Acid deposition: a select review 1852–1990

1. Emissions, transport, deposition, effects on freshwater systems and forests

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(*Received 26 August 1992; revised 22 September 1992*)

Part 1 of this review discusses the origins of acid deposition and its impacts upon freshwaters and forests. Part 2 will be concerned with the impact of acid deposition and its precursors on materials and human health, and with the control technologies and programmes introduced as a consequence of the environmental impacts of acid deposition.

(Keywords: emissions; acid deposition; environmental impact)

INTRODUCTION

The purpose of this paper is to review the sources and effects of acid deposition, and the technological and political initiatives that may be, or have been, taken to exercise control over emissions of acid deposition precursors.

The environmental implications of acid deposition and acidification remain the subject of extensive international and national research programmes addressed to the local, regional and global sources and effects of sulfur dioxide, oxidized and reduced nitrogen species, and volatile organic compounds.

The relation between cause and effect is by no means fully understood, owing partly to the extremely complicated interactions between pollutants and within atmospheric, terrestrial and freshwater environments, which have both direct and indirect effects upon susceptible targets. However, the broad relation between sulfur and nitrogen emissions and environmental change, particularly with regard to freshwater environments, is understood well enough to enable national and supranational control policies to be formulated, for example the European Community's Large Combustion Plant Directive and Cars Directive, or the United Nations Economic Commission for Europe (UNECE)'s protocols on emissions of sulfur dioxide or nitrogen oxides and their transboundary fluxes.

The scale and importance of the contemporary phenomena tend to suggest that acid deposition is a relatively recent environmental problem. However, it was first described in 1852 by R. A. Smith (see *Figure 1*) working in the city of Manchester[1], and it was Smith[2] who coined the term 'acid rain'. At this time, sulfur was clearly the main pollutant and the effects of acid deposition were clearly an urban and near-urban phenomenon. Environmental effects attributable to acid deposition either described or postulated by Smith included damage to building materials, metalwork and textiles, as well as the impact on vegetation[2]. The southern Pennines of England received acid precipitation as a direct consequence of acid emissions in Manchester and other cities and have continued to do so for over

Figure 1 R. A. Smith

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Figure 2 *Sphagnum papillosum*

200 years[3]. The combination of dry and wet deposition of sulfur compounds profoundly affected sphagnum moss communities (see *Figures 2* and *3*), leading to disappearance of species, habitat change, moorland drying and subsequent erosion[4].

Other contemporary measurements of precipitation chemistry include those begun at Rothamsted[5–7] in 1853, the initial interest being in the supply of nutrients to plants. Other historical measurements of precipitation composition are described by Cowling[8]. Although the distribution of pollutants from industrial and urban areas was generally no more than regional in scale, longer-range transport of pollutants to Norway from the UK is considered to have occurred as early as 1881[9].

Although Cowling[8] provides an extensive review of the historical interest in precipitation chemistry, only a fragmented data set of limited quality is available[10,11] for the period before the renaissance of interest in the subject of acid deposition in the late 1960s and early 1970s[12,13], when effects on freshwater were observed. From that time on, research into the causes and effects has been driven by a political impetus, as nations have individually and cooperatively responded to the challenges posed by acid deposition.

One of the most important of the subsequent phase of investigations was the Norwegian SNSF project that began in 1972[14]. Scientific and political interest in the USA was stimulated by a discussion[15] of the regional distribution of acid precipitation and its significance for terrestrial and aquatic ecosystems in North America. In Europe, as a consequence of the Swedish case study[13], the Organisation for Economic Cooperation and Development began its review of the long-range transport of air pollution in 1973[16]. Under the auspices of the United Nations, the European Monitoring and Evaluation Programme (EMEP) began in 1977[17], and shortly afterwards the National Atmospheric Deposition Program became operational in the USA[18]. The National Acid Precipitation Assessment Program was established by Congress under the Acid Precipitation Act (PL 96-294, Title vii) of 1980[19]. In 1982 Sweden convened an international conference on acid rain[20], and in 1983 the joint UK, Swedish and Norwegian Surface Water Acidification Programme was proposed[21]. Contemporary scientific understanding of the causes and effects of acid deposition was reviewed at the International Symposium on Acidic Precipitation held at Muskoka, Canada, in 1985[22]. The extensive Canadian contribution to scientific understanding of the causes and effects of acid deposition has been summarized[23]. More recently the UNECE has established the International Cooperative Programme to assess the effects of atmospheric pollutants on materials[24], on forests[25] and on lakes and rivers[26]. The policy implications of acid deposition have been the subject of several conferences and are summarized by Schneider[27] and the Office of Technology Assessment[28].

EMISSIONS OF ACID DEPOSITION PRECURSORS

The major gaseous precursors of acid deposition are sulfur dioxide (SO_2) and oxides of nitrogen (NO_x), but emissions of ammonia, volatile organic compounds, hydrochloric acid and alkaline dusts all influence the acidity of precipitation to some degree[10,29]. These species have both natural and anthropogenic sources. Anthropogenic sources include power generation, refineries, industry, transport and commercial and domestic activities[30]. Natural sources include biogenic emissions from terrestrial, tidal and oceanic areas and non-biogenic emissions from natural combustion, geothermal activity, lightning, airborne soil particles and water aerosols[10]. Emissions of sulfur dioxide, nitrogen oxides, ammonia and volatile organic compounds are discussed below.

Sulfur dioxide

Moller[31] estimates the natural and anthropogenic emissions of SO_2 to be of the same order, recent estimates of natural emissions[32] being in the range 100–1000 Mt a^{-1} and anthropogenic emissions[33] 120–160 Mt a^{-1}. Such estimates must be treated with caution, as they are extrapolations from a small number of measurements to the global scale.

However, although there is similarity in order of magnitude, there are distinct spatial differences between natural and anthropogenic sources of emission. Biogenic sources are concentrated in tropical areas[32], whereas anthropogenic sources are predominantly located in the industrialized northern hemisphere, covering $<10\%$ of the Earth's surface, where $\sim 68\%$ of anthropogenic emissions occur[34]. More than 90% of the sulfur in the

Figure 3 Blanket bog, North Wales: foreground, *Spagnum cuspidatum*; middleground, *Eriophorum* spp.; background, *Calluna vulgaris*

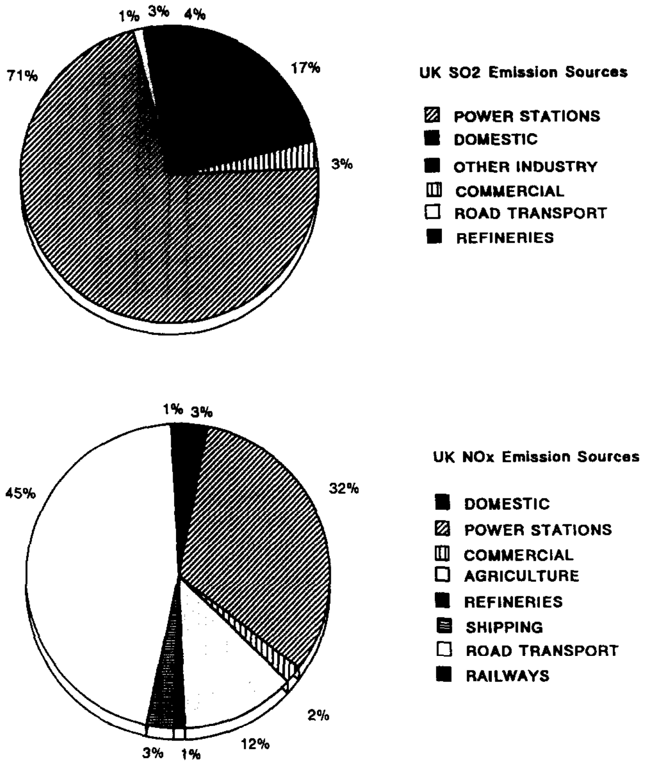

Figure 4 UK sulfur dioxide and nitrogen oxides emission sources (Ref. 30)

northern hemisphere atmosphere is of anthropogenic origin, and thus on the regional scale, man-made emissions predominate[35]. Recently, emissions of marine-derived sulfur species have attracted considerable scientific interest. Such emissions may be of local significance in spring and summer, and further work to assess this is required[29].

Although global anthropogenic emissions are estimated[34] to be ~160 Mt a^{-1} estimates of emissions are most detailed for industrialized areas where fuel combustion and other data are used as a basis. In 1987, according to the European Monitoring and Evaluation Programme[36], Europe is estimated to have emitted 42.942 Mt of SO_2, 18% less than in 1980. In 1988 Europe is estimated[37] to have released 41.856 Mt of SO_2.

On a national scale, where good estimates of consumption and sulfur content of fuels are available, and information on population density and other factors is reasonably reliable, emission calculations are more precise, but they still have an uncertainty of 10–20%[10,38]; for example, the UK emission estimate[30] for 1988 (see *Figure 4* and *Table 1*) shows a total of 3.664 Mt of SO_2, down 5% from the previous year. Some 71% of this is estimated to have arisen from power stations, 17% from industrial sources, 4% from the domestic sector, 3% from refineries, 3% from the commercial sector and 1% from road transport. Emissions from large combustion sources, >50 MW(th) and the subject of European Community Directive 88/609, were 84% of total UK emissions.

Major source areas of anthropogenic emission include the northern part of England, the Low Countries, the former Western Germany and Eastern Europe[10]. A large area of high emission density occurs in the eastern USA: 75% of all US emissions arise east of the Mississippi river[39].

