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Acidic Precipitation: Considerations for an Air-Quality Standard

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ACIDIC PRECIPITATION: CONSIDERATIONS OF AN AIR QUALITY STANDARD

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Acidic precipitation, wet or frozen deposition with a hydrogen ion concentration greater than 2.5 μeq ℓ^{-1} is a significant air pollution problem in the United States. The chief anions accounting for the hydrogen ions in rainfall are nitrate and sulfate. Agricultural systems are more likely to derive net nutritional benefits from increasing inputs of acidic rain than are forest systems when soils alone are considered. Agricultural soils may benefit because of the high N and S requirements of agricultural plants. Detrimental effects to forest soils may result if atmospheric H^+ inputs significantly add to or exceed H^+ production by soils. Acidification of fresh waters of southern Scandinavia, southwestern Scotland, southeastern Canada, and northeastern United States is caused by acid deposition. Areas of these regions in which this acidification occurs have in common, highly acidic precipitation with volume weighted mean annual H^+ concentrations of 25 μeq ℓ^{-1} or higher and slow weathering granitic or precambrian bedrock with thin soils deficient in minerals which would provide buffer capacity. Biological effects of acidification of fresh waters are detectable below pH 6.0. As lake and stream pH levels decrease below pH 6.0, many species of plants, invertebrates, and vertebrates are progressively eliminated. Generally, fisheries are impacted below pH 5.0 and are completely destroyed below pH 4.8. At the present time there are few studies that document effects of acidic precipitation on terrestrial vegetation to establish an air quality standard. It must be demonstrated that current levels of precipitation acidity alone significantly injure terrestrial vegetation. In terms of documented damages, current research indicates that establishing a standard for precipitation for the volume weighted annual H^+ concentration at 25 μeq ℓ^{-1} may protect the most sensitive areas from permanent lake acidification. Such a standard would probably protect other systems as well.

Introduction

Should a regulatory standard be set regarding the acidity of precipitation? The present level of understanding of all the physical and chemical atmospheric processes responsible for producing the acidity of "acid rain" is probably not adequate to predict how emissions of SO_X and NO_X might be altered to produce a change of two-tenths, one-half, or a full pH unit. But even if such information became available, does regulating the H^+ concentration of precipitation make sense with respect to reducing environmental impacts? Just what are the impacts attributable to precipitation acidity?

Acidic precipitation is here defined as wet or frozen deposition (i.e., rain and snow) with a hydrogen ion concentration greater than 2.5 μeq ℓ^{-1} (less than pH 5.6). Acidic precipitation has the same meaning as the commonly used term "acid rain." Much of what is known about its effects has been derived from investigations of acid deposition (acidic precipitation and dry deposition) problems in Canada and Scandinavia, as well as the United States. Significant interest has been aroused in both legislative and executive branches of the U.S. Government in determining the severity of problems relating to acidic precipitation. As a result of this interest an air quality standard may be established to limit the concentration or even the deposition of pollutants associated with acidic precipitation to prevent or ameliorate its impacts.

The first step in developing an air quality standard is the demonstration of a link between ambient pollutant levels and environmental damage.[1] Can such links be established for just the acidic precipitation component of acid deposition? This paper evaluates the current state of knowledge of the impacts of acidic precipitation on a variety of receptors and addresses the following questions:

a. What pollutants are responsible for precipitation acidity?

b. Where do the greatest impacts occur?

c. What receptors are at risk?

d. What is the current extent of damage and benefit?

e. What are the environmental costs and benefits of preventing acidic precipitation?

f. Should an air quality standard be developed now for acidity of precipitation?

Acidic precipitation has some unique problems for standard-setting. Expected effects may develop slowly and may be detectable only after several decades. Cause and effect relationships are difficult to establish. It is not clear in what media a standard should apply or what form it should take: e.g., airborne concentrations of precursors, or acidity in precipitation, or total amount of pollutant deposited per unit time. Standard-setting problems are compounded by the regional nature and multiple forms in which acid deposition occurs; dry aerosol, gas, rain, and snow. It is clear that the extent of damage in many cases is dependent upon the buffering capacity of the receptor. This results in highly regional aspects of damage per unit exposure. If acidic precipitation is a serious threat to the environment, it may not be prudent to wait for a thorough understanding of all chemical and physical mechanisms linking pollutant emissions to receptor damages before taking regulatory action. The problem, from emission of pollutants through atmospheric transport and transformation, to deposition and biological impacts may be considerably more complex than has been the case with other pollutants for which standards have been established.

This paper will be confined to the relationship between precipitation acidity and receptor effects and will not address the quantitative relationships along the entire chain from pollution emissions to environmental damage. We will first document the existence of acidic precipitation including temporal and spatial variability. Then we will present a review of receptor characteristics and responses to acidic precipitation, describing links between acidic precipitation and environmental effects, where such links are known. Information gaps will be highlighted and some suggestions made as to how to fill these gaps. Finally we will evaluate whether or not a critical threshold can be established for precipitation acidity, which can serve as a meaningful standard to protect terrestrial and/or aquatic biota.

Precipitation Characteristics of the Eastern United States

Precipitation Chemistry Patterns and Trends

Present Situation. Precipitation in the northeastern United States is acidic. Data from the National Atmospheric Deposition Program are plotted in Figure 1.[2] Results show that the median pH of precipitation in portions of Pennsylvania, Ohio, and New York is less than 4.2, while most of the northeastern portion of the United States has a median below pH 4.4.

The pH levels in Figure 1 are in good agreement with data of the MAP3S and EPRI Networks.[3] The pH of the precipitation in this region is explained by the presence of strong acids, sulfuric and nitric, with weak acids probably being relatively unimportant.[4] When the anthropogenic emissions of sulfur and nitrogen in the eastern United States were negligible, the pH of precipitation in the northeast would probably have been less acidic.

About 90% of the sulfur in the atmosphere of the northeastern United States is contributed by anthropogenic sources. These sources are predominantly power plant, industry, and area sources.[5] A similar budget estimate for nitrogen inputs to the atmosphere of the northeastern United States is unavailable. However, it has been estimated that 56% of the anthropogenic (fossil fuel combustion) NO_x emissions for the entire United States in 1972 were from sources within the northeastern United States.[6] Furthermore, the increases in nitrates in precipitation from the mid-1950s to the mid-1970s can be reasonably accounted for by increases in these anthropogenic nitrogen emissions.[7]

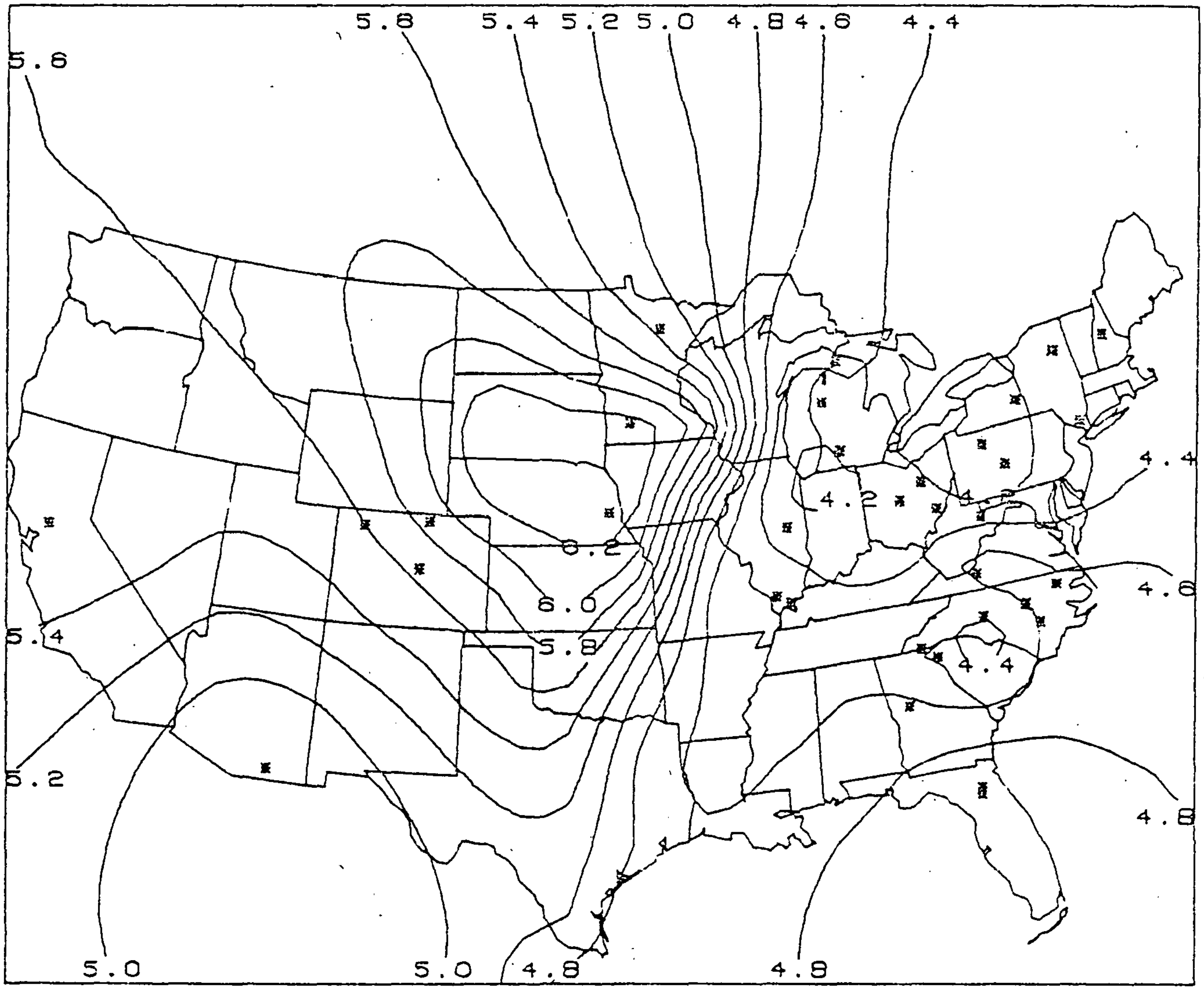


Figure 1. Isopleths of median precipitation pH for portions of 1978-1979.[2]

Acidity in precipitation events can be categorized by the percentage of events within various pH ranges. Precipitation samples collected at MAP3S stations at Ithaca, New York; University Park, Pennsylvania; and Charlottesville, Virginia were chosen for this categorization because they are three representative areas within the northeastern United States and data for three years (1976-1979) are available.[8] Most events had pH levels between pH 3.5 and 4.5. Less than 0.5% of all events were below pH 3.0 or above pH 5.5 (Table I).

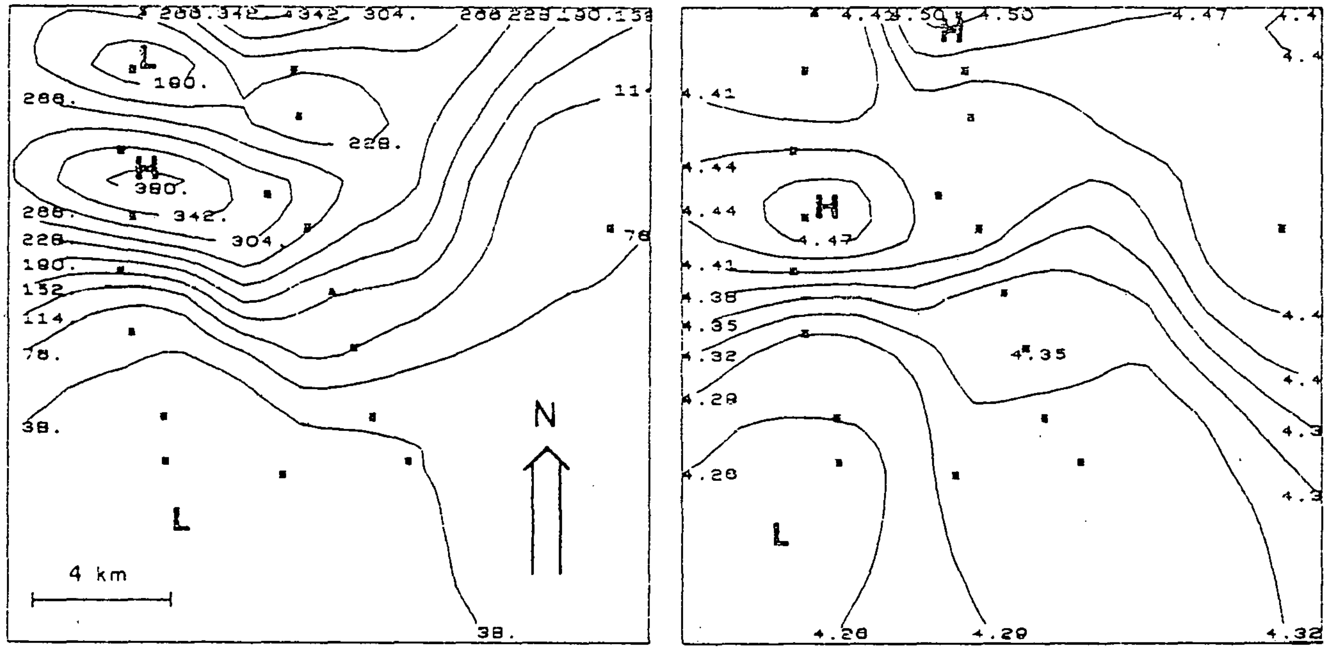
Table I. Percentage distribution of precipitation events in the northeastern United States by pH.[8] Volume weighted average pH of all samples was 4.12.

