Effects of nitrogen deposition on the acidification of terrestrial and aquatic ecosystems

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Abstract

The concentration of ammonium and nitrate in precipitation has increased during this century. The deposition of N compounds (wet + dry) is reaching 30 to 40 kg $ha^{-1}yr^{-1}$ in many areas in Central Europe and above 20 kg in the southern parts of Scandinavia. In extreme situations throughfall data indicate depositions above 60 kg $ha^{-1}yr^{-1}$ in Central Europe and above 40 kg $ha^{-1}yr^{-1}$ in south Sweden. Very high depositions are observed on slopes at forest edges and adjacent to areas with animal farms and manure spreading.
In areas with low N deposition almost all deposited N (>95%) will be absorbed in the tree canopies or in the soil. In areas with high deposition an increased outflow is observed which in some cases reach 10 to 15 kg $ha^{-1}yr^{-1}$. The increased output is an indication of N saturation of the ecosystem and it leads to acidification effects in soils, soilwater, groundwater and surface waters.

1. Introduction

The concentration of N compounds in the precipitation over North America and Europe has successively increased (N.A.S., 1983; Galloway and Dillon, 1983; Galloway and Likens, 1981; Söderlund and Granat, 1982; Rodhe, 1982). Data from the European Air Chemistry Network (EACN) show a significant upward trend in ammonia and nitrate deposition during the last decades (Söderlund and Granat, 1982; Rodhe, 1982; Kallend *et al.*, 1983). A doubling in nitrate concentrations from the end of the 50's to the beginning of the 70's has been shown (Rodhe, 1982). The upward trend for nitrate, however, is not observed after 1975 (Rodhe, 1985). The ratio of nitrate to sulphate in precipitation has also changed during the last decades. In south Scandinavia NO_3^-/SO_4^{2-} was 0.27 (on an equivalent basis) for the period 1955-59 and 0.47 for the period 1975-79 (Persson, 1982).

The upward trend in nitrate concentrations in precipitation corresponds well with the trend in NO emissions (W.S.L., 1983; Jernelöv and Lövblad, 1985). No similar study of the changes in ammonia emissions during this century has been made.
Since the wet deposition of nitrate and ammonium has increased both in absolute terms as well as in relation to sulphate it is important to evaluate the magnitude of this deposition, its interactions with the ecosystem and its possible effects. In this paper we will discuss these terms with respect to a coniferous forest ecosystem.

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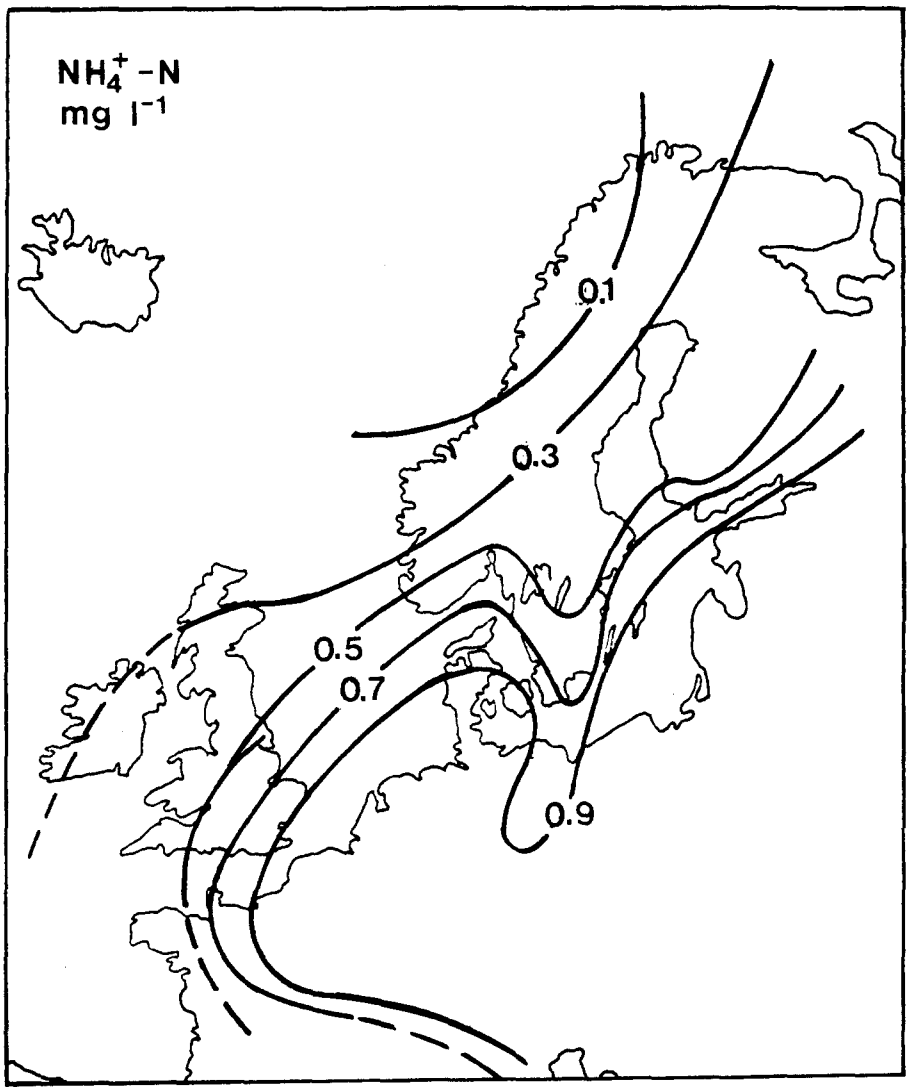

Fig. 1 Mean concentration of NH_4^+ in precipitation in Europe. Data from CCC/EMEP (SNV, 1984).

