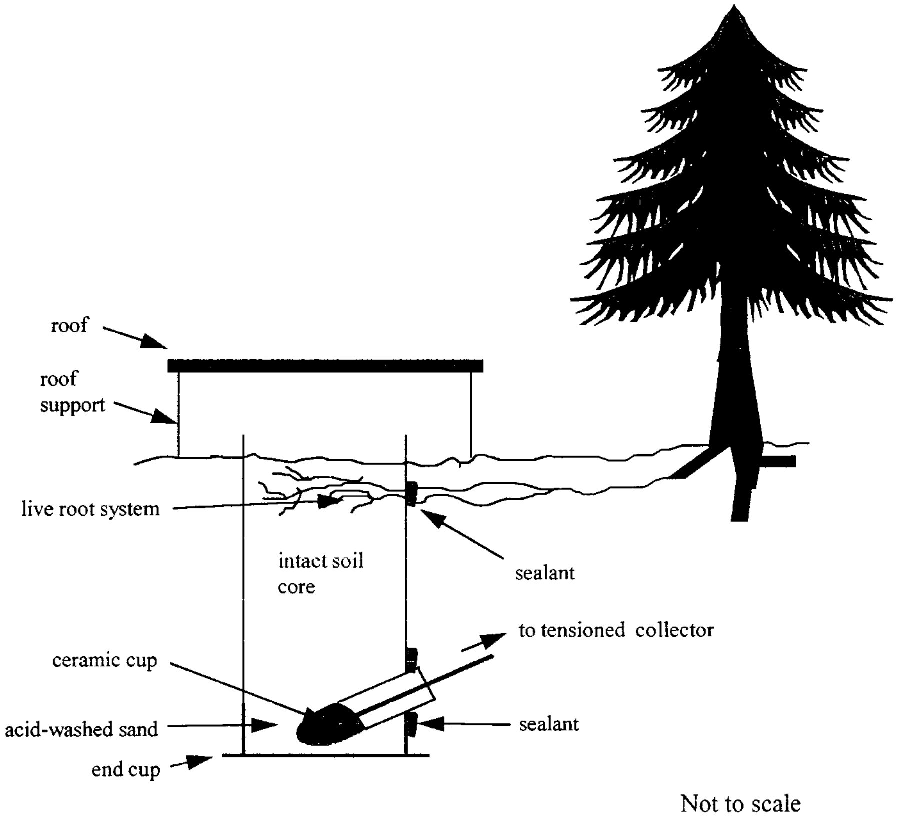

Figure 2. The CORE lysimeter design, showing a rooted lysimeter.

the lysimeter were sealed with silicone adhesive. Control lysimeters received the same treatment, but without roots being implanted. All lysimeters then received an initial watering in order to aid root establishment. In this way, root sinks which were attached to the adjacent mature trees were established in the '+R' treatments.

Throughfall applications (throughfall + $(NH_4)_2SO_4$ at 75 kg NH_4^+-N ha^{-1} a^{-1} for the '+N' treatments) were made to each lysimeter at two-week intervals, with volumes representing the particular throughfall volume of each individual site, collected and measured for this period. All lysimeters at the same site therefore received identical throughfall volumes, but throughfall volumes and compositions differed between sites. The $(NH_4)_2SO_4$ additions were made from 'stock' solutions, mixed with the throughfall in the field, and applied evenly over the year (2.88 kg N ha^{-1} added to the throughfall every two weeks). The resulting total NH_4^+-N and SO_4^{2-}-S inputs to the lysimeters are summarised in Table 2. No fresh litter was added to the lysimeters during

Table 2. Mean total NH_4^+-N NO_3^--N and SO_4^{2-}-S (kg ha^{-1} a^{-1}) inputs, outputs and percentage of input (%) leached from control lysimeters and lysimeters receiving high $(NH_4)_2SO_4$ deposition (+N) from October 1992 to October 1993. *29 September 1992-19 January 1993; #6 October 1992–1 June 1993.

			Clonegal	Fontainebleau#	Grizedale	Haldon*	Solling	Wekerom
Control	NH_4^+-N	input	11.5	2.4	8.5	2.7	12.3	29.0
		output	1.1	14.4	2.3	0.3	2.5	29.4
		%	9.6	600.0	27.1	11.1	20.3	101.4
	NO_3^--N	input	4.2	1.8	6.5	1.1	18.2	8.9
		output	94.7	1.7	77.0	48.7	92.6	98.6
		%	2254.8	94.4	1184.6	4427.3	508.8	1107.9
	SO_4^{2-}-S	input	13.6	5.8	34.3	5.6	37.4	15.3
		output	21.3	8.1	38.9	9.2	32.6	38.1
		%	156.3	139.7	113.4	164.3	87.2	249.0
+N	NH_4^+-N	input	84.0	42.1	79.3	21.5	71.0	92.6
		output	5.5	27.6	22.5	1.3	16.7	69.0
		%	6.5	65.6	28.4	6.2	23.6	74.5
	NO_3^--N	input	8.1	1.5	10.7	1.0	14.9	12.9
		output	102.6	3.8	113.4	64.1	95.0	90.0
		%	1266.7	253.3	1059.8	6410.0	637.6	697.7
	SO_4^{2-}-S	input	125.0	61.4	117.5	26.7	101.9	60.2
		output	50.3	25.1	106.9	18.3	61.7	76.2
		%	40.3	40.9	91.0	68.4	60.6	126.7

the experimental period. The leachates were collected every two weeks and analysed for volume, pH, major cations and anions.

All lysimeters were destructively sampled at the end of the experiment, and root condition and mass determined. The health of roots was evaluated using the methodology described in Ruess et al. (1996), in which roots were separated from the soil by washing over a 0.5 mm sieve and quantified using a counting grid. Root tips were classified as turgid (young tip assumed to be absorptively functioning), shrivelled (older tips; with apparently live and functioning vascular tissue but assumed to be not functioning absorptively) or dead (see Ruess et al. 1996, and references therein). Lysimeters were first established in the field in Spring 1992, but the roots failed to survive at most sites and had to be re-established in Autumn 1992. $(NH_4)_2SO_4$ additions started in September 1992 at all sites and results are reported for one year following the start of the $(NH_4)_2SO_4$ additions.

Chemical analyses

Soil and throughfall solutions were analysed for pH, and major cations (Na^+, K^+, Ca^{2+}, Mg^{2+}, Al^{3+}, NH_4^+-N) and anions (NO_3^--N, Cl^-, SO_4^{2-}-S). Methods used for chemical analyses at each laboratory were subjected to a calibration check, and included atomic absorption spectrophotometry, flame emission spectrophotometry, inductively-coupled plasma spectrometry, colorimetric methods and ion chromatography (Dionex). Soil pH was measured in water (1:2.5 fresh soil:water) after two hours of shaking. C and N were measured using a Carlo-Erba C:N analyser. Texture, CEC and base saturation have been determined by Coûteaux & Sallih (1994).

Statistical analyses

Output concentrations and volume data were used to calculate 2-weekly ionic fluxes for each lysimeter, and the resulting values were totalled to provide an annual flux for each ion and lysimeter. These yearly ionic fluxes were analysed for significant differences between treatments using analysis of variance (one-way ANOVA) and Tukey's studentized range test (SAS 1989), and means of replicate lysimeters are shown in the Figures. The relationships between concentrations of ions in soil solution were investigated using Pearson's correlation coefficient.

Soil solution concentrations are reported as arithmetic means of replicate lysimeters and comparisons of ionic concentrations between treatments over time were made using a two-way ANOVA with repeated measures on one factor (time), adjusting the error terms for mixed designs (SAS 1989; Cody & Smith 1991). Since interaction terms were significant, treatment effects were analysed at individual times, as recommended by Hatcher & Stepansky (1994), using one-way ANOVA and Tukey's studentized range test (HSD).