Percentage of all events	pH interval
0.9	3.00 - 3.49
31.4	3.50 - 3.99
56.4	4.00 - 4.49
10.5	4.50 - 4.99
0.6	5.00 - 5.49
0.2	5.50 - 5.99

Short-term Temporal and Spatial Variations. Individual precipitation events showed marked temporal and spatial variability in deposition volume and chemistry.[9] Generally, there is an inverse correlation between SO_4^{2-} concentration and rainfall sample volume (Figure 2). Sulfate and NO_3^- concentrations in rainfall deposited by a single storm at areas a few kilometers apart varied by a factor of 2 to 3 (Figure 2) while concentrations of Ca^{2+} and Mg^{2+} varied by a factor of 5 to 6 (data not shown). The pH levels within the event shown in Figure 2 varied from about 4.2 to 4.5 and the lowest pH levels coincided with the highest sulfate concentrations while the highest pH values coincided with the lowest sulfate concentrations.

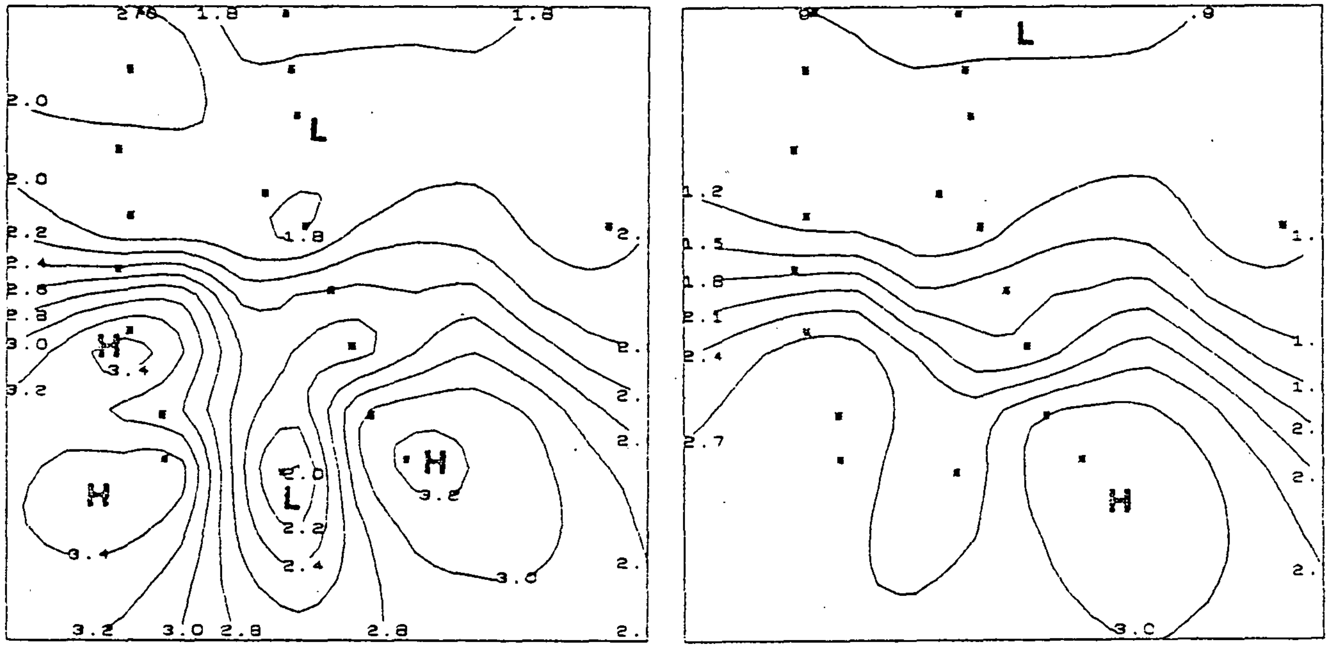
Temporal changes during the rainfall event shown in Figure 2 are shown in Figure 3. A sequential rainfall sampler was located at the north edge of the closed 1.8 contour of sulfate in Figure 2. All ions sampled exhibited the same pattern so only four ions are shown. When concentrations of each ion were normalized with values for the first rainfall sample, ion concentrations were high at the beginning, low during the middle, and high again near the end of the shower. Ion concentrations were inversely related to rainfall rates. Ion concentrations changed by more than a factor of 10 during the shower. This temporal variation in chemistry was greater than network spatial variation in chemistry (Figure 2). Furthermore, this large temporal variation occurred at a site where the spatial variation was small.

During the shower the pH rose from 4.1 to 4.9 and decreased to 4.5 at the end. The pH changed in response to the concentrations of all ions in the samples. In more acidic samples, nitrate and sulfate were the major anions present. In all samples, the concentration of protons was highly correlated with concentrations of sulfate and nitrate. These experimental results show that rainfall acidity occurs with large spatial and temporal variations. Similar temporal variations of the major inorganic ions have been reported by other investigators, with the sequential samples being collected hourly[10] or on the shorter scale of seconds to minutes.[11,12]



SCORE - GROUP 183 VALUE: SAMP. VOL.

SCORE - GROUP 183 VALUE: PH(MEAS)



SCORE - GROUP 183 VALUE: SO4(PPM)

SCORE - GROUP 183 VALUE: NO3(PPM)

Figure 2. Spatial patterns of rainfall sample volume (mℓ), measured pH, and concentrations of sulfate and nitrate for a shower of 2 July 1978 near Champaign, IL.[9] The points indicate sites of data collection. Contours are most reliable near collection sites. H = High values; L = Low values.

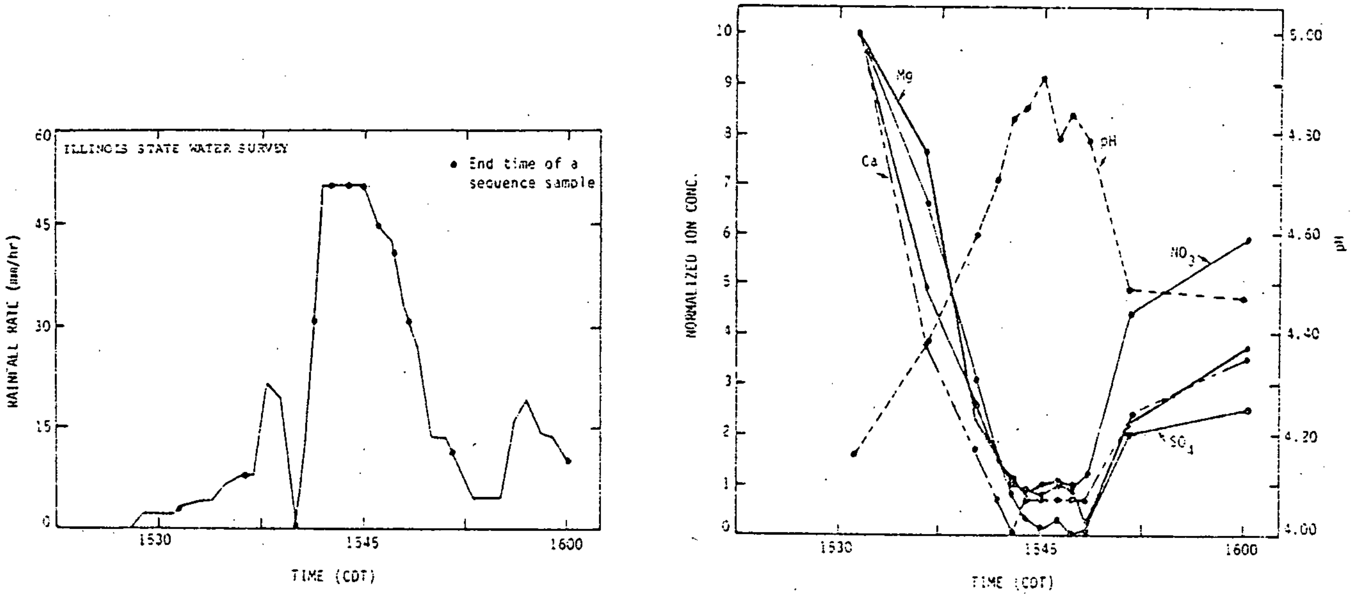


Figure 3. Rainfall rate, pH, and concentrations of calcium, magnesium, nitrate, and sulfate in rainfall samples taken sequentially during a shower on 2 July 1978 at a site near Champaign, IL.[9] Dots indicate the end time for the collection of the sequential samples.

Overall Evaluation of Precipitation Chemistry. These data show that precipitation within the northeastern United States is acidic and that there are large variations in chemistry within and among rain events and among nearby sites for individual events. However, as is implicit in Figure 1, averaging of samples over a longer period of time removes much of the spatial heterogeneity. This short-term variability in chemistry makes it impractical to establish a precipitation standard based upon data collected from a small number of samples.

Since 90% or more of the SO_x and a large portion of NO_x are of anthropogenic origin in the northeastern United States, there is little doubt that precipitation was less acidic at some earlier time. Lack of earlier data has prevented the calculation of an acceptable rate function for the acidification of precipitation.

Receptor Characteristics and Responses

Soils

Soil Chemistry. Due to their buffering capacity, soils are less responsive to short-term acidic precipitation events than exposed vegetation or streams whose flow rates and sources of inputs (i.e., surface runoff versus groundwater) may vary an order of magnitude following heavy rainfalls. Thus, effects of acidic precipitation on soil nutrition are probably best assessed on the basis of deposition rates rather than concentration in rainfalls.

There are two general areas of concern relative to the interactions of acidic rain with soils: 1) effects of acidic rain on nutrient leaching, soil acidification, and the nutrient status of terrestrial ecosystems, and 2) the impact of interactions between acidic rain and soils on the composition of waters reaching aquatic ecosystems. To some extent these concerns are mutually exclusive; i.e., if incoming H^+ displaces and leaches nutrient cations from basic soils, acidification of aquatic ecosystems is prevented. On the other hand, incoming H^+ may have little effect on the nutrient content of very acidic soils, but in sufficient amounts, H^+ could mobilize Al which produces toxic effects in aquatic ecosystems.[13]

Agricultural Systems. Within the realm of terrestrial effects, it is generally agreed that agricultural soils are less susceptible to adverse nutritional effects than forest soils.[14-17] Generally, agricultural soils have a better nutrient status, are better buffered, and most significantly, the effects of fertilization and liming far exceed the effects of acidic rain or acid deposition on such soils.[16]

On the other hand, there is mounting evidence that the deposition of sulfur from the atmosphere is beneficial to many agricultural soils.[15,17-19] Coleman[20] noted an increase in frequency of S-deficient agricultural soils since the advent of both low-sulfur home heating fuels (fuel oil and natural gas as opposed to coal) and S-free fertilizers. Tabatabai and Laflen[19] stated that 13-17 kg ha^{-1} S added by precipitation in Iowa is important to crop production because Iowa soils do not contain sufficient plant-available S to meet crop requirements under greenhouse conditions. Since no S-deficiency symptoms have been reported for crops under field conditions, it is suggested that atmospheric deposition fulfills crop S needs. In the final integrated assessment of acidic rain impacts, consideration must be given to S inputs (both via acidic rain and SO_2 absorption) to crop production and the potential costs of S fertilization if pollution abatement technologies are implemented.

