# THE IMPACTS OF ATMOSPHERIC N INPUTS ON THROUGHFALL, SOIL AND STREAM WATER INTERACTIONS FOR DIFFERENT AGED FOREST AND MOORLAND CATCHMENTS IN WALES

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**Abstract.** A study of inorganic-N concentrations in streams, soil waters, throughfall and rainfall was conducted for one year in five moorland and 20 Sitka spruce plantation catchments in upland Wales. The forest ages ranged from 10 to 55 yr. Highly significant positive relationships with forest stand age existed for inorganic-N concentrations in streamwater, B and O horizon soil waters and throughfall. Inorganic-N in streams and B horizon waters was entirely $NO_3^-$. Inorganic-N fluxes in throughfall also showed a significant, positive relationship with stand age. Throughfall flux of inorganic-N in the oldest stand was 25.1 kgN $ha^{-1}$ $yr^{-1}$, double that in incident rainfall.

The older forest stands appear unable to utilise the available N. Nitrification is very active in the soils of these older stands, resulting in significant soil acidification. The processes responsible for the observed $NO_3^-$ leaching losses, and the implications for the debate on Nitrogen Critical Loads are discussed.

## 1. Introduction

In recent years, the balance of interest concerning acid deposition, soil and streamwater acidification has shifted from the role of $SO_2$ to that of N compounds, primarily $NO_x$ and $NH_3$ (Grennfelt and Hultberg, 1986; Skeffington and Wilson, 1988; Henriksen and Brakke, 1988). Dry and cloudwater deposition of N has been recognised as a substantial input; for example, total N inputs of more than 20 kgN $ha^{-1}$ $yr^{-1}$ have been estimated for comparatively remote areas of Britain (Fowler *et al.*, 1989), 8–10 times that encountered in sites with a pristine environment (Aber *et al.*, 1989).

Despite these substantial N inputs, British upland systems, including plantation

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forests of exotic conifers, have been regarded as nitrogen limited. By implication, $NO_3^-$ leaching losses are expected to be small and much of the soil and streamwater data obtained from upland catchments has reinforced this view (Roberts *et al.*, 1989). However, studies we have conducted in a 50 yr old Sitka spruce (*Picea sitchensis* (Bong.) Carr.) plantation catchment at Beddgelert in North Wales provide evidence that the site has reached 'nitrogen saturation' (Stevens *et al.*, 1993b). At this site, wet deposition inputs (i.e. bulk precipitation but excluding most dry and occult deposition) of inorganic-N ($NO_3^-$-N plus $NH_4^+$-N) during the period 1982 to 1990 inclusive averaged 10.3 kgN $ha^{-1}$ $yr^{-1}$, and stream outputs averaged 14.6 kgN $ha^{-1}$ $yr^{-1}$, with stream $NO_3^-$ concentration averaging 0.7 mgN $L^{-1}$. Studies of streams draining from younger Sitka spruce plantations in Wales show lower $NO_3^-$ concentrations and loads: 0.32 mgN $L^{-1}$ and 6.4 $kg^{-1}$ $yr^{-1}$ in a 36 yr old crop at Hafren Forest, Plynlimon (Stevens *et al.*, 1990) and 0.25 mgN $L^{-1}$ or less in streams draining 12 and 24 yr old crops at Tywi Forest (Waters and Jenkins, 1992). The latter values are similar to those observed in moorland streams in Wales, which average 0.2 mgN $L^{-1}$ (Reynolds *et al.*, 1983; Roberts *et al.*, 1989).

These observations provide limited evidence of a relationship between spruce crop age and $NO_3^-$ leaching. However, if such a relationship exists more generally, then there is a potentially widespread problem of soil and possibly streamwater acidification at sites with old Sitka spruce plantations. This is because in situations (as appears to be the case at Beddgelert) where enhanced atmospheric N deposition is not immobilised or recycled, either by vegetation, the soil microbial population or in soil organic matter, nitrification of any excess $NH_4^+$ may occur and the $NO_3^-$ generated is likely to leach into freshwaters. Nitrification, followed by leaching of $NO_3^-$, may be either a soil or water acidifying process because the leached $NO_3^-$ is accompanied by an equivalent quantity of cations. If these cations are $Al^{3+}$ and $H^+$, streams will be acidified. If base cations are leached, then soils will acidify but not streams. It is likely, therefore, that the soils at Beddgelert are becoming acidified and depleted of base cations at a faster rate than that attributable to sulphur pollution alone.

A number of interacting processes may be responsible for the increased $NO_3^-$ leaching rates from older crops:

a) Increased dry and cloudwater deposition to the older forests, resulting, in these stands, in larger total N inputs. These inputs are then in excess of the gross uptake capacity of these older trees.
b) Increased rates of mineralisation and nitrification in forest floor and soil organic matter. This may be in response to larger N inputs from a).
c) Reduced uptake of available N by older, as compared with younger, trees (Miller, 1979).

The objectives of this study are, firstly to ascertain whether the relationship between Sitka spruce crop age and $NO_3^-$ leaching holds on a regional basis by investigating a set of 25 sites in Wales, and secondly to assess process a) above using throughfall inorganic-N fluxes as a surrogate for total atmospheric inputs of inorganic-N to

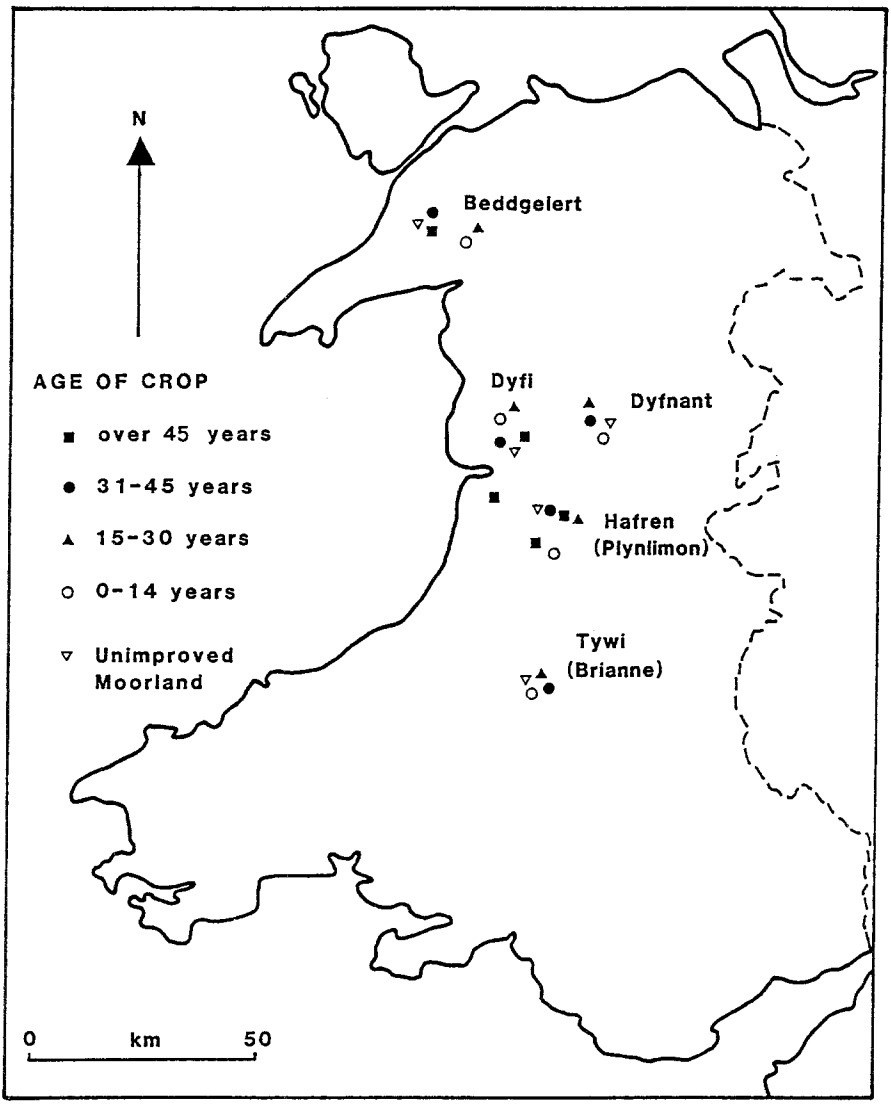


Fig. 1. Locations of the 25 study sites in Wales.

the forests. Further work is in progress to assess rates of mineralisation, nitrification and soil acidification.