Annual hydrological budgets were used to assess the hydrological integrity of each lysimeter and blocked or leaking lysimeters (output volume $<50\%$ input volume) were discarded from all subsequent analyses. Lysimeters with root implants in which the roots were not viable at the end of the experiment were also discarded. The resulting number of replicates per treatment therefore varied between 4 and 7.

Results

Soil solution concentrations

Ammonium

Under control conditions, NH_4^+ concentrations in leachates were around 0.5 meq l^{-1} for Wekerom and Fontainebleau, and below 0.1 meq l^{-1} at the other four sites (Figure 3). These values were in the same range as input concentrations at Wekerom, about half the input concentrations at Grizedale and Solling, and around ten times lower at Clonegal and Haldon. Only at Fontainebleau were leachates 8 times more concentrated than inputs. N treatments resulted in a similar input concentration range at the six sites, with the lowest annual mean input concentrations at Grizedale (0.72 ± 0.38 meq l^{-1}) and highest mean annual concentrations at Clonegal (1.90 ± 1.23 meq l^{-1}).

There were no consistent significant effects of the roots on leachate NH_4^+ concentrations in the +R and +R+N treatments across most sites. Only the Solling soil showed reduced NH_4^+ concentrations in rooted lysimeters (+R+N) compared to +N, significant ($p < 0.05$) for three sampling dates.

Responses of leachate NH_4^+ concentrations to increased deposition differed markedly in the six soils. Whereas all soils exhibited significantly increased NH_4^+ concentrations under +N treatments (Table 3), the extent of the increase was very different between soils. Despite the high input concentrations of NH_4^+ at Clonegal, soil solution concentrations remained low under all treatments, and the N addition treatment was reflected in significantly ($p < 0.05$) increased NH_4^+ concentrations during the winter months only (Figure 3). The Haldon soil (input concentrations 1.45 ± 1.03 meq l^{-1}) also showed NH_4^+ concentrations below 0.25 meq l^{-1} for all treatments, but significantly ($p < 0.05$) higher concentrations for +R+N lysimeters only in October and November. In contrast, the Wekerom and Fontainebleau (input concentrations 1.96 ± 1.0; 1.15 ± 0.49 meq l^{-1} respectively) soils showed clear increases in NH_4^+ concentrations within 2–3 months of the start of the $(NH_4)_2SO_4$ additions, reaching concentrations of 1.5 meq l^{-1}; and this effect was significant ($p < 0.05$) until the end of the experiment. However, at Fontainebleau, output concentrations were no longer greater than input concentration, as was observed under control conditions. Grizedale and Solling (input concentration 0.85 ± 0.6 meq l^{-1}) also showed significantly increased NH_4^+ concentrations in both $(NH_4)_2SO_4$ addition treatments within 1–3 months of the beginning of the experiment, but with concentrations below 0.4 meq l^{-1} for most sampling dates. As for Haldon, the +N+R treatment in Grizedale resulted initially in

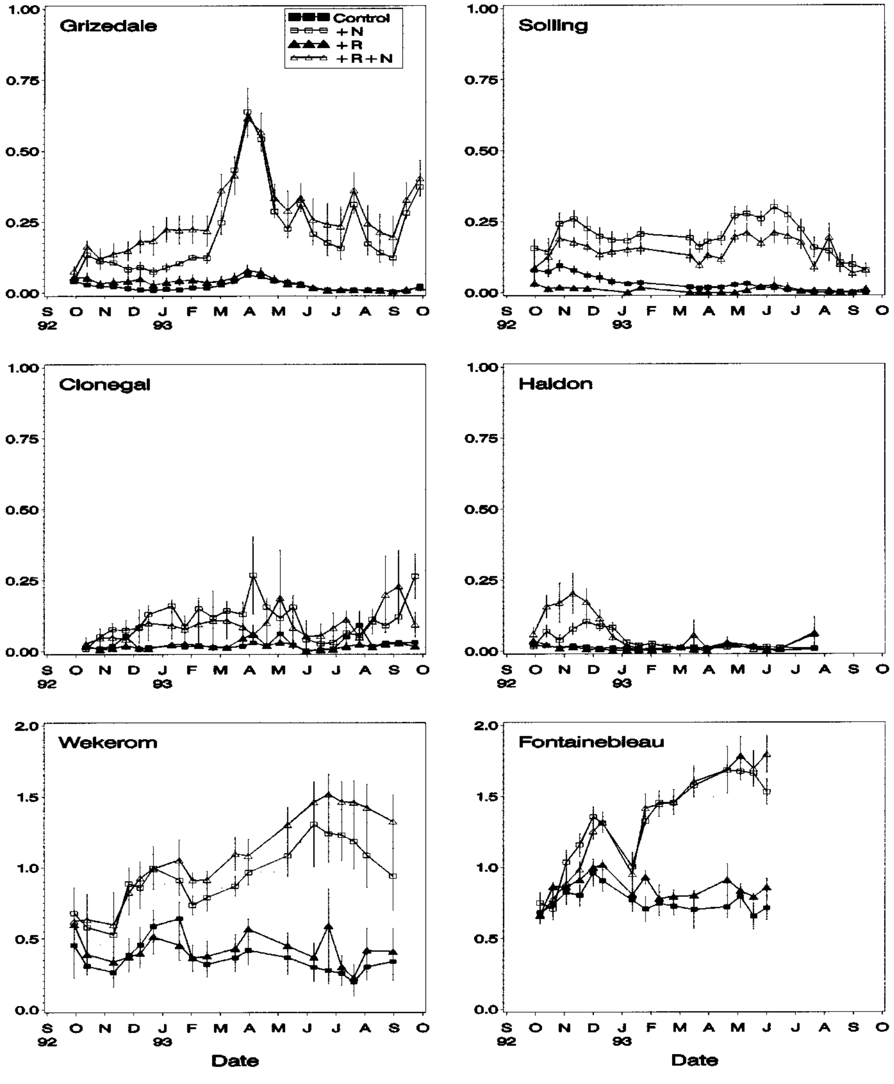

Figure 3. NH_4^+ concentrations (meq l^{-1}) in leachates of control (■–■), $(NH_4)_2SO_4$ treated (□–□), rooted (▲–▲) and rooted $(NH_4)_2SO_4$ treated (△–△) lysimeters (mean ± s.e.m.).

more N leaching than for the +N treatment alone. These soils both had C:N ratios below 16 (Table 1).

Hence, the six soils showed contrasting soil solution NH_4^+ concentrations, with Grizedale and Solling showing notable concentrations only when $(NH_4)_2SO_4$ had been applied (Figure 3). The Clonegal and Haldon soils displayed low NH_4^+ concentrations, even when additional NH_4^+ had been applied,

Table 3. Results of two-way ANOVA for time (TI) and treatment (TR) with repeated measures on TI and significant differences ($p < 0.05$) between treatments (a posteriory Tukey's studentized range test, HSD), for leachate NH_4^+, SO_4^{2-} and NO_3^- concentrations.

		TR	TI	TR∗TI	TUKEY
Clonegal	NH_4^+	0.009	0.0001	0.0001	N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R
	NO_3^-	0.1963	0.0001	0.0141	n.s.d.
Fontainebleau	NH_4^+	0.0001	0.0001	0.0001	R+N, N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R; R > C
	NO_3^-	0.6896	0.0552	0.0112	n.s.d.
Grizedale	NH_4^+	0.0001	0.0001	0.0001	R+N, N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R
	NO_3^-	0.0074	0.0001	0.0001	R+N, R > C
Haldon	NH_4^+	0.0008	0.0001	0.0001	R+N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R
	NO_3^-	0.0001	0.0001	0.0355	C, N > R+N, R
Solling	NH_4^+	0.0001	0.0001	0.0001	R+N, N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R; R+N > N
	NO_3^-	0.0035	0.0001	0.0001	N > R+N, R; C > R+N
Wekerom	NH_4^+	0.0011	0.0001	0.0001	R+N, N > C, R
	SO_4^{2-}	0.0001	0.0001	0.0001	R+N, N > C, R
	NO_3^-	0.5586	0.0001	0.0152	n.s.d.

whilst NH_4^+ concentrations were high in Wekerom and Fontainebleau under all treatments, with increases under additional NH_4^+ inputs.