Temporal trends in the European emission of sulfur since 1865 have been reviewed by Bettleheim and Littler[40] and from 1900 to 1980 in the USA by Gschwandtner *et al.*[39]. Emissions at the turn of the century were dominated by coal-burning for heating and industry, but as a consequence of the postwar economic boom in the 1950s, emissions sharply increased as a result of power station expansion and increased oil consumption[10]. By 1979 Europe accounted for 44% and North America 24% of global anthropogenic emissions[33]. Since that date the use of flue gas desulfurization and increased energy efficiency have resulted in decreases in emissions in many countries. Despite the reductions recorded in the northern hemisphere, global emissions continue to increase[29].

Oxides of nitrogen

Almost all oxidized nitrogen is released as nitric oxide (NO), which is oxidized at ambient temperatures to nitrogen dioxide (NO_2), the process being accelerated by the presence of volatile organic compounds and ozone[30]. The emissions of oxides of nitrogen are less certain than for SO_2 and the total emission is estimated by Irwin[10] to be in the range 25–99 Mt a^{-1} expressed as nitrogen. As with SO_2, there are both anthropogenic and natural sources of emission of NO and NO_2. The principal natural sources are biomass burning, lightning, microbial activity, biological processes, ammonia oxidation and stratospheric input[10]. Natural sources may account for ~33% of global emissions[41], although some authors[42] suggest rough equivalence between natural and anthropogenic emissions. Anthropogenic sources include high-temperature combustion of fossil fuels in power stations and motor transport. Estimation of emissions is complicated by the formation of NO_x from nitrogen present in the fuel and from thermal oxidation of atmospheric nitrogen[43]. Hence emissions depend on combustion conditions and fuel properties. The largest source is now likely to be vehicle emissions.

Major source areas are similar to those for SO_2, and in North America and Europe anthropogenic emissions may account for 75–93% of all emissions of NO_x[44]. The 1988 European emission of nitrogen oxides (as NO_2) was estimated[37] at 20.408 Mt. The UK emission in 1988 was 2.480 Mt, a 2% increase on 1987[30] (see *Figure 1* and *Table 1*). The major sources are road transport, responsible for 45% (23% from motor spirit and 21% from DERV), 32% from power stations, 12% from other industrial sources and the remainder from a variety of

Table 1 United Kingdom sulfur dioxide and nitrogen oxides emissions by source in 1988[a]

Source	SO_2 (10^3 t)	(%)	NO_x (10^3 t)	(%)
Domestic	155	4	72	3
Commercial/public service	100	3	59	2
Power stations	2618	71	792	32
Refineries	97	3	34	1
Agriculture	7	–	4	–
Other industry	618	17	297	12
Railways	4	–	35	1
Road transport	54	1	1108	45
Civil aircraft	–	–	9	–
Shipping	9	–	70	3

[a] From Ref. 30

sources including the domestic and commercial sectors and refineries. Emissions from large combustion sources, >50 MW(th), and the subject of European Community Directive 88/609, are 40% of total UK emissions.

Ammonia

Ammonia is the most important neutralizing compound in the atmosphere. Its major sources include animal wastes, fertilizer applications and industrial emissions. Smaller contributions arise from human respiration, traffic and uncultivated soil[45]. The European spatial pattern of ammonia emissions is quite different from that of SO_2 or NO_x, since agricultural rather than industrial areas dominate the emission profile[10]. The UK Review Group on Acid Rain[29] reported the European emission of ammonia to be 6.956 Mt a^{-1} expressed as nitrogen, and Kruse *et al.*[46] have recently estimated the England and Wales agricultural emission to be 302 000 t a^{-1}.

Temporal analysis of emissions indicates that in recent years they have increased as a consequence of intensive animal husbandry practices; for example, emissions from livestock are estimated[47] to have increased by 50% between 1950 and 1980.

Volatile organic compounds (VOC)

Certain VOCs are important in the formation of acidic species in the atmosphere[10] through photochemical generation of oxidizing species. VOCs include hydrocarbons, oxygenated and halogenated organics (excluding methane) emitted from traffic sources, industrial processes and evaporation of solvents. Natural sources are also important, particularly forests. Regional-scale emission estimates are available for the USA[48], Europe[49] and Western Europe[50], where the major sources are road transport and industrial solvents. As with other estimates of emissions, national data are considered to be more reliable; for example, the 1988 UK emission of VOCs is estimated at 1.846 Mt, 57% from industrial processes and 30% from road transport[30]. The uncertainty attached to VOC emission estimates is much larger than for SO_2.

TRANSPORT AND DEPOSITION

The processes of emission, transport and deposition are represented in *Figure 5*. On combustion of fuel, or other sources containing acid deposition precursors, emission to the atmosphere occurs. The subsequent mixing of the emitted pollutant with the receiving air is largely determined by the height of emission and the prevailing weather pattern. High-level emissions, such as those from power stations, leave the chimney at high velocity. Turbulence in the atmosphere, generated by convection and the friction between wind and surface features, acts to disperse an emission in both the vertical and horizontal planes[44]. Vertical dispersion is the principal mechanism by which emissions are diluted[51]. Mixing occurs with oxidants present in the atmosphere from other sources, and such interaction of precursors and oxidants may give rise to acidic sulfate and nitrate aerosols, for example.

A proportion of the emitted sulfur dioxide will be removed continuously by dry deposition, except during rainfall. Oxidation occurs, particularly in sunlight, to sulfuric acid, aerosol or sulfate, by reactions in the gas phase, in the liquid phase or on the surface of solids[52]. Husar *et al.*[53] report oxidation rates of up to 4% in plumes.

Removal from a plume may occur through dry (adsorption directly on a surface or uptake by plants), wet (rain, sleet, snow, hail) or occult (impaction of cloud or droplets on to vegetation or other surfaces) deposition. The term 'acid deposition' is used to encompass all these processes[11]. Garland[54] and Irwin and Williams[55] review deposition processes, and Hicks[56] discusses measurement methods for dry deposition. Factors affecting the dry deposition process include the characteristics of the airborne substances, meteorology–surface interactions and surface properties[57,58].

Dry deposition can be estimated from the product of ground level concentration and the deposition velocity of the species in question[51]. This latter quantity is defined as the flux to a surface per unit concentration in air. Dry deposition is thus assumed to be proportional to sulfur dioxide concentration[51]. Measurements[54,56,59–62] have shown that deposition velocities vary from surface to surface, seasonally[63,64] and diurnally[65].

Removal by dry deposition is more important for plumes emanating from low-level sources, but such emissions do become well mixed in the boundary layer within a few hours, and the major part of the emission is transported downwind in the same manner as for high-level sources[51]. Fisher[66] estimates that the concentration at long range from a low-level source is on average only 10–20% less than that from a high-level emission of the same initial concentration.

However, if a plume is subjected to rainfall, removal of precursors such as sulfur dioxide will occur; the amount removed depends inter alia on the amount of sulfur dioxide converted to sulfate. Rates of removal in wet deposition are greater than for dry deposition, the concentration of sulfur in rainwater being determined by the solubility of sulfur dioxide and the acidity of the rainwater. The process is self-limiting: as sulfur dioxide dissolves, it increases the acidity of rainwater, thus limiting further dissolution[51]. As sulfate forms a cloud condensation nucleus, wet deposition is a significant removal mechanism.

Gas-phase oxidation of sulfur dioxide by hydroxyl radicals to sulfur trioxide and then to sulfuric acid or ammonium sulfate is important; however, conversion is slow and amounts to only 1–4%[51]. Aqueous-phase

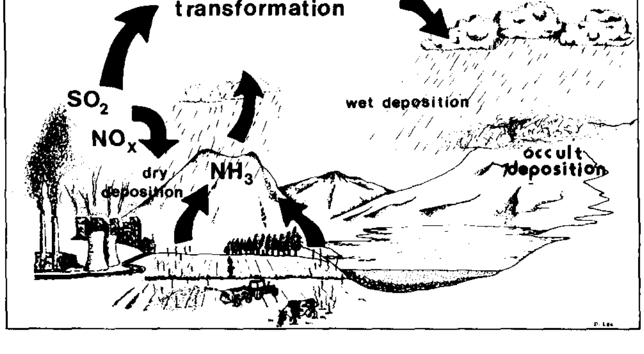

Figure 5 Schematic representation of emission, transport and deposition processes

reactions involving hydrogen peroxide and ozone are important[67]; their relative importance depends on their concentration and the time scale of removal. Oxidation by hydrogen peroxide is rapid and is limited by the availability of the oxidant. Oxidation by ozone is slower, owing to its low solubility, and the reaction rate is inversely related to acidity[51]. The extent to which this reaction controls the rate of removal of sulfur dioxide is determined by the amount of sulfur dioxide present, the duration of rainfall and the presence of species such as ammonia that affect the acidity of water[68].

Nitric oxide is a primary emission but is not removed to any degree as dry deposition; it is oxidized to nitrogen dioxide, nitrate aerosol or nitric acid. Little aqueous-phase oxidation is likely, owing to the low solubilities of nitric oxide and nitrogen dioxide. Oxidation degrees between 0.2 and 12% are reported[68]. Irwin and Williams[55] suggest that the conversion of nitrogen dioxide to peroxyacetyl nitrate and subsequent reformation of nitrogen dioxide far downwind can be an important reservoir for acidic oxidized nitrogen species. Nitric acid is volatile and will exist in the gas phase in the atmosphere, unlike sulfuric acid, which will be present as fine particles ($<0.2\ \mu m$)[69]. Consequently sulfuric acid particles may act as condensation nuclei or reduce visibility.

Both acids will react with alkaline materials in the atmosphere, such as calcium and magnesium compounds and ammonia[11,59], for example to form ammonium sulfate or ammonium nitrate. However nitric acid can volatilize from the latter[68].

