

Effects of acid deposition on soil and sensitivity of the soil to acidification

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Introduction

The pollutants emitted during combustion of fossil fuels and deposited on the soil surface or the vegetation will sooner or later enter the soil and react with the soil components. The pollutants will affect the solid soil material as well as the composition of the soil solution and the drainage water. Due to the acidity of the dominant pollutants, the deposition may cause an increased acidification of the soil. Soil acidification is, however, also caused by natural processes and, in cultivated soils, by processes induced by cultivation. Therefore, the extent of acidification caused by pollutants must be evaluated in relation to the acidification which is due to natural processes and cultivation.

In this respect it is important that the regions in North America and Europe, where the acid deposition is high due to a high density of industry, have a humid climate which promotes natural soil acidification. In a humid climate precipitation exceeds evaporation and the major part of the excess precipitation normally percolates the soil profile and causes leaching of soluble substances. This leaching, in combination with production of acids in the soil, is the major cause of natural soil acidification.

Natural soil acidification

As a result of biological processes a number of acids including carbonic, nitric, sulphuric and various organic acids are produced in the soil. In addition, plant uptake of nutrients as well as humus production and decomposition may affect soil acidity. The acids produced in the soil become neutralized by reaction with bases, and if the reaction products are soluble and hence subject to leaching, this causes a removal of bases from the soil.

In a natural ecosystem at equilibrium some of the processes are of minor importance; the main ones to consider are the effects of carbonic acid and soluble organic acids. In soils which are not strongly acid the major factor in natural soil acidification is carbon dioxide which is produced by plant roots and by organisms, mainly microorganisms, decomposing organic matter in the soil. As a result the carbon dioxide content in the soil air may be several times higher than in the atmosphere. While the major part of the carbon dioxide is released to the atmosphere a smaller amount is dissolved in the soil water, where it forms carbonic acid which reacts with bases in the soil. Due to the high carbon dioxide content in the soil air the carbon dioxide concentration in the soil water may become quite high.

The extent of leaching, i.e. the amount of bases that can be removed from the soil by carbon dioxide dissolved in the soil water, depends upon pH because carbonic acid is a weak acid. The pH of water in equilibrium with air containing 0.03% carbon dioxide (approximate content

in the atmosphere) is about 5.6. If the carbon dioxide content in the air is 10%, the pH of the water becomes 4.5. Intermediate levels of carbon dioxide content in the air result in intermediate pH values. From this it follows that a soil pH lower than about 4.5 cannot be caused by carbon dioxide unless the carbon dioxide content in the soil air exceeds 10%.

The amount of bases leached by water containing dissolved carbon dioxide decreases strongly with decreasing pH. At pH 7, water in equilibrium with air containing 1% carbon dioxide dissolves 1.5 me bases (metal ions) per liter, while at pH values less than 5 no bases are dissolved at all. Hence, leaching of bases proceeds rapidly only from soils with comparatively high pH values.

Despite the fact that pH values below about 4.5 cannot be caused by carbon dioxide, soils that are considerably more acid are common in humid climates. The reason is that soluble organic acids with an acid strength exceeding that of carbonic acid cause leaching of bases at lower pH values. Evidence for the occurrence and leaching of such acids is given, for examples in Bruckert³, Coulson et al.⁴ and Petersen¹².

Under natural conditions soil acidification is counteracted by bases produced by weathering of minerals. The rate of this base production depends on the content of weatherable minerals in the soil. However, in most soils this base production is insufficient to prevent soil acidification. At pH values about 4, aluminium compounds, which are abundant in most soils, become effective in preventing further decline in pH.

Soil acidification due to cultivation

In addition to the processes causing soil acidification under natural conditions, some processes induced by application of fertilizers and removal of plant material affect the acid-base status of cultivated soils. The effects of fertilizer application depend on the nature and amounts of fertilizer applied. Basically, fertilizers containing plant nutrients as anions such as nitrates and phosphates counteract acidification since uptake of anions by the plants is compensated by excretion of hydroxyl or hydrogen carbonate ions. On the other hand, fertilizers containing plant nutrients as cations, e.g. potassium, contribute to soil acidification because plant uptake of cations is compensated by excretion of hydrogen ions. In a closed ecosystem these effects are balanced when the plant material is returned and decomposed in the soil. This is, however, not the case in cultivated soils where the plant material to a larger or smaller extent is removed from the soil.