Sulphate

Unlike the responses of NH_4^+, the SO_4^{2-} concentrations showed clear and similar treatment effects across all soils (Figure 4). In the absence of $(NH_4)_2SO_4$ additions, all soils maintained soil solution concentrations of SO_4^{2-} between 0.2 and 0.5 meq l^{-1}. Throughfall concentrations were also quite consistent, falling between similar values (not shown). The presence of roots did not exert significant effects (Table 3) on soil SO_4^{2-} concentrations, compared to either control or +N treatments. This was the same across the different soil types, except Solling and Fontainebleau, where SO_4^{2-} concentrations significantly increased in +R+N and +R, respectively (Table 3). However, the SO_4^{2-} additions in the two +N treatments resulted, after a gradual increase in SO_4^{2-} concentrations, in marked and significant increases in soil solu-

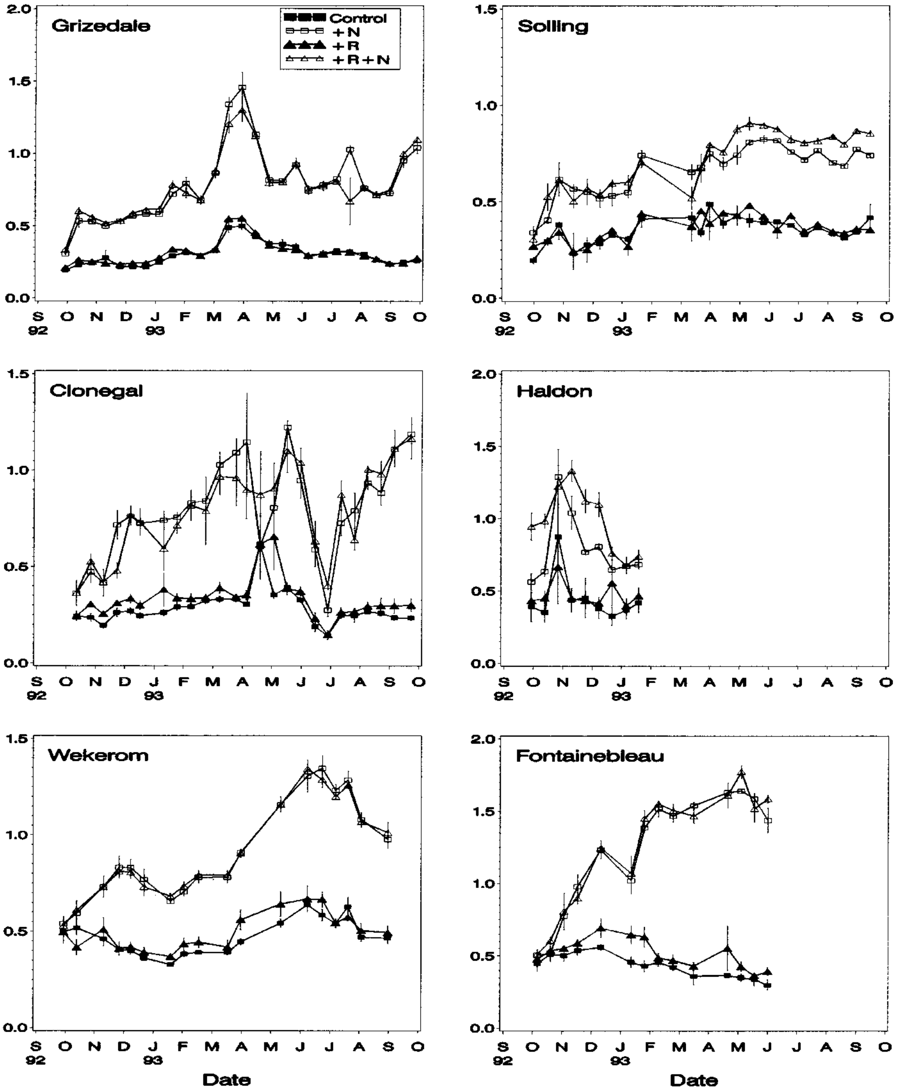

Figure 4. SO_4^{2-} concentrations (meq l^{-1}) in leachates of control (■–■), $(NH_4)_2SO_4$ treated (□–□), rooted (▲–▲) and rooted $(NH_4)_2SO_4$ treated (△–△) lysimeters (mean ± s.e.m.).

tion concentrations in all soils (Table 3). Mean annual input concentrations under $(NH_4)_2SO_4$ additions ranged from 0.90 ± 0.53 meq l^{-1} at Solling and 2.24 ± 2.41 meq l^{-1} at Clonegal. By the end of the experiment, all soils had concentrations of around 1 meq l^{-1} for the $(NH_4)_2SO_4$ treatments, with Fontainebleau reaching SO_4^{2-} concentrations of 1.5 meq l^{-1}.

Nitrate

With the exception of Fontainebleau, all the soils under study exhibited higher NO_3^- leachate concentrations than was input in throughfall, and could be regarded as nitrifying. Fontainebleau leachates showed NO_3^- concentrations similar to throughfall values (0.054 ± 0.048 meq l^{-1}) and this soil was considered to be non-nitrifying. NO_3^- dominated the inorganic anions in soil solution in the control lysimeters at the other five sites and these soils maintained a solution concentration in excess of 0.5 meq l^{-1} for the duration of the study (Figure 5). The soils differed in the seasonality of NO_3^- concentrations with, for example, Solling and Wekerom showing a similar and marked rise in NO_3^- concentrations in summer and autumn. The leachates from the Grizedale and Haldon control lysimeters were surprisingly consistent over the year of study, whilst Clonegal showed a marked concentration decline during the summer of 1993 (Figure 5).

The addition of $(NH_4)_2SO_4$ to the lysimeters at Wekerom did not significantly increase NO_3^- concentrations (Table 3), suggesting that the added N was not used as a substrate for nitrification (Figure 5). Similarly, the Fontainebleau soil failed to show a significant NO_3^- response to the N additions (Table 3) but, unlike the Wekerom soil, the soil from this site did not nitrify under control conditions. There was no significant effect of the N addition at Haldon, as NO_3^- concentrations in N-treated lysimeters were not significantly different from controls or +R (Table 3). These three soils not responding by increased NO_3^- leaching to the $(NH_4)_2SO_4$ additions differed in soil characteristic, past N inputs and vegetation cover (Table 1).

Grizedale, Solling and Clonegal soils responded four to nine months after the beginning of the N additions with significantly ($p < 0.05$) increased NO_3^- concentrations in +N or +N+R treatments, compared to controls (Figure 5). Differences at Clonegal were, however, not significant when analysed across the whole experimental period, and +N was not significantly different from C at the two other sites (Table 3), explained by the significant ‘time$*$treatment’ interaction. The rooted lysimeters (+R) behaved differently, and resulted in higher NO_3^- concentrations in Grizedale in comparison to controls, with no significant effects in Clonegal, Wekerom and Fontainebleau. The comparison of +R and +R+N to C and +N treatments in Solling showed a reduction in NO_3^- concentrations due to the presence of roots (Table 3). The Haldon soil showed significantly lower NO_3^- concentrations in both rooted lysimeter treatments than in the C and +N lysimeters.