The important oxidizing species, for example ozone, occur naturally, but their concentrations in the troposphere may be increased by anthropogenic emissions[51]. If there are no competing species, nitric oxide will react with ozone to form nitrogen dioxide, which in the presence of sunlight will decompose to NO and ozone. The reactions are fast and a photostationary state is quickly reached, so that no more ozone than originally present is produced[51]. However, in the presence of certain volatile organic compounds, nitrogen dioxide can be produced without consumption of ozone, and decomposition of nitrogen dioxide then provides a mechanism for ozone production[51,55]. Hydroxyl radicals are formed from the photolysis of ozone in presence of water or the photolysis of carbonyl compounds in the presence of NO[51].

In occult deposition, cloud and fog water droplets are deposited on surfaces by turbulent impact[70]. Recent studies[59,71] have shown that fog and cloud water may have much higher ionic concentrations than that of precipitation. Depending on location, occult deposition may represent a significant fraction of the total hydrological input to an area, particularly to tops of hills that are in cloud for much of the year. Dollard *et al.*[72] have shown that as much as 20% of the wet deposition of nitrate, non-marine sulfate and hydrogen ion input may come by this pathway at Great Dun Fell in north-west England. However, Irwin and Williams[55] consider occult deposition to be of minor significance in the sulfur budget as a whole. Great Dun Fell is also the site for studies of the seeder–feeder process of orographic rainfall enhancement mechanisms. This mechanism is the most important orographic enhancement operating in the UK and involves the scavenging of cloud formed as air rises over hills (feeder cloud) by precipitation originating from higher-level frontal cloud (seeder cloud). The process modifies the intensity and the chemical composition of rainfall[73,74]. Equally large concentrations of major ions are observed in cloud water at other sites[75].

DEPOSITION MONITORING

Monitoring methods

Wet deposition of acidity and other ions in precipitation can be monitored directly by collection of rainfall. Collectors are carefully designed to minimize the effect of the sampler on a sample[76]. Certain collectors are designed to open only during periods of precipitation – the so-called wet-only collectors. Deposition to collocated wet-only and bulk precipitation collectors is discussed by Stensland[77], Lee *et al.*[78] and Lee and Longhurst[79]. Whereas wet-only collectors are valuable in areas where wind-blown dust is a significant contributor to deposition, bulk collectors give valuable data if carefully sited, maintained and operated[59].

As mentioned above, dry deposition is calculated from the product of near-surface concentration and an appropriate deposition velocity[55]. The collection of occult precipitation is discussed by Unsworth and Fowler[75].

In most precipitation chemistry networks, hydrogen, ammonium, potassium, sodium, calcium, magnesium, zinc, chloride, nitrate, sulfate and carbonate are routinely monitored[11,78].

The natural acidity of precipitation is thought[11] to be due to the dissociation of carbon dioxide in rainwater, giving rise to a weakly acidic solution of pH ~5.6. Some workers[80] consider that natural fluctuations in the sulfur emission cycle can depress the pH of rain to 5.0. As mentioned above, the various ions routinely collected have different sources, including combustion, agriculture, terrestrial dusts and sea salts. This last source includes calcium, sulfate, sodium and chloride ions, and the marine contribution must be calculated to determine the anthropogenic input to precipitation composition. This is done by assuming that the sodium in bulk precipitation is of marine origin and that the ratio of sodium to, say, sulfate is the same in sea water and precipitation; the resulting calculation determines the non-marine contribution in precipitation.

Monitoring programmes and patterns of deposition

Measurement of precipitation chemistry is routinely carried out by a number of agencies on a regional or continental scale including, for example, the European Air Chemistry Network (EACN)[81–85], the European Monitoring and Evaluation Programme (EMEP)[86–93] and the World Meteorological Organization[94]. Patterns of precipitation chemistry show considerable year-to-year variation. Intra-network comparisons are validated by common operating protocols, identical collectors and interlaboratory comparison exercises. Lack of these controls on quality mitigates against inter-network comparisons.

In the current phase of EMEP, 92 monitoring stations[95] measure precipitation and acid deposition precursors according to a rigorous protocol that ensures comparability of data. Gaseous sulfur dioxide, nitrogen dioxide, ozone, nitric acid and ammonia, particulate sulfate, nitrate and ammonium, and precipitation amount, concentration of nine ions and conductivity are all regularly determined. In the period 1984–1986, maximum concentrations of aerosol sulfate and non-marine sulfate in EMEP precipitation were found in central–eastern Europe, with a smaller maximum of aerosol sulfate in western Europe.

In the second phase of EMEP the distribution of pH across Europe was calculated according to data supplied by participating nations for the period 1978–1982[84]. The resulting spatial pattern indicates that most of the UK (excluding Northern Ireland, north-west Scotland and south-west Cornwall), southern Scandinavia, northern France, northern Austria, Switzerland and north-eastern Europe is bounded by the pH 4.5 isoline. The most acidic precipitation occurs in central Europe, bounded by the pH 4.1 isoline, encompassing northern Belgium, the former Western Germany, the former DDR, eastern Czechoslovakia and Poland. These patterns cannot be considered robust. They are strongly influenced by changes in emissions and in meteorological conditions.

Precipitation chemistry measurements are also made on both national and smaller scales. For example, measurements have been reported for Fenno-Scandinavia[95], the UK[96,97], the USA[98–101], Alberta[102], northern Britain[103], Ontario[104], eastern Canada[105], northern Spain[106], Greece[107], Massachusetts[108], West Africa[109] and Hungary[110]. Measurements of precipitation chemistry have also been made in remote areas of the world[111], where the contribution of organic acids to the measured hydrogen ion concentration can be significant.

A comparison of North American and European precipitation chemistry measurements has been attempted by the RMCC[112], with the conclusion that sulfate and nitrate concentrations and wet deposition rates are greater in central Europe, but levels are comparable for wet-deposited sulfur in North America, east of the Mississippi, and southern Scandinavia at ∼20–40 kg of sulfate per hectare per annum.

Monitoring of acid deposition on a systematic basis did not begin in the UK until the beginning of 1986[10,96]. Data from the first three years' operation of the monitoring networks, established by Warren Spring Laboratory on behalf of the DOE, have been published[29], and broadly confirm previous observations at a smaller number of monitoring stations[11,59]. The concentration of H$^+$ increased from west to east, with a maximum between the Humber and the Wash; maximum concentrations of non-marine sulfate, nitrate and ammonium occurred in the east, particularly south of the Wash; and the greatest deposition occurred in areas of highest rainfall, particularly parts of Highland Scotland, North Wales, Cumbria and the Pennines, where acid deposition is comparable with that in areas of southern Scandinavia. The largest concentrations of non-marine sulfate occurred in low-rainfall easterly air flows, and the highest deposition in high-rainfall westerly air flow. The 1988 pattern of concentrations for selected ions is shown in *Figure 6*.

This broad pattern of increasing acidity concentration gradient from west to east in the UK is consistent with the hydrogen ion pattern described by the EMEP[84]. The

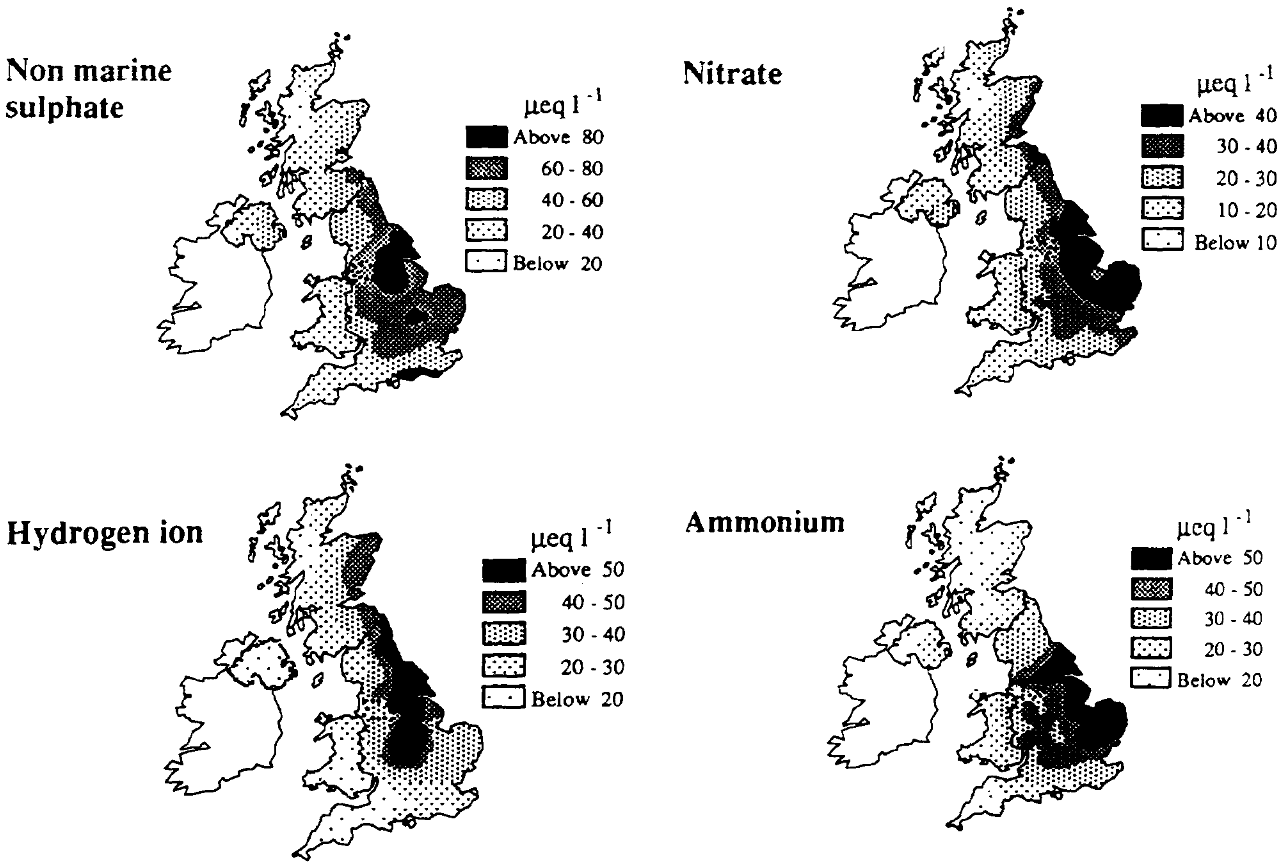

Figure 6 Concentrations of non-marine sulfate, nitrate, hydrogen and ammonium ions in UK rainfall in 1988 (interpolated data from the secondary national network). Original data supplied by Warren Spring Laboratory on behalf of the Department of the Environment

concentration gradients of other ions are also consistent with European-scale patterns.

