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LARGE SCALE IMPACTS OF ACID DEPOSITION ON FORESTS AND FOREST SOILS IN THE NETHERLANDS

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Abstract
Since the early eighties, effects of acid atmospheric deposion have received much attention in the Netherlands. Effects of elevated S and N deposition on soil solution chemistry is mainly manifested by increased concentrations of Al associated with increased concentrations of SO_4 and NO_3. Presumed critical Al concentrations (0.2 $mol_c\ m^{-1}$) and Al/Ca ratios (1.0 mol mol^{-1}) are generally exceeded below 20 cm soil depth. There is also ample circumstantial evidence that elevated N deposion during the last decades affected the forest nutrient status and caused large changes in forest vegetation. About half of the Dutch forests have foliar N contents exceeding a critical limit (1.8%). Field evidence for a relationship between soil acidification and nutrient imbalances in the soil and the foliage on one hand and the vitality of forests (mainly expressed by defoliation class) on the other hand is, however, lacking. This result implies that an exceedance of critical acid loads, based on critical Al concentrations and Al/Ca ratios observed experiments in relation to effects on root (uptake), do not imply visible effects or even the dieback of forests.

1. INTRODUCTION

Since the early eighties it is recognized that Dutch forests receive large inputs of NH_4 and SO_4 from the atmosphere (Van Breemen et al., 1982). At the same time the possible role of atmospheric sulphur (SO_x) and nitrogen (NO_x, NH_x) deposition in forest dieback in central Europe became a subject of wide public and political discussion. Since then, effects of atmospheric deposition of SO_x, NO_x and NH_x on forests have received much attention in the Netherlands. Research efforts were focused on the impacts of acid deposition on soil (solution) chemistry (e.g. Van Breemen and Verstraten, 1991; De Vries and Leeters, 1994), forest vegetation (e.g. Hommel et al., 1990; Van Dobben et al, 1994), forest nutrient status and forest vitality (e.g. Van den Burg et al., 1988; De Visser, 1994) and the relationship between soil solution chemistry and forest vitality (e.g. Roelofs et al., 1985; Hendriks et al., 1994).

Important effects of elevated SO_x, NO_x and NH_x deposition on the soil are: (i) a decrease on base saturation and pH and (ii) an increase in the concentrations of SO_4, NO_3, NH_4 and Al. Controlled laboratory experiments on seedlings have shown that increased concentrations of NH_4 and Al in relation to Ca, Mg and K affect the root system of trees and the uptake of these base cations. Because of the importance of soil mediated effects, much experimental research on the effects of acid atmospheric deposition on Dutch forests dealt with the impacts on the soil solution chemistry. Several local monitoring studies were performed in a total of eighteen forest stands. Furthermore, a one-time national survey was carried out in early spring in 150 representative forest stands on non-calcareous sandy soils across the country. The chemical composition of the soil solution in both the monitoring and survey sites was mainly measured to gain insight in (i) the fate of SO_4, NH_4 and NO_3 in forest soils and (ii) the mobilization of Al and base cations (Ca, Mg, K and Na), which mainly neutralize the acid input, based on input-output budgets (Van Breemen and Verstraten, 1991. De Vries and Jansen, 1994).

The one-time soil solution survey in 150 forest stands was also carried out to study the relationship between the soil and soil solution chemistry and forest vitality characteristics, such as needle loss and needle colour, (Hendriks et al., 1994). Consequenly, the 150 survey stands were selected from about 3000 stands belonging to the Dutch forest vitality inventory. According to the frequency distribution of tree species in the Netherlands, 45 stands of *Pinus sylvestris* (Scots pine; 30%), 30 stands of *Quercus robur* (oak; 20%) and 15 stands (each 10%) of *Pinus nigra* subsp *maritima* (Corsican pine), *Pseudotsuga menziesii* (Douglas fir), *Picea abies* (Norway spruce), *Larix kaempferii* (Japanese larch) and *Fagus sylvatica* (beech) were selected. More information on the selection procedure is given in De Vries and Leeters (1994). Other important aims of the one-time national survey were the (i) determination of the chemical composition of the foliage to assess the nutrient supply (Hendriks et al., 1994) and (ii) assessment of the chemical composition of the soil solution on a national scale from relationships with deposition level, stand characteristics (such as tree species) and site characteristics (such as soil type; Leeters et al., 1994).

This paper summarizes the impacts of acid deposition on forests in the Netherlands with respect to soil solution and groundwater chemistry, forest nutrient status, forest vegetation and forest vitality. Critical loads for nitrogen and acidity that have been derived in relation to several effects will be reviewed critically in the light of these results.