## 2. Sites and Methods

### 2.1. Sites

The study was conducted in five forest districts in north and mid Wales (Figure 1). Three of these districts were areas where nutrient cycling and biogeochemical studies have been carried out by the Institute of Terrestrial Ecology for a number

These are Beddgelert (Stevens *et al.*, 1989), Hafren, which includes Plynlimon (Reynolds *et al.*, 1989), and Tywi, which is the Llyn Brianne area (Waters and Jenkins, 1992). The remaining two districts, Dyfi and Dyfnant, contain extensive areas of upland conifer plantation typical of that found throughout this part of Wales. These five districts are abbreviated to BT, HN, TY, DI and DN respectively throughout the remainder of this paper.

In each forest district, five catchments were used, one conforming where possible to each of the following criteria:

- agriculturally unimproved moorland and grassland
- Sitka spruce planted 1975–1990 (0–16 yr old)
- Sitka spruce planted 1960–1974 (17–30 yr old)
- Sitka spruce planted 1945–1959 (31–45 yr old)
- Sitka spruce planted pre-1945 (>45 yr old)

These five categories have been coded 0–4, such that, for example, the Beddgelert moorland and >45 yr old forest sites are coded BT0 and BT4 respectively.

All forest sites were first rotation (except for BT2, which later was found to be second rotation) and originally planted on agriculturally unimproved moorland. The trees at site BT1 were also subsequently found to be 24 yr old, not 15 as originally thought. Data from this site have retained the site code BT1, however.

Moorland sites BT0, DI0 and HN0 were dominated by *Nardus stricta – Festuca ovina* grassland, TY0 by *Molinia caerulea* grassland and DN0 by heather moorland composed primarily of *Calluna vulgaris* and *Erica cinerea*. All moorland catchments were grazed by sheep. Sheep were also present at various times in some of the forest sites, but most noticeably in BT1 and TY1 where occasional contamination of soil water samples was experienced.

Catchments were required for our study as we intended sampling streamwater, in addition to soil water and throughfall. In practice, streamwater sampling sites defined the areas of the catchments. Great difficulty was experienced finding forested catchments occupied purely by trees of the required age. In some cases, catchments required to have old trees also contained small areas of young trees. However, we avoided those catchments where the requirement was for young trees, but old trees were also present. Areas of felling or extensive windblow in any catchment resulted in rejection of that catchment. Several forest catchments contained areas of moorland (Table I). All catchments were underlain by Ordovician or Silurian shale, slate or mudstone with little if any extraneous drift material and the soils were dominantly stagnopodzols (Avery, 1980), equivalent to Aquods (possibly Cryaquods) of the USDA system (Soil Survey Staff, 1975). A typical soil profile consisted of spruce litter, peaty O horizon, grey, leached, anaerobic E horizon, bright brown Bs and stony C horizons. In reality, a catenary sequence of soil types often occurred in the catchments, but wherever possible, the plot used for soil water and throughfall sampling was located on a stagnopodzol. The ground flora at the forest sites was completely absent (and therefore typical of spruce plantations in upland Britain), except in the oldest site at Beddgelert where a sparse bryophyte

TABLE I

Characteristics of the 25 sites

| Forest district | Site code | Age of trees (yr)[a] | Elevation of plot (m) | Mean tree height (m) | Crop basal area ($m^2$ $ha^{-1}$) | Thinning[b] | Moorland % in catchment |
|---|---|---|---|---|---|---|---|
| Beddgelert | BT0 | 0 | 360 | – | – | – | 100 |
| Beddgelert | BT1 | 24 | 380 | 7.9 | 46.9 | N | 0 |
| Beddgelert | BT2 | 29 | 290 | 15.6 | 48.2 | N | 30 |
| Beddgelert | BT3 | 44 | 250 | 17.5 | 56.7 | L | 0 |
| Beddgelert | BT4 | 55 | 380 | 18.9 | 65.8 | S | 20 |
| Dyfi | DI0 | 0 | 400 | – | – | – | 100 |
| Dyfi | DI1 | 15 | 400 | 5.9 | 13.3 | N | 50 |
| Dyfi | DI2 | 27 | 350 | 12.7 | 39.1 | L | 10 |
| Dyfi | DI3 | 40 | 400 | 22.7 | 67.1 | S | 23 |
| Dyfi | DI4 | 51 | 375 | 18.5 | 69.8 | S | 53 |
| Dyfnant | DN0 | 0 | 430 | – | – | – | 100 |
| Dyfnant | DN1 | 10 | 460 | 2.5 | 0.8 | N | 0 |
| Dyfnant | DN2 | 16 | 410 | 6.4 | 33.0 | N | 0 |
| Dyfnant | DN3 | 32 | 450 | 15.3 | 43.3 | N | 35 |
| Dyfnant | DN4 | 53 | 300 | 26.3 | 65.4 | S | 30 |
| Hafren | HN0 | 0 | 430 | – | – | – | 100 |
| Hafren | HN1 | 14 | 480 | 5.1 | 16.2 | N | 30 |
| Hafren | HN2 | 28 | 580 | 7.1 | 25.2 | N | 0 |
| Hafren | HN3 | 37 | 380 | 14.9 | 48.4 | L | 0 |
| Hafren | HN4 | 53 | 400 | 12.5 | 39.5 | N | 1 |
| Tywi | TY0 | 0 | 440 | – | – | – | 100 |
| Tywi | TY1 | 14 | 490 | 5.8 | 23.5 | N | 5 |
| Tywi | TY2 | 19 | 475 | 7.8 | 47.5 | N | 5 |
| Tywi | TY3 | 31 | 480 | 13.0 | 77.3 | N | 0 |
| Tywi | TY4 | 53 | 450 | 21.8 | 28.7 | N | 0 |

[a] Age 0 = moorland.

[b] N = no thinning; L = line thinning; S = selective thinning.

and fern flora had become re-established, and in the youngest sites at Dyfi, Dyfnant, Tywi and Hafren where canopy closure was incomplete and remnants of moorland vegetation survived.

One plot was laid out in each of the 25 catchments. These plots were 20 by 20 m and were located in a representative area of moorland or the appropriate age of forest (Table I).

### 2.2. Solute sampling and chemical analysis

Soil water samples were collected from the base of the O horizon and within the Bs horizon. Samples from the O horizon were collected using three simple PVC tensionless lysimeters per plot, each discharging into buried 10 L polythene carboys.

Water samples from the Bs horizon in each plot were obtained from three porous ceramic cup suction samplers, similar to those described by Wagner (1962). The three O horizon samples per plot were bulked prior to analysis, as were the three Bs horizon samples. Throughfall was obtained from the forest sites using 12 collectors per plot located at random along a 20 m transect. Each collector consisted of a 58 $cm^2$ polythene funnel and 5 L polythene bottle and supported such that the rim of the funnel was approximately 60 cm above the ground surface. The volumes of throughfall were measured and the twelve individual samples per site were bulked on each sampling occasion prior to analysis. Collectors were not relocated after each sampling interval. At moorland sites, throughfall was collected by three sets of three lengths of miniature guttering, discharging into buried 5 L bottles. These often suffered from contamination by sheep excreta. The volumes of water collected were also often affected by blockages and disturbance to the extent that we have been unable to calculate throughfall nutrient fluxes from these moorland sites, and the concentration data may be influenced by contamination.