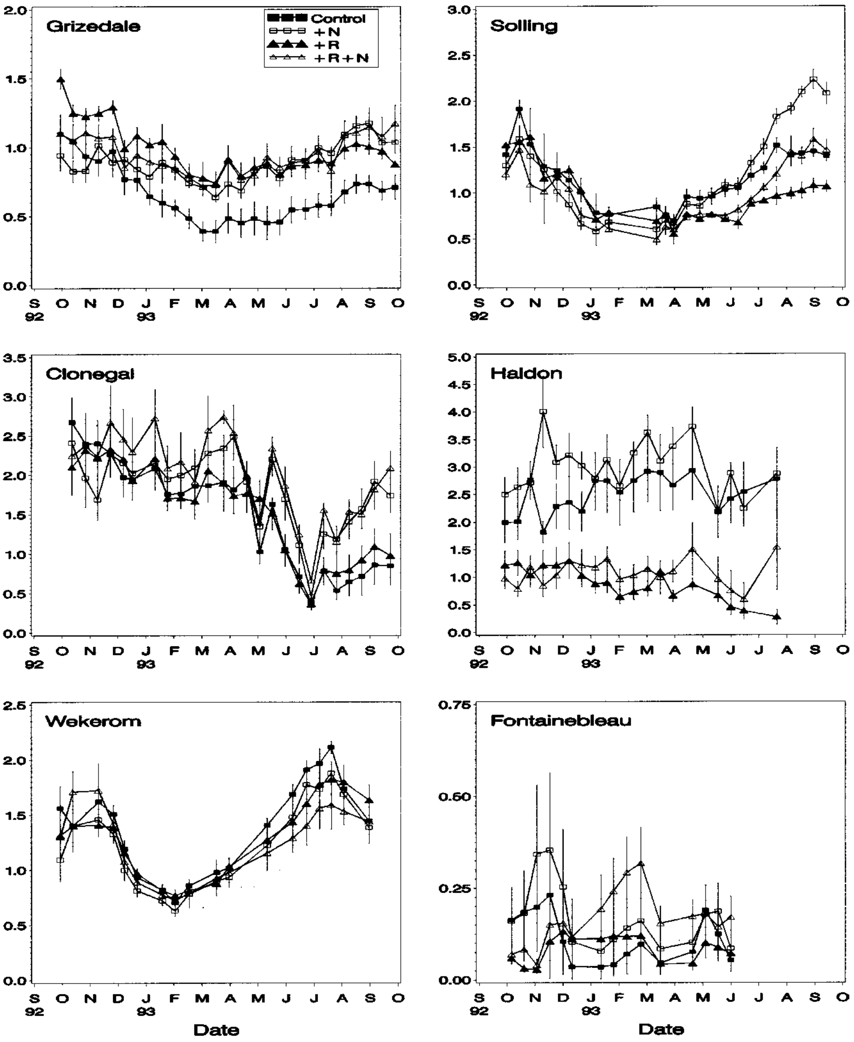

Figure 5. NO$_3^-$ concentrations (meq l^{-1}) in leachates of control (■–■), $(NH_4)_2SO_4$ treated (□–□), rooted (▲–▲) and rooted $(NH_4)_2SO_4$ treated (△–△) lysimeters (mean ± s.e.m.).

Cation/anion fluxes

Complete data on annual ionic inputs and outputs for Haldon were only available from September 1992 to January 1993 and, because of possible seasonal variability, the fluxes for this site were not extrapolated to represent yearly values. Similarly, fluxes for Fontainebleau were calculated for a period of only 8 months.

Untreated throughfall composition (Cin) showed highest NH_4^+ deposition at Wekerom of 2.1 keq ha^{-1} a^{-1}(29 kg N ha^{-1} a^{-1}) and less than 1 keq ha^{-1} a^{-1} (14 kg N ha^{-1} a^{-1}) at the other sites (Table 2, Figure 6). Other major cations in the throughfall were Ca^{2+} and Mg^{2+}, except at Wekerom where throughfall chemistry was dominated by NH_4^+. Throughfall cations were mainly balanced by SO_4^{2-}, NO_3^- and Cl^- (not shown) at all sites. The $(NH_4)_2SO_4$ addition resulted in total deposition of 5.0–6.6 keq ha^{-1} a^{-1} NH_4^+ (70–93 kg N ha^{-1} a^{-1}) and 3.7–7.7 keq ha^{-1} a^{-1} SO_4^{2-} (60–125 kg S ha^{-1} a^{-1}) at the different sites where annual fluxes were available (Figure 6, Table 2).

Under control conditions, NH_4^+ leaching was low at all sites, except at Wekerom, where outputs equalled inputs, and Fontainebleau, where outputs exceeded inputs, representing net mineralisation (Table 2). The six soils showed different responses to the additional $(NH_4)_2SO_4$ input, with Clonegal and Wekerom representing two extremes (Table 2). The Clonegal soil leached only 7% and 40% of the added NH_4^+ and SO_4^{2-}, respectively. Haldon also showed high NH_4^+ retention, leaching only 6% of input. This is surprising, as these two soils showed lowest C:N ratios (Table 1). They had, however, highest pH values. In contrast, Wekerom leached 75% and 127% of the added NH_4^+ and SO_4^{2-}, with a value exceeding 100%, representing SO_4^{2-} desorption or mineralisation. Fontainebleau also showed high NH_4^+ leaching, but outputs were lower than inputs, in contrast to the controls (see above). The other soils showed intermediate behaviour, leaching between 24–28% and 61–91% of the added NH_4^+ and SO_4^{2-}, respectively. These figures do not account for mineralisation, assuming that all leached NH_4^+ came directly from throughfall, and therefore represent the maximum % of NH_4^+ input which could have been leached. In the +N treatment, SO_4^{2-} behaved largely as a conservative ion in Grizedale and Wekerom (Table 2), but with some retention occurring in Solling and Haldon (± 40%), and notable retention occurring in Clonegal and Fontainebleau (± 60%). Roots significantly reduced NH_4^+ leaching only at Solling, as the comparison of +N and +R+N showed (see above, Figure 6). The fate of NH_4^+ therefore varied between high retention of the additional inputs in the two less acidic Picea abies sites with low C:N ratio and mean past N inputs (Clonegal, Haldon), slightly increased leaching in response to high inputs in two acid Picea abies sites with C:N around 16, but low and high past N deposition (Grizedale, Solling), and highly increased leaching in two acid, sandy Pinus sylvestris sites with high C:N (Wekerom, Fontainebleau). No clear relationship appeared between the NH_4^+ retention in the different soils and their past N inputs. The two sandy soils were distinct by their high percentage of leaching, and the two less acid soils by their high retention