2 EFFECTS ON FOREST SOILS

2.1 Soil solution chemistry

Exceedances of critical chemical values

The most pronounced reflections of atmospheric deposition of S and N on the solution chemistry of Dutch acid sandy soils are the high SO_4 and NO_3 concentrations in the soil solution and concomitantly high concentrations of acidity (H+Al). Results of linear regression analyses for the various monitoring and survey sites, showed that the concentration of H+Al gets closer to the concentration of SO_4+NO_3 with increasing depth for nearly all sites (de Vries et al., 1994b). Moreover, the linearity of the relationship increased with increasing depth as shown by an increasing value for the adjusted coefficient of determination (R^2_{adj}). A 1:1 relationship (on an equivalent basis) between [H+Al] and [SO_4+NO_3] in the soil layers below the rootzone, indicates that external inputs of N and S to the soil (corrected for N and S retention in the soil) will cause mobilization and leaching of equivalent amounts of H and Al.

In all monitoring sites with a non-calcareous subsoil (16 out of 18 sites), the calculated annual average Al concentration in the subsoil largely exceeded the Dutch drinking water standard (0.2 mg l^{-1} ≈ 0.02 mol_c m^{-3}) in the subsoil. The annual average NO_3 concentration exceeded the EC drinking water standard for NO_3 (50 mg l^{-1} ≈ 0.8 mol_c m^{-3}) in 14 sites and the Dutch target value (25 mg l^{-1} ≈ 0.4 mol_c m^{-3}) in 16 sites. Molar Al/Ca and NH_4/K ratios, which are regarded as indicators of potential reduction of nutrient uptake by roots, showed strong gradients with depth in the intensively monitored sites. The Al/Ca ratios, based on annual average Al and Ca concentrations, for these sites generally exceeded an assumed critical value of 1.0 at a depth of 20 cm, which is within the predominant zone of root activity. In contrast to Al/Ca ratios, NH_4/K ratios tended to decrease, and so become more favourable for biota, with increasing depth. In several intensively monitored sites molar NH_4/K ratios were above an assumed critical value of 5 in the upper 20 cm of the soil.

In the non-calcareous survey sites (147 since three sites appeared to be calcareous) the EC drinking water standards in the subsoil (60 - 100 cm below soil surface) were exceeded every where for Al (Fig 1A) and in 31% of the cases for NO_3, whereas 55% of the sites exceeded the Dutch target value for NO_3 (cf Fig. 1B). An Al concentration of 0.2 mol_c m^{-3}, which is considered critical with respect to effects of roots, was exceeded in 80% of the sites (Fig 1A). However, Al and NO_3 concentrations measured at the survey sites may deviate from annual average values. Values for the molar Al/Ca and NH_4/K ratio in the survey sites were generally lower than those at the monitoring sites. In the forest topsoil (0-30 cm depth), the critical Al/Ca ratio of 1.0 was exceeded in 57% of the 147 survey sites (Fig 1C), whereas only 4% of the