While nitrogen applied as nitrate counteracts acidification, the opposite is true when nitrogen is applied as ammonium. An acidifying effect occurs, both when the ammonium is taken up directly by the plants and when it

is subject to nitrification prior to uptake. If a fraction of the nitrate formed by nitrification of ammonium is lost by leaching, which will often be the case on soils receiving large dressings of fertilizers, the acidifying effect is enhanced, since the base production occurring in connection with plant uptake of nitrate is absent.

In practice the net effect of the processes mentioned is usually an increased soil acidification. An estimate of the acid production caused by the different processes in an average Danish agricultural soil is given in table 1⁸. The extent of the acidification depends on the intensity of the cultivation practice. It is large in intensive agriculture and horticulture, but it is not altogether absent in forestry, where fertilizers are occasionally applied and where removal of plant material takes place.

The cultivation practice in intensive agriculture and horticulture usually includes application of lime to the soil which reverses soil acidification. Accordingly, in humid climates, where natural soil acidification is important, cultivated soils are normally less acid than their uncultivated counterparts despite the increased acidification caused by cultivation.

Atmospheric deposition

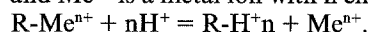
The nature and the amounts of pollutants deposited on the soil depends on a number of factors including the distance from the emitting sources and the climatic conditions. In addition to sulphur and nitrogen compounds the deposition includes mainly chloride, calcium, magnesium, sodium and potassium. The compounds deposited may be considered to be partly neutralized sulphuric, nitric and hydrochloric acids. On soils covered by vegetation the greater part of both wet and dry deposition occurs on plant surfaces where the acids may be further neutralized before entering the soil. However, the bases used in this neutralization will originate from the soil.

Reactions of acid pollutants in the soil

Being acid, the pollutants will react with bases in the soil. In calcareous soils, i.e. soils containing calcium carbonate, a complete neutralization of the acid will take place, and except for a reduction in the calcium carbonate content, which is equivalent to the amount of free acid entering the soil, no detrimental effects are to be expected.

In non-calcareous soils the acids will react with exchangeable cations, i.e. cations held by electrostatic forces at the surface of negatively charged clay and humus particles. Normally, the dominant exchangeable

cations are Ca^{2+} , Mg^{2+} , K^{+} and Na^{+} (so-called exchangeable bases), H^{+} and Al^{3+} . Since the cation exchange ability is mainly a property of clay and humus the amount of exchangeable cations in a soil (the cation exchange capacity) increases with increasing clay and humus content, although it also depends on the kind of clay. Soil pH, which is the pH measured in a suspension of a soil sample in water or salt solution, is a function of the fraction of the cation exchange capacity which is accounted for by exchangeable bases (the base saturation percentage). With increasing base saturation percentage, pH increases. Hence, adsorbed bases dominate in soils with high pH values while H^{+} and Al^{3+} are dominant in acid soils. Reactions between hydrogen ions from pollutants and exchangeable cations may be illustrated as follows, where R is a negatively charged soil component (clay or humus) and Me^{n+} is a metal ion with n charges:



Reactions of this type will proceed until an equilibrium is reached, which is governed by the composition of the adsorbed cations and the activities of the ions in solution. Hence, the absorption of hydrogen ions by the ion exchange complex will increase with increasing amounts of adsorbed metal cations, i.e. increasing base saturation percentage. On the other hand, it will decrease with increasing concentration of metal cations in the soil solution.

Wiklander²² expressed the exchange of adsorbed metal cations with hydrogen ions from the soil solution by $\frac{\Delta \text{Me}}{\Delta \text{H}}$ where ΔMe is the amount of metal cations (bases) released from the ion exchange material when an amount of hydrogen ions (ΔH) is added to the soil. Laboratory experiments²³ showed that $\frac{\Delta \text{Me}}{\Delta \text{H}} = 1$ when dilute sulphuric acid having pH 3.56 was added to an aqueous suspension of a sample from a cultivated soil with pH 5.7. An identical experiment with a sample from the A2 horizon

of a podzol soil with pH 3.98 resulted in $\frac{\Delta \text{Me}}{\Delta \text{H}} = 0.25$. Hence in this soil the removal of bases amounted to only about 25% of the amount of hydrogen ions added.

Addition of neutral salts to the sulphuric acid solution decreased $\frac{\Delta \text{Me}}{\Delta \text{H}}$ below 0.1 in the case of the acid podzol soil but had no effect on the cultivated soil where $\frac{\Delta \text{Me}}{\Delta \text{H}}$ remained at unity.