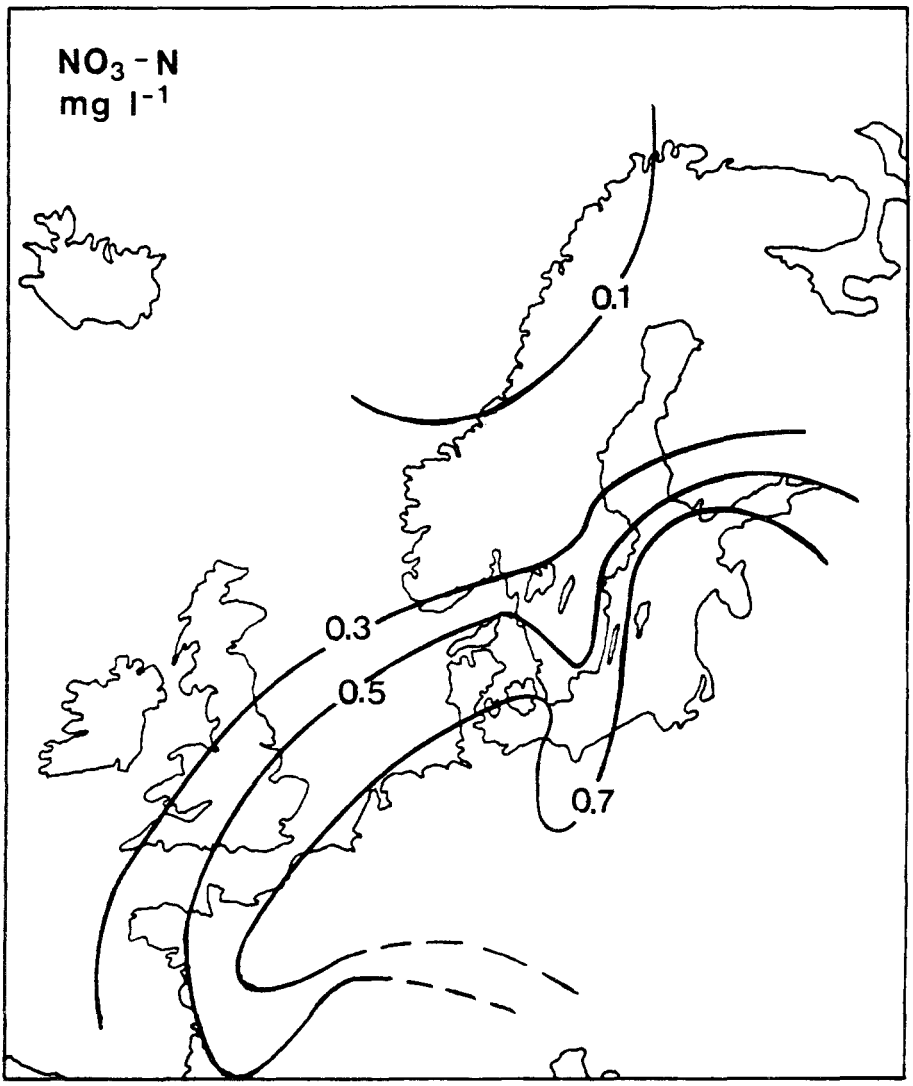

Fig. 2 Mean concentration of NO_3^- in precipitation. Data from CCC/EMEP (SNV, 1984).

2. Deposition of nitrogen species

The flow of N compounds from the atmosphere to the ground and further into the different compartments of a forest ecosystem is a complicated process and it is only known partially. The main reasons why it is difficult to describe qualitatively as well as quantitatively are:

i) Anthropogenic N compounds occur in many different chemical forms.

For long-range transport and deposition the most important compounds are NO_2, gaseous HNO_3, particulate nitrate, and particulate ammonium.

ii) Nitrogen is a nutrient and deposited N compounds will, to a large extent, be metabolised and retained in terrestrial and aquatic ecosystems.

iii) Natural exchange processes between atmosphere and soil/vegetation, for example fixation of N from air and denitrification in the soil, may influence the net flow of N as well as be influenced by the anthropogenic deposition.

The distribution of wet deposition of ammonia and nitrate in Europe is investigated within the EACN and the European Monitoring and Evaluation Programme (EMEP) networks. The precipitation in the most polluted areas in Europe has today a mean concentration of 0.9 mg L^{-1} or more of NH_4^+-N and 0.7 mg L^{-1} or more of NO_3^--N. The concentration decreases successively when approaching the less polluted areas and it is near 0.30 mg L^{-1} of NH_4^+-N and 0.15 to 0.30 mg L^{-1} of NO_3^--N in the north of Great Britain and in the middle of Scandinavia (SNV, 1984) (Figures 1 and 2).

Besides the wet deposition of ammonium and nitrate, a deposition of several kilograms ha^{-1} of soluble organic N has been found (Rosén and Lundmark, 1985; Matzner et al., 1982). The origin of this N input is so far unknown; it might be of anthropogenic origin or it might be natural. For the N budget it might in many cases be necessary to include this input.