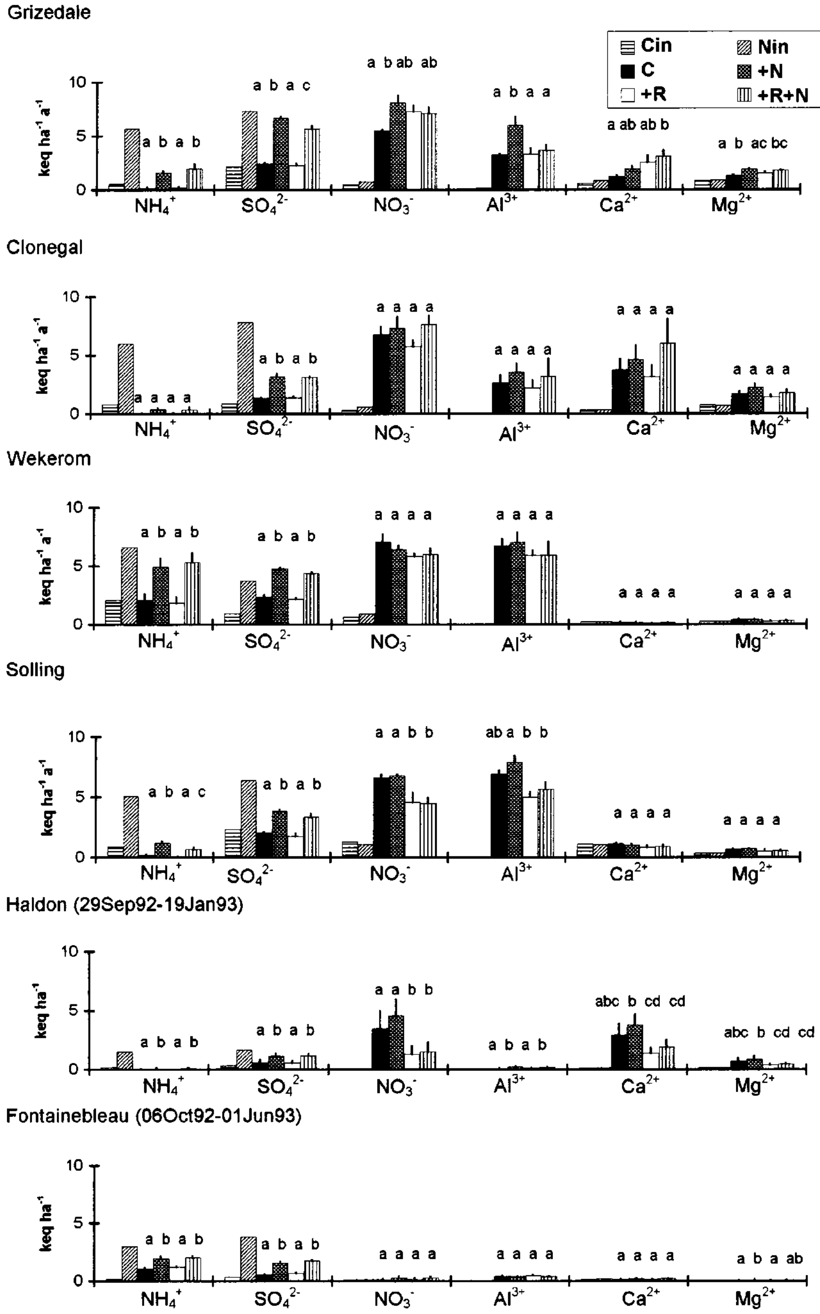

Figure 6. Annual ionic inputs (mean ± s.e.m.) for control (Cin) and $(NH_4)_2SO_4$ treated (Nin) throughfall, and outputs for control (C), $(NH_4)_2SO_4$ treated (N), rooted (R) and rooted $(NH_4)_2SO_4$ treated (+R+N) lysimeters (kg ha^{-1} a^{-1}). Values are means for 4 to 7 replicates (see text). Different letters denote significant differences between outputs at $p < 0.05$.

characteristics. However, these data have to be considered together with the NO_3^- results, as some of the NH_4^+ input could have been leached as NO_3^-.

NO_3^- dominated anionic fluxes at five sites and was associated with high rates of nitrification (Figure 6, Table 2). Only Grizedale responded significantly in terms of NO_3^- output to the $(NH_4)_2SO_4$ treatment (Figure 6) and, therefore, part of the low NH_4^+ recovery in leachates at this site, could be explained by nitrification of the added NH_4^+ input. Comparison of (NH_4^+ + NO_3^-) inputs and outputs under control conditions (Table 2) indicated that most of the NO_3^- leached originated from the internal N pool. The significant increases in NO_3^- concentrations detected at the end of the experimental period at Solling and Clonegal (see above) were not reflected in the yearly fluxes. Rooted lysimeters tended to show less NO_3^- leaching at the five nitrifying sites, and this effect was highly significant at Solling and Haldon (Figure 6), confirming results found for ionic concentrations.

The increased NO_3^- output at Grizedale was matched by increased Al^{3+} leaching for the +N treatments, but the increased (not significant) NO_3^- leaching in the +R treatments paralleled increased Ca^{2+} leaching (Figure 6). In Clonegal and Haldon, NO_3^- leaching was balanced predominantly by Ca^{2+}. The outputs of Ca^{2+} and Mg^{2+} from the Wekerom soil were very low, with a 1:1 relationship between NO_3^- and Al^{3+}. Al^{3+} also dominated cation fluxes from the Solling soil. Although Mg^{2+} played a significant role in ionic balances in certain of the soils (Grizedale, Clonegal), the overall contribution was small. Roots significantly reduced cation leaching in Solling and Haldon, with Solling showing a significant reduction in Al^{3+} leaching in the +R and +R+N treatments, compared to C and +N (Figure 6). Haldon exhibited significantly lower Ca^{2+} and Mg^{2+} leaching in both types of rooted lysimeters, compared to +N, and these reductions were probably associated with the reduction in NO_3^- leaching observed for rooted lysimeters at these sites. Cation leaching was very low at the non-nitrifying Fontainebleau site, remaining below 1 keq ha^{-1} for each of these three cations. Thus, the six systems differed considerably in cation leaching: Wekerom and Solling were Al^{3+} dominated; Grizedale and Clonegal showed intermediate behaviour, with Clonegal having the higher base reserves; Haldon was dominated by Ca^{2+} and Mg^{2+}, and Fontainebleau showed little cation leaching (see below).

Correlations

Correlations were made between soil solution NO_3^- and individual and combined cation concentrations for each soil. Soil solution cation concentrations were clearly driven by the N dynamics in the five nitrifying sites, as NO_3^- concentrations in lysimeter leachates were highly correlated ($p < 0.001$) with almost all cation concentrations. Significant correlations with the one or two cations (highest r^2) most important in balancing equivalent charges with NO_3^- (Figure 6), were selected (Figure 7). Soil solution concentrations of NO_3^- and Al^{3+} were highly correlated for Solling and Wekerom ($r^2 = 0.66$, $p < 0.001$; 0.61, $p < 0.001$, respectively), whereas Grizedale and Clonegal showed improved correlations when Ca^{2+} was also included ($r^2 = 0.45$, $p < 0.001$; $r^2 = 0.40$, $p < 0.001$, respectively; Figure 7). Variations in Ca^{2+} concentrations in soil solution at Haldon were explained by NO_3^-, without the inclusion of other cations ($r^2 = 0.85$, $p < 0.001$). In marked contrast, NO_3^- at Fontainebleau best correlated with Mg^{2+} ($r^2 = 0.36$, $p < 0.001$), but concentrations of both ions were extremely low.

These leaching characteristics could be interpreted as related to pH and base saturation (Table 1). Solling was the most acid soil (pH 3.3), and Wekerom, although less acid, showed lowest base saturation in the mineral soil horizons, thus explaining high Al^{3+} leaching. Grizedale, with a pH similar to Wekerom, showed a somewhat higher base saturation in the 1–5 cm of the mineral horizon, hence explaining some Ca^{2+} leaching. Clonegal, with a pH at the bottom limit of the exchange buffer range (pH 4.2), leached mainly Ca^{2+}, but already some Al^{3+}. Haldon was still in the exchange buffer range, and, compared to Clonegal, had a higher pH in the lower mineral soil (pH 4.8, not shown). Both NO_3^- and cation concentrations were very low at Fontainebleau, and establishing relationships to soil characteristics are not meaningful.