Rain was collected in simple plastic rain gauges (Stevens, 1981) mounted with their rims 1.5 m above the ground surface. Twenty-two sites were allotted a rain collector in a forest clearing or adjacent open area; three forest sites were served by collectors on adjacent moorland sites. Volumes of rain were measured prior to chemical analysis. A sample of streamwater was collected on each sampling occasion.

A bulked sample of each of the O and Bs soil waters and throughfall was collected from each of the 25 sites at four-week intervals for one year from November 1990 until October 1991. Rain was collected from the 22 sites at four week intervals from November 1990 until April 1991. From April 1991 until October 1991, analytical constraints dictated a reduction in sample numbers. During this period, rain was collected at four week intervals and individual volumes measured, but, prior to analysis, samples were bulked by forest district, resulting in a single rain sample for each of the five forest districts per four week period. Subsequent analysis of the initial six months' rainfall chemistry data for the 22 individual sites showed no significant within forest district differences, justifying the decision to bulk these samples.

On return to the laboratory, pH was determined and samples filtered through 0.45μ papers within four days of collection. Samples were then stored for up to 6 weeks at 4 °C prior to analysis. No preservative was used. $NO_3^-$ was determined by ion chromatography and $NH_4^+$ colorimetrically by autoanalyser using standard methods.

### 2.3. Data analysis

A small number of results were missing from data set as a consequence of breakages and contamination by bird and animal droppings. Because the concentrations of $NO_3^-$ and $NH_4^+$ tended to be very seasonal, simple averaging of the remaining records to produce annual means could be misleading if records had been lost

TABLE II

Regression equations for inorganic-N concentrations and fluxes with tree crop age. Linear, quadratic and cubic terms are in columns headed a, b and c respectively. Where the regression model is improved by inclusion of site physical or crop characteristics, these are described in the text

| | | Constant | a | b | c | $R^2$ | F | d.f. | p |
|---|---|---|---|---|---|---|---|---|---|
| Concentrations | | | | | | | | | |
| Stream | $NO_3$-N | 0.128 | −0.0296 | 0.00172 | −0.0000191 | 0.839 | 12.68 | 7,17 | <0.001 |
| Bs horizon | $NO_3$-N | 0.111 | −0.0072 | 0.00080 | | 0.651 | 20.52 | 2,22 | <0.001 |
| O horizon | $NO_3$-N | 0.069 | 0.0227 | | | 0.793 | 14.54 | 5,19 | <0.001 |
| O horizon | $NH_4$-N | 0.223 | 0.0122 | | | 0.558 | 29.07 | 1,23 | <0.001 |
| O horizon | inorg.-N | 0.423 | −0.0514 | 0.00477 | −0.0000613 | 0.939 | 37.52 | 7,17 | <0.001 |
| Throughfall[a] | $NO_3$-N | −0.127 | 0.0502 | −0.00057 | | 0.657 | 16.31 | 2,17 | <0.001 |
| Throughfall[a] | $NH_4$-N | 1.059 | −0.1019 | 0.00545 | −0.0000684 | 0.558 | 6.76 | 3,16 | <0.01 |
| Throughfall[a] | inorg.-N | 1.549 | −0.1269 | 0.00749 | −0.0000951 | 0.663 | 10.49 | 3,16 | <0.001 |
| Fluxes | | | | | | | | | |
| Throughfall[a] | $NO_3$-N | 2.81 | 0.115 | | | 0.889 | 8.89 | 9,10 | <0.01 |
| Throughfall[a] | $NH_4$-N | 5.54 | 0.067 | | | 0.808 | 4.68 | 9,10 | <0.05 |
| Throughfall[a] | inorg.-N | 8.34 | 0.182 | | | 0.894 | 9.38 | 9,10 | <0.001 |

[a] Moorland site data excluded from regression model.

during low or high concentration periods. To overcome these problems, least square annual means have been produced for each stratum (i.e. rain, throughfall, O horizon water, Bs horizon water, streamwater) of each of the 25 sites.

Modelling of inorganic-N concentrations (and fluxes in rain and throughfall) was undertaken in the GENSTAT package (Payne *et al.*, 1987) using a multiple regression approach. The data were examined principally for their relationship with stand age and with other physical and crop characteristics recorded at each site, such as elevation, aspect, slope, thinning etc. The possibility of forest district effects was also examined using forest as a 'factor' in the regression models. Model construction involved a mixture of stepwise techniques and eclectic philosophy (Gilchrist, 1984), i.e. only terms with biological relevance and which were implicitly meaningful were included, and the principle of parsimony was employed, i.e. the number of terms kept small. In all cases inspection of residuals to check for model adequacy was carried out using the methods suggested by Draper and Smith (1991). Interactions with forest district were included in the model where such an effect was significant. Model parameters have been presented after averaging coefficients, where appropriate, over the five forest districts.

## 3. Results

Stream water $NO_3^-$ concentration and age of forest were related in a highly significant manner (Table II and Figure 2). Annual mean $NO_3^-$ concentrations were less than 0.2 mgN $L^{-1}$ in the moorland sites (plotted as age zero in this and all sub-

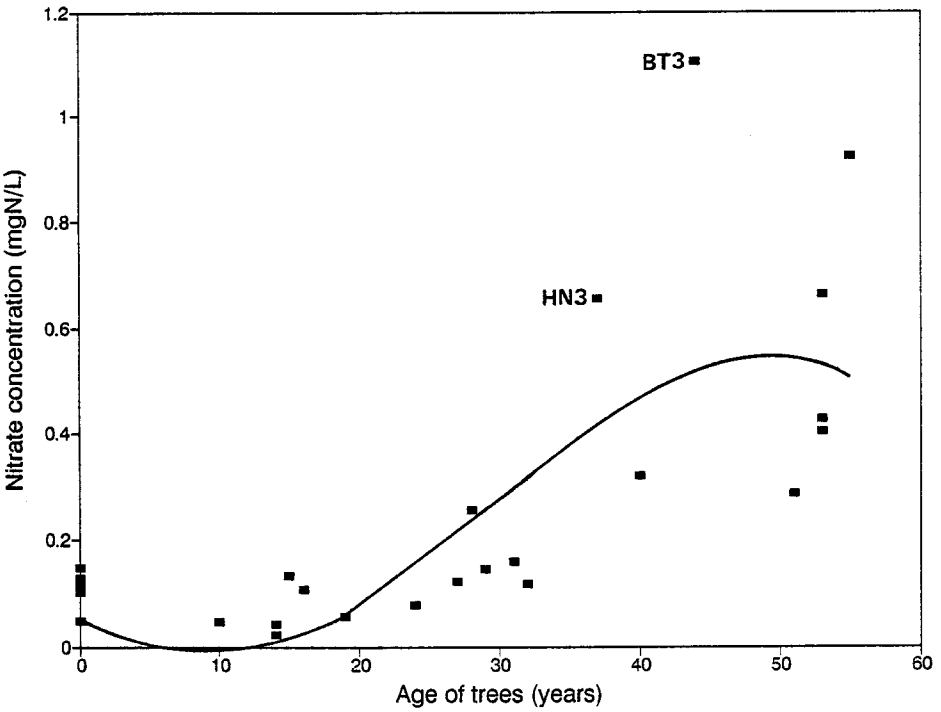


Fig. 2. Mean $NO_3^-$ concentrations in streams (mgN $L^{-1}$). The regression equation is $y = 0.128 - 0.0296x + 0.00172x^2 - 0.0000191x^3$.

sequent Figures) and in younger forests, and up to 1.1 mgN $L^{-1}$ in forests older than 32 yr (Figure 2), The regression equation (Table II) contained quadratic and cubic terms, indicating a 'tailing off' in concentration in the oldest forests and a slight reduction compared with the moorland streams in the 10–14 yr old forests. Data points on Figure 2 fit quite closely to the regression curve, apart for sites HN3 and BT3. However, much of the forest in the catchment area of HN3 was thinned shortly before our observations were made, possibly inducing a pulse of nitrification and $NO_3^-$ leaching. We have no explanation for the high concentration at site BT3, however – thinning at the site had taken place several years previously and any pulse of nitrification should have passed.

