Effects of acid deposition on crops and forests

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Summary. Acid deposition includes the gases, sulphur dioxide and nitrogen oxides, and their derivative acids which are dissolved in various forms of precipitation. The effects of both dry and wet deposition on crops and forests are reviewed, including interactions between these two categories of pollutant. Particular emphasis is given to the current forest decline in central Europe, including the possible role of ozone.
Key words. Acid deposition; crops; forests.

Introduction

Until recently, interest in the effects of acid deposition on vegetation has been focussed almost exclusively on the impacts of sulphur dioxide (SO_2) gas. Nevertheless, the problems of wet deposition have been recognised for a long time, with the term 'acid rain' having been coined in 1872 by R. A. Smith[74], who observed damage on vegetation and materials around cities in northern England. Research on the effects of SO_2 on vegetation dates essentially from the publication[75] in 1871 of Stöckhardt's studies on the effects of fumes on conifers in Germany and there is now a wealth of information available for crops and trees, but relatively little is known about impacts on wild herbaceous vegetation. Unfortunately, most of the earlier work concentrated on the effects of very high concentrations of the gas for a short duration in producing visible injury on plants. While this may have been appropriate for studying problems around severe point sources of SO_2, developments in emission control technology and more stringent legislation have brought about a considerable improvement in air quality in most western European countries in recent years. It is much more appropriate at the present time to ask the question as to what effects prolonged lower concentrations of SO_2 have on plant growth in rural areas around cities and industrial districts.

All high temperature combustion processes produce nitrogen oxides (NO_x), partly arising from nitrogen in the fuel, but mainly as a result of the combination of atmospheric oxygen and nitrogen. The importance of this pollutant has, until very recently, been completely overlooked, but in view of its normally accompanying SO_2 at equal or greater concentrations (on a volume/volume basis)[28] it must be taken into account when considering the phytotoxic effects of acidic dry deposition. A third pollutant, ozone (O_3), although not an acid gas, must also be taken into consideration, in view of its presence at levels potentially damaging to vegetation on occasions over large areas of western Europe. O_3 is produced as a result of photochemical reactions involving nitrogen oxides and hydrocarbons, and thus is essentially confined to the summer months.

During the last 15 years, increasing concern about the effects of acid precipitation on the environment, has led to many studies in which plants and soils have been subjected to artificial acidified rain. However, very few experiments have examined the impact of combinations of acid precipitation and gaseous pollutants, as will be discussed later. Nevertheless, such studies are vital in understanding the long-term impacts of acid deposition, as both wet and dry processes have the potential to contribute to vegetation injury.

Effects of SO_2 and NO_2 on trees

Problems have been experienced for many years in growing trees, particularly conifers, in industrial regions. Thus a survey of *Pinus sylvestris* (Scots pine) in the Ruhr region of Germany found that this species was absent from sites where the mean annual SO_2 concentration exceeded 75 ppb[45]. A similar investigation in industrial regions of the southern Pennine hills in northern England found a significant negative correlation between mean winter SO_2 levels and the frequency of occurrence of *P. sylvestris*[26] (fig. 1). This study was carried out in response to reports of poor performance in forestry trials in the district; a considerable improvement has taken place recently in these trials, accompanying sharply falling SO_2 levels[48,49] and this adds weight to the argument that air pollution was the causal factor.

It is probably more difficult to understand the effects of air pollutants on trees than any other group of plants. Commercial forestry practice in the U.K. involves growing conifers up to 60 years old, while in central Europe final harvesting of the crop is often carried out after 100 years. The longest fumigations which have been per-

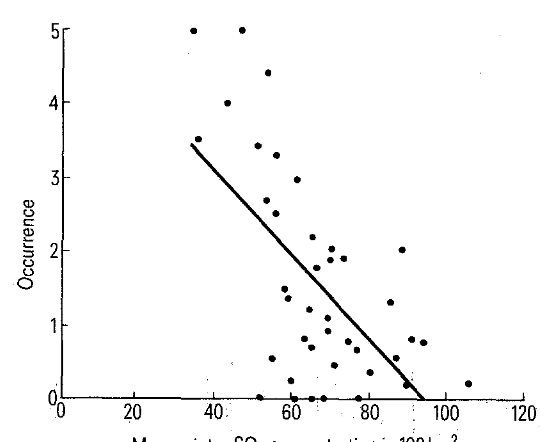

Figure 1. Relationship between occurrence of *Pinus sylvestris* and mean winter SO_2 concentrations (1968–1970) in the southern Pennine hills. (Figures for occurrence are the number of 2 km squares (4 km^2) occupied by at least one specimen per 10 km traverse of 100 km^2 in the study region[26].) Used with the publisher's permission.

formed with acid gases on trees are of only 3 years duration, while in most cases considerably shorter experiments have been performed. Thus it is only practicable to administer a pollutant under controlled conditions for a very small part of the tree's life-cycle. Furthermore, the dimensions of currently available fumigation systems necessitate the use of very young individuals, which may differ from mature trees in their susceptibility to pollutants.

