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ATMOSPHERIC ACID DEPOSITION: EFFECTS ON THE CHEMISTRY OF FOREST SOILS

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ABSTRACT

Enhanced acidification of surface horizons of forest soils takes place in areas with high inputs of acid atmospheric deposition. The evidence for enhanced soil acidification comes from measurements of old and new soil samples, from chemical analyses of soil solutions, form input-output budgets of soils and watersheds, and from geochemical studies on soil profiles. These results are supported by simulation models of soil acidification.

As a result of enhanced soil acidification, mineral acidity has largely replaced organic acidity as the dominant acid source in forest soils. Many forest soils are now characterized by lower pH values, higher levels of dissolved and adsorbed aluminum and lower levels of adsorbed base cations than their unpolluted counterparts. Saturation of ecosystems with atmospheric nitrogen from NO_x and NH_3 increases the risk of further soil acidification, particularly in situations of forest decline.

The higher levels of dissolved aluminum, combined with lower levels of base cations, especially calcium, are potentially harmful for most plants. Increased supply of nitrogen only temporarily increases primary production, but eventually leads to further soil acidification, nutrient disorders and vegetational changes. While it is difficult to prove or disprove the thesis that enhanced soil acidification plays a major role in the present forest decline, there can be little doubt that negative effects occur, and are likely to increase in the future unless deposition rates are drastically decreased.

Levels of atmospheric deposition that are acceptable for terrestrial ecosystems in the long run vary, but are in the order of 1 to 1.4 equivalent kmol/ha.yr for sulfur and 0.4 kmol/ha.yr for nitrogen. Current deposition rates in many forests in central and western Europe are 2 to 8 equivalent kmol/ha.yr for S and 1 to 3 kmol/ha.yr for N.

INTRODUCTION

The question whether atmospheric deposition significantly contributes to soil acidification is still debated. Some soil scientists consider these effects small or insignificant:

"Additions of acidity in precipitation to soils are insignificant in comparison with the acidity produced in natural processes and by agricultural soil amendmends. ...The information available to this time indicates that acid rain has not adversely affected soils. ...The pronounced intrinsic buffering capacity provided by soil minerals and organic matter tends to minimize changes in pH of soils receiving acid precipitation. The amounts of N and S added by precipitation are beneficial to agricultural, forested and unmanaged soils". (ref. 1).

In this article we will show that there can be no reasonable doubt that anthropogenic atmospheric deposition has strongly enhanced soil acidification in many forest areas in Europe, and has created chemical soil conditions not normally found in unpolluted soils. We will call attention to the potential problems associated with high inputs of atmospheric nitrogen in many forest areas in western Europe. Next we will consider the more difficult question whether the change in chemical soil conditions is related to the forest decline reported recently in western Europe. Finally we will briefly discuss what levels of atmospheric deposition of sulfur and nitrogen are acceptable in the long run.

RATES OF SOIL ACIDIFICATION

Soil acidication is defined here as a decrease in the acid neutralizing capacity (ANC) of soil material to a certain depth (e.g. to the lower boundary of the rootzone) below the soil surface (refs. 2 and 3). A decrease in ANC is often, but not always, associated with ecologically important changes in soil chemistry such as (1) a decrease in pH, (2) a decrease in the contents of exchangeable bases in the soil, and (3) an increase in the concentrations of potentially toxic metals (e.g. aluminum) in the soil solution. We will return to these ecologically important soil changes later.

Chemical input-output budgets for ecosystems and for associated soils can be transformed into proton budgets, which show the magnitude of proton sources and sinks attributable to various groups of processes (refs. 2 and 4). By comparing proton fluxes associated with (1) atmospheric deposition, (2) internal acid formation in the soil (e.g. dissociation of organic acids and CO_2), (3) uptake of cations and anions by plants, (4) mineralization of cations and anions from organic matter, (5) mineral weathering and ion exchange reactions, and (6) drainage export of dissolved acidity, the relative importance of atmospheric deposition to the proton budget of an ecosystem can be evaluated. Table 1 gives proton budgets for ecosystems with acidic and near-neutral soils, and at low and high levels of atmospheric acid deposition.