These results show that a soil with a low pH adsorbs only a fraction of the acid from the soil solution. The consequence is that only a fraction of the acid deposited on acid soils contributes to further acidification of such soils. In fact if the soil pH is low enough, no acid will be adsorbed at all by the soil.

Whether hydrogen ions are adsorbed or not depends on the base saturation percentage of the soil and the pH of the solution entering the soil. If the soil has a base saturation percentage low enough to cause pH of the solution to become lower than it is when the solution is added to the soil, no hydrogen ions will move from the solution to the ion exchange complex, i.e. the soil does not become more acid.

To what extent pollutants may acidify the water entering the soil is not known precisely. The pH of the precipita-

Table 1. Estimate of acid production and deposition in arable soil in Denmark

	me per m ² per year
Carbon dioxide	450
Nitrification	> 210
Plant uptake of nutrients	80
Wet deposition	29
Deposition of particles	3
Dry deposition	85
Total	> 857

tion is monitored, particularly in parts of Europe and North America. However, dry deposition affects the acidity of the water entering the soil towards a lower pH. Hence, a valid comparison is not obtained by comparing soil pH with the pH of the precipitation. Even allowing for a decrease in pH due to the dry deposition, there is little doubt, however, that the upper horizons of several natural soils will be so acid that they do not take up significant amounts of hydrogen ions from the percolating solution.

In humid areas soil pH normally increases with depth. The increase is due to the natural soil acidification being more intensive in the upper soil layers. Table 2 shows the pH of each horizon in two Danish soils which have never received fertilizer or lime. The podzol is on coarse glacio-fluvial sand while the luvisol is in clayey morainic material which was originally calcareous. In this soil calcium carbonate is still present from a depth of 75 cm. As the soil solution percolates such profiles its composition will become modified in each horizon by cation exchange processes, provided, of course, that equilibrium is obtained or approached.

The implication of this is that the cationic composition of the soil solution is governed by the composition of the adsorbed cations, while the total amounts of cations leached depend partly on the amounts of anions deposited as pollutants. The cations deposited, of course, affect the composition of the adsorbed cations and hence, in the long run, the cationic composition of the soil solution. However, this is of rather small importance since, normally, the amount of free acid deposited is relatively small compared with the amounts produced by natural acidification processes, at least in soils that are not very acid where the leaching of bases due to the natural soil acidification process is rather high (table 1).

In acid soils the dominant adsorbed cation is Al^{3+} rather than H^+ . Hence, cations released from the cation exchange complex to the soil solution in acid soils tend to be Al^{3+} . This has important implications since Al^{3+} is toxic to plants when present in sufficiently high concentration in the soil solution. It may also have implications for the water resources receiving drainage water from the acid soils, since a high content of aluminum affects the biological quality of the water.

However, the aluminum compounds in the drainage water may be removed in strata below the soil profile, provided these strata are less acid than the soil profile. The

possibilities and extent of this removal depend on the geological and hydrological conditions. Where the underlying strata consist of impervious igneous rocks the water may flow to water courses mainly through superficial soil layers and large fissures in the rock. Under these conditions the possibilities of aluminum neutralization and removal are small. On the other hand, if the water passes through porous layers of soft limestone or other calcareous materials a complete removal of aluminum is likely to occur.

Soil acidification due to acid deposition

A number of investigations have been carried out in order to establish whether the acid deposition has caused a decrease in soil pH. Ulrich, Mayer and Khanna¹⁹ found that pH measured in 0.01 M $CaCl_2$ in two forest soils in Central West Germany decreased 0.3–0.4 units during 5–7 years from 1966–68 to 1973. The decrease occurred in all soil layers above a depth of 50 cm. However, in the subsequent period up to 1979 a pH increase occurred²⁰. According to Troedsson and Nykvist¹⁸ and Troedsson^{16,17} it has not yet been possible to demonstrate that the pH of the topsoil in Swedish forests has declined due to acid deposition. The conclusion is based on 30,000 pH measurements carried out in 1961–63 and 1971–73.

Wiklander²² found no decrease in pH in a Swedish podzol soil from 1934 to 1974. Similarly, Linzon and Temple¹⁰ found no significant decreases in pH in 6 sandy soils in Ontario, Canada from 1960 to 1978. Most of the soils studied were podzols and pH was measured on samples from all pedogenetic horizons. Last et al.⁹ concluded that it still remains an open question whether pH in soils that have been subject to acid deposition has changed significantly during the last decades.