Root health

The fine root mass and vitality for the two rooted treatments at the end of the experiment are presented in Table 4 for each site, except for Fontainebleau where no root data were available. Lysimeters at each site contained live turgid root tips, but there were distinct differences between sites. Clearly, root establishment at Haldon exceeded that at other sites, whilst the number of turgid root tips was smallest in the +R treatment at Clonegal. The N addition treatment had no significant effect on the dry weight of fine roots at any of the sites, and the number of turgid root tips was only significantly different at Clonegal, where an increase occurred in response to the $(NH_4)_2SO_4$ treatment.

Discussion

The aim of this experiment was to determine the responses of soils with representative European N pollution histories to an increase of 75 kg ha^{-1} a^{-1} NH_4^+-N, in addition to their local NH_4^+-N input. N deposition levels resulting from $(NH_4)_2SO_4$ treatment in the current study were of the same order of

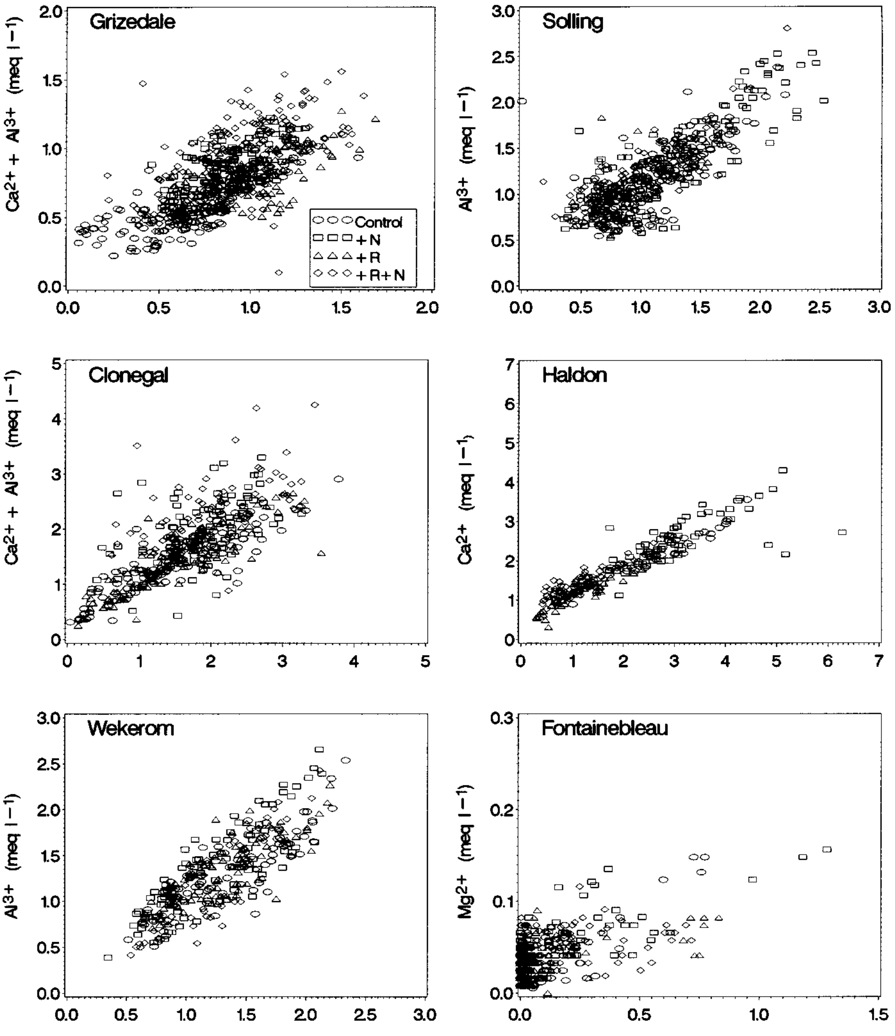

Figure 7. Cation versus NO_3^- concentrations in leachates of control (○), $(NH_4)_2SO_4$ treated (□), rooted (△) and rooted $(NH_4)_2SO_4$ treated (◊) lysimeters. $r^2 = 0.45$ (Grizedale), 0.40 (Clonegal), 0.61 (Wekerom), 0.66 (Solling), 0.85 (Haldon), 0.36 (Fontainebleau); $p < 0.001$.

magnitude as amounts found in areas of high deposition (Van Breemen & Van Dijk 1988), and two weekly N additions mixed with local throughfall were considered to be a realistic surrogate for enhanced N deposition. However, the resulting concentrations applied depended on the throughfall volume of each site and this has to be considered when comparing leachate concentrations across sites. Results from previous studies have demonstrated the validity of the ‘CORE’ approach with regard to transport processes and nutrient

Table 4. Mean fine root dry weights and numbers of turgid root tips per lysimeter for the two rooted treatments for five sites. The data are means ± s.e.m, and significant differences between the R and R+N treatments at each site are shown, n.s.d = no significant difference at $p < 0.05$.

Site	Treatment	Dry weight fine roots (g)	p	Total number turgid root tips	p
Clonegal	R	0.13 ± 0.08		49.0 ± 43.6	
	R+N	0.39 ± 0.23	n.s.d	363.0 ± 75.7	0.023
Grizedale	R	0.22 ± 0.07		292.8 ± 278.2	
	R+N	0.56 ± 0.31	n.s.d	65.7 ± 37.9	n.s.d
Haldon	R	12.43 ± 3.20		5726.7 ± 3118.9	
	R+N	6.84 ± 0.87	n.s.d	2685.8 ± 1096.2	n.s.d
Solling	R	1.89 ± 0.02		473.0 ± 293.0	
	R+N	1.66 ± 0.28	n.s.d	1117.3 ± 338.1	n.s.d
Wekerom	R	0.78 ± 0.26		198.8 ± 165.0	
	R+N	0.64 ± 0.08	n.s.d	60.0 ± 60.0	n.s.d

cycling (see Raubuch 1992, and references therein). For example, studies at the Solling site, using tension samplers not confined in soil cores, resulted in soil solution NH_4^+ concentrations similar to those found in this study (Beese 1989).

Since annual NO_3^- leaching was not increased by N deposition (except for Grizedale), the annual NH_4^+ balance gives an indication of the net amount of NH_4^+ retained by the soil. The value represents, however, the minimal percentage retained (or the maximum percentage leached) by the soil, as leaching of mineralised NH_4^+ cannot be quantified, and was set to 0 for percentage calculations. The added NH_4^+ seemed to leach readily through the two acid sandy soils, where output concentrations rapidly equilibrated with input concentrations, indicating that the N retention capacity of these systems was saturated. These two soils already showed a correspondence between NH_4^+ inputs and outputs under 'control' conditions and it can be hypothesised that sites already showing NH_4^+ outputs greater or equal to inputs would not be able to retain additional N inputs. The increased NH_4^+/cation ratio resulting from this situation could lead to nutrient imbalance of the trees (Roelofs 1985). Results are in close agreement with those of Coûteaux and Sallih (1994), who studied the same soils in laboratory experiments. They also found high ^{15}N retention for Grizedale and Kilkenny (a site similar to the Clonegal site studied here) and low retention for the sandy Wekerom soil. Incorporation of applied ^{15}N-NH_4^+ into the biomass has been reported by both Stams et al. (1990) and Coûteaux and Sallih (1994), and is thought to be a major mechanism of N retention. Differences across various sites in the ability of soils to sequester N have been explained by site history (Feger 1992), C availability (Verhagen et al. 1995), differences in N nutrition and microbial N turnover (Miller & Miller 1988; Feger 1992) or texture (Zoettl 1990). Through physical characterisation of the same soils than used in our study, Coûteaux and Sallih (1994), concluded that texture, C and N content, and level of acidification were the most important factors determining N retention in the soils under study. We found highest retention in the two less acidified soils ($\mathrm{pH} \geqslant 4.2$) with low C:N. The two acid soils ($\mathrm{pH} < 4.2$) of sandy texture and high C:N leached a high proportion of the added N, and the two acid clay-clay loam soils leached 20–30% of the added N. N leaching characteristics did not seem related to past N inputs. However, clear patterns could only be established with a bigger dataset permitting meaningful statistical regression analysis.