An additional source of variation in the concentrations of $NO_3^-$ in the streams draining older sites was that some catchments contained a proportion of moorland in addition to the areas of forest; water draining from moorland had low concentrations and would have diluted that draining the forested areas. However, the proportion of moorland could not have been an important factor as there was no significant relationship between the residuals from the regression analysis and percent moorland ($F_{1,16}=2.57$, $p>0.10$).

Inorganic-N in the streams was almost entirely as $NO_3^-$, as $NH_4^+$ concentrations were below the analytical detection limit of 0.1 mgN $L^{-1}$ on virtually all sampling occasions at all 25 sites.

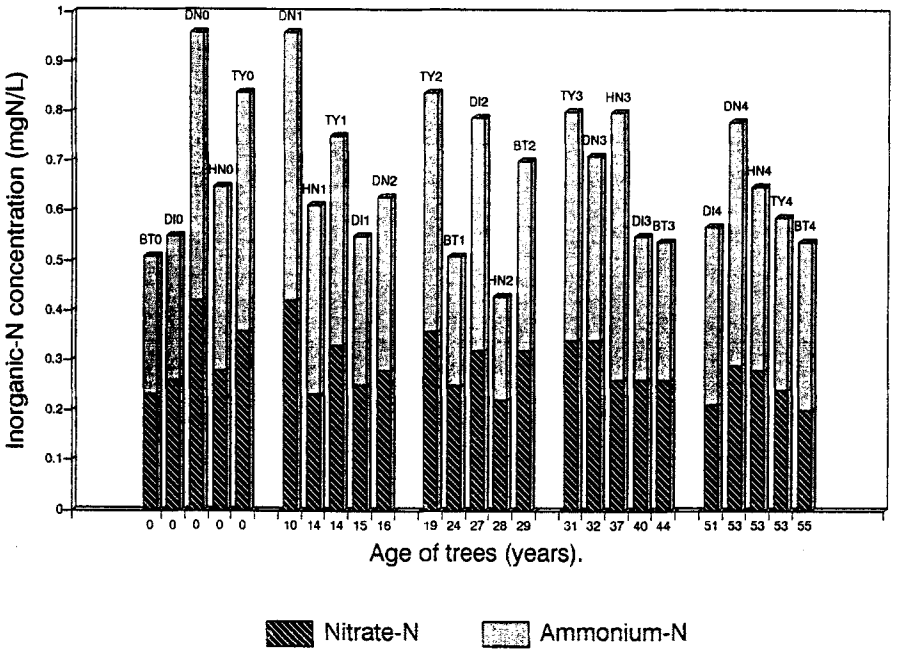


Fig. 3. Mean $NO_3^-$ and $NH_4^+$ concentrations in rain (mgN $L^{-1}$).

The observed relationship between forest age and $NO_3^-$ in stream water cannot be explained through site-to-site variations in the inorganic-N content of rain. There were no significant relationships ($p>0.05$) between inorganic-N concentrations and fluxes (or $NO_3^-$ and $NH_4^+$ individually) and forest age and this is confirmed by Figures 3 and 4.

Rain at all 25 sites was composed of approximately equal concentrations of $NO_3^-$ and $NH_4^+$, irrespective of the total inorganic-N concentration (Figure 3). Inputs ranged between 7 and 13 kgN $ha^{-1}$ $yr^{-1}$ (Figure 4), the largest being at sites BT0 and BT4 where the annual rainfall was substantial (almost 3000 mm $yr^{-1}$).

In contrast to the rain data, there were significant positive relationships between inorganic-N concentrations in throughfall (both $NO_3^-$ and stand age in the 20 forest sites (Table II). The regression analysis of throughfall data excluded that from the five moorland sites because we suspected contamination by sheep urine: inorganic-N in moorland throughfall samples was dominated by $NH_4^+$ (Figure 5) and although certain samples or analyses were rejected when contamination was obvious, some carry-over was inevitable. The moorland throughfall data must therefore be regarded with suspicion.

Inorganic-N concentration in throughfall was greater than that in rain at all 20 forest sites, except the 10 yr old site DN0 (Figure 6). The difference was small in the other young stands, but substantial (a 2- to 5-fold increase) in all other sites. Nitrate and $NH_4^+$ concentrations were approximately equal in throughfall

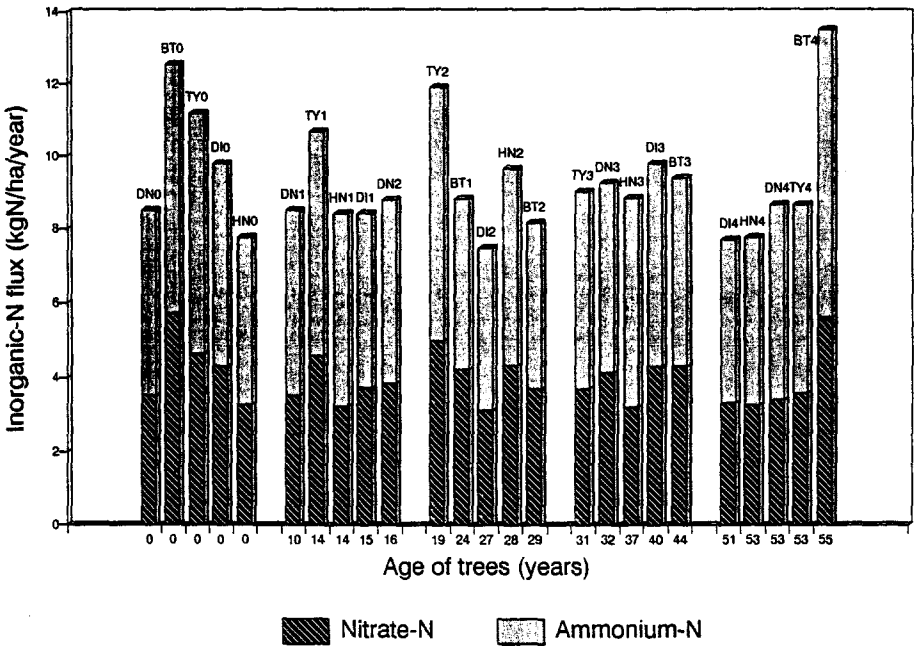


Fig. 4. Nitrate and $NH_4^+$ fluxes in rain (kgN $ha^{-1}$ $yr^{-1}$).

