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Deposition of acidifying and eutrophicating substances in Dutch forests

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SUMMARY

In order to qualify and quantify the acidifying and eutrophicating substances that contribute to atmospheric deposition in Dutch coniferous forests, open field precipitation and throughfall water have been sampled monthly at 14 locations in The Netherlands over an 18-month period. Spatial variation was studied by comparing the chemical composition of deposition in forests of four regions. Within the forests, throughfall fluxes of different tree species were compared. In this paper the deposition fluxes of sulphuric and nitrogenous compounds are emphasized.

This study proves that most forested areas in The Netherlands receive high loads of nitrogenous (5·6 kmol N ha^{-1} y^{-1}) and potentially acidifying (9·4 kmol H^+ ha^{-1} y^{-1}) compounds. Particularly, deposition of ammonium and sulphate is high, not only in areas with intensive agricultural activities, but throughout the country. Regional differences in deposition appear to be relatively small. With the exception of the two coastal locations, deposition shows little spatial variation. In the coastal region loads of nitrogenous and potentially acidifying compounds are lower than elsewhere in the country. Generally higher throughfall fluxes have been measured in Douglas fir stands than in pine stands. The larger surface area of firs probably accounts for an enhanced dry deposition. In the summer season throughfall fluxes are generally smaller than in the winter season.

Key-words: nitrogenous compounds, potentially acidifying compounds, seasonal variation, spatial variation, throughfall fluxes.

INTRODUCTION

The enormous increase of pollutants in the atmosphere during the last decade has caused a dramatic change in the chemical composition of atmospheric deposition. Ozone, nitrogen oxides, and sulphur dioxide are accepted to be the pollutants affecting the environment on a global scale.

In The Netherlands the first observations of forest damage were made in the south-eastern part of the country. The regional character of this forest decline indicated a regional cause. None of the above mentioned pollutants fitted the damage pattern. However, ammonia emission and ammonium deposition appeared to be extremely high in the affected area. Recent research has indicated that a high atmospheric ammonium input

leads to acidification and eutrophication of the forest soil and consequently adversely affects the vegetation (Van Breemen *et al.* 1982; Roelofs *et al.* 1985).

In The Netherlands, 91% of ammonia present in the atmosphere is directly emitted from livestock farms or evaporates from liquid-manured, arable land (Ivens 1990). While most ammonia emitting sources are situated near ground level, atmospheric transport of this compound was always supposed to be small compared to that of other pollutants. Consequently deposition of both ammonia and ammonium was expected to take place at a relatively short distance from the source. The local character of the initial forest damage confirmed these assumptions. Asman (1987) found that gaseous ammonia is indeed deposited close to the source, but that ammonium sulphate aerosoles can be transported over long distances.

On wet plant surfaces or in humid air ammonia reacts with sulphur dioxide to form ammonium sulphate. Consequently large amounts of ammonium and sulphate are filtered from the atmosphere by the tree canopy (Van Breemen *et al.* 1982; Grennfelt & Hultberg 1986; Draaijers *et al.* 1988). Apart from ammonium and sulphate, fluxes of various other compounds are altered due to down-wash of particles or gases deposited on the plant surface and exchange of substances by the tree canopy (Freiesleben *et al.* 1986; Lindberg *et al.* 1986). After the passage of rainwater through the canopy the actual load of pollutants on the forest floor can be estimated. Leaving aside canopy exchange, throughfall measurements give good estimates of atmospheric deposition as stemflow contributes at most 8% to the total load on forest soils and, therefore, can be neglected. (Van Breemen *et al.* 1982; Miller 1984; Bredemeier 1988).