Amounts of S required by plants vary greatly. Terman[15] reported annual S removals in crops ranging from 13 (rice) to 96 (sugarcane) kg S ha^{-1}, with an estimate of 18.5 kg S ha^{-1} for 10 principal crops in the U.S. as compared to S inputs ranging from a low of 3.7 kg ha^{-1} yr^{-1} in parts of Alabama and Arkansas to highs of 140 to 160 kg ha^{-1} yr^{-1} in industrial areas of Indiana and Wisconsin. Overlaying sulfate deposition isopleths onto maps of croplands may be a useful way of projecting where S deposition is beneficial or excessive from the perspective of crop nutrition.

Nitrogen deposition by acidic precipitation, while potentially beneficial to crops, is usually of insignificant magnitude to contribute to crop N status since crops require about 100 to 300 kg N ha^{-1} yr^{-1}.

Forest Systems. Acidic rain can have both beneficial and adverse effects on forest nutrient status. While acid deposition can result in accelerated nutrient cation leaching from forest ecosystems,[21] the deposition of associated sulfate and nitrate ions can benefit ecosystems deficient in S or N. A true evaluation of the effects of acidic rain on forest nutrition must include an objective analysis of the costs of accelerated nutrient leaching vs the benefits of accelerated nutrient input. Such an undertaking is complex and requires knowledge of forest nutrition, nutrient cycling, and nutrient leaching mechanisms.

Potential Benefits of Elevated S and N Deposition to Forest Nutrition. Since forests recycle nutrients their annual requirements are generally much lower than those for agricultural crops. Rates of N deposition insignificant to crops may be quite significant to forests. However, rates of S deposition sufficient to meet crop requirements are likely to exceed forest requirements substantially.

Rates of S and N deposition relative to forest requirements can be derived from nutrient cycling data. Sulfur growth requirements have been estimated from N cycling data based upon an S:N ratio of 0.03 (g - atom basis) determined by Kelly and Lambert.[22] In this assessment, net annual increments of N and S in foliage and fine roots are assumed to be zero (inputs = outputs, steady-state conditions) and only net nutrient accumulations in woody tissues are considered as a net annual loss from soils. In every case considered here except the West German (Solling) site and the Pinus echinata stand in Tennessee (Walker Branch), atmospheric N inputs (excluding gaseous inputs) are less than N increments (Table II). Atmospheric S inputs, on the other hand, exceed calculated S requirements for wood tissue increments by a large margin at all sites considered. Previous studies on S cycling in forests have shown that excess S (i.e. above the 0.03 S:N ratio) accumulates and cycles as SO_4^{2-}.[22-24] In fact, foliage SO_4^{2-} is a sensitive indicator of tree S status; low levels (<80 ppm SO_4-S) in needles of Pinus radiata indicate S deficiency whereas high levels (>400 ppm SO_4-S) indicate S excess coupled with probable N deficiency.[25,26] These results indicate that some excess S is necessary for adequate tree growth so that the calculated increment requirements in Table 2 are probably underestimates. Nonetheless, moderate atmospheric S inputs can fulfill forest S requirements; even very modest S inputs to the Douglas fir ecosystem in Washington resulted in large excesses in foliar SO_4^{2-}.[26]

Sulfur deficiencies can occur in forest soils remote from pollution inputs (e.g., pumice and basaltic soils of Oregon and Washington).[27,28] Several Douglas fir stands not responding to N fertilizer in the northwestern U.S. had deficiency levels of foliage SO_4^{2-}.[26] Also, widespread nitrogen fertilization in the northwestern and southwestern U.S.[35] places greater demands upon forest ecosystem S reserves.

Nutritional consequences of excess SO_4^{2-} inputs are unclear. Perhaps uptake of excess SO_4^{2-} aids in charge balance problems in ammonium-utilizing forest ecosystems where uptake of cationic nutrients exceeds uptake of anionic nutrients (necessitating either HCO_3^- uptake or H^+ release from roots). Thus, while atmospheric inputs of S sufficient for average crop requirements (13.5 kg S ha^{-1})[15] exceed forest requirements, this is not likely to cause nutritional harm to forest ecosystems. Inputs of N in polluted rain, while they do not fulfill forest N requirements, do appear to make a significant positive contribution.

Table II. Increments of N and calculated requirements of S in branch and bole components of several forest ecosystems compared with atmospheric N and S inputs.

Location	Species	Age (yr)	Nutrient Increments		Inputs[a]	
			N[b]	S[c]	N	S
			kg·ha^{-1}·yr^{-1}			
Washington	*Pseudotsuga menziesii*	42	10	0.7	1.7	4.1[d]
Washington	*Alnus rubra*	30	18.0	1.2	1.7	4.1[d]
Tennessee	Mixed deciduous	30-80	23.1	1.6	8.7	18.1[e]
Tennessee	*Pinus echinata*	30	8.2	0.6	8.7	18.1[e]
North Carolina	Mixed deciduous	60-200	9.1	0.6	4.9	12.2[f]
North Carolina	*Pinus strobus*	15	23.5	1.6	5.5	11.7[f]
New Hampshire	Northern Hardwood	110	18.4	1.3	5.8	12.7[g]
W. Germany	*Fagus sylvatica*	59	16.7	1.2	21.8	24.1[h]
W. Germany	*Picea abies*	34	8.1	0.6	21.8	24.1[h]

[a] Precipitation only
[b] From 29
[c] Calculated from S:N ratio 19
[d] From 30
[e] From 31
[f] From 32
[g] From 33
[h] From 34

Potential Detriments of Elevated Acid Inputs to Forest Nutrition.

Effects of acidic rain on soil leaching and acidification must be assessed within the context of natural H^+ production in the ecosystem.

Hydrogen ion budgets in forest ecosystems based upon measured mass balances of cations and anions have been calculated.[36] This model has been used for forest ecosystems in Sweden, West Germany, and Oregon.[37] Even in the most heavily impacted Solling site in West Germany, analyses show that atmospheric hydrogen ion inputs are small (~10%) compared to internal production.

A more conservative approach involves input-output balances of H^+ and metal cations to predict the long-term net acidification of soils.[38] This approach considers only a net result of various H^+ production and consumption processes involved in the Solling model (i.e., net cation removal from the site). Studies of the intensity required for this kind of analysis are rare (particularly in the area of carbonic/organic acid leaching), but some data are presented in Table III.

Cation removal by internal H^+ production and leaching (by carbonic and organic acids) exceeded atmospheric H^+ input by a factor of 2 in the acid rain-impacted Thompson, Solling, and Jadraas sites. Cation removal by either bole or whole-tree harvesting reduces the significance of atmospheric H^+ inputs even further, but atmospheric inputs could be considered negligible only at the unimpacted Findley Lake and H.J. Andrews sites (Table III).

Unfortunately, internal H^+ production varies considerably among various forest ecosystems, and generalizations about internal vs atmospheric H^+ inputs are hazardous. Furthermore, much of the above discussion is predicated on the assumption that H^+ inputs to soil via either acidic rain or natural mechanisms will result in an equivalent amount of base cation removal. Since anions must balance cations in any solution, this further implies that added anions and H^+ are mobile in soils. This assumption, while useful in conceptually exploring various scenarios, has been shown to be invalid in many instances, particularly with respect to sulfate.[39-42]

Sulfate is known to adsorb to iron and aluminum oxides in soils where it can create exchange sites capable of retaining incoming H^+ or other cations.[39-44] Thus, sulfate-adsorbing soils can be acidified by H_2SO_4 without an equivalent amount of cation leaching. Conversely, non-sulfate adsorbing soils can be leached by H_2SO_4 without an equivalent amount of acidification due to H^+-induced acceleration in the rate of cation weathering from soil minerals.

<u>Soil Interactions on Acid Transport to Aquatic Systems</u>. Although acid deposition does not seem to be rapidly affecting soils in our normal concept of time where 100 years is a "long time," it should be remembered that chemical and physical changes in soils are usually measured on a geologic time scale. On this time scale, changes which occur over a period of decades or even centuries may be considered to be rapid. Nyborg et al.[48] estimated the rate of soil acidification near a gas plant as a result of up to 50 kg ha^{-1} yr^{-1} SO_2 deposition to be approximately a one pH unit decrease in ten to twenty years. Results of a computer soil simulation model show that the loss of bases could be significant on a time scale of decades and that the pH of the soil solution would show a downward trend of 0.1 units per decade.[49] In geologic time, this would be considered a fast reaction rate, but this rate would not appear to be able to account for the observed drops of 1 pH unit in lakes and streams of Norway, the northeastern U.S., and Canada over the last two decades.

Rosenqvist[50,51] has argued that natural acid production could significantly affect the acidity of surface waters in Norway. His arguments have sparked further thinking and research as to possible mechanisms of acid transport through soils to aquatic ecosystems. One idea currently being explored is the application of the anion mobility concept to acid soils.[52-54] According to this model, inputs of mobile anions to acid soils result in a reduction in solution pH which causes an increase in all cations including H^+, to retain a charge balance in solution. Experimental acidification of both

Table III. Atmospheric H^+ inputs vs cation removal by internal H^+ production (carbonic and organic acids) and potential net annual cation removal in bole only and whole-tree harvesting in selected forest ecosystems.

Site	Species	Age (yr)	Precipitation H^+ input[a]	Cation leaching by internal acid production[b]	Cation removal by harvesting[c]	
					Bole	WTH
			$(eq \cdot ha^{-1} \cdot yr^{-1})$			
Thompson, Washington	*Pseudotsuga menziesii*	42	240[d] (4.8)	420[d] (5.9)	380[e]	660[e]
Solling, W. Germany	*Fagus sylvatica*	59	900[g]	1950[g]	220[e]	370[e]
Jadraas, Sweden	*Pinus sylvestris*	?	190[g]	226[g]	?	?
Findley, Washington	*Abies amabilis*, *Tsuga mertensiana*	175	90[h] (5.6)	1410[h] (4.5)	272[e]	460[e]
H.J. Andrews, Oregon	*Pseudotsuga menziesii*	450	28[g]	22700[g]	60[e]	106[e]

[a] Weighted average $[H^+]$ times precipitation amount; weighted average $[H^+]$ as pH appears in parenthesis where available.

[b] Calculated from net increase in weighted average HCO_3^- or organic anion concentration (the latter estimated by anion deficit) times water amount. Weighted average $[H^+]$ as pH for solutions appears in parentheses where available.

[c] Nutrient content divided by age; WTH = whole tree harvest, removal of all aboveground biomass.

[d] From 30.

[e] From 29.

[f] From 45.

[g] From 37. For comparison in this table, only H_2CO_3 production values are included.

[h] From 46,47.

monolith lysimeters and mini-catchments have verified the relationship between SO_4^{2-}, H^+, and Al^{3+} in effluent or runoff waters.[55,56] However, there remain questions as to whether the magnitude of this effect could also account for observed pH declines in Norwegian streams and lakes.[54] Clearly, further research in this area is needed.

Overall Evaluation of Effects on Soils. Current information, especially with regard to internal acid production in forest soils, is totally inadequate to set air quality standards for acid precipitation. From a forest nutrition perspective, one might arbitrarily consider annual H^+ deposition rates equal to internal acid production as an upper limit. Taking the lowest values from Table III, we have an input of approximately 200 eq ha^{-1} yr^{-1} (Jadraas site) as a minimum standard. It is evident, however, that this standard would have little meaning at the H. J. Andrews site. As sulfuric acid, this standard equals a minimum of 3.2 kg S ha^{-1} yr^{-1}, a figure likely to be adequate to fulfill most forest S requirements (Table II). As nitric acid, this equals a minimum of 2.8 kg N ha^{-1} yr^{-1}, an amount which could contribute appreciably to the N fertility of some forest ecosystems (Table II).

From the agricultural crop perspective, S inputs of 3.2 kg ha^{-1} yr^{-1} are probably too low to fulfill crop requirements and may necessitate S fertilization in some areas. Costs of fertilization must be factored into the final decision on an acidic rain standard.

Finally, from the perspective of acid transport from terrestrial to aquatic ecosystems, several considerations must be taken into account including the percent of land area covered with soil vs barren rock, the amount and accumulation of snow, the amount of surface runoff vs soil percolation, and the initial acidity of soils. Ideally, different standards could be set for different regions of the country, better yet, within states. Since acidic rain is a regional phenomenon, however, this receptor-based ideal is inconsistent with regional deposition patterns.

Soil Microbiology

Soil microorganisms play an important role in nature and are critical to ecosystem function and the well-being of plants, animals, and humans. They are responsible for transformations of various elements and occupy a significant place in carbon, nitrogen, phosphorous and sulfur cycles.