Rates of acid deposition in areas far from important pollution source areas, as in Indonesia and Washington State, USA (ecosystems 1 and 3) are generally below 1 kmol of H^+ per ha per year. In such areas atmospheric deposition does not significantly contribute to soil acidification (ΔANC). In the (naturally) acid soil that receives high loads of atmospheric acidity (ecosystem 2),

TABLE 1

Proton budgets of selected ecosystems. Proton transfers in kmol/ha.yr. Taken from van Breemen et al (refs. 5 and 6).

Ecosystem	sources				sinks				
	atmos-	internal							
	pheric input	CO_2	biomass+ weathering	sum	sum	biomass	weathering	drainage output	ΔANC
Moderately acid soil, low atmospheric input									
1. Agathis plantation, Indonesia	0.0	4.0	2.6	6.5	6.5	3.0	3.5	0.0	-2.1
Acidic soil, high atmospheric input									
2. Picea forestSolling, BRD	4.8	0.0	2.3	7.1	7.0	0.6	6.1	0.4	-5.9
Neutral soil, low atmospheric input									
3. Unvegetated till Wash., USA	0.4	7.2	2.2	9.8	9.9	0.0	9.9	0.0	-9.9
Calcareous soil under Quercus, moderate to high atmospheric input									
4. Castricum, Neth.	2.2	12.4	0.2	15.0	15.1	0.6	14.5	0.0	-14.3
5. Hackfort, Neth.	7.9	4.7	3.8	16.4	16.3	0.2	16.1	0.0	-15.4

however, most of the soil acidification is clearly due to atmospheric acid deposition. In near-neutral soils the rate of internal proton production due to dissociation of CO_2 can be very high, and normally exceeds the load of atmospheric acidity (ecosystems 3 and 4). However, in calcareous soils with excessive atmospheric deposition (ecosystem 5), CO_2 appears to be replaced by strong mineral acids (sulfuric and nitric acid) as the agent of soil acidification, while the toal proton load of the system remains about the same (compare ecosystems 4 and 5). Similar findings were reported for other ecosystems (ref. 5). That rates of present-day soil acidification in areas with high atmospheric deposition are indeed higher than during pre-industrial times also follows from the studies of the change in ANC with depth in soil profiles of known age developed in presumably homogenous parent material. By dividing the total amount of cations, that have apparently disappeared over the depth of the soil profile by the age of the soil, one obtains the mean annual decrease in ANC over the soils' life time. Over the past ten thousand years, pools of base cations plus Al have decreased annually by 0.1–0.5 (equivalent) kmol/ha.yr in podzolic soils in the Netherlands (ref. 7) to 1.1–3.2 kmol/ha.yr in (mineralogically richer) acid brown forest in the FRG (ref. 8). The present-day rates of removal of Al plus base cations in the same soils under forest with typical (high) rates of atmospheric deposition in these countries are two to ten times higher.

EVIDENCE FOR DECREASED SOIL pH AND INCREASED LEVELS OF DISSOLVED ALUMINUM

Whether the accelerated decrease in ANC has indeed affected soils ecologically, e.g. by lowering the soil pH and increasing the levels of dissolved aluminum, can only be decided by comparing affected ecosystems with clean(er) reference ecosystems. Reference data may come from (1) ecosystems in less polluted areas that are very similar to the polluted ecosystem, and (2) samples or analytical data of the same soil at a less advanced stage of atmospheric pollution (i.e. by comparing "old" and recent samples). Ecosystems in polluted and less polluted areas can never be completely comparable. Nevertheless, differences in soil chemical conditions of similar ecosystems are sometimes so outspoken that there can be little doubt about the effects of acid deposition. In clean environments such soils mainly acidify by loss of base cations (Ca, Mg, K and Na, plus small amounts of Al in podzolic soils) due to CO_2, organic acids and uptake by plants. With increasing atmospheric deposition, dissolution of inorganic Al in acidic soils becomes increasingly important, and Al may eventually become the dominant cation, with SO_4 and NO_3 as accompanying anions (Table 2).