Dry deposition of oxidized N compounds will essentially occur in the form of NO_2, gaseous HNO_3 and particulate nitrates. The deposition of these species goes along different pathways and different mechanisms regulate the deposition. For NH_3 and NO and sometimes also for NO_2 a "negative" deposition is observed indicating that the vegetation and the soil might be a source of N compounds.

In the Lake Gårdsjön project we have estimated the dry deposition essentially based on atmospheric concentration monitoring at Gårdsjön or adjacent sites on the Swedish west coast and of the best available estimates on dry deposition velocities (Grennfelt et al., 1985; Hultberg and Grennfelt, 1986). The estimates indicate that the dry deposition input is of the same order of magnitude as the wet deposition both for oxidized and for reduced N compounds at this site (Table I).

Table I.

Dry and wet deposition of N compounds to a forest area within the Lake Gårdsjön watershed. The dry deposition estimates are based on monitored atmospheric concentrations at the site or elsewhere on the Swedish west coast and literature data on deposition velocities.

		Deposition kg N ha^{-1} yr^{-1}
Dry		
	$NO_2 + HNO_3$	3.6 - 5.1
	particulate NO_3^-	0.3 - 1.1
	NH_3	?
Wet	particulate NH_4^+	1.4 - 5.5
	NO_3^-	4.6
	NH_4^+	6.3
		16.2 - 22.6

3. Extremes in impact of nitrogen compounds

Given that the effect of atmospheric pollution on an individual tree is likely to depend mainly on the deposition rate in the immediate vicinity, it is reasonable to ask whether there are locations which receive particularly high deposition and whether this effect is of practical importance.

Slopes and tops of hills at high elevations which are exposed to polluted air masses are shown to receive much larger quantities of pollutants than flat land due to interception of cloud droplets with high concentrations of pollutants (Lovett et al., 1982; Scherbatskoy and Bliss, 1983).

Exposed forest edges adjacent to open fields and clearcuts is another deposition situation where one might expect increased deposition. In south Sweden this deposition has been studied by means of throughfall along parallel lines from the forest edge into the closed forest (Hasselrot and Grennfelt, 1985). For many ions the throughfall at the forest edge will be 2 to 4 times higher compared to the closed forest (Figure 3.). The results often show much more pronounced edge effects for nitrate and ammonium than for sulphate, indicating that specific deposition mechanisms sometimes will dominate. Besides the cloud droplet incerception, the gaseous deposition of HNO_3 and ammonia might be very large at the edges.

Another situation where very large N deposition might be expected is close to ammonia emission sources from the ground or low