If there is a significant impact of acidic precipitation on soil microbial processes, then a potential would exist for reduced soil fertility and economic loss primarily in unmanaged range and forest soils. Thus, the impacts of acidic precipitation on soil microbes is an appropriate consideration for establishing an air quality standard.

Microbial growth, activity, and numbers are reduced by soil acidification.[57] Normally functioning plants, soils, and soil biological communities will be affected when a significant change in soil pH is observed due to acidic precipitation. Different soil types will respond to acidic rain differently and one may or may not see any change in soil pH.

Organic Matter Decomposition. A diverse group of microorganisms participate in the decomposition of natural organic materials in soil. Reduction in microbial growth, activity, and numbers may drastically affect the organic matter turnover, soil structure, and nutrient availability for plant growth in the ecosystem. Although an increase in soil acidity due to acidic precipitation may enable certain groups of organisms to proliferate, most microorganisms are sensitive to acidity. Depending on the nature of organic materials (type of vegetation), soil type, pH, temperature, moisture, etc., one should expect differences in the rate and extent of organic matter decomposition.

The effects of acidification on forest litter decomposition vary with type of materials studied. Exposure to rainfalls of pH 3.0 increased the rate of decomposition of lodgepole pine needles compared with needles exposed to rainfalls of pH 5.6.[58] Applications of acid to pine needles in litter bags in field plots also stimulated decomposition.[59] In contrast, no changes in decomposition rates were detected when spruce needles or aspen sticks were tested.[58]

Acidic rain application significantly reduced organic matter decomposition in forest soils where increases in soil acidity have been observed.[57,58,60] With a slight decrease in soil pH after applications of rainfall of pH 2.5, rates of cellulose decomposition were decreased. The rate of decomposition of humus decreased significantly with increased acidification; changes in pH of humus samples were observed.

Effects of acidity on microbial decomposition of oak leaves in naturally acid and acidified soils have been studied.[57] There was a 52% decrease in total CO_2 production in the pH-adjusted acid soil relative to the natural control soil (pH 4.6). Highly significant differences ($p < 0.01$) in the rates of CO_2 production were observed among soils amended with organic matter. A 37% reduction of total CO_2 evolution was observed in the acidified (pH 3.5) soil. Among the soils tested there was a significant ($p < 0.01$) correlation between the amount of CO_2 produced and the exchangeable hydrogen ion content of the soil.[57]

Nitrogen Transformation. Nitrogen is the major nutrient limiting plant growth in nature. Higher plants are known to assimilate nitrogen in the form of nitrate and ammonia. Ammonification, nitrification, nitrogen fixation, and denitrification are affected to varying degrees by soil acidity.

Ammonification. Organic nitrogenous compounds in soils, sediments, plants, and animals are converted to ammonium (ammonification) by a large group of heterotrophic bacteria, fungi, and actinomycetes. Ammonification is inhibited by acidification of some soils. Ammonification in a pH-adjusted acidic soil (pH 3.5) was about half that of a naturally acidic soil (pH 4.6). Highest rates of ammonification were observed in the pH-adjusted neutral soil.[57] However, ammonification is much less sensitive to acidification than is nitrification.[61]

Nitrification. Nitrification in acid soils is inhibited and nitrifying organisms are eliminated at high acidities. Nitrification is the sequential oxidation of ammonia to nitrite and then to nitrate by autotrophic and heterotrophic microbial communities. Autotrophic nitrification rates have been calculated to be ten times greater than heterotrophic nitrification rates. Autotrophic nitrification is principally accomplished by *Nitrosomonas* sp. and *Nitrobacter* sp. These organisms derive energy for growth from the oxidation of inorganic nitrogen compounds and carbon for cell synthesis from CO_2. Nitrification occurs optimally at neutral to slightly alkaline pH levels. In acid environments, nitrification proceeds slowly, even in the presence of an adequate supply of substrate, and responsible species are rare or totally absent at high acidities. Typically, nitrification decreases markedly below pH 6.0 and becomes negligible below pH 5.0,[61] yet nitrate may occasionally be present in field soils of pH 4.0 and below.[62] Some soils nitrify at pH 4.5 while others do not. The difference is possibly attributed to acid-adapted strains or to chemical differences in the two habitats. Neutral to alkaline soils have large nitrifier populations.[62] Accumulation of nitrate has been observed in acidic soils as low as pH 3.9.[63] However, Walter and Wickramasinghe[64] isolated pure cultures of ammonium-oxidizing autotrophic, nitrifying bacteria from acid soils at pH 4.0 to 4.5. Nitrite-oxidizing bacteria were detected in several acid soils but pure cultures were not isolated.

In a beech forest soil (≈ pH 3) very small numbers of nitrite- and nitrate-forming organisms were found but substantial amounts of nitrate were detected.[65,66] Formation of nitrate by heterotrophic nitrification in acid soils has been suggested.[58,67] Little autotrophic and heterotrophic nitrification in acidified soil (pH 3.5) was observed in a soil perfusion study.[57] Application of acidic rain (pH 4) reduced nitrification rates in a soil with pH 4.4.[68]

Decreased nitrification in acid and acidified soils would reduce nitrogen loss due to leaching and denitrification and would increase nitrogen availability in the form of ammonium to higher plants, however.

Denitrification. Soil bacteria are known to reduce nitrates to nitrogen gas under anoxic conditions in the presence of available carbon. This process is called denitrification. Of particular interest is the biogenic emission of N_2O and its subsequent effect in the depletion of atmospheric ozone or its contribution to the formation of nitrate in the atmosphere. Soil pH is known to affect the rate and the composition of the gaseous end products of denitrification. Recently, Francis et al.[57] reported that denitrification was rapid at soil pH 6.5 with little N_2O detection, indicating complete denitrification whereas in acid (pH 4.6) and acidified (pH 3.5) soils, N_2O was the major end product. Nevertheless, the significance of denitrification should be critically evaluated in light of reduction in the rates of nitrifying activity and the possibility of reduced nitrogen loss in acid soils and the amounts of N_2O released into the atmosphere.

Nitrogen Fixation. Nitrogen fixation by free-living organisms in acid forest soils may be insignificant. Nitrogen-fixing microorganisms differ in tolerance to acidity. Environments more acidic than pH 6.0 contain little or no nitrogen fixing bacteria. Nitrogen fixation by free-living bacteria in freshly collected soils (pH 4.6) was not detectable. Only soil samples of pH 5.7 amended with glucose and preincubated under aerobic or anaerobic conditions exhibited slight activity.[57] Blue-green algae, which also fix nitrogen, grow poorly and are found to be few in number in acid environments.

Soil acidification affects symbiotic nitrogen fixation in legumes. Although several physical and chemical factors contribute to efficient nitrogen fixation in legume-Rhizobium symbiosis, soil acidity affects (a) plant growth, (b) survival of rhizobia, and (c) the symbiotic relationship. In garden peas, for example, successful infection, formation of nodules, and thus efficient fixation of nitrogen are halted below pH 4.8.[69] Toxicity resulting from iron or aluminum in acidified soils also has a profound effect upon nitrogen fixation.

Overall Evaluation of Effect on Soil Microbiology. Further acidification of acid forest soils by acidic rain is perhaps a very slow process. Many years may be required for acidic rain to change the soil pH. Rapid adaptability of microbial populations to changing physical and chemical environments and substantial differences in the measured soil pH versus the actual pH in the microsite environments make it difficult to accurately monitor short-term changes which might be caused by acidic precipitation. Slow acidification may affect soil microbial communities which may gradually result in the selection of acid-resistant or tolerant organisms or elimination of certain species altogether. On a long-term basis, acidic rain may affect certain key processes catalyzed by soil microorganisms such as organic matter decomposition, and nitrogen transformation and ultimately the nutrient cycling in the forest ecosystem.

To date, studies with simulated acidic rain indicate overall reductions in several soil microbial processes. In some cases stimulatory effects on microbial activity have been observed. However, these have been assumed to be of a temporary nature. The physical and chemical characteristics of the soil and its response to environmental pollutants often determine the type, abundance, and activities of soil microorganisms. No generalizations can be made because of the diversity and complex nature of these systems. Therefore, based on the existing data, it is not clear to what extent and rate the current acidic precipitation is affecting soil microbial processes. Future studies should focus on obtaining quantitative information on the various microbial processes critical to ecosystem function.

Aquatic Ecosystems

Establishing a standard for precipitation acidity to protect aquatic ecosystems carries two principal sets of assumptions. First, it assumes that there is a cause-effect relationship between precipitation acidity, or other substances for which acidity serves as a surrogate, and freshwater acidification, and that acidification of fresh waters can be prevented or ameliorated by not allowing the acidity of precipitation to exceed a definable concentration. Second, it assumes there are ecosystem components damaged by acidification and that these damages are severe enough to warrant establishment of a standard to prevent or ameliorate them.

Precipitation and Freshwater Acidification. Evidence from a variety of investigations indicates that the deposition of anthropogenic substances, particularly oxides of sulfur and nitrogen, results in the acidification of streams, rivers, and lakes in widely separated areas of the world. Synoptic surveys (one or a few samples collected in a short period of time from each of many lakes) in Norway,[70,71] Sweden,[72-74] Scotland,[75] the northeastern United States,[76,77] and southeastern Canada[78] all indicate the wide-spread acidification of low conductivity, oligotrophic lakes in regions receiving acidic precipitation. More extensive studies, involving many observations in one lake or in each of many lakes in these regions confirm the observations of the synoptic studies.

The extent to which dry deposition of SO_x and NO_x contributes to the relationship between acid deposition and acidification of fresh waters is not yet known. This is largely because actual measurements of dry deposition (as distinguished from bulk minus wet-only collector concentrations), especially gaseous forms, are very difficult and not widely carried out. Estimates of deposition velocities vary greatly and there is no general agreement on the relationships between wet and dry components. Watershed mass balance studies at Hubbard Brook (NH) have shown 32% of sulfur inputs to the system are by dry deposition with SO_2-S gas impaction estimated to be twice as large as aerosol-S deposition.[79] In the absence of a complete understanding of relationships among sulfur species in the atmosphere and their deposition rates, using the concentration of H^+ in precipitation as a surrogate for SO_x and NO_3^-, deposition may be considered.

Relationships between measured precipitation acidity and regional freshwater acidification have been extensively studied as noted above. Stream and lake acidification in these areas has occurred in a short period of time, the past few decades. The rapidity of this acidification is demonstrated both by actual measurements of temporal changes in water chemistry and by the disappearance of fish. Fresh waters which formerly supported natural fish populations have become so acidic and aluminum contaminated, a consequence of watershed acidification,[13] that fish cannot survive in them.[70,77,80-83]

This rapid change in lake chemistry is associated with a general increase in sulfate and/or nitrate concentration of precipitation in the impacted areas.[70,73,84,85] Regional patterns of lake-water chemistry reflect regional patterns in precipitation chemistry.[70,75,86-88] Furthermore, many streams and lakes in the areas listed above also have rapid pH fluctuations due to acid deposition either directly in response to episodes of low pH rain or indirectly, following snow melting and the release of atmospheric pollutants stored in the snow pack.[70,82,89-92]

In some cases, acidification may be caused by direct deposition of acids onto the surface of lakes. For example, Honnedaga Lake in the Adirondack Mountains, which has been recently acidified, has a lake surface-to-watershed ratio of 1:4. Due to neutralization of atmospheric acids by the watershed, reducing acid inputs to this lake from run-off waters, 72% of the annual H^+ input to Honnedaga comes directly by precipitation to the lake surface. Similarly, Mirror Lake, New Hampshire, receives 80% by direct precipitation. In the Fyresdal/Nissedal region of southern Norway where chemical buffering in watersheds is least effective, many lakes are acidic due to acidic precipitation and about 24% of the acids are recieved by direct precipitation to the lake surface for an "average" lake.[93] More typically, however, a much higher proportion of acid inputs is derived from watershed runoff. Thus, complex interactions occur among the substances entering by atmospheric deposition and vegetation, soils and geology within the watershed, modifying the chemical composition of waters entering streams and lakes. Since the role each system component plays in modifying acid inputs differs from one watershed to another, it will be difficult to establish a standard for precipitation acidity which will protect aquatic ecosystems from acidification based on detailed models of these interactions.