The concentrations of ions in precipitation show considerable variation with season, with location of measurement, with type of precipitation and even within precipitation events[11,29,59]. Irwin[10] drew attention to the changing spatial importance of the ratio of nitrate to non-marine sulfate in European precipitation and cited the identification by Nodop[113] of the highest ratio in places of high emission density. Temporal changes in the importance of nitrate in precipitation have been identified in Sweden[113], the UK[114] and the USA[115]. This changing pattern is consistent with observed decreases in sulfur emissions and increases in nitrogen oxides emissions.

Distinct temporal patterns have been observed in emissions of acid deposition precursors and the composition of their reaction products in precipitation and deposition chemistry. In the UK and Europe, emissions of precursor gases are at a maximum in winter, and maximum concentrations of sulfate and nitrate are observed in precipitation in spring and early summer[11,59]. As many different ions exhibit distinct seasonal patterns despite different origins, a common meteorological factor such as wind direction[59] seems to be involved.

Smith and Hunt[116] identified the episodic nature of much of the deposition experienced in certain areas of western Europe, where a large proportion of the annual deposition can occur on a relatively small number of days. Such events are associated with air masses whose trajectories indicate passage over major emission source areas. Obviously such deposition has significant implications for receptor environments.

In certain areas of the world a considerable proportion of annual precipitation occurs as snowfall. Rain and snow have different mechanisms of pollutant removal, and their composition can be quite different[117]. Data presented[118] for the USA suggest that the concentration of nitrate is higher in snow than in rain; this is consistent with findings[119] in the Scottish Cairngorms. Lower concentrations of sulfate were found in snow samples than in rain[118,119]. Snowmelt in spring gives rise to an acid flush, and this has important implications for aquatic ecosystems.

The importance of reliable, quality-assured monitoring cannot be overemphasized. It is vital in defining patterns and trends and in providing data from which environmental effects can be assessed.

EFFECTS ON FRESHWATER SYSTEMS

This section deals briefly with general issues of freshwater acidification and more specifically with short-term acid pulses, which can have fatal effects on aquatic biota. In addition, the evidence of lake and reservoir acidification is reviewed to provide an insight into the recent time scale for acidification of some of these waters.

Concern about the effects of acid deposition on aquatic ecosystems arose for two reasons: its potential impact on aquatic biota[120–122] and on human health[123,124].

Recent studies show that acidification of surface waters may have serious implications for aquatic biota and that it may affect species diversity[125–127]. In particular, it appears to have a major effect on fish populations[128–132]. It is thought that the elevated concentration of aluminium in surface waters associated with low pH is the main toxic agent. Aluminium solubility is largely determined by pH, and various hydroxy-aluminium complexes are formed, depending on the level of acidity. Other aluminium complexes may also be formed with fluoride, sulfate and organic solutes present in surface waters. In laboratory tests, survival of brown trout fry has been shown to be reduced in a concentration of aluminium of $250\,\mu g\,l^{-1}$, especially if the associated calcium concentration is low. Salmonids are particularly affected and it is thought that mortality rates substantially increase during episodic acid events.

Human health can also be potentially affected by acidification of aquatic systems. Concern has been expressed over the mobilization of metals in domestic water supply systems[133], caused by the leaching of toxic metals (e.g. lead, cadmium, mercury, aluminium and copper) from catchments and from the corrosion of water supply pipes. In particular there has been recent concern over levels of aluminium in drinking water, and research has indicated that aluminium may be implicated in the development of Alzheimer's disease[134].

Up to the early 1970s, many researchers were of the opinion that acid deposition was the main factor responsible for the acid status of surface waters. In particular, the disappearance of fish from lakes in the southernmost counties of Norway and Sweden was seen to be associated with the increase in European emissions of sulfur. This led to the conclusion that acidity in precipitation was the cause of declining fish stocks. However, more recently a number of researchers have suggested that this status may be regulated by other processes[135–138], and believe that natural processes such as the internal generation of organic acids are primarily responsible for freshwater acidification, although it is more likely that both acid deposition and natural processes contribute to the acidification of surface waters[139]. A number of catchment studies have been undertaken, particularly in Europe and North America, to investigate the complex relation between the processes involved in the acidification of vulnerable surface waters[140–146].

Vulnerability of surface water to acidification

Acid deposition falling directly on to the surface of a free-flowing stream (see *Figure 7*) is unlikely to contribute to acidification. On the other hand, acid deposition falling directly on to standing water can potentially contribute to acidification. However, this largely depends on, among other things, the alkalinity of the catchment and the ratio of the drainage area to lake area[147]. Catchment studies in the Adirondack Mountains, for example, have shown that the vulnerability of both lake and stream waters to acidification by atmospheric deposition depends on the biogeochemistry and hydrology of the whole catchment. Precipitation follows various pathways through a terrestrial ecosystem before it reaches a stream or a lake, and it is important to discuss the respective roles of these factors in contributing to, or resisting, acidification[148].

Vegetation and land use. It has been shown by many researchers that forest cover modifies the quality of intercepted bulk deposition (both wet and dry deposition) as it moves through the forest canopy[149,150]. Trees, in particular coniferous trees, are very efficient scavengers of dry-deposited gases and aerosols, including oxidized

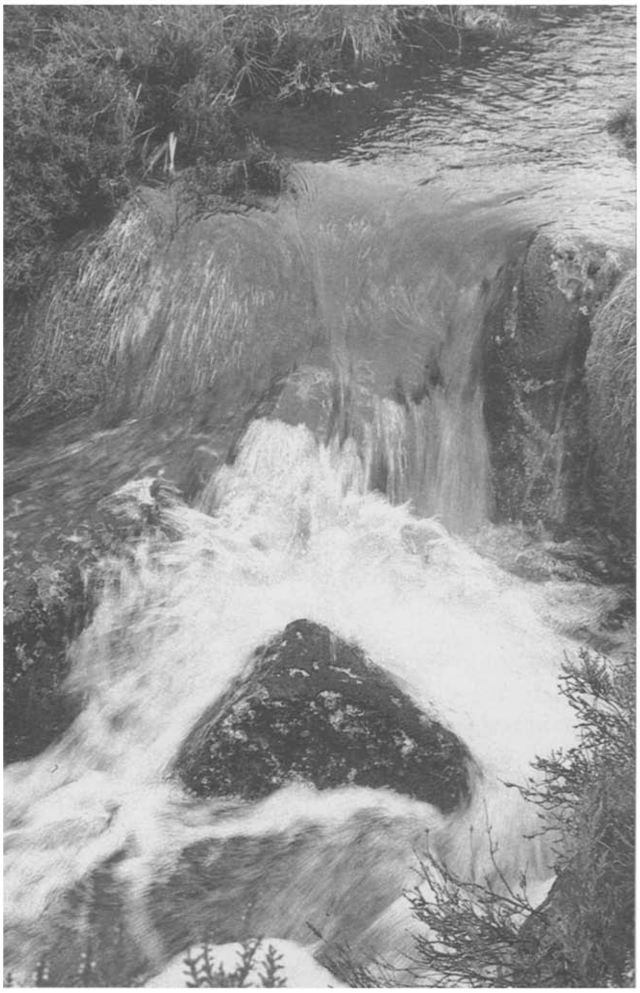

Figure 7 Upland stream, English Lake District

and reduced nitrogen species, sulfur dioxide and sulfate. Therefore the chemistry of throughfall is altered owing to the leaching of these deposits and of excreted metabolites on the leaf surface. In addition, evapotranspirational losses associated with forest land use have been shown to cause increased concentrations of, for example, sulfate in surface waters in the Derbyshire Peak District[151].

Rovenqvist[135,136] and Krug and Frink[137,138] have suggested that vegetation in catchments can have a profound influence on surface water quality. In particular, soils under coniferous forests can become particularly acidic owing to ion-exchange processes and leaching. Changed land use practices can also influence the susceptibility of surface water to acidification. For example, timber harvesting appears to decrease, and reforestation to increase, surface water acidification[152,153].

Bedrock geology. Bedrock geology is used most frequently to assess the potential susceptibility of surface waters to acidic inputs[154]. Areas underlain by granites and related igneous rocks and non-calcareous sandstones offer little buffering, because of their slow weathering properties. Catchments located in such geologies generally have low alkalinities ($<200\ \mu g\ l^{-1}$)[155,156] and are regarded as susceptible to acidification. However, areas underlain by bedrocks containing calcite or other carbonate material have the potential for considerable neutralizing or buffering capacity[157,158]. Water draining these catchments tends to have enriched concentrations of base cations; consequently there is little if any evidence to show that such catchments are vulernable to acidification processes[151].