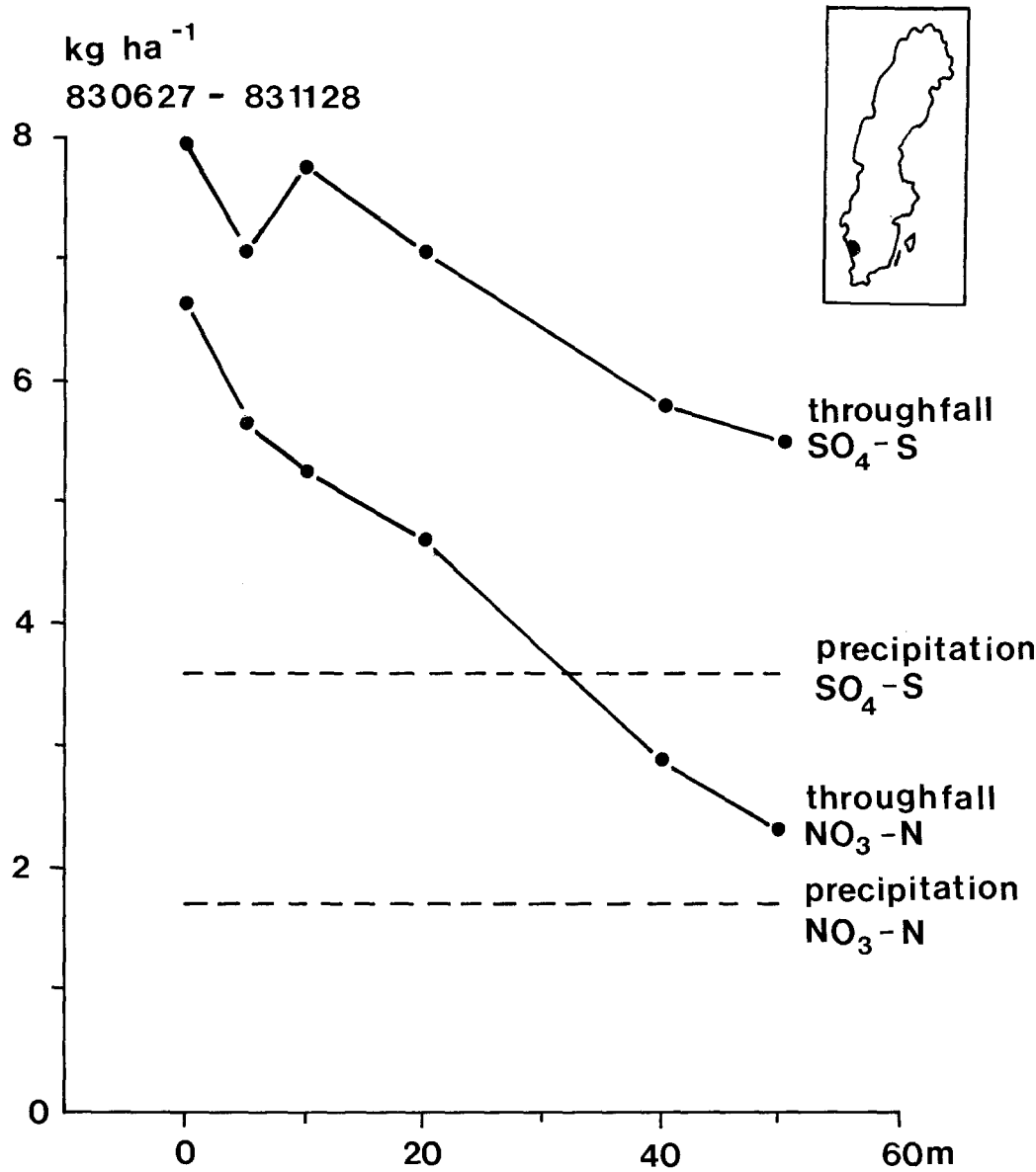

Fig. 3 Throughfall of nitrate and sulphate at a forest edge of a pine forest in south west Sweden (Hasselrot and Grennfelt, 1985).

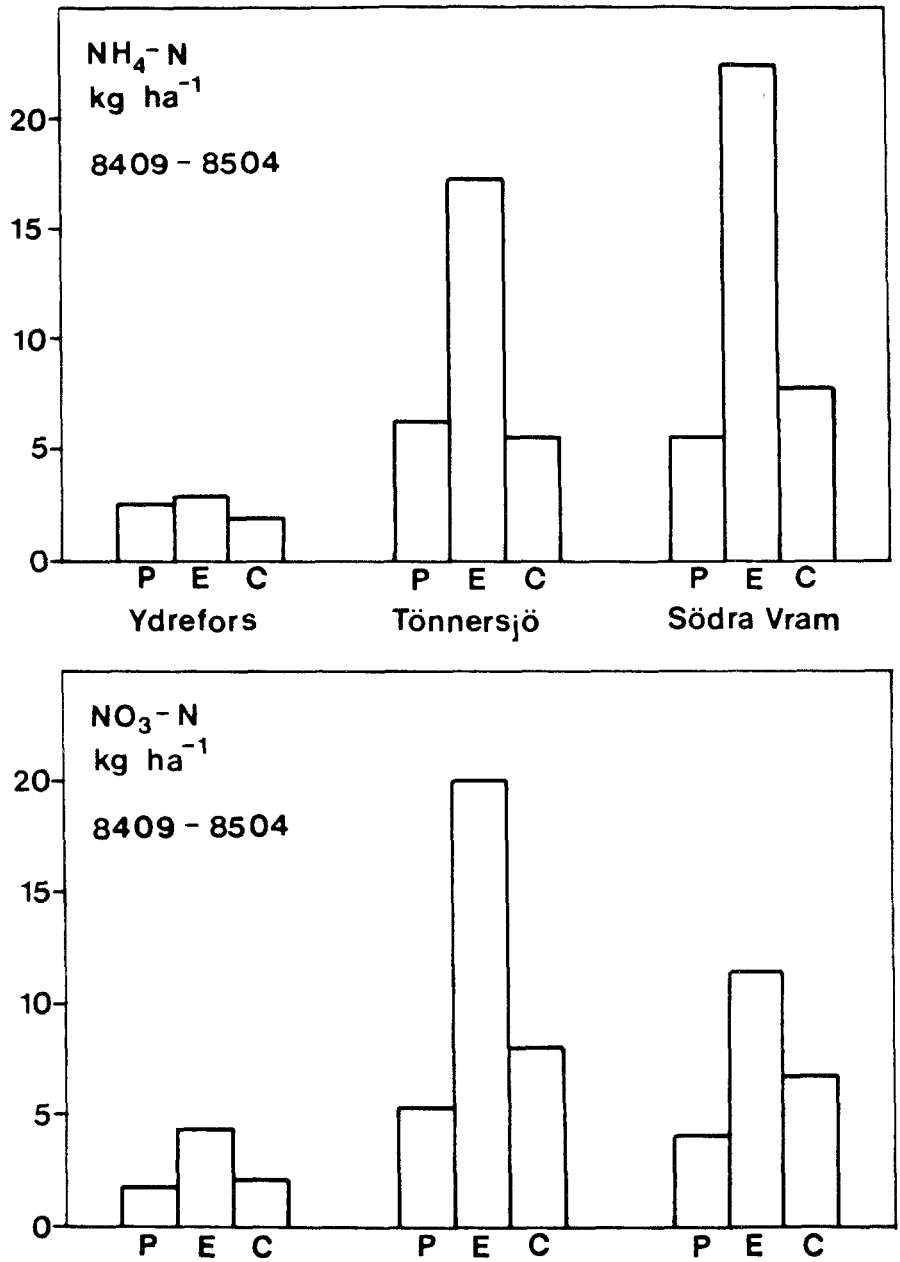

Fig. 4 Ammonium and nitrate flow in precipitation (P) and throughfall at the forest edge (E) and within the forest (C) of mature spruce forests at three sites in south Sweden. Data from 8 mo (Sept. 84 - April 85). Ydrefors is situated in the central part of south Sweden, Tönnersjö at the SW border of the south Swedish highlands, and Södra Vram in the agricultural lowlands in the very south of Sweden.

heights. Chicken farms and manure spreading are such sources. In the Netherlands very high ammonium depositions as well as ammonium concentrations in the soil have been shown (Roelofs et al., 1985; van Breemen and Jordens, 1983; van Breemen et al., 1982). A total input of inorganic N of 64 kg ha^{-1} yr^{-1} has been monitored of which 75% was in the form of NH_4^+ and 25% in the form of NO_3^- (van Breemen et al., 1982).