An alternative to a complex modeling approach is to treat the watershed as a black box, ignoring essentially all of the complex biogeochemistry within the black box, and examining the relationship between acid deposition and the chemistry of surface waters in affected areas. This is the approach taken by Henriksen[88] in deriving empirical models of lake acidification for geologically similar areas, having granite or other siliceous bedrock, thin podsolic soils, poorly buffered surface waters, and which receive acidic precipitation (pH 4.0 to 4.6).

First, Henriksen notes that lakes in which pH has declined significantly during the past 20 to 25 years are located in areas receiving precipitation more acidic than pH 4.6 (annual volume-weighted average H^+ concentration).

Second, Henriksen defines acidification as a loss of alkalinity (or acid-neutralizing capacity). Analysis of calcium and alkalinity concentrations in northern Norway and the Experimental Lakes Area (ELA) of Canada (regions of similar, slow-weathering bedrock with annual precipitation acidity above pH 4.6 and having slight evidence of lake acidification), show these two variables to be highly correlated. In acidified lakes, an empirically derived equation can be used to calculate pre-acidification alkalinity from the current Ca concentration. (This assumes a constant Ca/Mg ratio and that Ca concentration is not altered by acidification. Henriksen discusses reservations about these assumptions.)

Third, Henriksen has shown that for lakes in southern Norway, a region highly impacted by acid deposition, acidification is closely related to the volume-weighted average H^+ and SO_4^{2-} concentrations in precipitation. Henriksen's analysis indicates significant acidification in areas (with similar geologic features) receiving volume-weighted average H^+ concentrations near the range 20-25 $\mu eq\ \ell^{-1}$ (pH 4.7 - 4.6) and SO_4^{2-} above 20 $\mu eq\ \ell^{-1}$.

When Henriksen's model is applied to lakes in southern Norway, southwestern Sweden, southwestern Scotland, the Adirondack Mountains of New York, and southeast Ontario, it has successfully predicted observed pH levels in these widely separated areas, even when extrapolated to the very high loadings of acid observed in some lakes near Sudbury, Ontario.[94] Henriksen's model is still being developed and another form of it is likely to show a strong positive correlation of lake pH with both calcium concentration in lake water and sulfate in precipitation.

Acid deposition is widely but not unanimously accepted in the scientific community as causing the phenomenon of lake acidification. Alternative hypotheses have been proposed. Rosenqvist[95] incorporated some valid geochemical arguments into a hypothesis which suggested that changing demographic and land-use patterns in Norway could be responsible for the wide-spread acidification observed there. His hypothesis was examined by several investigators.[93,96-99] No evidence was found which supported the main assertion that changing demographic patterns and land use were responsible for the regional acidification of lakes in Norway.

It has also been suggested that aggrading forests may reduce acid-neutralizing capacity of soils as a result of the net uptake of base elements.[100] Presumably, there is not an equivalent return of these materials to the soil by decomposition of litter and dead-fall, as might be expected in a "mature" forest. A study of 72 lakes (pH 4.3 to 7.6) was conducted in a region of Scotland where reforestation is occurring and precipitation is acidic (pH about 4.3). Three categories of forest cover were defined for catchments of these lakes: reforested, partially reforested, or not reforested. Lake pH was found to be unrelated to forest cover. Furthermore, lake acidification estimated from excess SO_4^{2-} levels was not significantly correlated with percent forest cover.[101]

In summary, there is abundant evidence that lakes and streams have become acidified in widely separated geographic areas which have in common both precipitation pH of less than 4.6 and geologic formations consisting of slow-weathering bedrock and patchy or thin soils, deficient in chemical-buffering materials. Alternative hypotheses so far advanced have explained neither the geographic nor temporal characteristics of lake and stream acidification. While the interactions of materials deposited from the atmosphere with watershed biota, soils, and bedrock are complex and, for the most part, poorly understood, a high degree of understanding of these interactions appears to be unnecessary to a linking of acidic precipitation and freshwater acidification in the most sensitive areas. The simple empirical-modeling approach used by Henriksen[88] is currently rather successful in relating sulfate concentration and precipitation acidity to lake acidification in the most sensitive areas. Such models are being developed further. An annual volume-weighted hydrogen ion concentration of 25 μeq ℓ^{-1} (pH 4.6) in precipitation appears to be a critical threshold. At greater concentrations, freshwater acidification will probably occur in the most susceptible regions. There currently is insufficient information to determine if significant freshwater acidification is occurring in areas of somewhat lower geologic sensitivity, or if less acidic rain will cause acidification in sensitive areas given a longer period (decades) of exposure.

Biological Changes Caused by Freshwater Acidification. Standards are established because some form of injury is known or anticipated if the standard is exceeded. In aquatic environments, impacts on many forms of organisms have been demonstrated. The decline of fisheries in lakes and streams of Scandinavia and North America caused by the acidification of watersheds and water bodies remains as the most obvious and recognized environmental impact of acid deposition. Concern has centered around the complete elimination of fish from impacted waters and this complete elimination seems to be taken as the de facto definition of injury. However, a variety of other biological changes also occur in aquatic ecosystems at much lower levels of acidification than are required to eliminate all fish. Clearly, a new definition of what constitutes injury is required, one which will encompass the alteration of aquatic communities which occurs in the pH range 5.0 to 6.0. This will require quantifying the relationships between acidification and biological changes. Most of the available information concerning effects on aquatic biota is qualitative.

Microbial Activity and Litter Decomposition. Inhibition of microbial decomposition can have profound effects throughout an aquatic ecosystem on detritus removal, conservation of energy, nutrient recycling, primary production, and detrivore production. Production at higher trophic levels may be affected by changes in microbial activity. Several investigations have indicated that microbial decomposition is greatly inhibited at pH levels commonly encountered in lakes affected by acidic precipitation.[102] Neutralizations of acidified lakes in Canada resulted in a significant increase in aerobic heterotrophic bacteria in the water column.[103] Organic litter accumulation is accelerated in acidified waters.[102,104-106]

Reduction of microdecomposer activities may have a direct affect upon invertebrates by altering their food supply. Although certain benthic invertebrates appear to feed directly on allochthonous detritus material, it seems that "conditioned" (colonized by microorganisms) material is preferred, and that the nutritional value of the detritus is highly increased by conditioning.[107] Bacteria may also be a food source to be removed by the filtering apparatus of organisms such as Calanoida. An inhibition of microbiota or a reduction in microbial decomposition processes would therefore have a direct impact on the lakes' animal communities.

Bottom-Dwelling Plants. Aquatic macrophyte communities are altered by acidification causing changes in animal habitats and possibly affecting nutrient cycling. Plant growth and productivity (O_2 production) can also be reduced.[108]

In some acidic lakes in Sweden, Norway, in the Adirondack Mountains, and in acid mine drainage waters,[103,105,109-112] unusually dense mats of Sphagnum occur. Many other acidic, clearwater lakes do not have unusual amounts of Sphagnum. In developing their hypothesis on oligotrophication, Grahn et al.[104] have stressed two biologically important consequences of this Sphagnum expansion. First, Sphagnum has an ion-exchange capacity which results in the withdrawal of base ions such as Ca^{2+} from solution, thus reducing their availability to other organisms. Secondly, dense growths of Sphagnum form a distinct biotope which is unsuitable for many members of the bottom fauna.

Dense growths of attached algae occur on the bottoms of acidified lakes and streams. Under some acid conditions, unusual accumulations of algae may occur. In Swedish lakes, Mougeotia and Batrachospermum become important components of the benthos and large areas were made up of dense felts of filamentous algae. Lime treatment caused a rapid decomposition of organic litter as well as great reductions of the algal mat, indicating that an inhibition of bacterial activities had taken place at low pH.[104,113-115] In artificial stream channels in Tovdal, Norway, and in a natural stream in the Hubbard Brood Experimental Forest, extensive growths of filamentous algae resulted upon acidification with H_2SO_4 to pH 4.0.[102,116] Several factors may contribute to these unusual accumulations of certain algae. Intolerance of various species to low pH or to consequent chemical changes will allow just a few algal species to utilize nutrients available in these predominantly oligotrophic waters.[117] Many species of invertebrates are absent at low pH, and removal of algae by grazing is probably diminished.[106,116] It is hypothesized that the increased algal growths are due, in part, to the reduction of invertebrate grazing activity.[118]

Phytoplankton. In many lakes free-floating algae, or phytoplankton, form the base of the food chain. The process of algal growth (primary productivity) is regulated by several factors including nutrient availability, light penetration, and grazing by small aquatic animals (zooplankton). All of these factors are altered by lake acidification.

Species composition of phytoplankton is altered by acidification and the number of species decreases. However, there is no consistency among various investigations as to which taxa are likely to be dominant under conditions of acidification. Pyrrophyta may be more common (e.g., species of Peridinium and Gymnodinium) than others in lakes near 4.0. With decreasing pH in the range of 6.0 to 4.0, many species of Chlorophyta are eliminated, although tolerant forms are found in the acid range.

Evidence concerning lake acidification effects on phytoplankton biomass is not consistent. Lake clarity has been observed to increase in a few lakes concomitant with increased acidity. Humic substances are readily precipitated in the presence of Al in the pH range 4.0 to 5.0 and this probably contributes to increasing lake clarity.[114] The concentration of chlorophyll, an estimator of phytoplankton biomass, is not consistently decreased by acidification.[118,119] Low phytoplankton biomass (<1 mg/ℓ) has been correlated with the concentration of available phosphorus which generally decreases with lower lake pH. Whole-lake manipulation of total phosphorus and/or H^+ strongly supports the notion that altered H^+ per se does not change phytoplankton biomass, while phosphorus loading does. Lake manipulation, however, does not provide information concerning acidification impacts on the terrestrial portion of a watershed, which is the source of most nutrients to a lake or stream.[118]

Interaction with Aluminum and Phosphorus. Watershed acidification greatly increases Al leaching[13,120] and several studies taken together support the notion that increased Al in soil pore water may reduce total phosphorus loading.[118] While less phosphorus may be removed at fixed Al concentrations below pH 5 than pH 5-6,[114] an elevated concentration of Al may result in an increase in phosphorus removal below pH 5, by whatever means, not only in the lake water but in the watershed soil as well. This topic needs further investigation.

Invertebrates. Zooplankton community composition is simplified with species number decreasing in acidified waters, but effects on biomass are not well documented.[121-123] The distributions and associations of crustacean zooplankton have been found to be strongly related to pH and to the number of fish species present in the lakes. Food supply, feeding habits, and grazing of zooplankton will probably be altered following acidification, as a consequence of decreased biomass and species composition of planktonic algae and bacteria. Studies of zooplankton have not been sufficiently intensive to assess whether acidification results in reduction of zooplankton standing stocks.

Bottom-dwelling (benthic) invertebrate communities are simplified with species number decreasing in acidified waters, and biomass may also decrease. Surveys at many sites receiving acidic precipitation in Norway, Sweden, Great Britain, and North America have shown that waters affected by acidic precipitation have fewer species of benthic invertebrates than localities which are less acidic.[106,121,122,124-126] Some species, such as the amphipod Gammarus lacustris, which is an important element in the diet of trout in Norwegian lakes where it occurs, cannot tolerate 24 to 48 hr of exposure to pH 5.0.[127,128]. This kind of data is not available for most fish-food organisms.

As with zooplankton, there have been few intensive studies of benthic invertebrate biomass which could allow an assessment of acidification impacts. A study of 6 acidic and 8 less-acidic Norwegian lakes found invertebrate density and biomass to be reduced at low pH.[129] Quantitative data concerning the effects of low pH on the benthic fauna available for some acidified Norwegian lakes showed notably low standing crops,[102] but a few species are favored by conditions in acidified waters. The Notonectidae (backswimmer), Corixidae (water boatman) and Gerridae (waterstrider) are often abundant in acidified soft waters at pH as low as 4.0. This may, in part, be due to lack of fish predation. Chironomids have been found to be the most common animal group in acidic lakes[129] and may provide the food base for carnivorous invertebrates which are abundant in the absence of fish predation.

Water hardness is an important factor in determining invertebrate responses to acidification. The importance of water hardness has been discussed most recently by Økland and Kuiper[130] who found the number of species of Sphaeriidae (small mussels) is positively correlated to Ca^{2+} concentration up to 2 mg ℓ^{-1} hardness (as CaO) as well as with pH over the range 4.6-6.9. Gastropods may be present at water hardness values as low as 1.5 mg CaO ℓ^{-1} only if pH is >6. Low pH inhibits moulting progression for the tadpole shrimp (Lepidurus arcticus) and this may be due to interference with Ca^{2+} uptake.[128] Progression of the crayfish Oronectes virilis through moulting was also inhibited at low pH and uptake of Ca^{2+} was retarded.[131] Physiological information for effects of acidification on fish-food organisms is sparse.