TABLE 2

The chemical composition of soil solutions from the Bhs horizon in four podzolic soils (Typic Haplorthod) from Denmark (sites 1 and 2) and the Netherlands (sites 3 and 4), sampled in December 1985. Vegetation cover is Calluna vulgaris (1, 3), Picea abies (2) and Pinus silvestris (4). Al is mainly free Al^{3+} at all sites.

site no	estimated acid input kmol/ha.yr	pH	Ca	Mg	K	Na	Al	NO_3	SO_4	Cl
			equivalent ionc concentration, mmol/m^3							
1	0.3	4.5	88	227	15	596	141	0	126	989
2	1.0	4.4	10	135	41	1120	546	0	406	1458
3	2.0	4.2	84	39	3	202	960	0	892	349
4	3.5	4.1	74	117	72	250	2350	1440	1578	481

High concentrations of dissolved aluminum associated with strong acid anions (sulfate and nitrate) in forest soil solutions in polluted areas have also been described from the FRG (ref. 9) and Sweden (ref. 10). In Solling, FRG, a distinct event in the soil acidification process was observed in the course of the chemical monitoring carried out there since 1969: the concentration of aluminum in the soil solution at 100 cm depth in acid brown forest soils under spruce increased from between 2 and 8 mg/l before 1975, to between 14 and 24 mg/l after 1977 (ref. 12).

Evidence for a depression of soil pH by atmospheric deposition from "old" and "new" soil samples has been described from the FRG (refs 13 and 14), Austria (ref. 15) and from southern Sweden (refs 11 and 16). The pH values of slightly to strongly acidic forest soils (pH 5 to 3.5) in these areas have decreased further over the past 5 to 55 years. One problem with this approach is that the pH of the surface soil tends to decrease with the age of the forest stand, due to formation of an organic forest floor and the assimilation of base cations by the vegetation. In the work by Hallbäcken and Tamm (ref. 11) the effects of stand age and of year of sampling (resp. 1927 and 1984) have been separated (Fig. 1). The results show that between 1927 and 1982 the soil pH has decreased by 0.3 (in the humus layer), 0.5 (A2 and B horizons) and 0.7 (C horizon) by some process not related to the vegetation, most likely as a result of acid atmospheric deposition. This conclusion is further corroborated by a parallel study in northern Sweden, where atmospheric acid deposition is negligible. There the same relationship between stand age and soil pH was observed, but pH values of "old" and of "new" samples did not differ significantly (C.O. Tamm, pers. comm.).

Modelling work, for example at IIASA (ref. 17), shows that the observed decreases in pH in acidic forest soils discussed above can indeed be explained, given the ambient rates of atmospheric deposition, pools of buffering substances in the soil and reasonable estimates for rates of buffering processes.

EVIDENCE FOR INCREASED N LEVELS IN SOILS AND DRAINAGE WATER

On a molar basis, the wet and dry atmospheric deposition of nitrogen (as ammonia, ammonium and nitrate) in forest ecosystems in Western Europe (as indicated by fluxes in throughfall water) is in the same order as that of sulfur (ref. 18). Particularly high rates of N deposition as ammonia occur in areas with intensive animal production, e.g. in large parts of the Netherlands (ref. 19). Although historic data are lacking, ammonia deposition in the Netherlands has undoubtedly increased dramatically during the past decades. Over much larger areas in the industrial world, wet deposition of nitrate has increased strongly since last century (ref. 20). In some forest areas deposition of nitrogen may now exceed the capacity of the ecosystems to assimilate all or most of the nitrogen. This is indicated by increased concentration of nitrate in soil solutions, ground- and stream water draining forested areas (refs 21, 22 and 23). Acidification as a result of nitrate formation (microbial nitrification) is generally believed to stop when the pH falls below 4. However in most forest soils, contrary to arable soils, nitrification can continue to values as low as 3 (refs 24, 25, 26 and 27).

Therefore, "saturation" of the ecosystem by nitrogen involves the danger of increased soil acidification by nitrification. The situation becomes worse when, for whatever reason, the vitality of the trees (and hence the capacity of the ecosystem to assimilate nitrogen) decreases: nitrate (in fact nitric acid) formed by nitrification of mineralized organic nitrogen is not taken up by plants and microorganisms, but is free to mobilize base cations and aluminum, and to depress soil pH. Various studies (refs 22, 23 and 28) provide evidence that this chain of events does take place indeed.