It is, of course, problematic to compare pH values measured over large time intervals. The techniques of measurement have been changed, and soils are not homogeneous, which may result in considerable differences in pH, even within short distances. Furthermore, the problem of separating the effect of natural soil acidification from that of acid deposition remains unsolved.

A pH decrease is not likely to occur in the top layer of very acid soils, since these will not become more acid due to deposition of pollutants. Due to increase of pH with depth, and the different pattern of cation exchange processes at different soil pH values, it is indeed more likely that a decrease could be observed in the subsoil layers where the cation exchange processes cause exchangeable bases to be replaced by hydrogen and aluminium ions. A larger or smaller fraction of the acidity which is adsorbed by the subsoil layers originates, however, from the upper soil layers where it is produced mainly by natural soil acidification processes. Due to the higher concentration of anions, mainly sulphate, which is due to pollution, the transport of acidity to deeper soil layers is facilitated. Cronan and Schofield⁷ found that the aluminum content of the soil solution increased with depth in an acid soil under coniferous forest in New Hampshire, USA.

Results by Cronan et al.⁵⁻⁷ and van Breemen et al.²¹ show a high aluminum content in the soil solution in acid soils in polluted areas. In most of the investigations the dominant anion was sulphate but the soils investigated by van

Table 2. pH in two uncultivated soils in Denmark

Podzol			Luvisol		
Depth (cm)	Measured in water	Measured in 0.01 M $CaCl_2$	Depth (cm)	Measured in water	Measured in 0.01 M $CaCl_2$
0–5	3.6	3.0	0–9	4.8	3.9
5–8	4.2	3.1	9–20	5.0	4.0
8–13	4.3	3.3	20–29	5.4	4.2
13–18	4.5	3.8	29–38	5.8	4.6
18–23	4.7	4.0	38–50	6.1	4.9
23–33	4.7	4.3	50–62	6.4	5.3
33–43	4.8	4.6	62–75	7.6	6.1
43–62	4.6	4.6	75–88	8.2	7.6
62–86	4.9	4.8	88–101	8.3	7.7
86–130	5.0	4.8	101–114	8.3	7.8
			114–127	8.3	7.8

Breemen et al.²¹ showed a high content of nitrate in the soil solution. The mobility of aluminum may be increased by formation of complex AlSO_4^+ ions which, because of their low charge, are adsorbed less strongly by the cation exchange complex than more highly charged ion species¹⁴.

The investigations quoted show that a direct effect of the acid deposition on soil pH is yet to be demonstrated. However, many investigations indicate that the deposition of sulphate leads to an increase in the aluminum content of the soil solution in acid soils.

Other anions like nitrate and chloride may contribute to the increased content of aluminum in the soil solution. However, under natural conditions nitrate is probably removed rather completely through uptake by the vegetation. Ahl and Oden¹ found only a small leaching of nitrogen from areas covered with forest in Sweden. On the other hand, as mentioned above, van Breemen et al.²¹ found a considerable nitrate content in the soil solution of an acid as well as in that of a calcareous soil in the Netherlands. The nitrate was assumed to originate from ammonia evaporated from animal manure applied to areas near the soils studied and reacting with acid deposition products on the vegetation surface.

The main source of deposited chloride is seawater and the chloride will, therefore, be followed by an almost equivalent amount of sodium. Since sodium is weakly adsorbed by the cation exchange complex it has a low ability to replace adsorbed aluminum. Hence, the contribution of chloride originating from seawater to leaching of aluminum is probably low. In an investigation in an area near the coast in Norway Skartveit¹⁵ found the rates of deposition and leaching of sodium to be rather similar indicating that no significant net adsorption of sodium occurred in the soil.

Sensitivity of soils to acid deposition

The sensitivity of a soil to acid deposition could be defined as the decrease in soil pH or base saturation percentage caused by deposition of a certain amount of acidity. However, since the acid deposition causes additional effects such as changes in the composition of the soil solution the extent of such changes must also be included in a discussion of sensitivity.

Decrease in soil pH

As discussed in the preceding sections the acid deposition can only cause a decrease in soil pH when this is higher than the pH of the polluted water entering the soil. Hence, this effect will be absent in very acid soils. In soils that are less acid the ability of the soil to resist a decrease in pH depends on its capacity for neutralizing the acid deposited, i.e. on its buffer capacity.