Effects on Fish Populations. The following points are generally accepted for fresh waters of southern Norway, southwestern Sweden, southeastern Canada, and the Adirondack Mountains of New York.[114,132-136] Fish have been eliminated from many fresh waters which have become acidified. As a general rule, acidified lakes are fishless below pH 4.8. In the U.S. the most heavily impacted region known is in the Adirondack Mountains. A survey of 217 lakes above 610 m elevation revealed 51% had pH values below 5. Of these acidic lakes 90% had no fish. In the period 1929-37, only 4% of these lakes had a pH below 5 and were fishless.[135]

Very large geographic areas impacted by acidic precipitation have suffered reduced fish stocks. This has been most clearly demonstrated in Sweden and Norway, where the number of fishless lakes is increasing yearly, but the same appears to be true for southeastern Canada from Ontario to Nova Scotia. No comparable analysis of fish status exists for the eastern U.S. outside of the Adirondack Mountains.

Elimination of fish populations from lakes and rivers is primarily caused by reproductive failure. As young-of-the-year are eliminated and year classes fail in most acidic waters, older fish become larger due to reduced competition for remaining food supplies. Diet also changes as preferred prey are eliminated by acidification. Egg and fry mortality is the primary cause for reproduction failure.[114,134-137]

Fish species differ in their tolerance of acidic conditions. In a survey of 50 Swedish lakes, Almer et al.[121] found the following order of sensitivity: roach > minnow > arctic char > trout > cisco > perch > pike > eel. Extensive Norwegian fisheries studies found Atlantic salmon to be most sensitive followed by sea trout (Salmo trutta) > brown trout > perch and eel.[134] A similar but more detailed list is provided by Beamish.[138]

Mechanisms of Fish Injury. High acidity leads to a loss of salts from blood plasma and body tissues of fish. Freshwater fish have serum salt concentrations which are hypertonic relative to surrounding water. Osmotic uptake of water is balanced by excretion but this is accompanied by loss of salts. Salt balance is maintained by active uptake through specialized cells which, in fish, are on the surface of the gills. Acid-stressed trout had reduced Na^+ and Cl^- concentrations in plasma and reduced K^+ in muscle tissues in amounts equivalent to the reduction in plasma Na^+.[134,139]

Acid stress on fish increases with decreasing total salt concentrations of the ambient water. Since a major factor is loss of salt, stress should increase as the availability of salt decreases. Leivestad et al.[134] have shown that increasing salt concentration to 14.4 mg Na^+ ℓ^{-1} reduced mortality of eggs and yolk-sac larvae of brown trout at pH ca. 4.9 compared to unamended water with 1.6 mg Na^+ ℓ^{-1} at the same pH. In spring, snow-melt water caused a dilution of stream water and, with first melt, an increase in acidity due to leaching of accumulated acids from the snow pack. Sea-trout larvae were killed when exposed to melt water from snow in southern Norway and survival of sea-trout (Salmo trutta) larvae decreased with acidity. The effect of dilution was demonstrated by rapid mortality of larvae in distilled water at pH 5.6.[134] A study of fish status in 941 lakes in southern Norway found total salt content to be directly related to "good" fish status and inversely related to fishless lakes.[134] Experiments with brown trout in flow-through tanks with pH-adjusted river water also showed improved regulation of ionic balance by increasing Ca^{+2} from 0.4 to 0.9 mg ℓ^{-1} at pH 4.0 and 4.3.[140]

Elevated levels of aluminum caused by watershed acidification have serious effects on fish. Watershed acidification results in elevated concentrations of aluminum in runoff waters to the range of 1 mg Al ℓ^{-1}. Laboratory studies show Al to be toxic to salmon at 0.2 mg ℓ^{-1}. Fish kills of cisco in two Swedish lakes impacted by acid deposition occurred when stream and lake water Al concentrations were 0.5 mg ℓ^{-1} at pH 5.0 to 5.5. Fish-gill Al concentrations were 7 to 8 times higher than in gills of fish from uncontaminated lakes. Other factors, i.e., climatic, chemical, and biological also contributed to these fish kills.[141]

Aluminum speciation is an important factor in its toxicity to fish. Maximum toxicity of Al to brown trout tested over the pH range 4.0 to 6.0 and Al concentrations of 0.2 to 0.8 mg ℓ^{-1} occurred at pH 5.0 with no toxicity observed at pH 4.0 and 6.0.[140] Al forms strong complexes of OH^-, F^-, $SO_4{}^{2-}$, and dissolved organics, and additions of these materials in experiments with brook trout and white sucker eggs and fry at pH 4.0 to 5.5 and Al concentrations of 0.1 to 0.5 mg ℓ^{-1} reduced or eliminated the toxic effects of Al.[142]

Toxic aluminum concentrations are associated with disturbed ion exchange over gills and with respiratory distress. Analysis of venous blood from brown trout exposed to 0.2 to 0.8 mg Al ℓ^{-1} and pH 4.0 to 6.0 found a rapid loss of plasma Na^+ and Cl^- occurred with Al toxicity. Addition of Ca^{2+} reduced this ion loss. Massive mucous clogging of gills occurred and venous oxygen tension was lowered in these fish.[142]

Effects of altered fish food supply, especially in the pH range of 5.0 to 6.0, on fish condition or community composition have not been studied. Acidification of lakes causes drastic alterations of plant and invertebrate communities in lakes. Although there is little doubt that physiological effects of H^+ and Al are the principal cause of complete elimination of fish from most acidic waters, slight effort has gone into investigating the consequences of changing food quantity or quality at intermediate acidity levels (pH 5.0 to 6.0). This may be a critical factor to survival and growth of newly hatched fish.

Overall Evaluation of Effects on Aquatic Ecosystems. Freshwater acidification has resulted in the elimination of many species of aquatic organisms at essentially all trophic levels. Elimination of all fish from numerous lakes in southern Norway, southwestern Sweden, southeastern Canada, and the northeastern United States is well documented. This appears to be due primarily to the physiological effects of H^+ and of aluminum ions (aluminum concentrations are greatly elevated by watershed acidification). Furthermore, many fish-food animals are eliminated at pH levels below 6.0. Fishery records indicate that lake acidification became a severe problem in the past two decades and that ever-larger areas are being impacted. Several areas of the U.S. not yet severely acidified are especially susceptible (upper Minnesota and Wisconsin) and decreasing pH of surface waters is anticipated at current rates of acid deposition.

The first set of assumptions stated at the beginning of this section concerning freshwater acidification on which a standard for precipitation acidity might be based really poses two questions. First, is there a cause-effect relationship between precipitation acidity or substances for which acidity is a surrogate and freshwater acidification? The numerous studies cited, including the simple empirical model approach[38] indicate that in the most sensitive regions acidification has occurred when the volume-weighted average annual hydrogen ion concentration exceeded 25 μeq ℓ^{-1} (pH 4.6). These models also show that lake acidification is strongly correlated with the concentration of sulfate in precipitation. Second, can acidification of fresh waters be prevented or ameliorated by not allowing the acidity of precipitation to exceed a definable concentration? This has not yet been demonstrated but a "yes" answer is implicit in the response to the first question.

The second set of assumptions when phrased as questions asks, are ecosystem components damaged by acidification and are these damages severe enough to warrant establishment of a standard to prevent or ameliorate them? The first part of this second set is shown to be true. There are major biotic changes in freshwater ecosystems ranging from altered composition and supply of fishfood organisms to complete elimination of fish. The well-documented elimination of fish from lakes and streams in the most sensitive areas, which are also impacted by acid depostion, constitutes an undesirable change (damage) to freshwater biota in these most sensitive areas. The second of these two questions cannot be answered directly now because several questions are yet unanswered regarding the severity of this damage. Among these unanswered questions are the following:

Are areas in which fresh waters are acidified increasing? In Norway they are,[70] but evidence documenting this in the U.S. is not strong. Comparisons of data over several decades demonstrate acidification has occurred, but do not necessarily indicate an increasing area of impact. On the other hand, there is evidence that acidification of lakes may be beginning in areas of the U.S. outside of the Northeast, in Minnesota,[143] Florida,[144] and Washington State.[145]

What is needed to answer this question is an intensive effort to obtain and analyze historical water-quality and fisheries records. Most of these records are not automated and their retrieval will be a tedious task. This approach was used very successfully in the Norwegian study, however.

Although fisheries are damaged by elimination of species at low pH (pH <5.0), what impact does acidification have on fisheries in the pH range 5.0 to 6.0? In this range the quality of fish-food changes (reduction in prey species) and some evidence indicates the quantity of fish-food organisms may also be reduced. Studies of acidification impacts on population and community dynamics are almost totally lacking. It is conceivable that the extent of damage to fisheries is much greater than currently reckoned because the current practical definition of meaningful injury is complete elimination of fish. Integrated studies of populations and communities at intermediate pH ranges are needed. These should include life cycle studies of selected species, microcosm assemblages of simple communities and integrated studies of whole-lake ecosystems in acidified areas.

Are the demonstrated damages (and possibly as yet unknown damages) to fresh waters severe enough to warrant the establishment of an air quality standard on precipitation pH? This is a political, not a scientific question. If currently demonstrated injuries are to be avoided, then simple empirical models[88] may now be adequate to define a maximum allowable standard. At precipitation acidities over 25 $\mu eq\ H^+\ \ell^{-1}$, acidification will occur (according to evidence cited above) in the most sensitive areas, resulting in elimination of fish from some lakes. It is not known if damages less than the complete elimination of fish are occurring in somewhat less sensitive areas because no investigations of this possibility have been made.

If precipitation acidity decreases, will acidification be abated? If so, at what rate? The answers are not currently known. Experimental manipulation of small watersheds, e.g., application of dissolved lime in low volume but sufficient quantity to be equivalent to an increase in average annual acidity, is one experiment which could assist in obtaining an answer.

Terrestrial Vegetation

Establishing a standard for precipitation acidity to protect terrestrial vegetation involves several assumptions. The first of these is that acidity in precipitation causes injury to vegetation. Injury can be defined as: a) loss of crop yield and/or quality, b) visible injury which would reduce the market value of a crop, c) loss of forest yield or long-term growth of trees, d) visible injury to ornamental plants that would reduce their aesthetic value, e) substantial alterations of plant community composition leading to ecosystem simplification, f) changes in herbivore populations or communities leading to any of the injuries above, and g) altered sensitivity to other air pollutants and/or plant pathogens leading to any of the injuries above. There is also an assumption that controlling the acidity of precipitation by establishing a standard which is not allowed to be exceeded will prevent or ameliorate known or highly probable injuries.

The impacts, if any, that acidic precipitation is having on terrestrial plants outside of laboratory or controlled field conditions have yet to be determined. Experimental research to date has shown that plant response can vary greatly depending on the species, the environment, and the method of exposure. Due to this variability, it has been necessary to study the impacts of acidic precipitation on a plant species by plant species basis. From this approach an overall picture of which plant groups are most sensitive and the types of injury which occur is beginning to emerge.

Experimental exposures of plants to simulated acidic precipitation over days, weeks, or a growing season are conducted to evaluate relationships between treatment H^+ concentrations and plant responses. In evaluating experimental results, it is important to distinguish between effects observed at H^+ concentrations which are much above the volume-weighted mean H^+ concentration of ambient precipitation, near ambient H^+ levels, and so-called "control" H^+ levels which are much below ambient levels (e.g., $pH \geq 5.6$). Both extremes, unusually high and unusually low H^+ concentrations, represent conditions that do not occur in the northeastern United States. However, when a large range of H^+ concentrations are tested, linear or curvilinear functions, with known confidence limits, of plant responses versus H^+ concentration will provide information to estimate changes in plant responses due to ambient or anticipated levels of acidity.

Evidence of Injury to Terrestrial Vegetation.