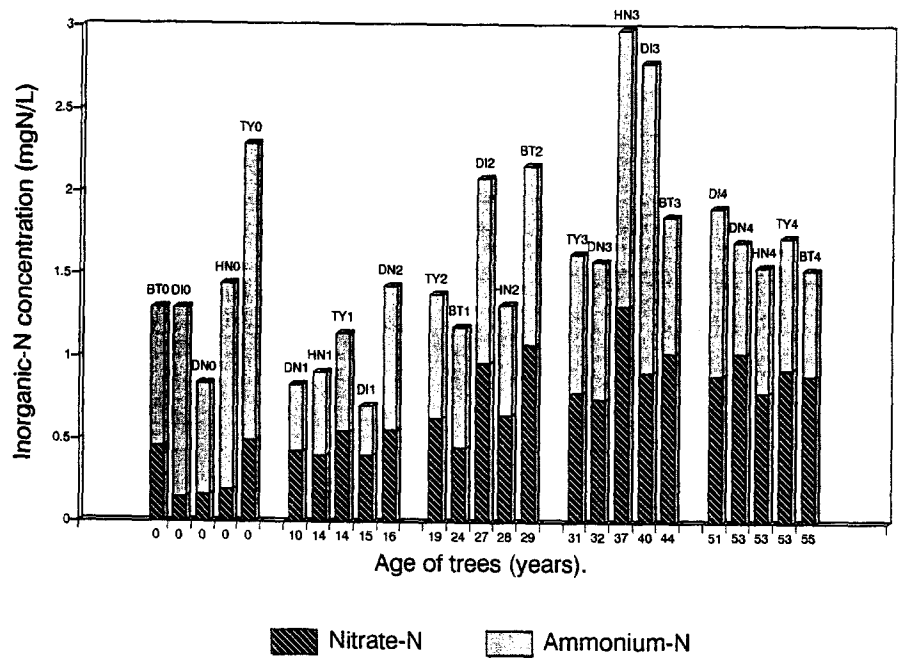


Fig. 5. Mean $NO_3^-$ and $NH_4^+$ concentrations in throughfall (mgN $L^{-1}$).

TABLE III

Rain and throughfall amounts (mm $yr^{-1}$) in the forest sites

| Site | Age of crop (yr) | Rainfall (mm $yr^{-1}$) | Throughfall (mm $yr^{-1}$) | Interception (%) |
|---|---|---|---|---|
| DN1 | 10 | 1786 | 1715 | 4 |
| HN1 | 14 | 2077 | 2172 | -5 |
| TY1 | 14 | 2000 | 1962 | 2 |
| DI1 | 15 | 2159 | 1715 | 21 |
| DN2 | 16 | 1795 | 1259 | 30 |
| TY2 | 19 | 2000 | 1153 | 42 |
| BT1 | 24 | 2289 | 1262 | 45 |
| DI2 | 27 | 1598 | 1012 | 37 |
| HN2 | 28 | 2482 | 1774 | 29 |
| BT2 | 29 | 2096 | 1270 | 39 |
| TY3 | 31 | 1783 | 1428 | 20 |
| DN3 | 32 | 1974 | 1294 | 34 |
| HN3 | 37 | 1927 | 910 | 53 |
| DI3 | 40 | 2253 | 1847 | 18 |
| BT3 | 44 | 2036 | 1398 | 31 |
| DI4 | 51 | 1692 | 1073 | 37 |
| TY4 | 53 | 1742 | 1127 | 35 |
| DN4 | 53 | 1662 | 1350 | 19 |
| HN4 | 53 | 1790 | 1444 | 19 |
| BT4 | 55 | 2874 | 2043 | 29 |

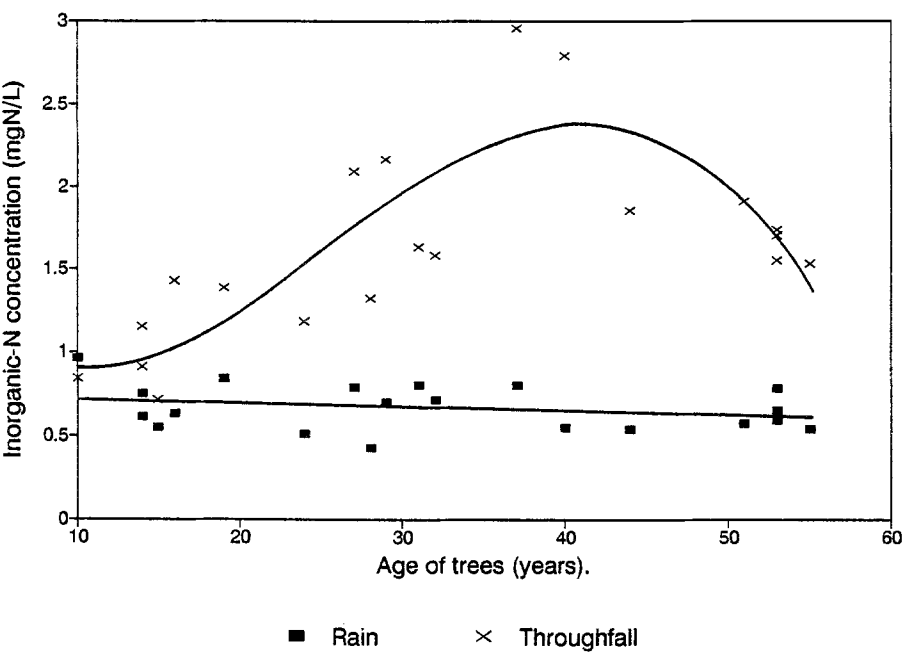


Fig. 6. Mean inorganic-N ($NO_3^-$ plus $NH_4^+$) concentrations in rain and throughfall (mgN $L^{-1}$), moorland sites excluded. Regression equations are $y = 0.748 - 0.00258x$ for rain and $y = 1.549 - 0.1269x + 0.00749x^2 - 0.0000951x^3$ for throughfall.

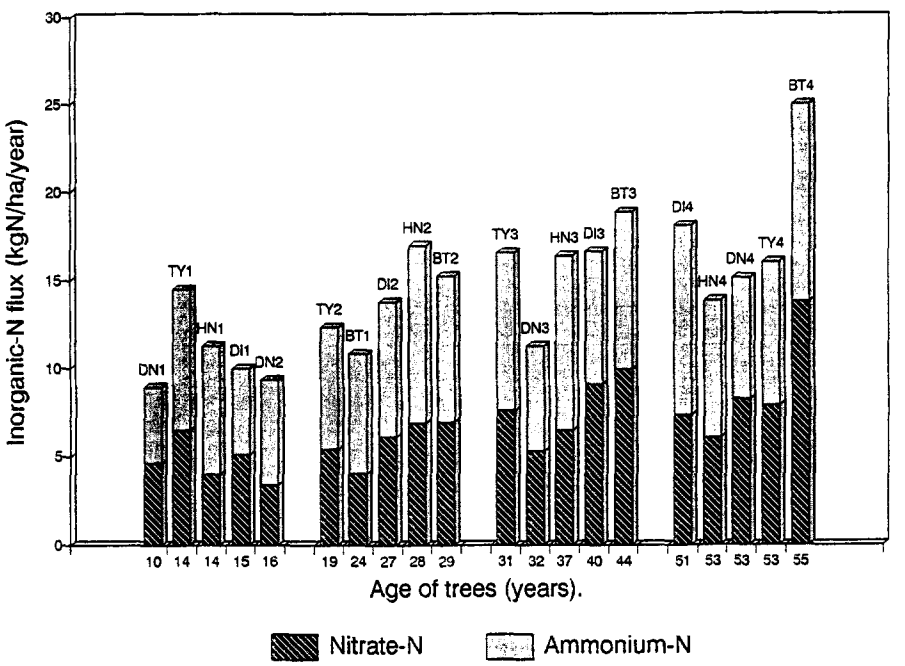


Fig. 7. Nitrate and $NH_4^+$ fluxes in throughfall (kgN $ha^{-1}$ $yr^{-1}$), moorland sites excluded.

of the forest sites, as in rain, despite the overall increase with stand age (Figure 5). There was considerable variation in inorganic-N concentration from site to site, especially in 31-45 yr-old sites. This variation may be related partly to the presence or absence of thinning: the regression models suggest that $NO_3^-$ and $NH_4^+$ were both higher in thinned stands.