With regard to the behaviour of added SO_4^{2-}, soils could be ordered according to the percentage of added SO_4^{2-} leached: Wekerom (>100% leached; SO_4^{2-} desorption or mineralisation), Grizedale (90%), Solling and Haldon (60%), Clonegal and Fontainebleau (40%). Significant SO_4^{2-} retention occurred at most sites, particularly Clonegal and Fontainebleau. Incorporation of SO_4^{2-} into plant and microbial tissue is believed to be low and SO_4^{2-} retention is generally attributed to anion adsorption (Johnson et al. 1985). In Grizedale and Wekerom, SO_4^{2-} behaved as a conservative ion, indicating that the capacity to adsorb anions was saturated. Similarly, catchment studies in which the deposition of SO_4^{2-} has been prevented, have shown rapid responses of SO_4^{2-} in streamwater and soil solution chemistry (Wright et al. 1990). However, SO_4^{2-} leaching was very poorly related to cation leaching in the current study, and SO_4^{2-} was not considered to be an important contributory factor to acidification in these soils.

None of the soils showed a dramatic increase in NO_3^- leaching in response to the consistently increased NH_4^+ supply. Similarly, in field studies using a ^{15}N pulse, nitrification of less than 4.2% of the applied $^{15}N\text{-}NH_4^+$ after 60 days (Emmet & Quarmby 1991) and less than 22% after one year (Stams et al. 1991) have been reported for coniferous forest soils. These studies have confirmed that a part of external NH_4^+ input can be oxidised to NO_3^-, some of it even within 24 hours (Emmet & Quarmby 1991), but cannot predict the long-term effects of chronically increased N deposition. Our study demonstrated an increase in NO_3^- concentrations due to increased NH_4^+ deposition after a time lag of approximately 4–9 months at three sites (Grizedale, Clonegal and Solling), emphasising the importance of long term studies in this area of research. This delay could be explained as a lag phase during which an initially low nitrifier population has developed (Vitousek & Matson 1985), or a first step of incorporation of NH_4^+ into the microbial biomass with subsequent heterotrophic or autotrophic nitrification, or, finally, autotrophic nitrification stimulated by local pH increase during mineralisation (Alexander 1961). The second hypotheses is supported by the high NH_4^+ retention of these three soils, as discussed previously.

Five sites could be considered as nitrifying, with highest soil solution concentrations of NO_3^- being found in the less acidic soils (Haldon and Clonegal). The two sandy soils (Fontainebleau and Wekerom) showed markedly contrasting nitrification characteristics, with the Fontainebleau soil failing to nitrify under any treatment. Kriebitzsch (1978) showed low nitrification for soils with high C:N ratio in the humus layer and the Fontainebleau site had the highest C:N ratio (28 for the humus layer, 27 in the mineral soil) of the six sites, being in the range in which Kriebitzsch reported no nitrification. However, non nitrifying coniferous forest soils have been described over a wide range of C:N ratios (Robertson 1982), and some nitrifying sites of our study also showed high C:N ratios in the humus layer (Solling: 24.5, Clonegal: 25.2, Wekerom: 25.2). High NH_4^+ leaching at the Fontainebleau site indicated that mineralisation was not limiting and that some other factor (e.g. nutrients, allelophatic inhibition, texture) was important. In contrast, Wekerom showed high nitrification with marked seasonality, but no response to NH_4^+ additions. This indicates that nitrifiers at this site could not use the NH_4^+ added in throughfall and were limited by another factor. This result is surprising since it had been reported that nitrification rates observed in acid forest soils may be high enough to transform deposition of NH_4^+ up to 60 kg ha^{-1} a^{-1} (Van Breemen & Van Dijk 1988; Stams & Marnette 1990).

The strong relationship between NO_3^- and cation concentrations across the five nitrifying sites confirmed the importance of NO_3^- leaching in controlling soil acidification (Reuss & Johnson 1986). The soils in the current study could be separated into four classes depending on pH and base saturation; the Haldon soil was situated in the CEBR (cation exchange buffer range sensu Ulrich 1981; pH 4.2–5.0), with NO_3^- and Ca^{2+} soil solution concentrations being directly related. Clonegal, pH = 4.2, showed a strong relationship between NO_3^- and (Ca^{2+}+ Al^{3+}) concentrations in soil solution, indicating that this soils was in the CEBR, but already exhibiting signs of Al^{3+} leaching. Grizedale (pH = 3.7) was situated in the ABR (aluminium buffer range, pH 2.8–4.2), but still leaching some Ca^{2+}, possibly because of higher base saturation than the other acid soils. The Wekerom and Solling soils ($pH<4.2$) showed a nearly 1:1 relationship between NO_3^- and Al^{3+} concentrations, being clearly situated in the ABR confirming the results of CORE et al. (1992).

In situ studies investigating the role of root uptake on nutrient cycling in forests reveal methodological difficulties and have often been based on root exclusion or budget approaches. In the current work, we have introduced root systems connected to mature trees into intact soil cores (Anderson et al. 1990) and measured changes in soil solution chemistry. The presence of live roots significantly reduced annual NO_3^- leaching (Figure 6) at Solling and Haldon, which had loamy soils. At Solling, roots also delayed the rise in NO_3^- and associated Al^{3+} concentration increases due to increased NH_4^+ deposition, but did not prevent acidification from taking place. In Grizedale, increased NO_3^- leaching was observed for all treatments when compared to the control and it appeared that the roots may have initially released N or may have altered physical conditions in the lysimeters, favouring more N mineralisation. Increased N loss and decomposition in the presence of live roots have also been reported by Faber & Verhoef (1991). At the other sites, no significant root effects were demonstrated.

Live roots were inserted into the lysimeters according to a standardised protocol at all sites. A first implant in spring failed, and it was thought that fine roots may establish more easily into lysimeters at a time when they are growing actively, which may be during autumn in mature stands (Flower-Ellis & Persson 1980). Maximum care was taken during root implant, but success of growth into the lysimeter may have depended on the health of root before implant and its ability to resist the manipulation. At the end of the experiment the Solling and Haldon lysimeters both contained higher number of turgid root tips. These differences in root health may be reflected in the relative nutrient uptake between sites and are considered to be indicators of tree health in the forest sites under study.

Our results showed clearly that under increasing NH_4^+ deposition, the fate of the NH_4^+ is central in determining possible acidification and nutrient imbalance consequences for forest soils. Soils capable of accumulating N will increase N in organic matter, with possible delayed increase in NO_3^- leaching and soil acidification. ‘Saturated’ soils will show high NH_4^+ leaching and high NH_4^+/cation ratios in soil solution. pH and texture seemed important in determining N retention capacities of soils, but no clear relationship was found with past N deposition. Nitrification was confirmed to be the key process in determining soil acidification, cation leaching and aluminium mobilisation under additional N loading, yet we are remarkably ignorant of both the organisms and the major edaphic and environmental controlling factors. Future research needs to be directed towards increasing our understanding of both the identity and the functioning of the nitrifiers, and new molecular techniques offer great potential (Nejidat & Abeliovich 1994). Our data show clearly that at some mature sites, root sinks can be important in delaying the acidification effect of enhanced N deposition, but fail to prevent acidification occurring. The very different responses of the six soils to increased $(NH_4)_2SO_4$ deposition emphasise that the establishment of N critical loads for forest soils need to allow for differences in N storage capacity and nitrification potential.

Acknowledgements

This work is part of the CORE project and was funded by the Commission of the European Communities (DGXII). We thank the referees for their constructive comments on the manuscript.