Investigations of the Dutch State Forest Service have shown that forest deterioration is no longer restricted to areas with dense concentrations of agricultural activities but also has expanded to other parts of the country (Anonymous 1984, 1990). The major aim of the present research was to find out whether forest damage in those areas could also be caused by an enhanced ammonium deposition. Therefore, qualification as well as quantification of atmospheric input to Dutch forests was essential. To investigate which acidifying and eutrophicating substances contribute to atmospheric deposition in forests, throughfall collectors were used. In order to establish regional differences in throughfall deposition, collectors were placed at 14 locations all over the country. Variation in throughfall fluxes within a forest was studied by comparing throughfall deposition under three coniferous tree species. The seasonal variability of atmospheric deposition will also be discussed.

MATERIALS AND METHODS

In November 1986, 14 forest locations in The Netherlands were selected. All forest stands consisted of coniferous trees planted on weakly buffered, sandy soils. The investigation concerned Corsican pine (*Pinus nigra* var. *maritima* (Ait.) Melville), Scots pine (*Pinus sylvestris* L.) and Douglas fir (*Pseudotsuga menziesii* (Mirb.) Franco). The selection of the sampling sites was primarily based on the occurrence of all three coniferous tree species within a distance of 5 km from each other. At each location one representative specimen of each species was chosen for the collection of throughfall water. The influence of local ammonia sources such as livestock farms and liquid-manured arable land was restricted by selecting relatively large forests and avoiding the edges of the forest. Furthermore, 25–40-year-old stands were chosen to diminish variations caused by age differences. However, neither tree vitality nor the occurrence of illnesses or infections were considered when selecting the sites.

Fig. 1. Geographical position of the sampling locations and their regional division in The Netherlands.
Coastal region: 1 = Terschelling, 2 = Schoorl
Northern region: 3 = Bakkeveen, 4 = Diever, 5 = Orvelte, 6 = Hardenberg
Central region: 7 = Lunteren, 8 = Loenen, 9 = Heumen
Southern region: 10 = Hulst, 11 = Ossendrecht, 12 = Breda, 13 = Maarheeze 14 = Meyel.

After selection of the stands, the locations were grouped into four regions according to their geographical situation; Coast, North, Centre and South. Figure 1 shows the distribution of the sampling locations.

Deposition collectors were placed in October 1986. For 18 months, open field precipitation (bulk) as well as throughfall water were collected. At each location, one rain sampler was placed in a clearing near the forest stand and two samplers were placed within the forest stand at 0·75 m distance from the stem of the selected tree, for this purpose.

The collectors consisted of a black, 2 l polyethylene bottle and a polyethylene gauge with an aperture of 74·5 cm^2. On the bottom of this gauge, a plastic filter was inserted in order to avoid organic matter and/or insects contaminating the water in the bottle. Bottle and gauge were placed in a plastic pipe dug into the ground. The opening of the gauge was

20 cm above ground level. In each collecting bottle, 2 ml of a solution containing 200 mg l^{-1} mercury chloride was added to inhibit microbial activity. After each month, the samples were collected and the bottles and gauges were cleaned with demineralized water. The volume was measured in the field, pH was determined in the laboratory within 24 h of collection. After that, samples were stored at −20°C until further analysis.

A Radiometer type PHM 82 standard pH-meter and a combined electrode was used to measure pH. Calcium, magnesium, aluminium, phosphorus and sulphur were measured with an Inductively Coupled Plasma Spectrophotometer(ICP), type IL Plasma 200. To check that the ICP could be used to estimate the sulphate content of the rainwater a colourimetrical method using a Technicon AAI-system (Anonymous 1969) was employed. Potassium and sodium were estimated using a Technicon Flame photometer IV. Colourimetrical determination with a Technicon AAII-system according to Technicon Methodology (Anonymous 1969) and Kempers & Zweers (1986) was used for nitrate and ammonium respectively, and a Technicon AAI-system according to Technicon Methodology (Anonymous 1969) and O'Brien (1962) for chloride.