The dangers of extrapolating from short-term fumigation experiments in predicting the effects of pollutants over the life-time of a tree have been demonstrated very clearly in recent years. The term 'latent injury' has been coined by Keller[42] who suggested that air pollution may produce a cumulative adverse effect on a plant's metabolism, which may only manifest itself in the form of reduced growth at a later date. Keller himself demonstrated the existence of such a phenomenon, when he fumigated *Fagus sylvatica* (beech) seedlings over the winter with 112 ppb SO_2 and recorded a reduction in the numbers of terminal buds developing the following spring[43]. More recently a fumigation of *Pinus sylvestris* (Scots pine) seedlings with 38 ppb SO_2 continuously for 650 days showed no reductions in growth after 275 days[33]; however, by the end of the experiment, the SO_2 treatment had reduced the dry weight increment by 14% compared with the controls. Growth analysis indicated that this depression in increment was almost entirely accounted for by a combination of reduced leaf production and accelerated loss of needles, rather than a loss of efficiency of the leaves in producing assimilate. The latent effect recorded in this experiment was confirmed in a field experiment, using three other conifer species in addition to *P. sylvestris*[32]. Seedlings were grown in the same soil in containers at a site in central London and at a rural site (Ascot), 34 km to the west. Over the first years the trees grew generally faster at the London site, probably reflecting the more favourable growing conditions arising from the higher temperatures encountered in the city (table 1). However, by the end of the second year there was no significant difference between the sites in the growth of *P. sylvestris* and *P. contorta* (lodgepole pine), while *Picea abies* (Norway spruce) and *Picea sitchensis* (Sitka spruce) showed substantial (25.0% and 45.6%, respectively) reductions in their relative growth rate in London compared with Ascot. The mean of SO_2 concentration at the London site was only 30 ppb, compared with 11 ppb at the Ascot site, although the presence of elevated NO_x levels (probably fairly similar to SO_2) must be taken into account in the urban area.

Further evidence of the sensitivity of conifers to low SO_2 concentrations was provided by a fumigation of two larch species *(Larix europaea* and *L. leptolepis)* with 22 ppb for 2 years, using an open-air exposure system, which demonstrated reductions in growth[16]. The lowest dose which has been shown to injure a conifer is 26 ppb × 6 h on sensitive individuals of *Pinus strobus* (eastern white pine)[36]. The clear demonstration of marked adverse effects on certain conifer species after exposure to air containing less than 40 ppb over periods of up to 2 years, inevitably raises the question as to the consequences of continuing such exposure throughout the life-time of the tree. It is not unreasonable to suggest that the magnitude of the effects might increase with duration of exposure over many years, and that growth reductions could be seen at SO_2 concentrations below those which have been reported so far in even the longest fumigation experiments. In fact, the International Union of Forest Research Organisations (IUFRO) currently recommends that 19 ppb SO_2 should not be exceeded as an annual mean in order to protect forest trees[80]. Furthermore, observations at exposed sites in central Europe have led IUFRO to make a further recommendation viz. that this limit be lowered to 9 ppb at locations where severe winter conditions predispose trees to injury. In order to place these recommendations in context, it should be noted that estimates have been made that 5% of western Europe experiences annual mean SO_2 concentrations between 19–38 ppb and a further 18% is subjected to 11–19 ppb[28].

In general, it appears that hardwoods are less susceptible to SO_2 than are conifers, although there are a limited number of species which do not follow this pattern[30]. Thus a fumigation of *Acer pseudoplatanus* (sycamore) and *Quercus robur* (pedunculate oak) with 64 ppb SO_2 for 71 weeks continuously between 1975 and 1976 produced relatively few signs of adverse effects: some stimulation of height growth occurred in both species, but senescence of the *Q. robur* leaves was advanced by 12 and 9 days, respectively, at the end of the two growing seasons[31]. A very similar effect of ambient air containing c. 30 ppb SO_2 has been demonstrated recently: in a 26 months experiment, *Q. robur* seedlings grown in chambers in central London ventilated with ambient air lost their leaves 3–9 days earlier than when grown in charcoal-filtered air[78]. Very little information is available on the long-term effects of SO_2/NO_2 mixtures on trees. Freer-Smith[29] has fumigated a range of hardwood species with 62 ppb of SO_2, NO_2, or their mixture over 60 weeks, starting in March 1981, with an intermediate harvest in the summer after 21 weeks. The magnitude and direction of the response by the different species to the various pollutant regimes were extremely variable (table 2), emphasizing the complexities inherent in understanding the effects of

Table 1. Relative growth rates (g g^{-1} $week^{-1}$) of trees grown for 2 years in central London and a rural site (Ascot)[32]. (Modified from Garsed and Rutter[32] with the publisher's permission)

Site	Mean SO_2 concentration (ppb)	*Pinus sylvestris* (Scots pine)	*Pinus contorta* (Lodgepole pine)	*Picea abies* (Norway spruce)	*Picea sitchensis* (Sitka spruce)
Ascot	11	0.0175	0.0165	0.0144	0.0158
London	30	0.0151	0.0156	0.0108	0.0086
Reduction in London cf. Ascot		NS	NS	25.0%	45.6%