References

Aber JD, Nadelhoffer KJ, Steudler P & Melillo JM (1989) Nitrogen saturation in northern forest ecosystems. BioScience 39: 378–386

Agren GI & Bosatta E (1988) Nitrogen saturation of terrestrial ecosystems. Environmental Pollution (series A) 54: 185–197

Alexander M (1961) Introduction to Soil Microbiology. John Wiley & Sons, New York

Anderson JM, Leonard MA & Ineson P (1990) Lysimeters with and without tree roots for investigating the role of macrofauna in forest soils. In: Harrison AF, Ineson P & Heal OW (Eds) Nutrient cycling in terrestrial ecosystems (pp 347–355). Elsevier Applied Science, London

Beese F (1989) Wirkungen von Kalkungs- und Düngungsmassnahmen auf die chemische Zusammensetzung der Bodenlösung. In: Ergebnisse neuerer, ökosytemarer Untersuchungen an Buchenbeständen im Solling (pp 27–48). Berichte des Forschungszentrums Waldökosysteme, Reihe A, Bd. 49, Universität Göttingen

Cody PR & Smith JK (1991) Applied Statistics and the SAS® Programming Language. Elsevier Science Publishers B.V., Amsterdam

CORE ESF, Anderson JM, Beese F, Berg MP, Bolger T, Coûteaux M-M, Henderson R, Ineson P, McCarthy F, Palka L, Raubuch M, Verhoef HA & Willisson T (1992) Mechanisms of nutrient turnover in the soil compartments of forest. In: Teller A, Mathy P & Jeffers JNR (Eds) Responses of forest ecosystems to environmental changes (pp 342–350). Elsevier

Coûteaux M-M & Sallih Z (1994) Fate of inorganic ^{15}N in the profile of different coniferous forest soils. Biology and Fertility of Soils 17: 101–107

De Vriess W, Posch M, Reinds GJ & Kamari J (1992) Critical loads and their exceedance on forest soils in Europe. Wageningen (The Netherlands), DLO The Winand Staring Centre. Report 58, 126pp

Emmet BA & Quarmby C (1991) The effect of harvesting intensity on the fate of applied ^{15}N-ammonium to the organic horizons of a coniferous forest in N. Wales. Biogeochemistry 15: 47–63

Faber JH & Verhoef HA (1991) Functional differences between closely-related soil arthropods with respect to decomposition processes in the presence or absence of pine tree roots. Soil Biology and Biochemistry 23: 15–23

Feger KH (1992) Nitrogen cycling in two Norway Spruce (*Picea abies*) ecosystems and effects of a $(NH_4)_2SO_4$ addition. Water Air and Soil Pollution 61: 295–307

Flower-Ellis JGK & Persson H (1980) Investigations of structural properties and dynamics of Scots pine stands. In: Persson H (Ed) Structure and Function of Northern Coniferous

Forests – An Ecosystem Study (pp 125–138), Ecological Bulletin 32. Swedisch Natural Science Research Council, Stockholm

Grennfelt P & Hultberg H (1986) Effects of nitrogen deposition on the acidification of terrestrial and aquatic ecosystems. Water Air and Soil Pollution 30: 945–963

Gundersen P & Rasmussen L (1990) Nitrification in forest soils: effect from nitrogen deposition on soil acidification and aluminium release. Reviews of Environmental Contamination and Ecology 113: 1–45

Hatcher L & Stepansky J (1994) A Step-by-Step Approach to using the SAS System for Univariate and Multivariate Statistics. SAS Institute Inc., Cary, NC

Hornung M & Skeffington RA (1993) Critical loads: concepts and applications. ITE Symposium 28, HMSO, London, pp 134

Johnson DW, Richter DD, Lovett GM & Lindberg SE (1985) The effects of atmospheric deposition on potassium, calcium, and magnesium cycling in two deciduous forests. Canadian Journal of Forest Research 15: 773–782

Kriebitzsch WU (1978) Stickstoffnachlieferung in sauren Waldboeden Nordwestdeutschlands. Scripta Geobotanica, Goettingen 14: 1–66

Miller HG & Miller JD (1988) Response to heavy nitrogen applications in fertilizer experiments in British forests. Environmental Pollution 54: 219–223

Nejidat A & Abeliovich A (1994) Detection of Nitrosomonas spp. by polymerase chain reaction. FEMS Microbiology Letters 120: 191–194

Nihlgard B (1985) The ammonium hypothesis-an additional explanation to the forest dieback in Europe. Ambio 14: 2–8

Nilsson J & Grennfelt P (Eds) (1988) Critical loads for sulphur and nitrogen (Report 1988: 15) Copenhagen: Nordic Council of Ministers

Raubuch M (1992) Wechselwirkungen zwischen mikrobiologischen und chemischen Parametern in Waldböden unter Berücksichtigung des atmosphärischen Eintrags. Berichte des Forschungszentrums Waldökosysteme, Reihe A, Bd. 89, Universität Göttingen

Reuss JO & Johnson DW (1986) Acid deposition and the acidification of soils and waters. Ecological Studies 59. 120 p

Robertson GP (1982) Nitrification in forested ecosystems. Philosophical transactions of the Royal Society London series B 296: 445–457

Roelofs JGM, Kempers AJ , Houdijk ALFM & Jansen J (1985) The effect of air-borne ammonium sulphate on *Pinus nigra* var. *maritima* in the Netherlands. Plant and Soil 84: 45–56

Ruess L, Sandbach P, Cudlin P, Dighton J & Crossley A (1996) Acid deposition in a spruce forest soil: effects on nematodes, mycorrhizas and fungal biomass. Pedobiologia 40: 51–66

SAS Institute Inc. (1989) SAS/STAT® User's Guide, Version 6, Volume 1, Cary, NC: SAS Institute Inc., 943 pp

Skeffington RA (1993) Problems with the critical loads approach: a view from industry. In: Critical loads: concepts and applications. ITE Symposium 28, HMSO, London, pp. 134

Skeffington RA & Wilson E (1988) Excess nitrogen deposition: Issues for consideration. Environmental Pollution 54: 159–184

Stams AJM & Marnette ECL (1990) Investigation on nitrification in forest soil percolation column. Plant and Soil 125: 135–141

Stams AJM, Lutke-Schipholt IJ, Marnette ECL, Beemsterboer B & Woittiez JRW (1990) Conversion of ^{15}N-ammonium in forest soils. Plant and Soil 125: 129–134

Stams AJM, Booltink HWG, Lutke-Schipholt IJ, Beemsterboer B, Woittiez JRW & Van Breemen N (1991) A field study on the fate of ^{15}N-ammonium to demonstrate nitrification of atmospheric ammonium in acid forest soil. Biogeochemistry 13: 241–255

Ulrich B (1981) Oekologische Gruppierung von Boeden nach ihrem chemischen Bodenzustand. Zeitschrift fuer Pflanzenernaehrung und Bodenkunde 144: 289–305

Van Breemen N & Van Dijk HFG (1988) Ecosystem effects of atmospheric deposition of nitrogen in the Netherlands. Environmental Pollution 54: 249–274

Verhagen FJM, Laanbroek HJ & Wolendorp JW (1995) Competition for ammonium between plant roots and nitrifying and heterotrophic bacteria and the effects of protozoan grazing. Plant and Soil 170: 241–250

Vitousek PM & Matson PA (1985) Causes of delayed nitrate production in two Indiana forests. Forest Science 31: 122–131

Wright RF, Cosby BJ, Flaten M. & Reuss JO (1990) Evaluation of an acidification model with data from manipulated catchments in Norway. Nature 343: 53–55

Zoettl HW (1990) Remarks on the effects of nitrogen deposition to forest ecosystems. Plant and Soil 128: 83–89