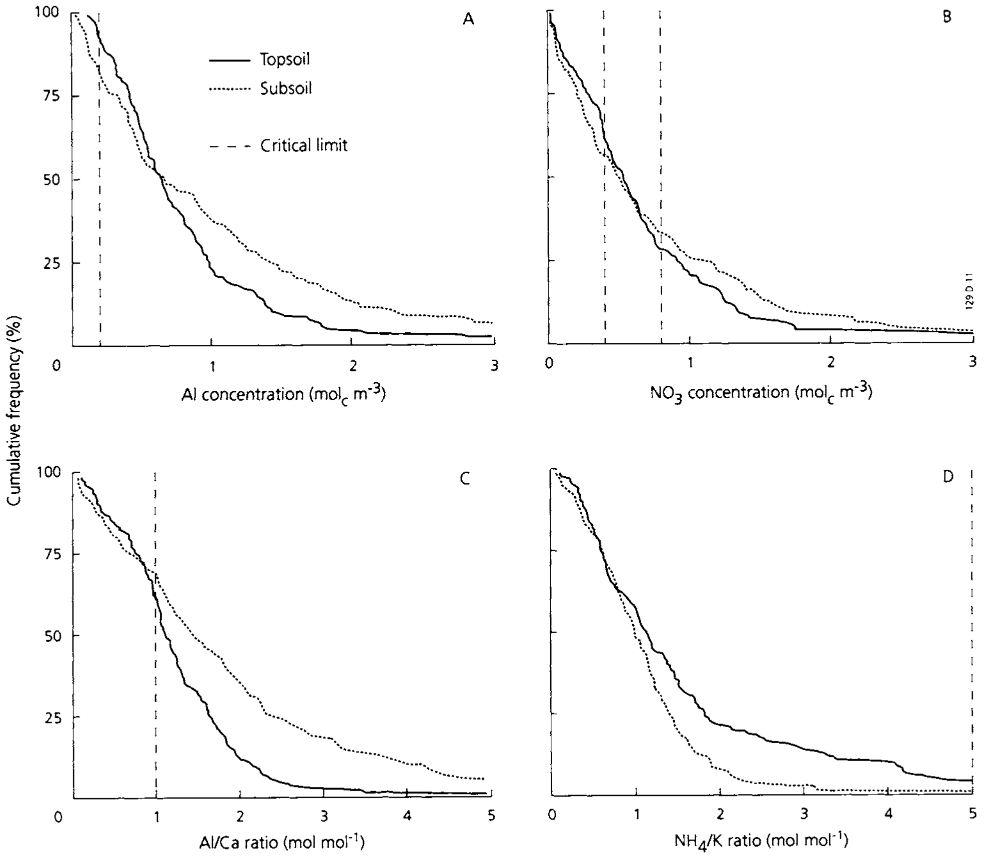

Figure 1. *Inverse cumulative frequency distributions of Al concentrations (A), NO_3 concentrations (B) Al/CA ratios (C) and NH_4/K ratios (D) in the topsoil (0-30cm) and subsoil (60-100cm) of 147 survey sites*

survey sites exceeded the critical NH_4/K ratio of 5 (Fig. 1D). The relatively low Al/Ca ratios were mainly a result of a high Ca concentration. This may partly be the result of a high Ca input from the atmosphere due to strong filtering of base cations by the forest canopy, especially near forest edges. The most important explanation might, however, be the different methodology to obtain soil solution (cf De Vries et al., 1994b).

The various soil solution parameters in the survey sites were largely influenced by tree species. Lowest pH values and highest concentrations in Al, SO_4 and NO_3 occurred below Douglas fir. The reverse was true for oak and beech, whereas Scots pine en black pine occupied an intermediate position.

The increase in solute concentrations between tree species, going from deciduous forest to pine forests to spruce forests, is most probably caused by increased dry deposition and evapotranspiration. Concentrations of Al, SO_4 and NO_3 also increased with increasing tree height and canopy coverage. This is most likely due to an increase in atmospheric S and N deposition (cf De Vries et al., 1994 b). Relatively good regression relationships ($R^2_{adj} > 0.5$) were found between the SO_4 and Al concentration and the tree species, tree height, percentage of forest in the surrounding area and acid atmospheric deposition. Even tree species and tree height alone already explained nearly 40% of the variation in SO_4 and Al concentration in the forest topsoil. For NO_3, the relationships were slightly worse ($R^2_{adj} \approx 0.4$; cf Leeters et al., 1994). The regression relationships that were found were used to produce SO_4, NO_3 and Al concentration maps at a grid resolution of 0.5 km x 0.5 km. Relatively low concentrations of SO_4, NO_3 and Al were generally predicted in the large forest complexes in the central part (Veluwe area) and the northern part (Drenthe) of the Netherlands, where atmospheric deposition is comparatively low. High concentrations were generally predicted in the small forest complexes in the eastern and southern part of the Netherlands. Small scale variation was caused by variation in tree species and tree height (cf Leeters et al., 1994)

input - output budgets

Soil solution chemistry data can be used to calculate drainage outputs from the soil. Comparison of such outputs with atmospheric inputs gives quantitative information on the retention or mobilisation of elements. Regarding S and N, it also gives insight whether the deposition of potential acidity (SO_x, NO_x and NH_x) is actually realised in the soil. This is only the case when all deposited S and N leaves the soil in the form of SO_4 and NO_3. Input - output budgets for ten intensively monitored sites (Van Breemen and Verstraten, 1991) and the 147 survey sites (De Vries and Jansen, 1994) showed that. Dutch forest soils are SO_4 saturated, but N is still largely retained (Table 1)

Table 1. *Average atmospheric inputs and drainage outputs of SO_4, NH_4, NO_3 and total N in ten intensively monitored soils between 1981 and 1990 and 147 soils in which a single measurement took place in 1990*

Type of research	Flux (kmol$_c$ ha^{-1} yr^{-1})							
	SO_4		NH_4		NO_3		N	
	in	out	in	out	in	out	in	out
Monitoring	2.77	2.69	2.98	0.12	0.87	1.78	3.85	1.90
Survey	1.74	1.77	3.19	0.13	0.97	0.70	4.16	0.83

Both the input and output of SO_4 was lower in the 147 survey sites than in the ten monitoring sites, which is most probably due to the decrease in SO_4 deposition during the eighties. However, in both types of research, average

SO_4 inputs equalled average SO_4 outputs, implying that SO_4 deposition contributes for 100% to the actual acidification of the soil,

Atmospheric deposition of NH_4 and NO_3 was comparable for the monitoring sites and survey sites. In the survey sites retention of N in the forest (by uptake) and the soil (by immobilization) was, however, larger than in the monitoring sites, since NO_3 leaching was considerably lower; even below the NO_3 input (cf Table 2). Theoretically, NH_4 deposition may not contribute to the acidification of these sites since it is either taken up a immobilised. In the intensively monitored sites, however, the difference between NO_3 leaching and NO_3 deposition of ca 0.9 $kmol_c$ ha^{-1} yr^{-1} is due to nitrification, and ammonia at least contributes ca 20% to the actual acidification of these soils (ca 4.5 $kmol_c$ ha^{-1} yr^{-1}).