Soil type and depth. Except when precipitation falls directly on to the surface water in a catchment or on to exposed rock, precipitation either flows across or percolates through soils before entering streams or lakes. The movement of precipitation through soil horizons has the effect of changing its chemistry and ultimately can potentially influence surface water chemistry. In particular, throughflow occurring in upper soil horizons that are naturally acidic has the potential to wash out internally generated acidity, although most of the acid input from this process to a lake or stream probably occurs during episodic events[159], as described in more detail later. Soil depth can also be important in determining the quality of surface water; this has been shown, for example, to be an important factor in the observed difference between the quality of water in two Adirondack lakes. The Woods Lake and Panther Lake catchment areas are practically homogeneous and ~30 km apart. Each receives the same amount and quality of precipitation, but Woods Lake is considerably more acidic. It has been shown that water reaching Woods Lake flows principally through shallow acid organic soil horizons, whereas water reaching Panther Lake flows through deeper, more acid-neutralizing, inorganic horizons[160].

Episodic acidification

This section reviews in some detail selected research on episodic acidification events in surface streams.

Significant short-term pulses or flushes of acid in small upland catchments (see *Figure 8*) can occur on an hourly time scale and relate to hydrological changes in the catchment[161,162]. These short-term acid flushes have been linked to detrimental impacts on fisheries in Scandinavia[163] and north-eastern USA[164].

The Acid Waters Review Group (AWRG) has outlined the hydrological pathways by which precipitation reaches surface waters and the chemical reactions that occur between water, soil and rock[165,166]. The three main hydrological pathways are overland flow, lateral flow through the soil horizons (throughflow) and flow through the bedrock (groundwater flow). Stream flow may be divided into two primary components: storm flow, which reaches the stream rapidly and comprises overland flow and rapid throughflow, and base flow, which comprises slow (unsaturated) throughflow and groundwater flow[167]. In many upland catchments, groundwater flow and throughflow from the lower soil horizons are enriched in base cations, derived from mineral weathering and cation exchange, which add to the alkalinity or acid-neutralizing capacity (ANC) of surface waters. During periods of intensive precipitation, the moisture contents of soils increase, and water will flow laterally as throughflow. Throughflow in the upper soil horizons which are naturally acidic has the potential to wash out internally generated Al and H[168]. In addition, rapid throughflow prevents slow neutralization reactions with weathering rock in the lower, less acid subsoil horizons. Also, cation exchange reactions between atmospheric

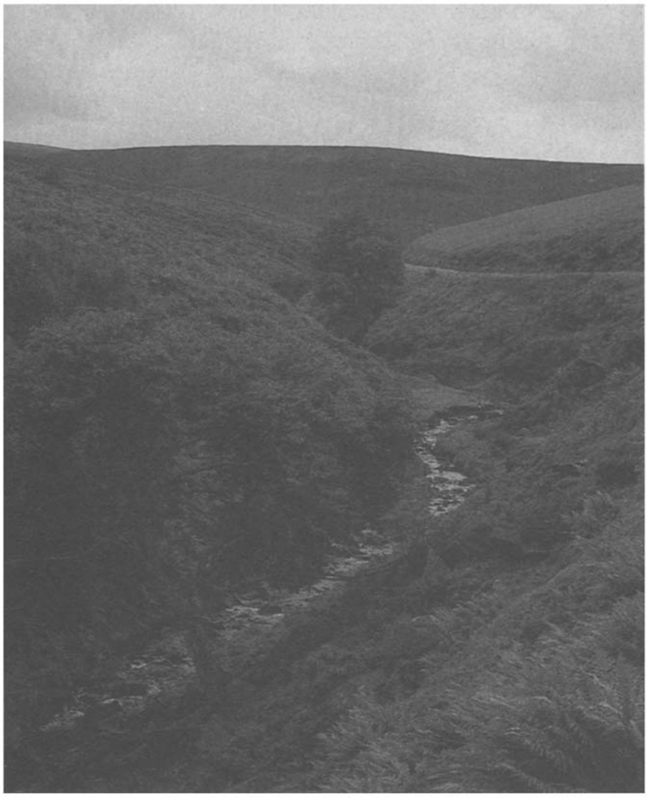

Figure 8 Upland stream, English southern Pennines

deposition and soil can occur rapidly on the humus and peat colloid surfaces within organic soils, and hence the concentration of acidifying ions in waters draining them may increase. When catchment soils become saturated to the surface, saturated overland flow occurs. Because the water does not infiltrate soil horizons, its chemistry is likely to be similar to that of precipitation. The AWRG has suggested[165,166] that these processes determine the composition of surface waters and the extent to which wet and dry deposition is modified through catchments. In particular, the reports emphasize the importance of catchment size and precipitation amount, particularly as they contribute to sudden episodes of acidified surface water.

The importance of hydrological processes in regulating the solution chemistry of acidic surface waters has also been stressed by many other workers. For example, studies from the Adirondack Mountains of New York, USA[160,169,170], Hubbard Brook, USA[142,171], Birkenes Creek, southern Norway[135], north-east Scotland[168], mid-Wales[172], and Cumbria, UK[173] have shown the importance of hydrological pathways in episodic surface water acidification. In many upland catchments, base flow is supplied primarily from mineral soils rather than bedrock. Such water is often enriched in base cations, even on acidification-susceptible parent materials such as granite or granite tills. However, as the water flux to the soils increases, throughflow in the O and A horizons becomes increasingly important. Such surface horizons tend to be naturally acidic and contain internally generated H and Al, which are flushed out during storm events.

Short-term elevated H and Al concentrations in surface waters have been investigated by several workers in a range of environments and it is generally agreed that they are attributable to heavy rainfall, snowmelt or a combination of the two[174]. A close relation of H and Al to discharge was observed[175] at Birkenes Creek, Norway; when soil water content was high, the hydrological response occurred within a few hours of the start of a precipitation event. However, Lawrence *et al.*[176] in a study of the hydrological control of aluminium and hydrogen in an acidic headwater stream at Hubbard Brook, suggested that the relation between surface water chemistry and hydrologic flow paths is probably more complex than previously thought. They observed that spatial variation in catchment biogeochemistry complicated the relation between discharge and the concentration of both Al and H. They concluded that the interrelation between spatial and temporal variation of catchment processes makes an evaluation of the factors controlling surface water chemistry extremely complex.

Precipitation deposited as snow has been shown to affect surface water quality during snow melts[177–181]. Johannessen *et al.*[177] observed that during and after snowmelt the concentration of ionic species in stream water differed from that at base flow in 37 Norwegian catchments. They suggested three possible stages of snowmelt. First, expulsion of stored soil water by piston flow at the onset of snowmelt may result in an increase in the concentrations of Ca, Mg and HCO_3, which are mainly supplied by weathering. This process is followed by dilution of these ions as a result of mixing with snowmelt water. However, there is a concomitant increase in the concentration of the major snowmelt ions (SO_4, NO_3, Cl, H, Na and NH_4). The remaining snow contains proportionately less of the snowmelt ions, because of preferential elution. Hence, in the third stage the snowmelt becomes more dilute with respect to these ions, and a reduction in the concentration of the snowmelt ions in the stream water is observed. Johannessen *et al.*[177] and Christophersen and Seip[175] have postulated that because the initial wave of meltwater is enriched with ions such as H and SO_4, it could be the cause of acid flushes. Abrahams *et al.*[181] monitored stream water chemistry during snowmelt in an upland catchment in the Cairngorms and discussed their results using the model of stream water chemistry suggested by Johannessen *et al.*[177] They acknowledged that interpretation of this model is not necessarily straightforward, because of the effects of soils in either promoting or retarding the supply of ions to the stream. However, they suggested that acid flush events during snowmelt are associated with both an increase in stream discharge and raised concentrations of major ions. They postulated that the hydrological pathways of meltwaters operative at the time of snowmelt largely determine stream water chemistry.

Like precipitation quantity, precipitation quality can also affect surface water acidification. Christophersen *et al.*[175,182,183] used simple hydrological reservoir models to describe daily and seasonal variations in surface water chemistry. They described the importance of atmospherically derived SO_4 for the transport of cations including H and Al through soil in the Birkenes and Storgama (Norway) catchments. This mobile-anion effect drives cation exchange[184,185] within the soils of the catchment. This process is very rapid compared with most geochemical reactions and thus surface water chemistry can be very sensitive to short-term changes in the composition of deposition reaching the land surface,

Figure 9 Acidified lake, south-west Sweden

provided that flow pathways are suitable. Langan[186,187] observed the episodic acidification of streams at Loch Dee, south-west Scotland; the apparent mechanism for these episodes of surface water acidification is one of ion exchange in the catchment soils following precipitation inputs laden with sea salts. The percolation of waters enriched with sodium chloride results in the exchange of Na for H on the humus and peat colloid surfaces within both peat and peaty podzol soils. Other workers who have investigated the role of sea-salts include Skartveit[188], who studied a remote catchment on the western coast of Norway and identified similar processes as the cause of short-term episodic acidity.

Evidence of acidification

There is considerable difficulty in establishing trends of acidification in streams and rivers. However, historic pH levels for lakes and reservoirs can be reconstructed from records of fossil diatom assemblages present in lake sediments[189–196]. Diatoms are microscopic algae which live free-floating in lake water. They have hard skeletons made of silica which are characteristic of each species and are relatively resistant to decay. Different diatom species have different pH preferences. Dead diatoms sink to the bottom of the lake, where the remains accumulate as sediment. Thus if a core of sediment is taken, the stratification of the diatoms will detail the acid history of the water. If sediment layers can be dated, then acidification can be associated with known historical events. Diatom analysis is used widely in Europe and North America[197]. In the UK Battarbee *et al.*[198] have systematically used diatom analysis to provide evidence of acidification in a number of regions including Galloway, Cumbria and Wales. Lake sediments also contain a record of land use change (pollen analysis), air pollution (trace metal and soot analysis) and other changes in the aquatic ecosystem (from analysis of algal, cladoceran and chironomid remains)[165].