Acidifying sulphuric and nitrogenous compounds amount to the total load of potentially acidifying compounds (potential acid) in throughfall water. As part of these acidifying compounds are counteracted by the deposition of alkaline substances, the total flux of potential acid can be calculated as the summed volume weighted deposition of $2 \times SO_4 + NH_4 + NO_3 - 2 \times Ca$ in kmol ha^{-1} y^{-1} (Buysman 1990).

Leaving aside canopy interaction, throughfall deposition is equivalent to total atmospheric deposition in forests. Total deposition can be subdivided into wet and dry deposition (Ivens 1990). According to Bredemeier (1988) for some constituents the canopy may act like an inert sampler, all dry deposited material is washed off the surfaces during rain events and appears in throughfall water. This can be assumed for elements that are deposited at very high rates compared to their internal cycle (e.g. sulphate in Central Europe). Ivens (1990) suggests that this probably also applies for the nitrogen deposition in The Netherlands. Based on these assumptions, dry deposition of sulphuric and nitrogenous compounds can be calculated as the difference between throughfall and bulk deposition.

Annual deposition fluxes were calculated as the sum of all measured monthly concentrations multiplied by the monthly volume of precipitation. Seasonal fluxes were studied by comparing two winter seasons (November 1986–April 1987 and November 1987–April 1988) with one summer season (May 1987–October 1987). Throughfall deposition per tree was calculated as the mean flux of two similar collectors. From annual or seasonal fluxes at the sampling sites, 50 percentile (median) values were calculated. Prior to statistical analysis, values of these fluxes were log-transformed to make the variance independent of the mean (Sokal & Rohlf 1981). Scheffé's multiple-comparison procedure was performed to establish the influence of regional and temporal variability on various components of the deposition. These statistical analyses were performed with the General Linear Models (GLM) procedure available in the Statistical Analysis System (SAS) software package (Anonymous 1985).

RESULTS

Regional differences

Between November 1986 and November 1987 the amount of precipitation measured in The Netherlands by using bulk collectors was 1060 mm (Table 1). In forests, 669 mm of

Table 1. Median values (50% percentile) of the annual amount of precipitation (mm) collected in bulk and throughfall samplers between November 1986 and November 1987 on a regional and national scale based on 14 locations in The Netherlands. None of the differences within the columns are significant according to Scheffé's multiple-comparison procedure

	Bulk precipitation	Corsican Pine	Scots Pine	Douglas Fir	Median throughfall
Coastal region	812	627	539	—	608
Northern region	1067	779	695	674	728
Central region	1125	710	753	550	715
Southern region	1044	511	719	669	663
National	1060	626	719	652	669

Table 2. Median fluxes of potential acid and total nitrogen in bulk and throughfall collectors and the amount of dry deposited potential acid in mol $ha^{-1} y^{-1}$ between November 1986 and November 1987 on a regional and national scale based on 14 locations in The Netherlands. Different letters within each column indicate statistical difference at the 5% level according to Scheffé's multiple-comparison procedure

	Potential acid			Total nitrogen	
	Bulk	Throughfall	Dry deposition	Bulk	Throughfall
Coastal region	1516a	5382a	3458a	925a	3548a
Northern region	2151ab	9990ab	7729ab	1415b	5978ab
Central region	2748b	10130b	7644ab	1480b	5952ab
Southern region	2416b	11222b	8842b	1325b	6544b
National	2154	9453	7187	1366	5556

throughfall water was collected, indicating that 33% was lost due to interception by the canopy. In the coastal region the amount of precipitation was lowest in both bulk and throughfall samplers. However, the difference between this and the other regions was not significant. The variation in the amounts of precipitation in stands of different species within the same area was often larger than the variation between the regions.

In the coastal region the deposition of potential acid was lowest, both in bulk and in throughfall. Significant differences in bulk and throughfall could only be established between the coastal region and the central and southern regions (Table 2). In all investigated forests about 75% of potential acid ($\pm 7{\cdot}2$ kmol H $ha^{-1} y^{-1}$) was deposited in a dry form.