ECOLOGICAL EFFECTS OF SOIL ACIDIFICATION

The cause of the widespread forest decline reported from various mid- and western European countries is now generally thought to be related to air pollution. It is not clear, however, whether air pollutants exert their influence mainly directly by affecting above-ground parts of the vegetation (leading to decreased photosynthesis), or mainly indirectly through soil acidification followed by impairing the functioning of roots. For instance, the low levels of Mg , K and Ca observed in many damaged forest stands in the FRG, and the positive effects of fertilizing affected trees (see e.g. the review by Baule, ref. 29) can be explained either by direct effects of pollutant on leaves, or by the effects of increased soil acidification (increased leaching of base cations; Al toxicity). Root growth seems to be invariably depressed in affected forest stands in the FRG (ref. 30), and could also result either from high levels of dissolved Al in the soil, or from decreased photosynthetic activity due to direct effects of pollutants on leaves. But measurements of root biomass dynamics and of the starch content of fine roots of norway spruce indicate that adverse soil conditions are probably more important than depressed photosynthesis in affecting root growth (ref. 30).

Effects of soil acidification on plants, microbes and soil fauna are highly complex. We will briefly discuss some of the results obtained with (1) experimental acidification of soils (2) pot- or nutrient solution studies with plants, and (3) correlative field studies on vegetation and atmospheric depostion.

Many field- and pot experiments about the effects of soil acidification on vegetation, soil micro-organisms and soil biota have been done by applying simulated acid precipitation and by recording changes in the biota. Often, the maximum deposition rates applied are up to two orders of magnitude higher than in actual field conditions (e.g. simultated acid rain of pH 2.5 in stead of 4.5) to mimmick the effect of prolonged (decades to centuries, rather than years) acid deposition. A review of many of such studies has been prepared by the VDI (ref. 18). Both positive and negative effects of artificial acid rain on trees (refs 31 and 32) and soil fauna (ref. 33) have been reported. This ambiguity is not surprising considering the complicated mix of potentially beneficial effects (increased supply of N, S, and, at least on the short term, base cations) and potentially harmful effects (increased levels of dissolved Al and, on the longer term, decreased levels of base cations) of adding large amounts of acid to a soil in a short period.

Most pot- or nutrient solution studies on the effects of acid deposition on plants deal with the effects of aluminum. It is generally agreed that high levels of dissolved inorganic Al (i.e. Al not complexed by organic ligands) are harmful to plants. This knowledge plus the observation that soil solution levels of inorganic Al have increased as a result of acid atmospheric deposition, is the basis of Ulrich's hypothesis of Al toxicity as a major factor affecting forest ecosystems (ref. 9). The aluminum toxicity question is, however, very complex. Particularly with trees that grow normally or often in acidic soils (norway spruce, scots pine, birch, beech), levels of dissolved aluminum reported to have harmful effects varied widely, from lower than those found frequently in strongly acidified soils (2 to 10 mg/l) to high concentration rarely observed in the field (20-50 mg/l) (ref. 36). These results always refer to seedlings. Low concentrations of dissolved Ca increase the susceptibility for Al of spruce and beech (ref. 34), and ratios of dissolved Ca/Al or (Ca+Mg)/Al may be ecologically more relevant than absolute Al-concentrations. High concentrations of nitrate increased the susceptibility of different grasses for dissolved Al (ref. 37). Whereas these results do not prove that Al toxicity plays an important role in forests affected by acid rain, they certainly do not exclude Al as an important adverse factor. Concentrations of other potentially toxic cations such as Mn and various heavy metals can also increase to dangerous levels as a result of soil acidification, and these may be involved locally in depressed growth of trees and herbaceous plants, for instance at sites of stemflow infiltration adjacent to beech trees (ref. 38). In the Netherlands high concentrations of ammonium in water in contact with leaves and shallow roots may induce deficiencies of other nutrients in trees, in particular of K and Mg (ref. 39).

For epiphytic lichens, certain mushrooms (Cantharellus cibarus) and a number of herbaceous plants, the decline in occurrence between 1950 and 1980 in the Netherlands, and the concentrations of SO_2 in the air were spatially correlated, suggesting a causal relationship involving direct or indirect effects of air pollution (ref. 40). Wittig and coworkers (refs 41 and 42) have observed a significant increase in acidophilic plants in the herb layer of Westphalian beech forests between 1976 and 1983.