Forest edge studies in areas with a high density of cattle in south Sweden also indicate a very large influence by ammonia (Figure 4). From an ongoing study data from 11 mo showed a throughfall flow of 26.5 kg NH_4^+-N ha^{-1} and 14.9 kg NO_3^--N ha^{-1} at a forest edge, while the corresponding figures for the wet deposition (open gauges) were 6.9 and 4.9 kg ha^{-1}, respectively (Hasselrot and Grennfelt, 1985).

4. The interaction of nitrogen with tree canopies

Studies in areas with fairly low deposition of N show that the nitrate and ammonium flow beneath the canopy is smaller than the precipitation (Rosén and Lundmark, 1985; Horntvedt et al., 1980; Richter and Granat, 1978). If the dry deposition of N is then added one might assume a substantial absorption of the deposited N in the canopy. Fertilizing experiments show that N (ammonium and nitrate) applied to vegetation surfaces will easily be taken up and metabolized (Verry and Timmons, 1977). The same will happen when ammonium and nitrate are deposited to vegetation surfaces with dry processes as well as when the precipitation passes the canopy. From throughfall studies in Sweden and Norway it is shown that in the summertime more than 70% of the ammonium flow and more than 50% of the nitrate flow in precipitation may be absorbed in the canopy (Horntvedt et al., 1980; Rosén and Lundmark, 1985).

In more polluted areas in south Sweden and on the European continent these relations are changed and higher nitrate flow and sometimes also higher ammonium flow is measured in the throughfall than by precipitation. In these areas the deposition to the canopies is larger than the trees are able to absorb. The soil may also supply the trees with N in such quantities that it will affect the uptake. This effect was shown in an experiment where throughfall was studied in a stand of Corsican pine before and after fertilization of the soil (Miller, 1984).

The interaction of the canopy on the N flow is also dependent of the time of the year. In winters, when the biological activity is small, there seems to be no uptake in the canopy. This effect is clearly seen in Figure 5, where monthly throughfall data of ammonium at a forest edge is presented. Observe that in the summer (May-July) ammonium in throughfall is less than in the open precipitation while it in March is five times that in precipitation.

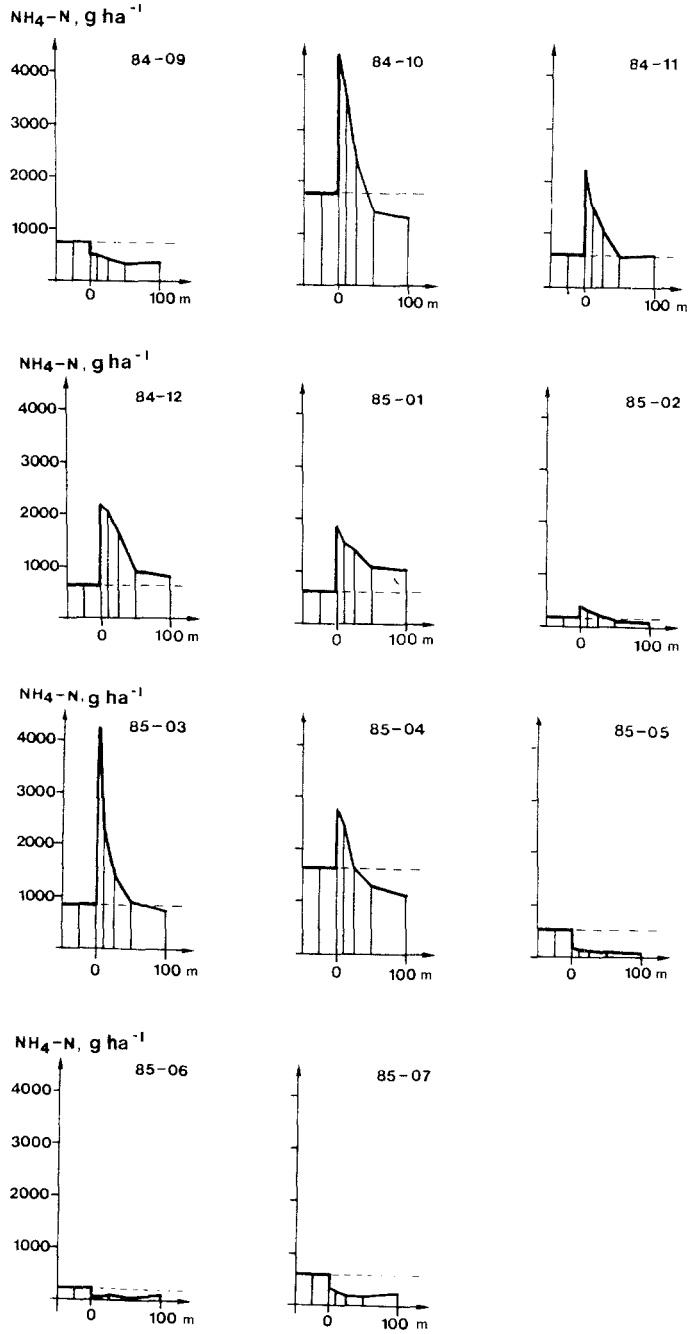

Fig. 5 The monthly variation in throughfall of ammonium at a forest edge of a spruce forest (Tönnersjöheden) in south west Sweden (Hasselrot and Grennfelt, 1985).