Throughfall inorganic-N fluxes (and $NO_3^-$ and $NH_4^+$ individually) also increased with stand age (Table II, Figure 7), despite substantially greater canopy interception of incident rainfall in intermediate-aged and older stands (Table III). Interception was negligible in the youngest crops, and as much as 53% in the older stands, but there was also considerable site-to-site variation. The latter is not surprising considering the range of canopy structures present; these included unthinned and thinned crops, one site which was partially windthrown during the study year (DI3), and also forest canopies partly defoliated by aphid attack.

Comparison of inorganic-N fluxes in throughfall with those in rain (Figure 8), shows that the former were greater at all forest sites. The difference was small in the younger forests, but in those older than 20 yr, inorganic-N fluxes in rain averaged 9.1 kgN $ha^{-1}$ $yr^{-1}$ whereas throuhgfall inorganic-N fluxes averaged 16.1 kgN $ha^{-1}$ $yr^{-1}$. The comparatively extreme figure for site BT4, where throughfall inorganic-N flux was 25.1 kgN $ha^{-1}$ $yr^{-1}$, is not unexpected. This was the site where 23 kgN $ha^{-1}$ $yr^{-1}$ of inorganic-N was measured in throughfall in the early 1980's (Stevens *et al.*, 1989).

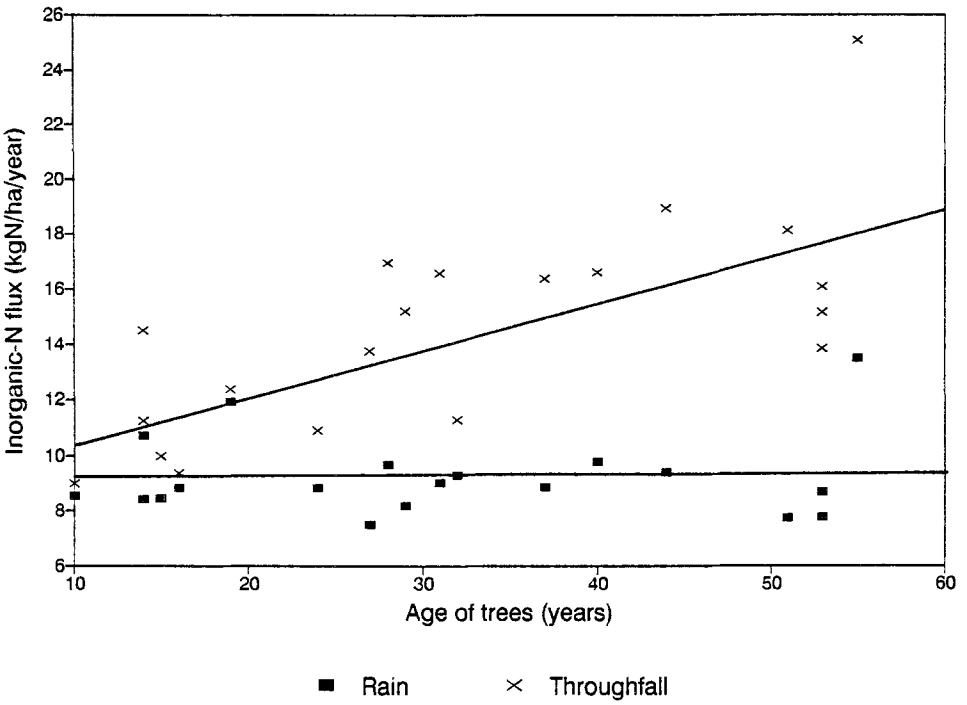


Fig. 8. Inorganic-N fluxes ($NO_3^-$ plus $NH_4^+$) in rain and throughfall (kgN $ha^{-1}$ $yr^{-1}$), moorland sites excluded. Regression equations are $y = 9.029 + 0.005x$ for rain and $y = 8.34 + 0.182x$ for throughfall.

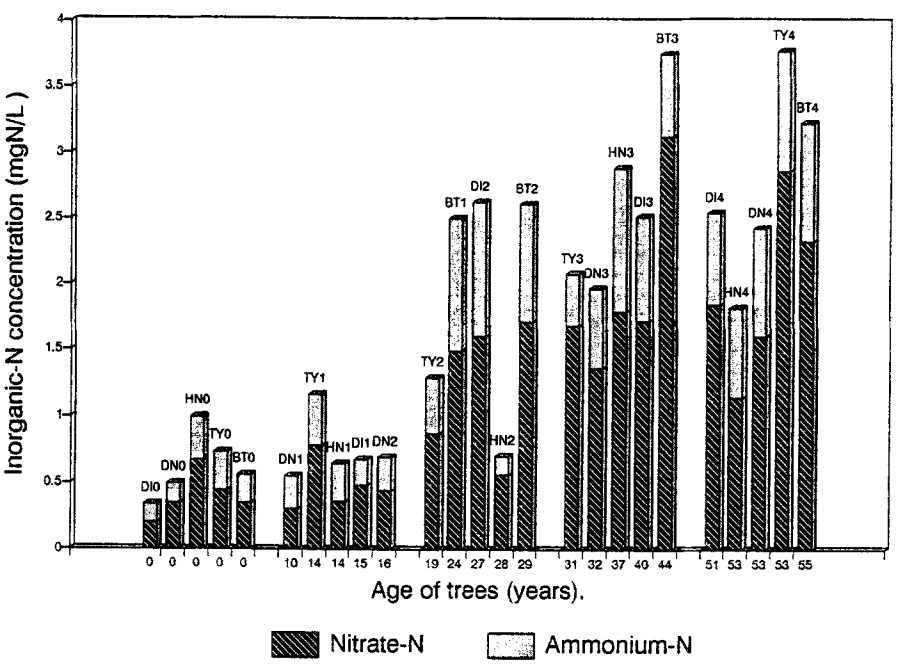


Fig. 9. Mean $NO_3^-$ and $NH_4^+$ concentrations in O horizon soil waters (mgN $L^{-1}$).

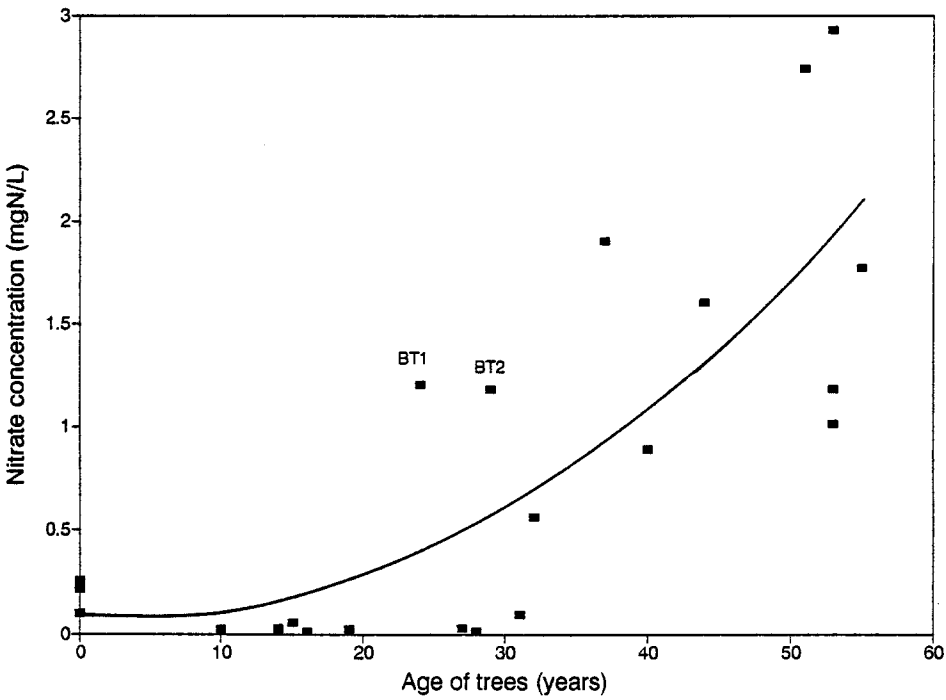


Fig. 10. Mean $NO_3^-$ concentrations in Bs horizon soil waters (mgN $L^{-1}$). Regression equation is $y = 0.111 - 0.0072x + 0.0008x^2$.