In almost all the waters examined in Scotland, England and Wales, analysis has shown stable diatom floras up to 1850. After this date, it has been suggested that atmospheric pollution from industrial sources has been responsible for subsequent acidification and diatom changes. Further evidence for the acidification of these waters is given by increases in both heavy metals and soot contained in sediment cores.

Data from the six lakes studied in Galloway[198] (Loch Enoch, Loch Valley, Round Loch of Glenhead, Loch Dee, Loch Skerrow and Loch Grannoch) were consistent with a decline in pH of between 0.5 and 1.2 within the last 150 years. The onset of recent acidification varied from lake to lake from about 1850 to 1925, although in general, acidification in the nineteenth century was slow and an acceleration occurred in the twentieth century between 1930 and 1950. *Figure 9* shows an acidified lake in south-west Sweden.

THE EFFECTS OF AIR POLLUTION AND ACID DEPOSITION ON FORESTS, AND THE PHENOMENON OF 'FOREST DECLINE' IN EUROPE

Forest decline has captured the imagination of the public and scientists alike, and also of the popular press. The idea that vast tracts of forest around Europe and North America are dying as a result of 'acid rain' is dramatic, or perhaps more realistically, melodramatic.

The problem is undoubtedly real, that there is an unprecedented decline of European and North American forest ecosystems. The question remains, however, whether air pollution is the cause of the decline in these two continents. Public attention was drawn to the problem in the former Western Germany by the press, particularly the weekly magazine *Der Spiegel*[199], in which Prof. B. Ulrich warned that Germany's forests were suffering a widespread decline. The terminology of forest decline needs some explanation, as some confusion has arisen from the different terms used. There is a vast amount of literature referring to the direct effects of air pollution on forests and forest soils[200], which in the older German literature is referred to as *Rauchschaden* (smoke damage). The German expression for what we understand here to be forest decline is *neuartige Waldschäden* (new types of forest damage – note the plural), more popularly referred to as *Waldsterben* (forest death). The more correct phrase, *neuartige Waldschäden*, gives some clue as to a possible definition of forest decline, that the nature of the damage is essentially new. A tentative definition that will be used here is that forest decline refers to the unprecedented widespread decline of forest ecosystems in both Europe and North America. The inclusion of the word 'ecosystems' is also important, as decline phenomena of individual tree species are well known[201].

Forest decline was first noticed in the south of the former Western Germany. Silver fir (*Abies alba*) has suffered sporadic declines over the years[202,203], which can be traced back as far as around 1500[203]. An outbreak of silver fir decline, or *Tannensterben*, occurred in the early 1970s, but by about 1980 similar symptoms of defoliation were noticed in south-west Germany on Norway spruce (*Picea abies*)[204], a forest tree of great economic importance. Subsequently, other forest trees such as Scots pine (*Pinus sylvestris*), European beech (*Fagus sylvatica*), ash (*Fraxinus excelsior*) and oak (*Quercus* spp.) were affected by the decline. Despite the fact that many different tree species are affected, research has concentrated on Norway spruce, beech, and to a lesser extent Scots pine, because of the economic importance of these species.

Forest damage surveys

One of the first problems encountered by scientists and foresters was the measurement of forest decline. The first survey of forest damages was conducted in the former Western Germany in 1982. This took the form of a questionnaire in all the states, with the exception of Baden-Württemberg and Bavaria, where a survey proper was conducted. This survey consisted of assessing the amount of defoliation in individual trees in four classes. From 1983 onwards, all the (former) Federal states conducted surveys similar to those conducted in Baden-Württemberg and Bavaria, except that defoliation was now classified into five intervals; 0–10%, 11–25%, 26–60%, >60% and 100%. This system is now adopted in the ECE survey, along with a four-class discoloration classification which is optionally combined with the defoliation system[205].

There are three organizational levels of survey. Individual countries conduct their own surveys, which may differ in methodology. The EC countries are required to conduct surveys as a direct result of European Community legislation[206]. Finally, 35 countries, signatories to the International Cooperative Programme on the Assessment and Monitoring of Air Pollution Effects on Forests, launched in 1985, participate in a monitoring exercise for which the United Nations Economic Commission for Europe carries out secretariat functions. The former Federal Republic of Germany acts as lead country for the Programme Task Force on Assessment and Monitoring of Air Pollution Effects on Forests, which is responsible for detailed planning and coordination of the programme. The Programme Coordinating Centre West (PCC-W) is the Federal Research Centre for Forestry and Forest Products, Hamburg, and the Programme Coordinating Centre East (PCC-E) is the Institute for the Environment and Utilization of Natural Resources, Ústí and Labem, Czech Republic. The PCC-W is responsible for the Western countries and the PCC-E for the Eastern countries.

As previously mentioned, forest damage surveys are principally conducted on the basis of defoliation. The ECE has published a manual[207] for use in its surveys; individual countries may use standard reference diagrams or photographs. One of the earliest set of diagrams was devised by the Federal Forestry Research Institute of Vienna[208], some of which were subsequently incorporated in the ECE manual. The best photographic document, which is used in many countries, is produced by the Birmensdorf Federal Forestry Research Institute of Switzerland[209] as part of their coordinated research programme, SANASILVA. Much effort has been put into the harmonization of forest damage surveys, although much work is still required.

Forest damage surveys are fraught with inherent problems, such as internal consistency, observer error (which is affected by many factors) and inappropriate sampling design[210]. Samples are usually obtained from the intersections of a 16 × 16 km grid. However, this varies from country to country and from state to state, depending on time and material resources, and on forest density. Considerable effort is put into training of foresters in some countries, in an attempt to maintain objectivity[211].

To remove some of the errors inherent in terrestrial surveys of forest damages, considerable efforts have been made in the field of remote sensing, using both colour infrared photography (CIR) and digital satellite images[212–215]. CIR involves taking photographs from suitable aircraft or helicopters using infrared film. Once the images are developed, a grid system is overlaid on the images and trees at grid intersections are examined. Although this might initially seem attractive, several problems exist. The photographs need to be taken under good weather conditions to obtain optimal images, and the colour quality of the images may vary between developing laboratories. Once the images are obtained, trained photo-interpreters classify the samples by species, social status, topography and damage class/type. Different types of damage can be differentiated for some tree species on the basis of CIR photography[216,217]. Maintaining consistency in classification between photo-interpreters is a problem in the same way as it is for terrestrial surveys. Satellite images may be analysed by a more objective technique, using multispectral computer scanning of the images. This may seem more attractive, but there is debate over whether this technique for surveying forest damages is sufficiently well developed to be reliable[215].

Whether surveys are made by terrestrial methods or elaborate remote sensing methods, it must be remembered that the results from any survey cannot be used to identify causes of the damages involved in forest decline. However, surveys are a valuable aid in the development and testing of hypotheses in the determination of causal factors[218,219].

Most European countries report the results of forest damage surveys every year (e.g. Refs 220–224) and, as previously mentioned, these are compiled to form the basis of a European survey by the UNECE[205,225]. The validity of comparing data from different countries has been seriously questioned, because of the different data collection methods and the wide geographical area concerned. A more fundamental point is that although some symptoms are 'new', they are without exception non-specific with regard to air pollutants[210,226]. For example, defoliation is universally assessed as a 'symptom' but may be the result of many biotic and abiotic factors, not necessarily related to air pollution at all. Misplaced emphasis on the results of forest damage surveys can bring about sensationalization of the subject by the media, and misunderstanding between scientists[227–229].

Symptomatology

Despite the reported prevalence of forest decline across central and northern Europe, the actual symptomatology of the disease(s) has been the subject of remarkably little research. As already mentioned, the presence or absence of forest decline is 'measured' in terms of leaf loss and/or yellowing, the two main symptoms of decline. None of the symptoms of forest decline is specific to air pollution, and leaf loss in coniferous species may be caused by a number of abiotic factors including light regime, temperature, water status, nutrition and frost damage[230]. Yellowing of needles generally occurs only at higher elevations and is often associated with nutritional deficiencies[231]. However, yellowing of needles may also be the result of abiotic factors other than air pollution, including nutrition and metal toxicity, water balance, temperature and frost stress, wind and salt damage[232].

The essential problem faced by foresters and scientists alike, when forest decline first became an issue, was the lack of knowledge concerning tree morphology – in other words, what does a normal tree look like, if indeed there is such a thing? Different branching patterns for Norway spruce had been documented in earlier literature[233,234], but less was known about important broad-leaved species such as beech and oak.

The German Federal Advisory Council on Forest Decline/Air Pollution, in its second report[231], defined five different disease types that it had identified for Norway spruce:

Type 1: the phenomenon of needle yellowing at higher elevations, prevalent in central and southern Germany. Older needles become chlorotic and are subsequently shed. This disease is associated with nutrient deficiencies, particularly of magnesium.

Type 2: the thinning of crowns at medium and high elevations, where much of the leaf biomass may be lost but little chlorosis occurs. This disease type is prevalent in central and northern Germany.

Type 3: the so-called needle reddening disease, which is a chlorosis of older needles which subsequently turn red and are shed. This occurs in older stands of Norway spruce, mainly in a limited geographical area, the Bavarian Alps.

Type 4: the chlorosis of needles in the Bavarian Alps, similar to Type 1, except that it occurs in the youngest needles and is associated with potassium and/or manganese deficiencies.