Loss of Crop Yield and/or Food Quality. Exposure to acidity in precipitation may conceivably decrease yield and/or food quality of crops. Since it is generally accepted that managed soils are less susceptible to perturbations by acidic precipitation due to normal applications of fertilizers and lime,[16] any effects of acidic precipitation on crop plants would probably result from exposure of foliage. Significant reductions (19% and 11%) in dry weight of trifoliate leaves and dry weights of pods plus seeds, respectively, of pinto beans occurred after exposure to acidic mists even though there was no visible leaf injury, when plants were grown in pots in a greenhouse.[146] In contrast, simulated acidic mists of pH 3.0 did not reduce plant growth rates of yellow birch even though all leaves exhibited leaf pitting and curling.[147] With many crop plants yield is determined by the development and survival of reproductive organs as well as by cumulative injury to foliage. Because relationships between growth and yield (plant biomass, economic crop yield, etc.) and visible plant injury, or any other index of injury, remain unknown for most plant species, no unified view of visible injury versus yield impacts is available at present.

Simulated acidic rain of pH 3.1 and below decreased the dry mass of seeds, leaves, and stems of pinto beans grown in pots under greenhouse conditions. On a percentage-mass basis the decrease in seed yield was comparable with reductions in biomass of leaves and stems. This decrease in yield was attributed to both (1) a decrease in the number of pods per plant and, (2) a decrease in the number of seeds per pod.[148] Simulated acidic rain decreased the dry mass of both stems and leaves of soybeans grown in a greenhouse. However, an increase in seed yield occurred when plants were exposed to rain of pH 3.1. A larger dry mass per seed was responsible for the larger dry mass of seeds per plant.[148] Effects of simulated acidic rain on 27 crop plants grown in pots were determined. The marketable yields of five crops (radish, beet, carrot, mustard greens, and broccoli) were reduced, while yields of six crops (tomato, green pepper, strawberry, alfalfa, orchard grass, and timothy) were increased when they were treated with acidified rain at various pH levels between 3.0 and 4.0 compared to pH 5.7 treatments. No consistent effects were observed for 16 other crops.[149]

Simulated acidic rain caused significant reductions in soybean yield and quality under experimental field conditions using standard agronomic practices.[150] Plants exposed to simulated acidic rainfalls of pH 4.0, 3.1, and 2.7 in addition to ambient rainfalls decreased seed yields 2.6, 6.5, and 11.4%, respectively, compared to plants exposed to ambient conditions only. On a per-plant basis, total seed protein decreased 10, 19, and 23% in plants exposed to simulated rainfalls of pH 4.0, 3.1 and 2.7, respectively, compared to plants exposed to ambient conditions only.[150] At the present time, this is the only adequately replicated experiment known that demonstrates significant alterations in crop productivity and quality under standard agronomic conditions due to precipitation acidity. Additional experimentation of this type using conventional agronomic practices is needed to evaluate injuries that may occur due to precipitation acidity at or close to current acidity levels.

<u>Visible Injury which would Reduce the Market Value of a Crop.</u> If the foliage or fruits of some crops exhibit blemishes, the market value would be reduced substantially. Simulated acidic rain has induced lesions on leaves and reproductive structures. The highest experimental treatment pH at which visible lesions on leaves have been observed is near pH 4.0.[149] A large percentage of the leaf area may exhibit lesions at pH 3.1, but at pH levels above 3.1 less than 5% injury is observed.[146,147,151-156] No relationship between crop yield reduction and foliar injury has been established, so this indicator of injury should not by itself be used to set a standard for precipitation acidity.

There is no evidence that current acidity levels injure field or forest plant foliage. Simulation experiments at low pH, however, indicate that this may be possible. Most injury to foliage by simulated acidic rain occurs on expanding or recently expanded leaves and needles. This is a critical stage in leaf development. Young (14 day old) birch seedlings are much more sensitive than older (6 week old) seedlings.[147] Leaves that had just reached full expansion were more sensitive to simulated acidic rain than unexpanded and already fully expanded birch leaves.[147] Needle elongation was inhibited if simulated acidic rain solutions of pH levels below 3.0 were applied to immature pine fascicles.[151,157] Applications of acidic rain may also inhibit leaf expansion.[158] More research is needed to determine the relationship between visible foliar injury and plant growth and/or yield.[156] At the present time there are no published experimental data linking realistic levels of acidic precipitation to visible injury to foliage or fruits that would reduce their marketability.

Loss of Forest Yield or Long-term Growth of Plants. There have been conflicting reports as to whether acidic precipitation can influence forest productivity. Tamm[159] concluded that except in areas where forest trees exhibit visible pollution symptoms, ambient acidic precipitation or other types of atmospheric acidity have no effect on tree growth. Other researchers found a statistically significant difference in tree growth between areas exposed to acidity and areas more remote from transported atmospheric acidity.[152,160] They concluded that there was no reason to attribute the growth reduction to anything other than acidic rain, but this by itself does not prove a cause-effect relationship. Researchers in Norway[161] and the United States[162] have not detected any consistent decreases in tree growth which could be attributed to acid deposition.

At the present time there has been no demonstration of losses of forest yield or forest productivity due to acidic precipitation. Experiments should be performed with adequate numbers of experimental replicates that could demonstrate economically significant differences between treatment means within 95% confidence limits. Experiments with adequate randomization of a large number of treatment replicates will avoid local soil and other microenvironmental problems that occur in agricultural fields and in natural terrestrial ecosystems.[150]

Alterations in photosynthesis and respiration could influence both forest and crop productivity. Photosynthesis and respiration in foliage may be influenced by exposure to acidic precipitation; however, research results are inconclusive. Leaves exposed to unrealistically low pH simulated rainfalls (pH 2.3) had lower sugar and starch contents compared with plants exposed to simulated rain of pH 5.7.[153] Foliage exposed to acidic rain may be stressed but short-term measurements of photosynthesis and respiration may not be sensitive enough to detect the degree of stress. Any relationship between the changes in carbohydrate status, on long- and short-term bases, with loss of plant and seed biomass, must be more firmly established before photosynthesis becomes a useful indicator of injury for the process of standard-setting.

Computer simulations of forest production using the JABAWA model, tested the effects of different levels of leaf area injury. Results showed that 10 to 20% per year of the leaf area of a tree species must be injured to change the role of that species or to reduce total forest biomass by a significant amount.[163] There are no indications that such extensive injuries do or will occur as a consequence of precipitation acidity.

Alterations in the nutrient content of foliage may influence the growth and productivity of both forests and crops. Moreover, nutrient levels in harvested portions of crops may affect the quality of foodstuffs. Acidic precipitation influences nutrient leaching from plant surfaces. Wood and Bormann[164] demonstrated that K^+, Ca^{2+}, and Mg^{2+} were leached from leaves of pinto beans more rapidly at pH levels of 3.0 and 3.3 than at pH levels of 4.0 and 5.0. Ca^{2+} leached faster from foliage of sugar maple than K^+ or Mg^{2+} at pH 3.0 than at pH 3.3. The leaching rates of K^+ and Mg^{2+} were significantly higher at pH 3.0 than at pH 4.0. In tobacco leaves, Ca^{2+} leached faster from foliage exposed to simulated rain of pH 3.0 than from foliage exposed at pH 6.7.[165] In pinto beans exposed to simulated rain for 5 days, more calcium, nitrate, and sulfate were leached from foliage at pH levels of 2.7, 2.9, and 3.1 than at a control pH of 5.7[166] In contrast, the amount of potassium leached was greater from leaves exposed to pH 5.7 than leaves exposed to pH levels between 3.4 and 2.9. The amounts of ammonium, magnesium, and zinc were the same at all pH levels tested. At the present time, insufficient experimental data are available to relate nutrient leaching and precipitation acidity with changes in vegetation growth or yield. As a result, nutrient leaching is not now a useful indicator of injury for the standard-setting process.

Acidic precipitation may change the surface characteristics of foliage. It has been suggested that acidic precipitation may effect the submicroscopic structure of the epicuticular wax layer(s) of leaves.[167] Some electron micrographs support this idea. This preferential erosion of the epicuticular waxes may be important for the penetration of rainwater. The effects of acidic precipitation on the cuticle itself are probably minimal, however. The most widely used methods of isolating cuticles involve exposure to strong acids.[168,169] Penetration rates of elements through isolated cuticles without epicuticular waxes were similar to rates through intact cuticles.[169] At the present time, there is no conclusive evidence that the leaf surface is altered by acidic precipitation to cause loss of crop or natural ecosystem productivity.

Nitrogen is, in general, the nutrient limiting productivity of temperate forests. Biological nitrogen fixation plus the deposition of ammonium and oxides of nitrogen are the routes by which this critical nutrient becomes available to forests. At Hubbard Brook, New Hampshire, bulk precipitation provides 6.5 kg N ha^{-1} yr^{-1}, an amount equivalent to nearly 40% of the total nitrogen added to forest biomass annually.[79] The proportion of nitric acid in rain over the northeastern U.S. and in Colorado has been increasing and now accounts for one-third of the acidity in precipitation.[33,170-172] Thus there is a potential for positive effects of acidic precipitation on forests by fertilizing with nitrogen as well as the potential for injury due to acid effects on vegetation by inhibition of the nitrogen cycle and other microbial processes in soils, or increased leaching rates of nutrients from soils.

Computer simulation exercises were performed to test some of these effects. The JABAWA computer model of forest growth was modified to include effects of precipitation acidity on nitrogen availability and tree growth. The model found a direct positive relationship between N availability and biomass accumulation.[173] The relationships between forest production, litterfall, and decomposition of litterfall were analyzed using the FORTNIT forest nitrogen model. The simulation found lower rates of litter decomposition would be compensated by higher forest floor biomass accumulation so that N availabilities would not be greatly altered over a period of 15 to 40 years. After 40 years, slower litter decomposition rates provide higher N availabilities due to the decomposition of the larger litterfall biomass. However, no field data are available to confirm these simulations.[173]

Precipitation acidity may preferentially affect unicellular organisms that are very important in ecosystem function. Most multicellular organisms that have evolved from aqueous environments onto dry land have specialized surfaces to retard desiccation. Organisms with protective coverings may be less sensitive to acidic rain than organisms that lack these coverings. Bacteria and other single-celled organisms are very sensitive to pH changes, presumably because they lack protective coverings. Procaryotic organisms seem to survive within a narrower range of pH levels than most multicellular organisms. Motility of most procaryotic organisms is greatest between pH 6.8 and 9.0.[174-176] Hoeninger[177] showed that a pH decrease results in an increase in abnormal wave motions in flagella of *Proteus*. Below pH 5.0 almost all motility ceased. The activities of bacteria are very important for the decomposition of leaf litter and thus the cycling of nutrients. However, although unicellular bacteria are very sensitive to the acidity in rainfall, with the present level of knowledge, the overall effects of natural levels of precipitation acidity on leaf decomposition and other related processes are unknown.

Visible Foliar Injury to Ornamental Plants that would Reduce Aesthetic Value. At the present time there is no evidence showing visible foliar injury to ornamental plants due to realistic precipitation acidities (excluding point source studies) that would reduce their market value. However, little research has been focused on this area.

Substantial Alterations of Plant Community Composition Leading to Ecosystem Simplication. If one or more plant species are preferentially affected by precipitation so they become greatly reduced or eliminated from plant communities, then the ecosystem may become simplified. This ecosystem simplification may eventually influence overall ecosystem productivity. Precipitation acidity could inhibit one or more critical stages in the life cycle of an organism. Experimental results show that fertilization and spermatozoid motility in ferns are very sensitive to pH levels below 5.7 and additions of sulfate and nitrate (43 μM).[178-180] These results suggest that the acidity of ambient precipitation in the northeastern United States inhibits the reproduction of ferns. The overall impact(s) that these injuries may have on ecosystem productivity is unknown.

Alterations in Herbivore Populations or Communities. It is conceivable that precipitation acidity might directly or indirectly alter populations of herbivores either positively or negatively, and such changes could be reflected in crop or forest productivity. However, at present, no data are available on the effects of precipitation acidity on terrestrial herbivores, let alone on subsequent impacts on plants.

Altered Sensitivity of Vegetation to Other Air Pollutants and/or Plant Pathogens. Acidic precipitation may affect gas exchange in plants which could alter plant sensitivity to other air pollutants. Individual cells of the epidermis may be injured upon initial exposure to acidic rain at pH levels of 3.4 and below.[154-156] Moreover, acidic rain may cause some alterations in the cuticle and/or in the functions of guard cells.[166] Foliage exposed to acidic rain may be more subject to wilting or water stress as well as becoming more sensitive to gaseous air pollutants. Knowledge of the effects of acidic rain in combination with gaseous pollutants such as ozone and sulfur dioxide is needed in order to understand the impacts precipitation acidity might have on vegetation in nature.