Inorganic-N concentrations in waters from the peaty surface O horizon also increased significantly with stand age (Figure 9). Concentrations in moorland sites and in the 0–16 yr old forest age group were generally less than 1.0 mgN $L^{-1}$, but were almost 4.0 mgN $L^{-1}$ in some older stands. Some of the variation in concentration is explained in the regression model: lower $NH_4^+$ concentrations occurred at lower elevations, but total inorganic-N concentration was greater at sites where trees were taller. At all sites, approximately 30% of the inorganic-N was $NH_4^+$, a lower proportion than in throughfall (around 50%).

In soil water from the mineral soil (Bs horizon), $NH_4^+$ concentration was below the analytical detection limit of 0.1 mgN $L^{-1}$ on most sampling occasions at all sites. Concentrations of $NO_3^-$ were less than 0.4 mgN $L^{-1}$ in moorland and forest sites younger than 28 yr (with the exception of site BT1 which we believe was contaminated by sheep), but increased to nearly 3 mgN $L^{-1}$ in some older forest stands (Figure 10). In the youngest forests, $NO_3^-$ was virtually undetectable and even lower than in the moorland sites. Complete uptake of available nitrogen by these young forests seems to have occurred, aided perhaps by uptake in ground flora which had become quite rank prior to canopy closure. As soon as canopy closure occurred at around 15 years, all ground flora was eliminated by shading and $NO_3^-$ concentrations became more typical of moorland sites.

In the older forests, Bs horizon soil water $NO_3^-$ concentrations showed considerable scatter (Figure 10). Apart from the variations in crop and site conditions already referred to, the regression analysis indicates that concentrations were lower at high elevation sites and in sites on steeper slopes.

The pattern observed for $NO_3^-$ concentration with stand age in the Bs horizon soil waters was very similar to that already seen in the streams (Figure 2). Unlike the regression equation for streamwater $NO_3^-$ and stand age which included a cubic term indicating a 'tailing-off' in concentration in the oldest forests, the regression equation for Bs horizon $NO_3^-$ concentration and stand age does not include a cubic term, so a 'tailing off' in the oldest sites is not indicated.

## 4. Discussion

Forests in the temperate zone have conventionally been regarded as N limited, which by implication suggests that leaching losses to streams will be small. The limited number of studies of the hydrochemical effects of afforestation in the UK tended to reinforce this view. For instance, $NO_3^-$ concentration in a stream draining a 'mature' spruce plantation at Loch Chon in Central Scotland averaged only 0.14 mgN $L^{-1}$ (Harriman and Morrison, 1982). In contrast, our results from Wales show that streams draining first rotation Sitka spruce plantations older than 30 yr contained significant concentrations of $NO_3^-$.

Relationships between forest stand age and streamwater $NO_3^-$ concentration have also been reported in the north-east of the United States by Vitousek (1977) and Leak and Martin (1975). The former author attributes his results to changes in successional status of the forest stands, with younger stands taking up large quantities of nutrients and a progressive reduction in biomass increment and nutrient uptake occurring in the older stands. These conclusions are in accordance with the theory of Miller (1979) that nutrient uptake is greatest in younger stands. If this theory is applied to the sites in our study, then a reduction in demand for nitrogen by the older crops may increase the availability of nitrogen in the soil, especially if deposition of N from the atmosphere increases with stand age. Nitrification has been shown to occur freely in these Welsh forest soils if $NH_4^+$ is available as the substrate (Emmett *et al.*, 1991; Stevens and Wannop, 1987), despite the low pH, so available nitrogen is likely to be nitrified and leached as $NO_3^-$.

Of the three processes described in the Introduction as providing possible explanations for increased leaching losses from older crops, we have shown that increased throughfall concentrations and fluxes of inorganic-N indeed occur. Inorganic-N concentrations in these older crops (32 yr or older) range from 0.8 to 1.8 mgN $L^{-1}$ in throughfall and 0.5 to 3.0 mgN $L^{-1}$ in the Bs horizon soil water, and sites with high throughfall inorganic-N concentrations tend to have high Bs horizon soil water $NO_3^-$ concentrations ($r_s$ = 0.809, $p<0.001$). Throughfall N may therefore be controlling $NO_3^-$ concentrations in the soils of the older crops, but not necessarily directly. An indirect control may be through the effect of enhanced

throughfall N concentrations on the rates of mineralisation and nitrification in the forest floor and soil organic matter. Any change in forest management or air pollution conditions which increases, even slightly, the rate of mineralisation of this organic matter could have a substantial effect on the availability of N. For instance, stagnopodzol soils in Wales contain large amounts of N in an 'unavailable' form in organic matter. Unpublished data for site BT4 show that there is 300 kgN $ha^{-1}$ in the accumulated spruce litter and F layer, and 4700 kgN $ha^{-1}$ in the surface peat and organic-rich mineral horizons.

The presence of comparatively large amounts of inorganic-N in throughfall tends to be viewed as an indicator of large inputs. However, increased availability of inorganic-N in the soil may, of course, result in increased tree uptake and foliar leaching of inorganic-N. It is not possible, therefore, to conclude that the increasing inorganic-N concentrations and fluxes in the throughfall of the older crops are entirely the results of increased atmospheric nitrogen deposition.

Studies of rain and throughfall in areas remote from sources of N pollution normally record net uptake of N by the canopy, even in very old spruce stands (eg. Carleton and Kavanagh, 1990). At all of our forest sites, throughfall inorganic-N flux was greater than that in rain. The most likely explanation for this phenomenon is that amounts of dry and occult deposition are substantial, even though the study sites in Wales are remote from the most intensive sources of $NO_x$ and $NH_3$. Support for this explanation is provided by Fowler *et al.*, 1989, whose modelling studies showed that at Kielder forest in northern England (an area similarly remote from industry, roads and intensive agriculture), atmospheric nitrogen inputs to a 15 m high Sitka spruce forest were almost double those to adjoining moorland. Most of the increase was in the form of of dry deposition, especially of $NH_3$. Further modelling studies are currently in progress (B. Reynolds, personal communication) which are attempting to estimate dry and cloudwater deposition to the 20 forest sites in the authors' study.

The source of the $NO_3^-$ leached from the soils and streams of the older crops remains undetermined. However, since uptake of N by older crops is almost certainly less than in younger crops, then the high soil inorganic-N concentrations in the older crops probably indicate a pool of soil N which is surplus to the vegetation requirements. This excess may be intensified if atmospheric inputs and rates of mineralisation and nitrification also increase in the older crops. The importance of rates of uptake of N in the standing crop may be illustrated using data from site BT4 at Beddgelert. Mean annual input of inorganic-N in wet deposition at this site was measured as 10.3 kgN $ha^{-1}$ $yr^{-1}$ from 1982-1990 inclusive (Stevens *et al.*, 1993b). Assuming a similar rate of input throughout the 50 yr period of crop growth, 515 kgN $ha^{-1}$ would enter the system. Over the same period, the crop accumulated only 428 kgN $ha^{-1}$ in above-ground biomass (Stevens *et al.*, 1988). Allowing for some N immobilisation in roots and forest floor, it is likely that the crop can theoretically keep pace with this level of atmospheric inputs. However, if the true N input is greater than that measured in rain, for example if throughfall

in the older crops contains dry and occult deposited N, then extra will be entering the system. The crop may be able to respond with increased growth and nitrogen uptake, but there are likely to be both climatic constraints to faster growth rates, and those caused by deficiencies of P and K which limit the ability of these older plantations to take up more N (Stevens *et al.*, 1993a).