5. Nitrogen and ammonium interactions with the soil

Forest growth is generally limited by N deficiency and N deposition will have a fertilizing effect. The retention of N is therefore high in forest ecosystems. High N deposition may, however, cause a N saturation which in turn may cause a considerable output of nitrate (Matzner et al., 1982; Hauhs, 1984) along with leaching of basic cations and/or Al and H^+ from the soil. This will in turn cause a decrease in base saturation of the soil (soil acidification) and/or increased acidity in runoff water.

The acid-base relationships of the N cycle in forest systems are quite well known. Oxidation of ammonium to nitrate produce acid and H^+ is released by root uptake of ammonium. Ammonium formation from amino-groups during mineralisation of organic nitrogen and uptake of nitrate either consume H^+ or release OH^- ions.

The chemical and biochemical reactions of N, from the acidification point of view, are normally simplified to the H^+-producing and H^+-consuming reactions (Reuss, 1976; van Breemen, 1985):

Consuming H^+	NO_3^- + H^+	->	bio-organic N
"	NO_3^- + H^+	->	N_2, N_2O (denitrification)
Producing H^+	NH_4^+	->	NO_3^- + $2H^+$ (nitrification)
"	NH_4^+	->	org.N + H^+.

If we assume that these processes are specific for the N-deposition effect on the H^+ budget of a soil the process is simplified to the following equation representing the net flow:

$$\Delta H^+ = (NH_4^+{}_{(in)} - NH_4^+{}_{(out)} + (NO_3^-{}_{(out)} - NO_3^-{}_{(in)}).$$

Small errors in the estimate of reactions involving H^+ in the N cycle may cause serious errors in H^+ budget. Reuss (1982) therefore suggests that the acidification of soils due to acid deposition is quantified through reduction in soil base saturation. Nitrogen uptake increases biomass production which increases uptake of base cations and reduces base saturation (see also Hultberg, 1985). Nitrate leaching (primarily NO_3^-) occur along with an equivalent amount of cations, which at high base saturation are Ca, Mg, Na and K, but with decreasing base saturation Al-species and H^+ increase.

A consequence of this approach is that the acidifying effect is independent of the ionic form of the N deposition. One mole of ammonium nitrate have twice the potential to acidify the soil as one mole of HNO_3. One mole ammonium sulphate will have twice the acidifying effect as one mole of H_2SO_4. The potential to reduce base saturation by N and S inputs is therefore independent of form.

With this approach the potential acidity can be much larger than that obtained from the H^+ activity. The potential acidity from ammonium sulphate deposition in a woodland in the Netherlands was estimated to 7.14 kmol ha^{-1} while actual acidity in throughfall

and stemflow was about 3% (pH = 4.6) of the potential (van Breemen and Jordens, 1983). The N-deposition reached at this site 64 ± 15 kg ha^{-1} yr^{-1} and both calcareous and acid soils leached NO_3 ions along with primarily Ca (calcareous soils) and Al (acid soils) as dominating cations. In the acid soils 65% of the nitrate in soil solution came from ammonia in throughfall (30% in the calcareous soil).

Wiklander (1983) stresses that N from deposition which is retained in the soils may increase nitrification after clear-cutting and induce high nitrate leaching along with decrease in soil base saturation and leaching of Al.
The increased Al-levels in soil solutions may result in precipitation of Al-P compounds in the soil (Broberg and Persson, 1985) which may cause P deficiency to trees and decreased catchment transport by runoff to lakes and streams. Decreased output of P and increased output of Al from the terrestrial system due to increased N inputs are two effects which have the most severe consequenses, oligotrophication and toxicity to fish and other organisms, in the aquatic ecosystem (Hultberg, 1985).

6. Mass flow of nitrogen compounds

Mass flow of N including precipitation and catchment runoff are studied at many forest sites in Europe. In some cases throughfall is also monitored but only few studies exist where the dry deposition is estimated (Table II). These input/output studies may be separated into three different categories

1. studies in low-polluted areas, mostly in central Sweden and Norway,

2. studies in moderately polluted areas in south Sweden and at some areas on the European Continent,

3. studies in highly polluted areas in central Europe.

The first category is characterized by a very high absorption of N in the canopy and a low runoff of N (<0.3 kg ha^{-1} yr^{-1}).

The second category is an intermediate phase where the N deposition affects the N circulation in the ecosystem. The load of N to the ground by throughfall reaches 5 to 10 kg N ha^{-1} but the runoff is still very low.

The third category is when nitrate leaching from the soil is elevated. The input at these places is larger than the ecosystem will be able to absorb.