Soil acidification is mainly manifested by Al mobilization from secondary Al compounds (De Vries et al., 1994b). Model calculations indicated that forest soils may become depleted in these compound within several decades at ongoing present atmospheric inputs (De Vries and Kros, 1989). This may cause a strong decline in pH, which may be an important stress to the forest ecosystem. However, the risk of Al depletion will strongly decrease at expected emission reductions. De Vries et al. (1993) calculated that the time period to reach complete Al depletion will increase from approximately 100 years at present acid loads to 2000 years at expected emission reductions in 50% of all forest soils. In the near future (2050), a relative Al depletion above 50% was only predicted in small areas in the southern and eastern parts of the Netherlands in soils with low present amounts of secondary Al compounds (De Vries et al., 1993). Model calculations also showed that emission reductions will lead to a fast improvement of the soil solution quality (decreased concentrations in SO_4, NO_3 and Al and increased pH; De Vries et al 1994a).

2.2 Groundwater chemistry

As with the soil solution, elevated atmospheric deposition has increased the concentrations of SO_4, NO_3 and Al in groundwater. This can be derived from a study on the chemical composition of phreatic groundwater in 156 gridcells of 0.5 km x 0.5 km , containing at least 0.1 ha of forest or heathland, that was carried out between 1989 and 1990 (Boumans and Beltman, 1991). Part of the measurements coincided with the 150 forest stands where the chemical composition of foliage, soil and soil solution was measured (a total of 71 stands) Median SO_4 concentrations in phreatic groundwater at these sites were comparable to those in the soil solution, but concentrations of Al and NO_3 were lower (Table 2). Still, Al concentrations exceeded the EC drinking water standard of 0.2 mg l^{-1} in ca 80% of the sites (forest and healthlands), whereas NO_3 concentrations exceeded the EC drinking water standard (50 mg l^{-1}) and the Dutch target value (25 mg l^{-1}) in 20% and 37% of the sites respectively (after Boumans and Beltman, 1991). The quality of drinking water, pumped up at much greater depths, will, however, be less affected because of Al retention and denitrification in the groundwater aquifer and because of mixing with various other watertypes.

Unlike soil solution, the NO_3 and Al concentration in groundwater was best 'explained' by a regression model including soil type and to a lesser extent by tree species (coniferous deciduous forest), tree height, surrounding land use and atmospheric deposition (Leeters et al., 1994; Boumans, 1994). NO_3 concentrations, for example, increase according to peaty soils < moderately drained, poor sandy soils < well drained rich sandy soils. (Boumans, 1994). As with the soil solution, however, lowest concentrations are generally found in the large forest complexes in the central part of the Netherlands and in the low deposition areas in the northern part of the Netherlands (Leeters et al., 1994).

Table 2. *Median concentrations of SO_4, NO_3, NH_4 and Al in the soil solution and in phreatic groundwater of 71 Dutch forest stands, sampled between 1989 and 1990. (After De Vries and Jansen, 1994)*

Element	Median concentration ($mol_c\ m^{-3}$)		
	0 - 30 cm	60 - 100 cm	groundwater
SO_4	0.97	1.08	1.04
NO_3	0.53	0.48	0.24
NH_4	0.20	0.09	0.00
Al	0.69	0.67	0.54

3. EFFECTS ON THE FOREST ECOSYSTEM

3.1 Foliar composition

Elevated atmospheric deposition of N and S compounds in the Netherlands during the last decades has led to an increase in the N content and a decrease in the P, K and Ca content in foliage. This can be derived from a study by van den Burg and Kiewit (1989), who compared the foliar composition of stands of Scots pine, black pine and Douglas fir in 1956 and 1988 in the 'Peel' area with intensive animal husbandry. Surprisingly, the Mg content did not decrease during that period However, even in 1956, the Mg content was already low. Furthermore, as with P, K and Ca, the Mg supply relative to N decreased (Table 3).

Table 3. *Average N contents and ratios of P, K and Mg to N in half years old foliage of stands of Scots pine, black pine and Douglas fir in 1956 and 1988 (After Van den Burg and Kiewit, 1988).*

Tree species	Nutrient content (%)				Nutrient ratio x 100 (gg^{-1})					
	N		P		P/N		K/N		Mg/N	
	1956	1988	1956	1988	1956	1988	1956	1988	1956	1988
Scots pine	1.5	2.3	0.15	0.14	9.9	6.1	34	27	3.0	2.7
Black pine	1.2	1.7	0.16	0.11	12.2	6.7	58	35	4.0	3.8
Douglas fir	1.4	2.2	0.25	0.12	18.1	5.1	68	24	6.1	5.0

The average increase in N content between 1956 and 1988 varied from 0.5% (black pine) to 0.8% (Scots pine and Douglas fir). According to criteria that have been given to judge the nutrient supply of trees in relation to growth, the content changed from shortage to excess (cf Table 5). The high N contents may also increase the susceptibility to frost (Aronsson, 1980) and fungal diseases such as Gremmenielle abitiena and Sphaeropsis sapinea (Roelofs et al., 1985; Boxman and Van Dijk, 1988). The largest changes in the foliar composition occurred for Douglas fir. Nutrient ratios in this tree were generally optimal for growth in 1956, whereas they were near the level of deficiency in 1988.