In the coastal region the contribution of sulphate and nitrate to the amount of total potential acid in throughfall deposition was highest, 55% and 16% respectively (Table 3). In this region significantly higher sulphate and nitrate and lower ammonium deposition was measured. On an average, sulphate contributed 46%, ammonium 44% and nitrate 10% to the load of potential acid in Dutch forests.

Table 3. Median values of the contribution of acidifying sulphuric and nitrogenous compounds to the total potential acid flux in bulk and throughfall collectors (in percentages) between November 1986 and November 1987 on a regional and national scale based on 14 locations in The Netherlands. For the explanation of significance symbols see Table 2

	Bulk			Throughfall		
	SO_4	NH_4	NO_3	SO_4	NH_4	NO_3
Coastal region	48a	30a	22a	55a	29a	16a
Northern region	44a	38ab	18a	43c	47b	10b
Central region	43a	40b	17a	45bc	45b	10b
Southern region	41a	41b	18a	49b	42b	9b
National	43	39	18	46	44	10

Table 4. Median fluxes of total nitrogen and potential acid in throughfall collectors of Corsican Pine (cp), Scots Pine (sp) and Douglas Fir (df) in kmol $ha^{-1} y^{-1}$ between November 1986 and November 1987 on a regional and national scale based on 14 locations in The Netherlands. Different letters within a line indicate statistical difference at the 5% level according to Scheffé's multiple-comparison procedure

	Total nitrogen			Potential acid		
	cp	sp	df	cp	sp	df
Coastal region	3·9	3·1	—	5·6	4·8	—
Northern region	5·8	5·2	6·2	9·9	8·9	10·0
Central region	5·1	4·6	9·3	8·8	7·9	14·7
Southern region	4·7	6·0	7·1	7·9	10·4	12·0
National	4·9ab	4·8a	6·7b	8·6a	8·3a	11·3a

In all regions, in throughfall as well as in bulk deposition, annual mean $NH_4/2SO_4$ molar ratios were 1·0 (data not shown).

In the coastal region, the nitrogen flux in bulk deposition was significantly lower than that in the deposition in the other regions (Table 2). In throughfall, nitrogen deposition was only significantly lower in the coastal area compared to the southern region. However, the amount of ammonium deposited at the coast was significantly different from all other regions in bulk as well as in throughfall. In the stands studied, the total nitrogen deposition amounted to $\pm 5{\cdot}6$ kmol $ha^{-1} y^{-1}$ ($=78$ kg N $ha^{-1} y^{-1}$) of which 81% was deposited as ammonium (data not shown).

Differences between tree species

The median annual amounts of nitrogen and potential acid deposited under each tree species per region are shown in Table 4. On a regional scale none of the differences between fluxes per tree species were significant. The median loads of nitrogen and potential acid were highest in the Douglas Fir stands (6·7 kmol N $ha^{-1} y^{-1}$ and 11·3 kmol H $ha^{-1} y^{-1}$

Table 5. Median fluxes per season in bulk and throughfall collectors in mol ha^{-1} 0·5 y^{-1} on a national scale based on 14 locations in The Netherlands. w86 = Winter 1986, s87 = summer 1987 and w87 = winter 1987. Different letters within a row and per sample type indicate statistical difference at the 5% level according to Scheffé's multiple-comparison procedure

	Bulk			Throughfall		
	w86	s87	w87	w86	s87	w87
Potential acid	874a	1290b	890a	5470a	3728b	5013ab
NH_4	418a	571b	420a	2652a	2027a	2755a
NO_3	151a	284b	172c	358a	596b	379a
SO_4	237a	278b	184a	1541a	816b	1222a
Mg	49a	48a	50a	183a	113b	156a
Ca	72ab	77a	50b	242a	163b	207ab
K	69a	123a	104a	342a	512b	315a
Na	440a	215b	520a	1293a	589b	1084a
H	225a	195a	117a	153a	50b	46b
Cl	457a	309a	803b	1183a	676b	1432a