In addition, since the crop N requirement is theoretically satisfied by wet deposition inputs, any mineralised N will be surplus to tree requirements. This surplus N may be immobilised by the soil microbial population, but this is only a short term sink as the death of these microbes will only add to the soil organic matter pool and this will subsequently mineralise.

Our results show that available N (from whatever source) in the soil of younger crops and moorland sites is immobilised, resulting in negligible $NO_3^-$ leaching losses. In these younger crops, rapid increase in biomass occurs and available N is fully utilised. In older crops, atmospheric inputs combined with mineralised N are present in quantities in excess of tree demand, nitrification is stimulated, $NO_3^-$ leaches and soil and water acidification occurs. In the long term, if current forest practice is continued, only a reduction in atmospheric N inputs can ameliorate this situation.

The importance of these results is that the soils and streams at the older forest sites are becoming acidified. There is no direct human health problem with the enhanced $NO_3^-$ concentrations in streams: the highest annual mean for any site was 1.1 mgN $L^{-1}$, and the highest concentration of any one individual streamwater sample (collected during a storm event) was 1.5 mgN $L^{-1}$. These values are well below the 5.7 and 11.3 mgN $L^{-1}$ guide level and maximum admissible concentration respectively for drinking water stipulated by the EEC (Commission of the European Communities, 1980). Even after harvesting older spruce forests, peak nitrate concentrations rarely, if ever, reach the EEC maximum admissible concentration (Stevens *et al.*, 1988; Neal *et al.*, 1992). Also, $NO_3^-$ comprises only a small proportion of the total anion concentration in streams even in the older sites (eg. 9% at Beddgelert site BT4, Stevens *et al.*, 1993b). There should only be a small increase in cation leaching to accompany the elevated anion concentrations, although this might be sufficient to increase $Al^{3+}$ leaching.

Of greater concern is acidification of the soil as a result of N transformations. For each mole of $NO_3^-$ produced from $NH_4^+$ inputs or mineralisation and nitrification of soil organic matter, and subsequently leached from the soil, two $H^+$ are generated. The rate of acidification from this process may be estimated as the net ammonium input plus the net nitrate output (van Breemen *et al.*, 1983; Berdén *et al.*, 1987). For site BT4, we estimate from data in Stevens *et al.* (1993b) that the rate of acidification due to these N transformations is 1.01 keq $ha^{-1}$ $yr^{-1}$, substantially more then acidification due to sulphate deposition in rain (0.37 keq $ha^{-1}$ $yr^{-1}$) or base cation uptake by the tree crop (0.51 keq $ha^{-1}$ $yr^{-1}$) at this site.

These results need to be assessed in relation to the current debate on Critical Loads (Nilsson, 1986; Nilsson and Grennfelt, 1988; Sverdrup *et al.*, 1990). Critical Loads for *acidity* are estimates of the ability of soils to neutralize acidity, what-

ever its source. The acidity is neutralized by weathering of soil minerals and the rate of neutralization clearly depends upon the mineralogical composition of the soil and its parent material. Maps showing the Critical Loads for acidity of soils in the UK (Hornung, 1991) are based upon five mineralogical classes of soil materials (Nilsson and Grennfelt, 1988). Soils at Beddgelert and in many other upland areas of Wales have a Critical Load of 0.2 or 0.5 keq $ha^{-1}$ $yr^{-1}$. From the rates of acidification given earlier, it can be seen that the Critical Load for acidity is exceeded by the sulphur component of acid deposition plus base cation uptake in trees, with N transformations exacerbating the problem considerably.

There may be an additional problem associated with second rotation and subsequent crops. Of the 428 kgN $ha^{-1}$ in the above ground biomass of the 50 year old trees at Beddgelert, only 128 kgN $ha^{-1}$ is in the bole. This is removed when conventional harvest takes place, leaving 300 kgN $ha^{-1}$ in brash. This brash is a source of organic-N not present in first rotation crops. If readily mineralised, availability of N in second and later rotations will be greater than in first rotation crops.

To ensure nutritional requirements of the crop for nitrogen are not exceeded, and nitrate leaching losses and acidification minimised, it may be argued that inputs of N from the atmosphere should be no larger than the amount of N which is removed in conventional harvest, or 2.5 kgN $ha^{-1}$ $yr^{-1}$. This has significant implications for the current debate on Critical Loads for nitrogen (Hettelingh *et al.*, 1991), and especially attempts to set a Critical Load for nitrogen as a *nutrient*. This has been defined (Rosén *et al.*, 1992) as:

$$CL(N) = N_u + N_i + (Q \times [NO_3^-]_{crit})$$

where

CL(N) = Critical Load for nitrogen as a nutrient
$N_u$ = Net uptake of N in biomass (kgN $ha^{-1}$ $yr^{-1}$)
$N_i$ = Long-term immobilisation of N in the soil (kgN $ha^{-1}$ $yr^{-1}$)
$Q$ = Precipitation surplus (mm)
$[NO_3^-]_{crit}$ = Critical nitrate concentration which protects against harmful effects. (mg $NO_3^-$ $L^{-1}$)

Applying this equation to data from Beddgelert ($N_u$ for conventional harvest = 2.6; $N_i$ = 0.5; $Q$ = 1770; $[NO_3^-]_{crit}$ = 0.3), a Critical Load for nitrogen as a nutrient of 4.3 kgN $ha^{-1}$ $yr^{-1}$ is obtained. This amount is clearly significantly less than the amount of N entering the system in wet deposition, and substantially less than in throughfall. To guard against problems associated with over-supply of N from the atmosphere, a substantial reduction in inputs to a level more characteristic of pristine areas of the globe is required.

The rain and throughfall data presented here clearly demonstrate that, assuming throughfall provides a reasonable estimate of the total atmospheric nitrogen inputs, these inputs to mature Sitka spruce plantations are double those in young stands. Presumably dry and cloudwater deposition (Fowler *et al.*, 1989) are responsible.

Because of the problems encountered in measuring quantities and chemical composition of throughfall beneath vegetation canopies on the moorland sites, we cannot however conclude from our data that atmospheric nitrogen inputs to forests are greater than to moorland systems, although this is almost certainly the case.

## 5. Conclusions

A clear relationship has been established between Sitka spruce crop age and concentrations of inorganic-N (entirely $NO_3^-$) in streams and Bs horizon soil waters. Inorganic-N concentration is negligible in moorland and crops younger than 30 yr, but increases substantially in older crops. A similar trend is observed in O horizon soil waters, but approximately one third of the inorganic-N is $NH_4^+$. Nitrification is active in the soils of these older crops. Concentration and fluxes of inorganic-N both increase approximately linearly with increasing tree age in throughfall of the forest sites, whereas there is no relationship between rain concentration and fluxes with crop age. There are approximately equal amounts of $NH_4^-$ and $NO_3^-$ in both throughfall and rain. The amount of inorganic-N in throughfall is substantially more than in rain in older crops, but equal to, or only slightly more, in younger crops. The most likely explanation is that dry and cloud-water deposition of N is greater than in younger crops.

Nitrogen is available in soils of the older crops in excess of the ability of the system to immobilise it. Leaching of $NO_3^-$ will result in acidification of the soil if accompanied by leaching of base cations, or acidification of water if accompanied by $H^+$ and $Al^{3+}$. Current N inputs to old forests are in excess of those which can be immobilised in the above ground tree components, and substantially in excess of that immobilised in the harvested portion. Critical Loads for Nitrogen as a nutrient are exceeded in these forest systems. A reduction in inputs to those more characteristic of pristine sites is theoretically necessary to ensure soil acidification is minimised.

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