A single measurement of the foliar composition of the forest stands in the survey of 1990 also indicated nutrient deficiencies in several tree species (Table 4). In eight stands with too high trees no measurements were made. Information on the criteria that were used to judge the nutrient contents and nutrient ratios in foliage is given in Hendriks et al (1994).

Table 4 Exceedances of the lower limit of critical foliar nutrient contents and ratios of seven tree species in forest stands (After Hendriks et al., 1994)

Tree species	Nr	Exceedance (%)							
		N1)	P	K	Ca	Mg	P/N	K/N	Mg/N
Scots pine	43	91	37	2	28	23	9	23	95
Corsican pine	14	14	79	14	36	7	7	7	57
Douglas fir	16	69	75	50	0	0	50	38	19
Norway spruce	15	67	33	67	33	27	7	33	60
Japanese larch	13	36	92	23	31	0	23	23	23
Oak	27	55	26	4	63	59	30	4	44
Beech	14	13	100	43	21	86	64	29	71
All	142	49	63	29	30	29	27	22	53

1) For N it is the upper limit

The most striking conclusions from Table 4 are (i) the absolute excess of N, (ii) the absolute shortage of P and, (iii) the relative shortage of Mg (and to a lesser extent also K) compared to N. Foliar nutrient contents were most significantly related to tree species, and to a lesser extent to nutrient contents in mineral soil, soil solution and humus layer. A high heavy metal content (Pb, Zn, Cd) of the humus layer negatively influenced the foliar nutrient content of P and Mg. Part of the 150 stands, i.e. four stands of Scots pine, Douglas fir and oak, have been monitored since 1992. However, the time period is too short to derive reliable trends. Simulations with the integrated dynamic forest/soil model SOILVEG indicate that expected N emission reductions in the coming decade may lead to a decrease in the foliar N content of ca 0.2 - 0.3% (Van Grinsven et al., 1991).

3.2 Forest vegetation

Circumstantial evidence is available for large changes in forest understory in the Netherlands over the period 1950-1990. These changes entail: (i) a decline of terrestrial lichens ('reindeer lichens') and of ectomycorrhiza mushrooms; (ii) an increase of grasses, notably Deschampsia flexuosa and (iii) a general increase of mosses and vascular plants that typically occur on nitrogen-rich soils. However, detailed information at site level is scarce.

A pilot study was carried out by De Vries (1983) at 'Boswachterij Kootwijk'. This is an area of pine forests on dry, sandy soil, where vegetation maps from. 1957 were available. This study showed a complete changeover in understorey vegetation, from a moss- and lichen-dominated type to a grass-dominated type. Scattered information on other sites shows that the changes observed at this site are probably typical for most Dutch pine forests on poor soils. A comparable study was carried out by Hommel et al. (1990) in a neighbouring area ('Speulderbos'), with both deciduous and coniferous forest. Comparable results were obtained in this study; in general, the nitrogen indicator value of the vegetation has strongly increased. A statistical evaluation of the geographical distribution of the vegetation changes showed that these changes were stronger at shorter distances to agricultural areas. Thus, this study yielded a direct indication for agriculturally derived ammonia as a cause for the observed changes.

Changes in the Dutch mushroom flora have been studied by comparing old (circa 1950-1970) excursion reports with recent inventories. Extensive studies of this type, carried out by Arnolds (1991) and others, showed a strong decline of fruitbodies of ectomycorrhizal fungi, and an increase of fruitbodies of wood-inhabiting saprotrophic and parasitic fungi. Among the soil-inhabiting saprotrophic species those of nutrient-rich soils had increased, while those of nutrient-poor soils had decreased.

Changes in the understory of Dutch pine forests in a more recent period were studied by comparing vegetation descriptions made in 177 permanent plots in 1984 and in 1993 (Van Dobben et al. 1994). This study showed a significant decrease in the cover of Erica tetralix and Calluna vulgaris and a strong increase of many nitrophilous species. As a consequence, a highly significant increase in Ellenberg N-indicator value was observed. Interestingly, the Ellenberg pH-indicator value had also significantly increased, indicating soil alkalization. This might be due to the decrease in S deposition during this period.