The studies cited above do not, and could not, prove conclusively that soil acidification by air pollution has important and often adverse effects on forest ecosystems, but in our opinion there can be little doubt that such effects occur.

CRITICAL LOADS OF ACIDITY AND NITROGEN ON TERRESTRIAL ECOSYSTEMS

Because inorganic dissolved aluminum is generally thought to play a key role in toxicity problems in both the terrestrial and aquatic environment, inorganic aluminum in soil solutions is often used as an indicator for potential adverse effects from atmospheric acidic deposition. Total dissolved aluminum is not suitable for this purpose, because the organically complexed aluminum commonly present in soil solutions of naturally acidic soils (Spodosols) is generally not toxic.

At the workshop on "Critical Loads for Effects of Sulphur and Nitrogen Deposition on Forest Soils, Groundwater and Surface Water" held in Oslo (ref. 43) it was agreed that input of potentially acidic substances in soils should be limited to levels which will only cause a negligible mobilization of inorganic aluminum in most acidic soils. Negligible mobilization of inorganic Al mobilization in soils is essential in areas where soil drainage water directly enters a surface water body, because aquatic organisms are generally more sensitive than terrestrial organisms to dissolved inorganic aluminum. Mobilization of inorganic aluminum is insignificant if total acid inputs (anthropogenic and natural) do not exceed the rate of weathering of base cations from primary minerals. At the Oslo Workshop, acceptable atmospheric acid loads were estimated from current base cation weathering rates in acidic soils (Spodosols, Acid Brown Forest Soils), using input/output studies (both for catchments and individual soil profiles) carried through in Scandinavia, central Europe and the Netherlands. Reported values for acceptable atmospheric acid loads range from 0.05 to 0.75 kmol H^+/ha.yr (ref. 43); 0.1 to 0.2 kmol H^+/ha.yr would probably not cause inorganic aluminum mobilization in most European acidic soils. It should be realized, however, that the weathering rate of primary minerals decreases upon prolonged soil acidification, so that in the long rund the acceptable acid load will decrease. For comparison, the current atmospheric acid input in southern Scandinavia is about 1 kmol H^+/ha.yr, while in Central Europe the acid inputs often exceed 3 kmol H^+/ha.yr.

The crital values indicated above may not apply to the very sensitive soils in large parts of the Netherlands, northern FRG and western Jutland, which have developed in sandy deposits, that are extremely poor in weatherable silicate minerals. Such soils show essentially no base cation leaching. In fact, atmospherically supplied nutrient base cations are retained in the ecosystem (Mulder, unpublished). In these acidic soils any atmospheric acidity is neutralized by aluminum solubilization (ref. 44). So, to prevent Al mobilization, the atmospheric acid load would have to be close to 0 kmol H^+/ha.yr! Because (1) terrestrial organisms appear to be less suceptible to inorganic aluminum than aquatic organisms and (2) acidic, sandy soils in the Netherlands hardly ever generate suface water, mobilization of limited amounts of aluminum by atmospheric acidic deposition in the Netherlands is probably acceptable. Molar ratios of dissolved (Ca+Mg)/Al < 1 have been proposed to indicate potential adverse effects on tree growth in sensitive sandy forest soils in the Netherlands (ref. 45). Dissolved (Ca+Mg)/Al ratios < 1 in the rooting zone occur when total atmospheric acid loads exceed 1 kmol H^+/ha.yr in the most sensitive sandy forest soils, or 1.4 kmol H^+/ha.yr in acid brown forest soils in the Netherlands. Current total acid loads in forest soils in the Netherlands are 3 to 6 kmol/ha.yr.

In most terrestrial ecosystems in the world increased atmospheric deposition of NOx and NH_3 would increase primary production. With continued high N deposition, however, other nutrients or toxic factors would limit or decrease primary production, and essentially all added nitrogen would be leached from the system. Information so far indicates that in most N-saturated systems nitrogen is leached in the form of nitrate. Many woodlands in the Netherlands, are saturated with nitrogen due to a prolonged high deposition of NH_3, and nitrification and the subsequent leaching of nitrate cause severe soil acidification. Soil acidification, imbalance in nutrients and increased nitrate concentrations in groundwater, can be decreased when N-saturation is avoided.