Abrahamsen (1980) presented a regession between N-input by precipitation and catchment runoff of N, showing that 30% of the wet deposition was leached from the terrestrial system.
Runoff data from Central Europe (Table II), however, show in five cases that the output of nitrate is 53 to 92% of the input by precipitation when this input exceeds 13 kg inorganic N ha^{-1}. Nitrogen saturation may already have occurred in these forest ecosystems (Figure 6).

TableII.

Mass flows of NO_3-N and NH_4-N in some forested catchments kg $ha^{-1}yr^{-1}$

Place	Country	Wet depositon		Throughfall		Outflow		Reference and comments
		NH_4-N	NO_3-N	NH_4-N	NO_3-N	NH_4-N	NO_3-N	
Jergul	N	0.35	0.42			-	0.04	Wright, 1982
Birkenes	N	7.4	7.8			<1.0	<1.0	-"-
Hälsingland	S	1.04	1.05			0.05	0.08	Rosén and Lundmark, 1985
Jädraås	S	1.3	1.3	0.8	1.1	0.05	0.03	Bringmark, 1980 (Data from May-Nov)
Kloten	S	2.7	3.2			0.03	0.1	Grip, 1983
Gårdsjön	S	6.3	4.6	4.7	5.7	0.023	0.24	Grennfelt et al., 1985, Hultberg, 1985
Söderåsen	S	~7.0	~5.0	~10	~9	-	5.0	Wiklander, 1983, beech and 1/2 year budget of spruce stand
Langen Bramke	D	14.8	8.9	10.0	9.7	0.2	1.9	Hauhs, 1984
Dicke Bramke	D	(14.8)	(8.9)			0.3	6.4	-"-
Wintertal	D	(14.8)	(8.9)			0.3	16.0	-"-
Solling	D	12.6	8.1	16.3	16.0	0.18	11.9	Matzner et al., 1982
Waroneu	B	12.4	11.0	17.9	16.5	0.8	12.0	Buldgen and Remack, 1984
Robinette	B	12.4	11.0	19.5	21.5	0.8	16.5	- " - - " -
Vysoca Pec	CZ	7.5	5.5			0.0	12.0	Paces, 1985

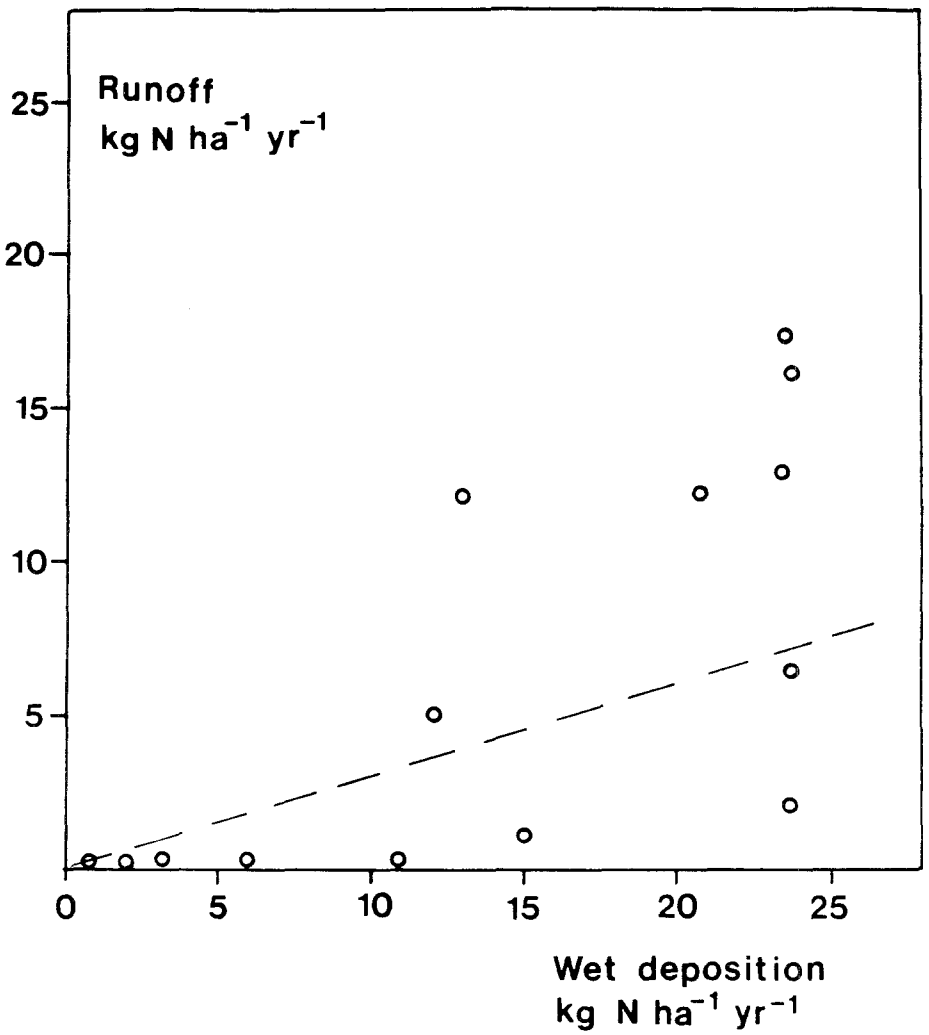

Fig. 6 Annual impact of $NH_4^+ + NO_3^-$ (kg ha^{-1}) by precipitation versus the leaching losses from watersheds and lysimeters in coniferous forests. Data from north and Central Europe. The line indicates the regression line obtained by Abrahamsen (1980).