Although direct information on the cause of these changes is lacking, most authors agree on natural succession and atmospheric N deposition as the most important factors. However, at present it is hardly possible to estimate the relative importance of these two factors. Studies comparing vegetation development in untreated and fertilized pine forests in areas with a low background deposition of nitrogen can be used to estimate the effects of nitrogen on forest vegetation. Such studies have shown that the changes in Dutch pine forests can be rather well simulated by the addition of nitrogen fertilizer at rates comparable to the rate of atmospheric nitrogen deposition (Van Dobben 1993). On the other hand, vegetation changes provoked by experimental soil acidification are generally unrelated to those presently observed in the Netherlands. The observed increase in Ellenberg pH-indicator value also implies that soil acidification is not the main cause for the vegetation changes, In that case a decrease in pH-indicator value would be expected. However, note that the increase in pH-indicator value may also be an artifact due to succession.

Studies on the significance of the changes in the undergrowth for the tree layer are virtually lacking. There are no indications that the dense grass cover hampers tree juvenation or succession in the tree layer. The decline of ectomycorrhizal fungi may be a factor contributing to a general decline of tree vitality, but a hard proof for this hypothesis is lacking.

3.3 Forest Vitality

The health of forests in the Netherlands has been monitored since 1983. To describe the state of health of a forest the term "vitality" is used. The vitality, is influenced by "traditional" factors such as fertility of the soil, provenance of tree species, rainfall, frost and drought, and pests and plagues, as well as by "new" anthropogenic factors such as air pollution and acid deposition. The annual vitality survey mainly has as an indicator function. It only gives an indication of the possible occurrence of combined stress. The commonly used indicators for the vitality of trees are defoliation and foliar discolouration. The defoliation class is judged to be the most important aspect of vitality. Since the beginning of the forest vitality inventories in 1984, the vitality of Dutch forests decreased until 1989. In 1990, the year in which the field inventory in 150 stands was carried out, the vitality was stabilised at the level of 1989. The vitality in 1991 was about the same. In 1992 the vitality decreased strongly, followed by a steady increase in 1993 and 1994.

Forest vitality characteristics, such as defoliation and foliar discolouration, are not only a function of the nutrient status of the tree and of factors influencing nutrient availability (e.g. the heavy metal contents in the humus layer and the Al concentration in the soil solution, which affect nutrient mineralisation and nutrient uptake respectively), but also of site factors such as rainfall, soil type, groundwater level (all affect soil water supply) and stand age. Results from a recent forest vitality inventory in Europe shows that especially stand age can be very important (UN-ECE/CEC, 1992). The defoliation class measured in 150 forest stands was related to these, so called, explanatory variables with multiple linear regression. Apart from tree species, soil type, groundwater level and stand age, stand characteristics which influence the input of elements by atmospheric deposition, i.e. tree height, canopy coverage and distance of the trees to the forest edge, were also included. Chemical variables that were used to predict the defoliation class were limited to the N content in foliage, humus layer and soil solution, the ratios of P, K, Ca and Mg to N in foliage, heavy metal contents in the humus layer and the pH, Al/Ca ratio and NH_4/K ratio in soil solution. This limitation was based on the assumption that the chemical composition of the foliage is the best reflection of the nutrient status of the forest, whereas elevated Al/Ca and NH_4/K ratios in solution and heavy metal contents in the humus layer may limit nutrient uptake due to root damage.

Tree species and stand age appeared to be very important explanatory variables. Together they explained about 44% of the variation in defoliation class (cf Table 5). The defoliation class becomes higher (worse vitality) with an increasing stand age. Using Mallows Cp as criterium to select the best subset of explanatory variables, the models 2, 3 and 4 gave the best description of the relation with the defoliation class. The percentage variance accounted for was 46% for all of the three equations. Using the percentage accounted for as criterium, the best description of the defoliation class was obtained by model 5. The value of R^2_{adj} was 53%.

According to model 5 the defoliation class increases (worse vitality) with an increasing foliar N content and/or a decreasing pH of the soil solution. This is a well acceptable explanation in view of existing theories on forest vitality (e.g. Ulrich and Matzner, 1983; Boxman en Van Dijk, 1988, Van den Burg and Olsthoorn, 1994). The nutrient status of the foliage and soil was, however, relatively unimportant in explaining forest vitality compared to tree species and stand age. A decrease in vitality could only partly be related to a relative P deficiency, due to N excess in the foliage, and to a decreasing pH of the soil solution. Results of this study do not confirm the theory that an increased Al concentration causes a decreasing vitality (Ulrich and Matzner, 1983), which is mainly based on laboratory experiments, showing root damage and limiting nutrient uptake at increased Al levels.