In the long term, the critical nitrogen load in ecosystems must be determined by the net uptake in plants in the standing biomass (ref. 43). Andersen (ref. 46) estimates the net growth increment in the woody vegetation of coniferous trees to be 0.4 to 1.0 kmol N/ha.yr at sites with low to medium productivity in Europe and north America. In the Netherlands the net annual nitrogen uptake in the woody biomass of pine is in order of 0.4 kmol/ha.yr for pine, and 0.4 to 1.3 kmol/ha in an oak-birch woodland (ref. 47). So, safe deposition rates of nitrogen (NH_3 + NO_3) at sites with a low productivity would be around 0.4 kmol/ha.yr. In south Scandinavia the annual total N deposition now exceeds 1.5 kmol/ha, while in central Europe the annual N deposition is between 2 and 3 kmol/ha. In the Netherlands the annual deposition of N on forests ranges from 3 to 4.5 kmol/ha. Natural background values, e.g. in remote areas in Canada, are less than 0.1 kmol N/ha.yr (ref. 48).

As indicated above it is important for both forest vitality and the quality of drinking water supplies to reduce the current N deposition in the Netherlands by 90%. At such levels of N deposition, nitrogen can be completely assimilated by the standing biomass and therefore can no longer cause soil acidification. The current deposition of 2.4 to 4.0 equivalent kmol S/ha.yr in the Netherlands should be reduced by at least 75% in order to keep aluminum concentrations in the soil solution within acceptable limits.

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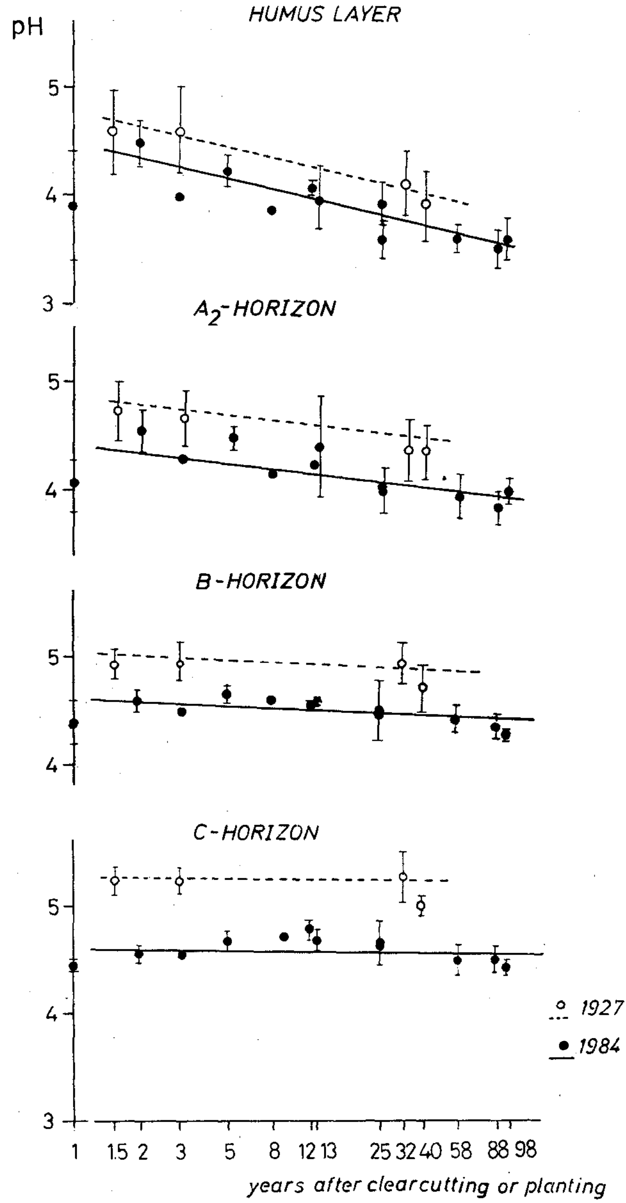

Fig. 1. Soil pH (in water) against time after clearcutting or planting in forest soils in Southern Sweden, for samples taken in 1927 and 1984 (Reproduced by permission from Hallbäcken and Tamm, ref.11).