7. Effects on forests of nitrogen deposition

Investigations of pine forests in the Netherlands show that needles of pine take up ammonia and excrete K, Mg and Ca which often results in K and/or Mg deficiencies. The high levels of N in needles are also correlated to fungal diseases. Field studies of forest damage to pine stands were correlated to increased ratios of NH_4 to K, Mg, and Ca in the soil solution (Roelofs et al., 1985).

Nihlgård (1985) also stresses that the ammonium deposition is an additional explanation to forest decline. In his hypothesis, he assumes that high uptake of ammonia stimulates biomass production which may cause relative deficiencies in Mg, K, P, Mo, B and water, and accumulation of non-protein N may increase. Increased non-protein N and waste products from assimilation may cause the tree to release the leaf or the needle. High levels of N in leaves and needles cause a decrease in frost hardiness and an increased susceptibility to insect attacks, fungi, bacteria and viruses.

8. Effects of nitrogen deposition in aquatic ecosystems

Generally, sulphate is the most important strong acid anion in acidified lakes and streams and on a yearly basis only <5 to 15% of the acidity in nutrient-poor lakes is explained by nitrate (Tirén, 1980).

The nitrate concentration has, however, increased during the last decades in both lakes and streams (Dickson, 1980) and in deep nutrient-poor lakes this progressive increase has been shown to cause a decline in pH and alkalinity. Figure 7 show that the acid Lake Gårdsjön have had an increase in both the lake inlet and lake outlet during the last years.

Acid surges in spring co-incides with elevated concentrations of nitrate in runoff from forested catchments (Likens et al., 1977) and in lakes (Galloway et al., 1980). During late autumn and winter when no nitrate uptake by vegetation occur, nitrate in autumn rains and/or accumulation in snowpack may be transported from the catchments. High concentrations may also occur in streams draining catchments with forest dieback (Paces, 1983) along with high concentrations of Al. A positive correlation between Al and NO_3^- have been demonstrated by Schofield (1984) in Woods Lake outlet and inlet during snowmelt.

9. Conclusions

The deposition of N compounds to forest ecosystems is in most cases higher than previously considered. Deposition estimates and throughfall data indicate that the atmospheric input in many areas in Central Europe reaches 30 to 40 kg N $ha^{-1}yr^{-1}$ and in extremes above 60 kg N $ha^{-1}yr^{-1}$. In south Sweden the deposition exceeds 20 kg N $ha^{-1}yr^{-1}$ and in extremes above 40 kg $ha^{-1}yr^{-1}$.

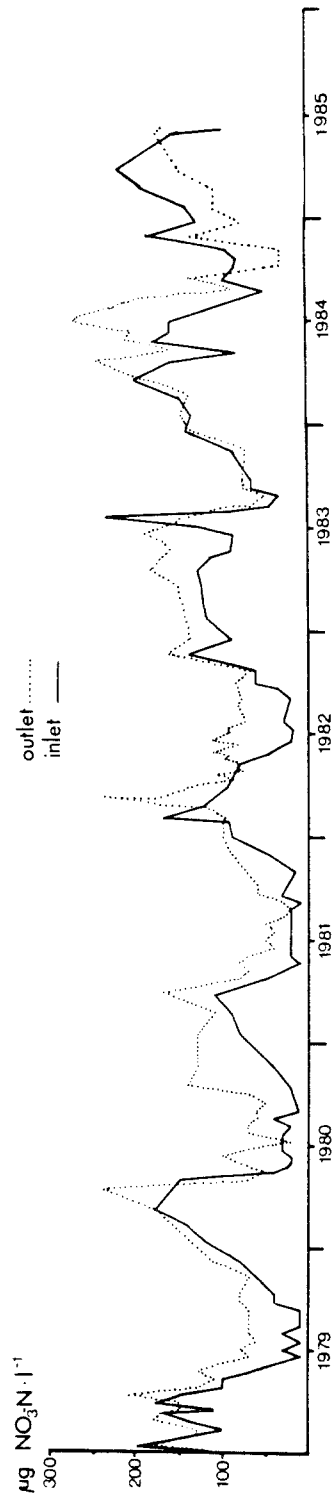

Fig. 7 Changes in the concentration of nitrate in Lake Gårdsjön in south west Sweden 1979 - mid 1985.

The consequence of an increased N input with respect to N utilization and forest growth was analyzed by Ågren (1983). With a theoretical C/N model, he found that with a deposition of 30 kg N ha^{-1} N saturation will be reached in fertile soils within 20 to 25 yr. For poor soils this saturation level will be reached within approx. 50 yr.

Our opinion is that many sites in Central Europe have had a yearly deposition of 30 kg N ha^{-1} for at last 25 yr and according to the model, these sites should be nitrogen saturated. The high leaching of nitrate (5 to 16 kg N $ha^{-1}yr^{-1}$) from many soils with a high N loading is probably the best evidence of this N saturation.

Moreover, there are indications of ecosystem effects related to high loadings of N, especially such loadings that exceed the absorption capacity of the ecosystem. Together with S deposition the N deposition may be an essential factor for soil acidification and forest damages as well as for acidification of lakes and streamwaters during snowmelt episodes.

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