Table 5 *Percentage of variance accounted for (R^2 adj) in several regression models between defoliation class, site characteristics and chemical variables*

Explaining variable	Explaining models				
	1	2	3	4	5
Tree species	*	*	*	*	*
Stand age	*	*	*	*	*
Canopy coverage					*
Soil type		*			*
Foliar content N				*	*
Soil pH				*	*
R^2 adj	0.44	0.46	0.46	0.46	0.53

4 DISCUSSION AND CONCLUSIONS

Critical loads

In the Netherlands, critical loads for nitrogen and acidity have been derived using (i) empirical data that directly relate loads to effects and (ii) steady-state soil models that calculate critical loads from critical chemical values for ion concentrations or ratios in foliage, soil solution and groundwater. Critical loads that thus have been derived are shown in Table 6 (cf De Vries, 1993). Based on the data in Table 6, a target acid load of 1400 mol_c ha^{-1} yr^{-1} has been set for the year 2010 with an N input below 1000 mol_c ha^{-1} yr^{-1}.

The uncertainly in critical loads given in Table 6 is strongly influenced by the reliability of the critical values for the chemical parameters. A critical review on the various criteria for Al (Sverdrup and Warfvinge, 1993) shows that the Al concentration criterion of 0.2 mol_c m^{-3} is very unreliable, whereas the molar Al/Ca ratio (or better Al/(Ca + Mg + K) ratio) should be defined as a function of tree species. Sverdrup and Warfvinge (1993) derived critical Al/(Ca + Mg + K) ratios of 0.2 for Willow, 0.5 for larch, ash and black alder, 0.8 for Scots pine and Norway Spruce, 1.2 for Birch, 1.7 for Oak and Beech and 3.3 for Douglas fir, which are all common tree species in the Netherlands. They took a 20% reduction in either biomass growth, root length or root growth as the criterium to derive such ratios while using a large compilation of literature data on the effects of Al on trees. Apart from Al, a critical NO_3 concentration related to vegetation changes is also very unreliable. A nitrogen mass balance for a calcareous grassland in the Netherlands indicates that vegetations changes may take place in a situation where N leaching hardly increases above natural background values (Van Dam, 1990). Similarly, N leaching is nearly negligible in Dutch heathlands changing into

Table 6 *Average critical loads for acidity and nitrogen for forest ecosystems in the Netherlands (After De Vries, 1993)*

Compound	Effects	Criteria[1]	Critical loads ($mol_c\ ha^{-1}\ yr^{-1}$)	
			Coniferous forests	Deciduous forests
Acidity	Root damage	$Al < 0.2\ mol_c\ m^{-3}$	1100[2]	1400[2]
	Inhibition of uptake	$Al/Ca < 1.0\ mol\ mol^{-1}$	1400[2]	1100[2]
	Al depletion	$\Delta Al(OH)_3 = 0\ mmol_c\ kg^{-1}$	1200[2]	1300[2]
	Al pollution	$Al < 0.02\ mol_c\ m^{-3}$	500[2]	300[2]
Nitrogen	Inhibition of uptake	$NH_4/K < 5\ mol\ mol^{-1}$	1250-5000[3]	
	Increased susceptibility	$N < 1.8\ \%$	1500-3000[4]	
	Vegetation changes	$NO_3 < 0.1\ mol_c\ m^{-3}$	500-1400[5]	800-1400[5]
	Nitrate pollution	$NO_3 < 0.4\text{-}0.8\ mol_c\ m^{-3}$	900-1500[6]	1700-2900[6]

[1] Background information on the various criteria is given in De Vries (1993). Critical Al and NO_3 concentrations and critical Al/Ca and NH_4/K ratios related to root damage, inhibition of nutrient uptake and vegetation changes refer to the soil solution. Critical Al and NO_3 concentrations related to pollution refer to phreatic groundwater. Critical N contents related to an increased risk for frost damage and diseases refer to the foliage.

[2] Derived by a steady-state model. Al pollution refers to phreatic groundwater. For ground water used for the preparation of drinking water, a critical acid load of 1600 $mol_c\ ha^{-1}\ yr^{-1}$ was derived (cf De Vries 1993).

[3] Derived by a steady-state model assuming to nitrification (first value; worst case) and 50% nitrification in the mineral topsoil (second value).

[4] Empirical data on the relation between N deposition and foliar N contents.

[5] The first value is derived by a steady-state model ('worst case') and the second value is based on empirical data.

[6] Derived by a steady-state model using a critical NO_3 concentration of 0.4 and 0.8 $mol_c\ m^{-3}$ respectively. NO_3 pollution refers to phreatric groundwater. For deep groundwater, the critical load will be higher because of denitrification.

grasslands. It is the increase in N availability through enhanced N cycling that triggers the vegetation changes (Berendse et al., 1987). The second value in Table 6 based on empirical data is this likely to be more reliable.