In calcareous soils acid is neutralized through reaction with calcium carbonate: $\text{H}^+ + \text{CaCO}_3 \rightarrow \text{Ca}^{2+} + \text{HCO}_3^-$. As long as the soil contains calcium carbonate no reduction in pH will occur, and the amount of acid that can be neutralized is proportional to the content of calcium carbonate. A content of say 1% CaCO_3 in a 20-cm-thick soil layer enables the soil to neutralize about 250 ke acid per ha or 25,000 me per m^2 . This is a large amount compared

with the extent of acid deposition on most locations. Hence, even a small content of calcium carbonate makes the soil insensitive to acid deposition.

In non-calcareous soils the buffer capacity depends on the cation exchange capacity and the base saturation percentage. When both are high, large amounts of acid may be neutralized by the soil. The cation exchange capacity may vary from almost nil in sandy soils with low humus contents to values above 100 me per 100 g in organic soils.

A soil containing, for example, 4% humus and 20% clay will have a cation exchange capacity of 18 me per 100 g assuming the cation exchange capacity of humus and clay to be 200 and 50 me per 100 g, respectively, and disregarding any contribution to the cation exchange capacity from particle size fractions other than clay. A 20-cm layer of this soil will have a cation exchange capacity of 450 ke per ha or 45,000 me per m^2 . A reduction of the base saturation percentage by, say, 10% requires 4500 me acid per m^2 which is a large amount compared with the extent of acid precipitation. A 10% reduction in the base saturation percentage is likely to cause a pH decrease of about 0.4 units. The acid deposition will have the largest effect on soil pH where the cation exchange capacity is low, i.e. in sandy soils with low humus contents.

In soils with high base saturation and high pH a considerable leaching of exchangeable bases occurs due to natural acidification processes, mainly production of carbon dioxide. Hence, the relative importance of acid deposition is rather small for soils with high pH. For the same reason soils with a high pH and a low buffer capacity are rarely found under climatic conditions promoting natural soil acidification. Due to the natural soil acidification such soils are likely to become acid rather fast.

Since the leaching of bases due to carbon dioxide declines with declining pH, the relative importance of acid deposition increases with declining pH, and it becomes of particular importance in soils with a pH of about 5, where the leaching of bases due to carbon dioxide is rather small. Hence, the acid deposition is likely to have a comparatively large effect on the pH of slightly acid soil, especially if the soils have a low buffer capacity due to sandy texture and low humus contents. Similar conclusions were drawn by Bache², McFee¹¹, Petersen¹² and Wiklander²⁴.

Strongly acid soils are not subject to any appreciable acidification since the acid deposition causes only an insignificant or no net leaching of bases from such soils.

Increase in aluminum content of soil solution

Adsorbed aluminum ions which are abundant in acid soils originate from minerals in the soil. Since most minerals present in the soil contain aluminum, a scarcity of aluminum is not to be expected except in some organic soils and some soils developed on pure quartz sand. As soil pH drops below a certain limit, aluminum ions are liberated from the minerals and become adsorbed by the negatively charged clay and humus particles.

Due to the low solubility product of aluminum hydroxide, the aluminum content in the soil solution will be below 1 ppm as long as the pH is above 5. However, if the pH declines below this value the maximum aluminum

concentration increases strongly¹². Normally, measurements of the aluminum concentration in the soil solution show values well below the maximum values as calculated from the solubility product of aluminum hydroxide. This shows that the soil solution is not saturated with respect to aluminum hydroxide.

The aluminum content of the soil solution depends on the aluminum saturation percentage of the cation exchange complex and the total salt concentration of the soil solution. The latter is to a large extent influenced by the amounts of anions originating from pollutants. The aluminum saturation percentage of the cation exchange complex is small at pH values above 5 but increases strongly as the pH declines below 5. The aluminum content of the soil solution, therefore, also increases strongly with declining soil pH. Hence, the more acid the soil is, the more severe are the problems caused by aluminum ions in the soil solution. The actual aluminum content in the soil solution is not likely to depend to any large extent on the absolute amount of adsorbed aluminum in the soil, since the aluminum ions removed from the adsorption complex will be rapidly replaced by mineral-bound aluminum. The only exception to this are the soils mentioned above which are low in minerals containing aluminum.

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Anthropogenic and natural acidification in terrestrial ecosystems

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Key words. Soil acidification; acid deposition; terrestrial ecosystems; H⁺ budget; silicate weathering.

Acids and bases in terrestrial ecosystems

Before discussing acidification, the nature of acids and bases has to be defined. The most appropriate approach is that of Brønsted. According to Brønsted, an acid is a

substance that can donate a proton, and a base is a substance that can accept a proton. In ecosystems, the acids and bases can exist as dissolved components in solution or as solid components, and they can be separated in different compartments (e.g. in soil or in plant).