Type 5: the thinning of crowns and loss of increment, occurring mostly in the northern German plain.

This differentiation is intended to discriminate between different diseases, where different stressors, or combinations of stressors, are at work but under the umbrella term of 'forest decline'.

One of the properties of forest decline is that it is not only degenerative, i.e. there is a loss of foliage and a loss of increment, but that it also produces growth modifications.

Growth modification of crown architecture has also been studied for Norway spruce and to a lesser extent for Scots pine[235–240]. The degenerative symptoms of decline are similar to some of the natural processes of ageing, and discrimination between the two can be problematic[235]. Norway spruce exhibits three basic branching types – brush, comb and plate – which are genetically determined[233,234], although it is possible that other intermediate types also exist[241]. These three branching types centre around the disposition of the secondary and higher-order branches. Comb spruce shows the greatest degree of branching plasticity[239] and can change its form to other branching types in response to the environment.

Extensive research by Roloff[242,243], of the University of Göttingen, showed that the visible decline symptoms in beech could be accounted for by a change in the crown architecture, from modified branching patterns. However, the cause of the change is still unknown. Roloff has recently used his model of branching architecture to model the crown architecture of 18 other broad-leaved tree species in relation to forest decline[244].

It is important to realize that symptoms of decline exist other than those that are visible. Annual increment may be reduced or increased[245,246] (see below, on nitrogen deposition). The formation of wetwood, or pathological wet-core, occurs in the trunk of silver fir and may disrupt transpiration[247,248]. Mycorrhizal associations (symbiotic associations between certain fungi and roots of some higher plants) may be reduced[249,250]. Nutrients may be leached from the foliage[251,252], and the cuticular wax of needles and stomatal plugs eroded or destroyed[253,254].

The definition of symptomatology of forest decline raises enormous problems. Symptomatology can be considered on the macroscale, i.e. the appearance of the tree, and also the microscale, i.e. physiological response of the tree, or on combinations of the two. At present, our knowledge is rather sparse on the subject overall, although it is good in parts. The symptomatology of forest decline was the subject of an entire symposium in 1988, and the reader is referred to this for a more comprehensive discussion[255]. Two recent publications[256,257] use high-quality colour photographs to illustrate forest decline symptomatology.

Hypotheses to explain forest decline

(*1*) *Soil acidification/aluminium toxicity.* This hypothesis was first proposed by Ulrich *et al.*[258] of the University of Göttingen, based on Ulrich's studies of nutrient cycling in the Solling ecosystem (northern Germany), from which he predicted a large-scale decline of European forests[259]. The hypothesis suggests that the natural process of acidification of forest soils is rapidly accelerated by the input of anthropogenic acids from the atmosphere, from the combustion of fossil fuels[260,261]. The natural buffering capacity of forest soils is eventually exhausted and the soil becomes acidified. The leaching of base cations such as Ca, K, Mg and Mn means that these ions are not available to the trees, and this can result in nutrient deficiency (Mg deficiency is often associated with yellowing of needles). Furthermore, as the soil is acidified, Al is released and disrupts the uptake of Ca/Mg by replacing Ca/Mg at bonding sites on the cell walls of roots[262,263]. At higher concentrations, Al is actually toxic to roots and inhibits elongation[264,265]. Although this hypothesis has been very well researched and undoubtedly explains some of the stress on trees, it does not explain the decline of trees on calcareous soils, where different factors must be at work.

(*2*) *Complex high-elevation disease.* This hypothesis involves the combined effects of ozone gas, acid deposition and nutrient deficiencies at high elevations, with possible interactions with frost[266–270]. High tropospheric ozone concentrations can occur at higher elevations[267,271,272], particularly in summer, causing damage to cell walls and the waxy cuticles of needles. Wet deposition, particularly fog and cloud water, may leach nutrients from the damaged leaves. Chlorophyll is then partly broken down. This results in reduced assimilation and reduced root growth. Therefore fewer nutrients are taken up, the physiological activity of the tree is reduced and it becomes more sensitive to frost damages[269,273]. Although this hypothesis appears to be attractive, the evidence for it is circumstantial, and field and laboratory fumigation studies in the UK and elsewhere have failed to replicate symptoms observed in the field[274–276]. So, although ozone may be important, it will occur in phytotoxic concentrations only at certain times and places, which are highly variable and dependent on prevailing meteorology.

(*3*) *Needle reddening disease.* Reddening of needles in parts of southern Germany has been documented for the autumns of 1982, 1983 and 1984[231,277,278] in older stands of Norway spruce (>60 years). The process usually starts in late September, when older needles turn a red colour within 2 weeks. The reddening symptom was found to be independent of soil type, nutrient supply and position in the stand[278]. The reddened needles show black bars and black dots indicating infestation by the fungi *Lophodermium picea* and *Rhizosphaera kalkhoffii*[277,278]. It has been suggested that climatological factors such as frost may predispose the trees to the above pathogens, and that the pathogens are the primary cause of this type of decline. This hypothesis for this particular 'disease type' was proposed by Rehfuess and his colleagues of the University of Munich[277,278]. However, there is disagreement over whether the pathogens are the primary factors involved in this type of decline[279,280]. This hypothesis still appears to be largely speculative, and at a recent major conference[281] no new evidence was presented.

(*4*) *Excess nitrogen deposition.* 'Excess' nitrogen deposition arising from the deposition of anthropogenic pollutants may be injurious to forest trees[94]. Airborne nutrients may be absorbed by foliage as well as by the root system. In the past, most forests were limited by nitrogen availability. Now, with the increase of nitrogen deposition, some soils have become nitrogen-saturated[282,283]. The effects of this are numerous. Increased nitrogen deposition increases biomass, shown by an increase in annual increment, so that Mg/P/K may become limiting factors for tree growth[282]. The fertilization effect may delay the hardening-off process in autumn and result in frost damage[284]. Furthermore, the fertilization effect, in combination with decreased base cation availability, may lead to increased susceptibility to pathogens[285]. Increased nitrification and leaching of nitrate lead to acidification of the soil, and therefore base cation loss[286]. The balance of shoot and root growth may be disrupted by the fertilization effect of nitrogen deposition[287]. This hypothesis is quite plausible, and in conjunction with other hypotheses may go some way towards explaining forest decline. Much research effort is currently being put into this aspect of forest decline.

(*5*) *Other hypotheses.* (*a*) *Radioactivity.* It has been suggested that forest decline can be linked with emissions from nuclear installations[288,289]. This hypothesis was based upon the assertion that the incidence of damages was higher around nuclear installations; it is not widely accepted[231,290] and the evidence for it has been refuted[291–293].

(*b*) *High-frequency electromagnetic radiation.* The energy associated with electromagnetic fields from television and radio repeater stations may disrupt ion fluxes within leaves by depolarization of the cellular membranes in the presence of an electrolyte (acid rain), owing to a resonance effect[294–296]. As yet, very little attention has been paid to this hypothesis as a local stress factor on forests, and more work is required to confirm or reject it.

(*c*) *Viruses.* It has been suggested that an unidentified virus or viruses may be responsible for the decline of European forests[297–299]. However, although viruses have been identified from declining trees[300] and may be predisposing factors, no satisfactory explanation for the supposed 'virus(es) vector' has been given and it is inconsistent with the spatial development of the problem[301–303]; foliar discolorations have been demonstrated to be the results of nutrient deficiencies rather than viral infestations[304].

(*d*) *Halogenated hydrocarbons.* It has been shown that spruce trees exposed to chloroethene, mimic some of the symptoms of forest decline, i.e. the discoloration of needles[305,306]. Halocarbons are used extensively as solvents in the textile and metal industries, and may escape into the atmosphere, where they have relatively long lifetimes[307]. Halocarbons have also been found in forest soils[308]. Although this hypothesis is plausible, more measurements are needed, along with carefully planned dose–response experiments.

(*e*) The above is not an exhaustive account of all the hypotheses that exist; there have been others involving climate[203,309], silvicultural errors[231], heavy metals, photooxidants and organic chemicals[310–315]. One of the most popular but less specific hypotheses has been that of 'stress', the main proponents of which have been Schütt and his colleagues of the Institute of Forest Botany at Munich University[204,303,316,317]. This hypothesis has been summarized[318] as follows: 'Permanent exposure to low concentrations of air pollution, composed of an unknown number of phytotoxic compounds and transported over long distances, leads to reduced vigour of trees and also affects other members of the ecosystem. It is manifested in lower production or lower availability of carbohydrates and secondary substances. This again increases the predisposition to climatic extremes and pathological events'. The idea of a stress hypothesis has been criticized as being too non-specific and untestable. However, it is almost universally accepted that forest trees are stressed by various stressors, which vary in magnitude from location to location. The criticism of this hypothesis may be understandable, but the issue raised by this debate, namely that it is difficult, if not impossible, to determine cause–effect relations and interrelations remains unresolved.

Early diagnosis of forest decline

Recently, some research has been directed towards finding diagnostic tests for detection of the onset of forest decline. Ideally, such tests need to be independent of species, tree type and age, location of sample and time taken. To be indicative of the cause, the diagnostic test should also ideally depend on such factors as air pollution, soil acidification and climatic/pathogenic stress factors[319]. In reality, it is unlikely that any tests that fulfil all these ideal requirements exist, but there are some that may fulfil at least some of the criteria. In 1986, a pilot survey was conducted to investigate the utility of some diagnostic tests. Samples were taken from 12 sites along a transect from south-west Germany to north-east Scotland[320]. The tests included investigation of the surface properties of spruce and Scots pine needles[321], ethene emissions and pigment ratios[322] and buffer capacities[323]. Conifer needles are covered by a layer of wax that may degrade in response to environmental conditions; the degree of erosion may be determined by the 'wettability' of the surface. The surface properties of conifer needles have proved to be a good indicator of the exposure to the atmospheric environment, but in the 'early diagnosis' study[320] no clear relations could be demonstrated between decline symptoms and leaf surface properties[321]. Ethene emissions from plant needles are known to be a good but unspecific indicator of stress[324]. In this study, rates of ethene emission were found to be low from sites that were relatively unpolluted, and higher from those that experienced frequent ozone episodes, higher rainfall acidity and elevated levels of primary pollutants[323]. Although this study produced promising results, none of the tests was pollution-specific, and differences could be attributed to other factors such as genotype, soil type and climate. It was concluded that it was necessary to develop the most successful diagnostic tests under controlled conditions in order to minimize the variability that arises from tree age, genotype and climatic conditions, and to determine specific response(s) to individual pollutants[325].