Table 6. Median values of the contribution of acidifying sulphuric and nitrogenous compounds to the total potential acid flux in bulk and throughfall collectors (in percentages) per season on a national scale based on 14 locations in The Netherlands. w86 = winter 1986, s87 = summer 1987 and w87 = winter 1987. For the explanation of significance symbols see Table 2

	Bulk			Throughfall		
	SO_4	NH_4	NO_3	SO_4	NH_4	NO_3
w86	47a	38a	15a	51a	42a	7a
s87	40b	41a	19a	40b	45a	15b
w87	46a	39a	15a	47c	46a	7a

respectively). The median nitrogen fluxes in Corsican and Scots Pine stands were almost similar and amounted to 4·9 and 4·8 kmol N ha^{-1} y^{-1} respectively. The deposition of potential acid in Scots Pine stands was lower (8·3 kmol) than the deposition in Corsican pine stands (8·6 kmol H ha^{-1} y^{-1}). However, significant differences existed only in nitrogen deposition between Douglas Fir and Scots Pine (Table 4).

Seasonal differences

In the summer season potential acid in bulk deposition was significantly higher than in the winter seasons (Table 5). However, in throughfall, significantly lower fluxes of potential acid were measured in the summer season. In bulk deposition the amounts of ammonium, sulphate and nitrate were significantly higher in summer. In this season throughfall contained a significantly higher amount of nitrate and a lower amount of sulphate. Potassium, magnesium and calcium showed also seasonal variation in throughfall. Depositions of sodium and chloride were significantly lower in summer than in winter in bulk and in throughfall.

The relative contribution of nitrogen oxides to potential acid in throughfall deposition was significantly higher in the summer season (Table 6). This increase was accompanied by a significant decrease of the relative contribution of sulphate. The contribution of ammonium was not significantly affected by the change of season. The relative contributions of ammonium and sulphate showed similar seasonal trends in bulk and throughfall deposition.

DISCUSSION

Regional differences in deposition appeared to be small. Actually, only the composition of the deposition in the coastal area differed from that of other parts of the country. In this region, the lowest amounts of potential acid and of total nitrogen were measured. This lower load was primarily due to the lower ammonium fluxes. The prevailing south-westerly wind mainly transports air from the sea which contains hardly any NH_x. At all other sites, provided they are not too close to sources, background emission density probably determines deposition (Schuurkes *et al.* 1988). In the forests at the coast, total nitrogen deposition amounted to $50\,kg\,N\,ha^{-1}\,y^{-1}$ of which 64% was due to the deposition of ammonium. However, in all investigated Dutch forests, nitrogen deposition amounted to $80\,kg\,N\,ha^{-1}\,y^{-1}$ of which 84% was deposited as ammonium. Similar throughfall fluxes of ammonium have been measured by other investigators in The Netherlands (Kleijn *et al.* 1988; van Breemen & van Dijk 1988; Ivens 1990). Throughfall measurements in Scots Pine stands in the UK revealed a much lower nitrogen deposition of 15 kg to 18 kg $N\,ha^{-1}\,y^{-1}$ (Alcock & Morton 1985; Skeffington 1983). Nilsson (1987) reported N-deposition to 30–40 kg $ha^{-1}\,y^{-1}$ with peak values of above 60 kg $ha^{-1}\,y^{-1}$ in Central Europe.

Duyzer *et al.* (1988) calculated deposition fluxes by estimating deposition rates and using emission data. They estimated the NH_x contribution to the load of nitrogen and potential acid at 60% and 30%, respectively. Only in the coastal area those estimated NH_x contributions equalled the measured contributions. In all other regions the measured contributions were higher, 84% and 44% respectively. These discrepancies can be explained by the fact that in calculations by Duyzer *et al.* (1988) co-deposition of ammonium and sulphate is not taken into account. Particularly in polluted areas concentrations of ammonium and sulphate mutually enhance their deposition rates (Adema 1986).