Acidic precipitation can affect microorganisms that inhabit surfaces of higher plants. Simulated acidic rain produced an 86% inhibition of the number of telia of *Cronartium fusiforme* on willow oak (*Quercus phellos*) and a 29% decrease in the percentage leaf area affected by bean rust (*Uromyces phaseoli*) on *Phaseolus vulgaris*.[181] Halo blight caused by *Pseudomonas phaseolicola* infections of leaves of *Phaseolus vulgaris* was stimulated or inhibited by simulated acidic rain depending upon the timing of application. Simulated acidic rain stimulated initial infection but inhibited development of the pathogen if applied after infection began. The overall effects that ambient levels of precipitation acidity have on plant host-plant pathogen interactions remain unknown at present. However, if rainfall acidity should increase or decrease, the degree to which plant pathogens affect either crop or forest plants may have significant economic impacts.[182,183]

Overall Evaluation of Effects of Acidic Precipitation on Terrestrial Vegetation. At the present time there is little unequivocal evidence that acidic precipitation, at ambient levels, is having deleterious effects upon terrestrial vegetation within the United States. The above statement is made more from a lack of concrete information than from an evaluation of large amounts of experimental data. Experiments that have been conducted in laboratory or greenhouse settings have yielded some information that may point to mechanisms of injury. However, few field experiments have yielded definitive results. Although decreases in yields of various crops have been documented in controlled-environment and field experiments at pH levels below pH 4.0, only well-designed field experiments will document changes in plant productivity or survival that may be expected from actual acidic rain exposures. It is essential that experiments using standard agronomic practices with large numbers of replicates be conducted so that results can be extrapolated to actual crop values.

Acidic precipitation may affect the productivity of crop and forest plants by direct impaction. If significant alteration in productivity of either crops or forests occur, significant economic loss may result. Currently, evidence linking levels of rainfall acidity to crop yields is meager. However, from present data it is clear that the level of injury, if injury occurs, is less than year-to-year changes due to differences in natural climatic factors such as precipitation volume or temperature. This does not mean, however, that possible injuries from acidic precipitation can be ignored. It is important to determine even small changes in productivity. Modeling exercises show that a growth reduction of 10 to 25% per year of one tree species would be required to reduce overall forest productivity. Below this 10 to 25% range a decline in productivity of one or more species could be compensated for by an increased productivity of one or more other species.[163,184] However, it should be noted that while such compensations may occur in complex ecosystems, they would not occur in agricultural systems. A 1.0% soybean yield reduction[150] would have represented a loss to growers in the northeastern United States of 50 million dollars in 1979 (ignoring price elasticity).[185] This reduction would affect yields independent of other sources of variation (except drought). Such losses from acidic precipitation, while contributing to an overall reduction in the annual harvest, may go undetected because annual variations in soybean yield (1714.2 to 2131.0 kg ha^{-1} in the decade 1970-1979) are much larger.[186]

In order to standardize results from future experiments, a uniform expression of data should be utilized. To date, the most meaningful relationships between rainfall acidity and plant responses have been obtained when the responses are plotted as a function of hydrogen ion concentration of the test solution applied.[158,180] This expression or some other more meaningful expression should be used so that data can be compared with ambient precipitation acidities.

Precipitation Acidity Standard for Protecting Forests and Crops. It has not yet been demonstrated that current regional levels of precipitation acidity alone significantly injure terrestrial vegetation (excepting areas affected by specific point sources). There are some suggestions that crop injury may be occurring, but the evidence is not conclusive. Since cause-effect relationships have not been shown, there is no information available on which to establish an acidity standard for precipitation that would prevent or ameliorate injuries. In the area of effects of acidic precipitation on terrestrial vegetation a much larger data base is required before a standard could be established, if indeed a standard is necessary.

Overall Evaluation

This paper evaluates the current state of knowledge of the impacts of acidic precipitation on a variety of receptors and addresses the following questions: (a) what pollutants are responsible for precipitation acidity, (b) where do the greatest impacts occur, (c) what receptors are at risk, (d) what is the current extent of damage and benefits, (e) what are the environmental costs and benefits of preventing acidic precipitation, and (f) should an air quality standard be developed now for acidity of precipitation?

Acidic precipitation, wet or frozen deposition with a hydrogen ion concentration greater than 2.5 $\mu eq\ \ell^{-1}$ (less than pH 5.6), is a significant air pollution problem in the United States. The chief anions accounting for the hydrogen ions in precipitation are nitrate and sulfate. Within the United States, precipitation acidity is greatest in the Northeast where about 90% of all sulfate and a significant fraction of all nitrate is attributed to anthropogenic emissions. Potentially, ecosystems within this area are at greatest risk. Data show that precipitation within the northeastern United States is acidic and that there are large variations in chemistry within and among rain events and among nearby sites for individual events. This variability in chemistry makes it impractical to establish a precipitation standard based upon data collected from a small number of samples. However, even with such variations, precipitation at representative sites within the Northeast show that very few events fall outside of the pH range of 3.00 to 5.49.

Agricultural systems may derive net nutritional benefits from increasing inputs of S in acidic rain but inputs of N are usually not great enough to contribute substantially to crop needs. Agricultural soils are generally better buffered and heavily fertilized, making the dangers of soil acidification much less than in forest soils.

Moderate inputs of acidic rain can be of nutritional benefit to forests by supplying N and S, but detrimental effects can occur if atmospheric H^+ inputs significantly add to or exceed H^+ production by mechanisms internal to the forest soil. Estimates of internal H^+ production in forests are very rare, but the available literature gives values ranging from approximately 200 to 23,000 $eq \cdot ha^{-1} \cdot yr^{-1}$. Taking the low figure as an arbitrary maximum allowable H^+ deposition rate and assuming an equivalent input of 67% SO_4^{2-} and 33% NO_3^- as counter-ions provides ~3 kg S and 1 kg N per hectare per year. This amount of S input is unlikely to fulfill agricultural needs but will probably meet forest requirements. Nitrogen inputs of this magnitude are probably insignificant relative to crop needs, but may supply as much as 40% of forest needs.

It must be kept in mind that effects may vary widely, and inputs beneficial to one system may be harmful to another. It would be ideal to assess the nutritional costs and benefits of different levels of acidic rain inputs over different biomes, physiographic regions, and even soil series before setting standards. Then a decision must be made as to whether the nutritional cost/benefit relationship over an entire region is positive or negative at a given input level and, more importantly, whether the standard will be based on the worst-case situation within the region or on some more "average" factor for the region as a whole. Unfortunately, the current data base--especially for forests--is totally inadequate for this type of analysis.

Acid deposition accelerates rates of mineral weathering from watersheds. Three principal factors mediate this process: (1) soil volume and porosity, (2) soil minerology, and (3) runoff rate. Hydrogen ions are exchanged for cations, particularly Ca^{2+}, Mg^{2+}, and Al^{3+}. As a result, aluminum concentrations may reach levels (> 0.2 mg ℓ^{-1}) known to be toxic to aquatic organisms. To date, there has been no documentation of decreased soil pH as a consequence of acid deposition although groundwater acidification is reported in Sweden. Information relating acidic precipitation to changes in soil chemistry alone is insufficient to justify the development of an air quality standard unless such changes are clearly linked to actual or potential injuries to biota, including humans.

If the pH of soils is changed by inputs of acidic precipitation, a reduction in several soil microbial processes may occur. Some processes, e.g., organic matter decomposition, may be stimulated but these stimulations have been assumed to be of a temporary nature. With the present level of knowledge, it is not clear to what extent or at what rate current levels of precipitation acidity affect soil microbial processes.

Acidification of freshwaters in southern Scandinavia, southwestern Scotland, southeastern Canada, and the northeastern United States is caused by acid deposition. The areas of these regions in which this acidification occurs have in common highly acidic precipitation with volume weighted mean annual H^+ concentrations of 25 µeq ℓ^{-1} (pH 4.6) or higher and slow weathering granitic or precambrian bedrock with thin soils deficient in minerals which would provide buffer capacity. Correlation analyses of data from these regions indicate that 25 µeq H^+ ℓ^{-1} is a critical threshold, above which permanent lake acidification occurs in the most sensitive areas. Episodic acidification may occur in small streams at less acidic pH's. There are indications that acid deposition may cause freshwater acidification in Florida, the Blue Ridge Mountains, upper Wisconsin and Minnesota, and Washington State, as well.

Biological effects are detectable at pH < 6.0 in freshwaters. As lake or stream pH decreases below this level, many species of plants, invertebrates and vertebrates are progressively eliminated. As a general rule, fisheries are severely impacted below pH 5.0 and they are completely destroyed below pH 4.8. Currently, there is a tendency to define only the complete elimination of fish as a meaningful injury, thus only the most acidic lakes with pH values below 5.0 and no fish are counted as casualties. A new definition of injury which considers general biological damage as well as elimination of fish should be accepted. This will require evaluating biological changes on a quantitative basis in freshwaters in the pH range 5 to 6. It is likely that more widespread injury to freshwater ecosystems will be found if this new definition of environmental injury is applied.

Information available to date indicates that establishing a standard for precipitation acidity for the volume weighted annual H^+ concentration at 25 µeq ℓ^{-1} may protect the most sensitive areas from permanent lake acidification. Such a standard for protecting lakes might be applied on a regional basis. For example, northern New York, New England, upper Wisconsin, and upper Minnesota are, at this time, recognized as the most sensitive areas of the United States having large numbers of lakes at risk. Canada is not included in this analysis, but has much larger regions highly sensitive to acidification.

Before a standard can be set, however, it must first be decided whether or not the known damages (or perhaps the anticipated damages) attributable to precipitation acidity, are severe enough to warrant their prevention or amelioration. An evaluation of the magnitude of the environmental injuries in freshwaters must await a quantification of those injuries known only qualitatively in the pH ranges 5 to 6.

There is an inadequate amount of information that shows decreases in crop growth except for one field study. Most studies with plants (crops and forests) are inadequate for standard setting because they are not conducted in the field with adequate randomization of plots coupled with rigorous statistical analyses.

Although visible injury to foliage by acidic precipitation has been documented in a variety of greenhouse studies, no experimental evidence demonstrates loss of field crop value or reduction in plant productivity due to visible foliar injury.

Acidic precipitation can contribute nutrients to vegetation and could also influence leaching rates of nutrients from vegetation. Although these processes occur, there are no data that show changes in nutrient levels in foliage that relate to crop or natural ecosystem productivity.

Experimental results show that fertilization of ferns is inhibited by current levels of acidic precipitation in the northeastern United States. However, the overall impacts of inhibited fertilization on perpetuation of the species or ecosystem productivity have not been evaluated.

Simulated acidic precipitation has been shown to effect plant pathogens in greenhouse and field experiments. Simulated acidic precipitation inhibited pathogen activities under some circumstances and promoted pathogen activities under other circumstances. No conclusion can be drawn about the effects of current levels of precipitation acidity on plant pathogen-host interactions.

From these data it must be concluded that research on the effects of acidic precipitation on terrestrial vegetation is too meager to draw any conclusions with regard to an air quality standard. It has yet to be demonstrated that current regional levels of precipitation acidity alone significantly injures terrestrial vegetation. Although there are some suggestions that crops might be injured, the evidence is not conclusive. More research data need to be evaluated in this area of effects of acidic precipitation on terrestrial vegetation before a standard could be established, if one is necessary.

From available research data it seems reasonable that organisms and/or single cells that are exposed to acidic solutions within a poorly buffered environment, in the broadest sense, would be most sensitive to protons in rainfall. Good examples of this situation would be organisms that live in fresh waters, bacteria and fungi on plant foliage, sperms of lower plants such as ferns and mosses, and organisms in leaf litter. In contrast, it seems reasonable that organisms and/or cells of organisms would be less sensitive in environments that would resist changes in pH. Examples would be highly buffered soils and cells within plant foliage with thick protective coverings that would retard penetration by acidic precipitation.

If we are to alleviate the effects of acidic precipitation, we must first decide whether or not the known or highly probable damages are severe enough that we are willing to pay the costs of reducing SO_X and NO_X emissions. However, it should be recognized that precipitation acidity is just one part of the much larger problem of air pollutants emitted by fossil fuel combustion. This larger problem involves considerations of human health, corrosion of structures, damages to forests and crops, and climate change. Taken together the environmental costs from fossil fuel combustion may be quite significant. The costs to prevent or ameliorate the effects of acidic precipitation in the United States are uncertain because the quantitative physical and chemical relationships between SO_X and NO_X emissions and a specified precipitation acidity level are unavailable.

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