Amelioration of declining forest ecosystems

Nutrient deficiencies have been found to have a variable role in the five types of forest damage which affect spruce[326]. From foliar analyses of Norway spruce and silver fir stands in the southern Black Forest, it was shown[327] that Mg availability in the soil had decreased markedly between 1975 and 1983. A decline of K availability was also found at specific sites in south-west Germany. In a comparison of foliar analyses from Norway spruce stands, reductions of K availability of >60% between 1961 and 1983 were demonstrated[3]. Nutritional disturbances of forest ecosystems are now widespread in both Europe and North America[327].

The idea of an imbalance of nutrient supply is important, as it is thought that the concept of 'nutritional disharmony' is more important than simple nutrient deficiencies[328,329]. Although acid deposition is thought to be the principal cause of nutritional disharmonies, other minor causes such as inappropriate silvicultural practices may have contributed to but cannot have caused the decline[330].

If nutritional disharmonies are held to be responsible for some of the forest damages prevalent, then fertilization and liming provide possible strategies for amelioration of forest ecosystems. It has been found that fertilization of younger chlorotic conifers will restore the colour to their needles within a few months, and to the needles of older trees after two vegetative periods. However, there appears to be a point at which trees will not respond to either fertilization or liming.

In 1984 the Federal German Government established a programme subsidizing up to 80% of the costs, of both materials and application, of liming and fertilization to correct for 'new types' of forest damage.

The application of fertilizers and nutrients can be divided into two main categories: compensative and ameliorative. Compensative measures usually involve the application of dolomitic (Mg-containing) limestone to neutralize acidified soil and to provide buffering against further acidification of forest soils. Ameliorative measures involve the specific application of fertilizers and nutrients which contain readily available Mg and K, and sometimes P. For ameliorative measures, soil and foliar analyses are required in the diagnostic process. The application of readily soluble fertilizers such as $MgSO_4$ produces rapid revitalization, whereas the application of dolomitic lime produces sustained restabilization[326]. In practice, combinations of the two strategies may be necessary.

Figure 10 Conifers, south-east Sweden

Despite the beneficial effects of liming and fertilization, there are some problems associated with these practices. Losses may be incurred when fertilizers and lime are applied in mountainous terrain, during heavy precipitation, or to shallow soils[326]. Liming has been shown to increase nitrogen mineralization and nitrogen displacement in the soil, which can lead to increased concentrations of nitrate in soil and groundwaters[326,331]. Another possible harmful effect of liming is that better rooting of spruce has been shown to occur in the humus layer, with a decrease in the fine root mass in the upper mineral soil horizons[332,333]. This shallow rooting system makes the tree more susceptible to drought damage. However, the opposite effect was observed for beech, in that application of lime stimulated root growth in the upper mineral horizons[333].

Other side effects of liming include increased mycorrhizal associations, increased earthworm populations and increased populations of microscopic fungi and bacteria[333], as acidification tends to reduce mycorrhizal associations and soil respiration.

Differential responses to growth have been recorded for spruce. Young plantations have shown increased growth, whereas older stands have shown no response or a negative growth response. In stands that are nitrogen-deficient, the decreased growth may be due to decreased nitrogen availability as a result of the liming[334]. However, this should not be a problem for many areas in central Europe which are nitrogen-saturated. Effects of growth response noted on sites judged to be silviculturally 'poor' have been found opposite to those that are 'good'[335].

Whilst forest decline continues to be an important issue in the ecological effects of air pollution and acid deposition, it is highly unlikely that one decisive 'factor' will be identified which universally explains forest decline. In more recent years, site-specific studies have become more important[336,337] and many valuable data have been acquired. These studies have re-emphasized the importance of considering local stress factors, the balance of which varies according to location. *Figure 10* shows conifers in south-east Sweden, and *Figure 11* Sitka spruce in north-west England.

SUMMARY OF PART 1

This paper has reviewed the sources and effects of acid deposition, and the technological and political initiatives

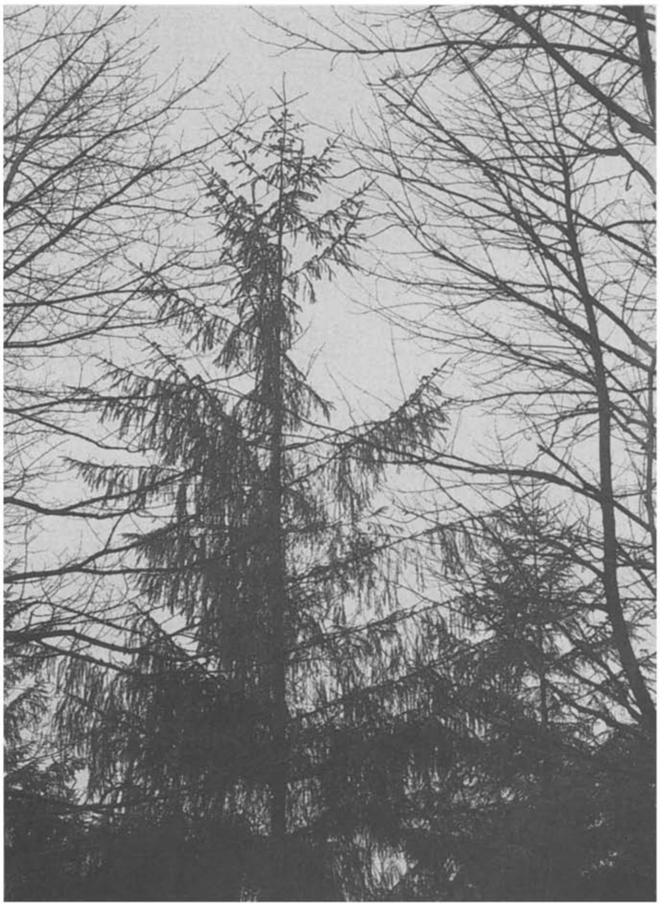

Figure 11 Sitka spruce, north-west England

that may be, or have been, taken to exercise control over emissions of acid deposition precursors up to 1990.

The environmental implications of acid deposition and acidification remain the subject of extensive international and national research programmes addressed at the local, regional and global sources and effects of sulfur dioxide, oxidized and reduced nitrogen species, and volatile organic compounds.

The relation between cause and effect is by no means fully understood, owing in part to the extremely complicated interactions between pollutants and within atmospheric, terrestrial and freshwater environments, which give rise to both direct and indirect effects on susceptible targets. However, the broad relation between sulfur and nitrogen emissions and environmental change, particularly with regard to freshwater environments, is understood well enough to enable national and international policies to be formulated.

The scale and importance of the contemporary acid deposition phenomena tend to suggest that acid deposition is a relatively recent environmental problem. However, it was first described by R. A. Smith working in the city of Manchester in the nineteenth century, and it was Smith who coined the term 'acid rain'. Hence early industrial regions such as the Manchester conurbation have probably experienced more than 200 years of acid deposition.

The major gaseous precursors of acid deposition are sulfur dioxide (SO_2) and oxides of nitrogen (NO_x), but emissions of ammonia, volatile organic compounds, hydrochloric acid and alkaline dusts all influence the acidity of precipitation to some degree. These species have both natural and anthropogenic sources. Anthropogenic sources include power generation, refineries, other industry, transport and commercial and domestic activities. Natural sources include biogenic emissions from terrestrial, tidal and oceanic areas, and non-biogenic emissions from natural combustion, geothermal activity, lightning, airborne soil particles and water aerosols.

European emissions of SO_2 and NO_x in the late 1980s were in the region of 43 and 20.5 Mt a^{-1} respectively. Measurements of concentration and deposition are made by most European nations as part of the European Monitoring and Evaluation Programme (EMEP).

Effects

Concern about the impact of acid deposition on aquatic ecosystems has arisen because of its potential effects on aquatic biota and on human health. Recent studies show that acidification of surface waters has serious implications for aquatic biota and that it may affect species diversity. In particular, acidification of surface waters appears to have a major effect on fish populations. It is thought that aluminium at elevated concentrations in surface waters, associated with low pH, is the main toxic agent. Salmonids are particularly affected and it is thought that mortality rates increase substantially during episodic acid events.

Concern over the effects of acid deposition and air pollution on forest systems in Europe is of more recent origin than that for freshwater. A number of hypotheses have been suggested to explain the observed decline in forest health in many European countries. Proposed explanations for forest decline include: soil acidification and aluminium toxicity; needle reddening disease; and the combined effects of ozone, acid deposition and nutrient deficiencies at high altitude and increased anthropogenic nitrogen deposition.

Whilst forest decline continues to be an important issue as regards the ecological effects of air pollution and acid deposition, it is highly unlikely that one decisive 'factor' will be identified which universally explains forest decline. In more recent years, site-specific studies have become more important and many valuable data have been acquired. These studies have re-emphasized the importance of considering local stress factors, the balance of which varies according to location.

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