Observed effects

The past decade of acidification research in Dutch forests has unequivocally shown that acid atmospheric deposition causes large changes in the chemical composition of foliage, (soil) solution and groundwater and in the understorey of forests. Critical loads related to these effects (cf Table 6) are strongly exceeded (the average acid load on Dutch forests exceeds 4000 $mol_c\ ha^{-1}\ yr^{-1}$) and this does have clear impacts on the forest ecosystem (cf Table 7).

Effects of elevated S and N deposition on soil (solution) and groundwater chemistry are most evident. Field studies showed that SO_4 behaves conservative in Dutch forest soils, whereas N is largely retained. Despite the high N

Table 7 Possible effects when critical loads are exceeded and observed effects in Dutch forests.

Possible effects	Average Critical load ($mol_c\ ha^{-1}\ yr^{-1}$)	Observed effects in the field
Rootdamage	1100-1400[1)]	Large exceedances of critical Al concentrations
Inhibition of uptake	1100-1400[1)]	Large exceedances of critical Al/Ca ratios
	1250-5000[2)]	Small exceedances of critical NH_4/K ratios
Al depletion	1200-1300[1)]	Depletion of secondary Al compounds
Groundwater pollution	300-500[1)]	Large exceedances of critical Al concentrations
	900-2900[2)]	Substantial exceedances of critical NO_3 concentrations
Increased susceptibility	1500-3000[2)]	Substantial exceedances of critical N contents; Nutrient imbalances; Increased shoot/root ratios
Vegetation changes	500-1400[2)]	Strong increase in nitrophilous species
Defoliation	------	Decline in past ten years, but no clear relationship with abiotic effects

[1)] Refers to acid loads
[2)] Refers to nitrogen loads

deposition, actual soil acidification, which is mainly manifested by leaching of Al associated with SO_4 and NO_3 leaching, is dominantly caused by S deposition. SO_4, NO_3 and Al concentrations increase in the order: deciduous forests < pine forests < spruce forests. Presumed critical Al concentrations (0.2 $mol_c\ m^{-3}$) and Al/Ca ratios (1.0 mol mol^{-1}) related to effects on roots are mostly (ca. 60 - 80%) exceeded below 20 cm soil depth. Concentrations of Al and NO_3 in groundwater often exceed EC drinking water standards (80% for Al and 20% for NO_3). The present atmospheric input has also caused a decline in the content of readily available secondary Al compounds, that mainly buffer the acid input (Wesselink et al.,1994). This may cause a further pH decline of the soil, which in turn may strongly affect the vitality of the forest(Houdijk,1993).

The relative small contribution of nitrogen to the acidification of Dutch forest soils as compared to sulphur does not imply that sulphur has a larger impact on the vitality of Dutch forests since the relation between soil acidification and forest vitality in the field is not very evident. This can be derived from both correlative field research (cf section 3.3) and from liming experiments in stands of Douglas fir and Japanese larch (Van den Burg and Olsthoorn, 1994). Most probably, at this moment the eutrophying impact of nitrogen is more important than the acidifying impact. Firstly, elevated N deposition strongly affects the forest nutrient status. Field studies showed an increase in N content of more than 50% in the last three decades. At present, about half of the Dutch forests have foliar N contents exceeding a critical limit related to an increased risk for frost and fungal diseases. Compared to N there is a considerable relative Mg deficiency (observed in 53% of the forests) and a large absolute P deficiency (observed in 63% of the forests). Recent results from fertilization experiments have shown that Douglas fir and Japanese larch reacted positively on P fertilization. Secondly, a large N input initially stimulates above ground growth but it leads to a stronger sensitivity to drought due to an increase in shoot/root ratio (De Visser,1994). Finally, there is ample circumstantial evidence that elevated N deposition during the last decades has caused large changes in forest undergrowth. Comparison of recent and old vegetation descriptions at hundreds of sites indicates an increase in nitrophilous species (both mosses and vascular plants) and grasses and a decrease of lichens and ectomycorrhiza mushrooms.

Unlike the effects on forest undergrowth, circumstantial field evidence for a relationship between soil acidification and nutrient imbalances in the soil and the foliage on one hand and the vitality of forests (mainly expressed by defoliation class) on the other hand is lacking. Tree species and stand age explains most of the variation in the defoliation class. Stand age may indirectly be related to air pollution since the period of exposition increases with stand age. Defoliation increased with an increase in N contents (relative P deficiency) and a decrease in pH, but the explanation of the defoliation class increased only very slightly when these variables were included. Aluminium in the soil solution appeared to have no significant effect on the defoliation class, even though there is ample evidence for its toxic effect in laboratory experiments. This result implies that an exceedance of critical acid loads, based on critical Al/Ca or Al/(Ca + Mg + K) ratios observed in laboratory experiments in relation to effects on root (uptake), do not imply visible effects or even the dieback of forests. However, an exceedance of critical loads does affect the long-term sustainability of forests due to depletion of secondary Al compounds. This risk increases when the rate at which present loads exceed critical loads is higher and the duration is longer.

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