According to Bücking & Steinle (1987) forest structure characteristics such as species distribution and crown cover account for spatial distribution of deposition within a forest. Catching of aerosols and small dry particles is influenced by the shape and size of the catching surface. Turbulence and deposition rate are small on a homogeneous surface and deposition is large in closed stands (Bücking & Steinle 1987). Draaijers *et al.* (1988) ascribe higher throughfall fluxes in Douglas Fir stands compared to those in deciduous stands to the relatively larger foliage surface. In this study the local variation was probably also primarily due to differences in dry deposition as a result of variation in the size of the catchment surface between pine and fir stands.

On a local scale variations in deposition are often larger than the variation between the regions. The amount of nitrogen and potential acid in throughfall deposition appeared to be more dependent on the individual characteristics of the stands at a location than on the geographical situation of the region. At places where the stands border an area with less high vegetation (forest edge) throughfall fluxes are enhanced (Ivens 1990). Investigations

of Ivens (1990) showed that in stands near emission sources enhanced throughfall fluxes can be measured at 100 m from the forest edge. According to Hasselrot & Grennfelt (1987) deposition fluxes are already enhanced at the transition of a stand with smaller to a stand with larger trees. This phenomenon probably also contributes to the local variation in deposition fluxes within one region, as forest structure characteristics like forest size and species composition differ widely between the locations.

Apart from affecting the amount of deposition due to height differences, tree age can also affect the chemical composition of throughfall deposition. Canopy exchange processes can strongly vary with tree age. In areas with a high input of actual acid, more acid is measured in throughfall deposition of older stands (Miller 1984). N-demands of older stands appear to be smaller than those of young stands (Wilson & Pitcairn 1988). In areas with low N deposition, amounts of ammonium and nitrate in throughfall deposition may even be smaller than those in bulk deposition (Lovett & Lindberg 1984; Persson & Broberg 1985; Grennfelt & Hultberg 1986).

Bulk and throughfall displayed opposite seasonal trends for sulphate and ammonium fluxes (Table 5). The enhanced throughfall fluxes during winter are related to high emissions in this season and to the increased co-deposition under wet conditions. Bulk fluxes of ammonium and sulphate are also expected to be higher in winter. The enhanced bulk fluxes in the summer season of this study are probably due to the high amount of precipitation in the summer of 1987. These findings are in agreement with measurements of the Netherlands Precipitation Chemistry Network. In that research increased amounts of precipitation and correspondingly enhanced fluxes of ammonium and sulphate were measured in summer 1987 compared to the summer seasons in 1986 and 1988 (Anonymous 1987, 1988). In bulk as well as in throughfall, nitrate deposition is higher in summer than in winter. This difference can be caused by higher emission of nitrogen oxides but also by partial nitrification of dry deposited ammonium. Potassium, magnesium and calcium only varied in throughfall deposition. The seasonal difference for these cations is probably caused by canopy exchange in summer. During dormancy in winter, this process is supposed to be negligible.

In conclusion, this study has proved that most forested areas in The Netherlands receive high fluxes of pollutants, in particular of ammonium and sulphate, and that this problem is not restricted to areas with intensive agricultural activities. In fact only the coastal area receives lower loads of nitrogen and potential acid. However, even in this region, the critical loads for acid (1600 eq) (Schneider & Bresser 1988) and nitrogen (1400 eq) (Boxman *et al.* 1988) are severely exceeded. Throughfall fluxes of nitrogen and potential acid varied with tree species. Species composition of the forest seems to cause more spatial variation in the deposition than the geographical position of the forested area. The relative contributions of ammonium and nitrate to total nitrogen as well as those of ammonium and sulphate to potential acid varied little. The equal contributions of ammonium and sulphate indicate co-deposition of those compounds. Seasonal and annual variations in deposition indicate that long-term monitoring of throughfall fluxes remains necessary for the estimation of the load of pollutants to the forest soils.

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