NS, not significant.

acid gases on forests. In the case of *Tilia cordata* (small-leaved lime) and *Alnus incana* (alder), NO_2 alone produced significant stimulations in shoot dry weight after 21 and 60 weeks, respectively. On the other hand, SO_2, generally reduced growth, while there were several instances of the pollutant combination producing a synergistic response. For all species, except *A. incana,* the growth reductions increased between the first and second harvests. Very recently, the same author[30] has reported the results of an even longer fumigation experiment with *Pinus sylvestris*, using 62 ppb of the pollutants, singly or in combination over 138 weeks. After 86 weeks, all three pollutant treatments produced decreases in the relative growth rates, compared with the controls, but the SO_2/NO_2 mixture showed an antagonistic interaction. However, a further 52 weeks fumigation did not result in an increased reduction in relative growth rates, but the effects of the pollutant combination changed from antagonistic to additive. The gas concentrations used by Freer-Smith are higher than those generally encountered in forested regions of western Europe. The only information available on the effects of SO_2/NO_2 mixtures more typical of rural areas is based on the results of fumigation experiments in the Netherlands, using six *Populus* (poplar) cultivars: treatment over only 42 days with a mixture of 23 ppb SO_2 and 30 ppb (day)/12 ppb (night) NO_2 produced up to six-fold increases in leaf-fall compared with clean air controls[56]. The significance of this effect for growth is not clear, but it seems likely that a deleterious effect on the overall performance of the plants could take place over a period of many years.

While there is some knowledge on the effects of acidic gases on individual tree species, there is scarcely any information on their impacts on forest ecosystems as a whole, other than in the vicinity of severe point-sources where there is large-scale destruction of vegetation. There are clearly serious logistical problems involved in determining such effects, with difficulties in performing controlled experiments and in establishing a natural baseline for comparison with changes occurring in the presence of pollution. A study of changes in species composition of a forest subjected to different levels of SO_2 and

Table 2. Effects on hardwood species of 62 ppb SO_2, 62 ppb NO_2, and 62 ppb SO_2 + 62 ppb NO_2 over 21 and 60 weeks continuously. (Modified from Freer-Smith[29] with the publisher's permission)

Species	Treatment	21 weeks (23 March 1981–19 August 1981)	p <	60 weeks (23 March 1981–17 May 1982)	p <
Tilia cordata (small-leaved lime)	NO_2	+ 42.2%	0.05	− 1.1%	NS
	SO_2	+ 31.5%	NS	− 36.8%	0.05
	$SO_2 + NO_2$	+ 8.1%	NS	− 32.7%	0.05
Populus nigra (black poplar)	NO_2	+ 18.4%	NS	− 13.2%	NS
	SO_2	− 3.3%	NS	− 16.0%	NS
	$SO_2 + NO_2$	− 39.9%	0.001	− 41.6%	0.05
Betula pubescens (birch)	NO_2	+ 7.8%	NS	− 5.2%	NS
	SO_2	− 20.1%	0.05	− 52.3%	0.001
	$SO_2 + NO_2$	− 52.2%	0.001	− 60.8%	0.001
Alnus incana (alder)	NO_2	− 32.0%	NS	+ 64.9%	0.05
	SO_2	− 54.2%	0.05	− 24.9%	NS
	$SO_2 + NO_2$	− 55.7%	0.05	− 42.0%	NS

NS, not significant.

NO_x in the Ohio River Valley suggested that air pollution caused both primary and secondary effects[51]: the tree canopy showed increased mortality, but this apparently permitted an increase in sub-canopy and shrub species density, which in turn suppressed species diversity in the herb layer. In contrast, a similar study in Pennsylvania[70] showed that, while changes took place in the species composition of the canopy, greater adverse effcts occurred on the sub-canopy, shrub, and herb layers. There is an obvious need for many more studies of this nature, if the significance of acid deposition for forest ecosystems is to be understood.

Effects of acid precipitation on trees

Studies on the effects of acid precipitation on trees have only developed on a large scale over the last decade. Abrahamsen and Tveite[3] have recently reviewed this topic and have noted the considerable difficulty in making generalizations, in view of a bewildering range of different types of experimental procedures used. Extrapolations of fumigation experiments to the field have often been criticized on the grounds that the pollutants are admininistered in an unrealistic manner, but it is apparent that this situation is much worse in the case of experiments with simulated acid precipitation. Thus experiments have been performed with a wide range of droplet sizes, varying intensities and amounts over different periods, continuously or episodically, above or below the canopy, with or without exclusion of ambient rainfall, and with artificial rain made up according to many different formulations. Furthermore, pH 5.6 rain is normally used as a control, this representing the acidity of distilled water in equilibrium with atmospheric carbon dioxide; however, such a figure is questionable, as there is evidence that unpolluted rain may reach pH 4.5 on occasions due to fluctuations in the natural sulphur cycle[18].

In general there is very little experimental evidence for rainfall of pH 3.2 either reducing growth of trees or producing visible injury on foliage. Indeed, there is abundant evidence for stimulation of growth in the presence of acid rain e.g. pH values between 3.0 and 4.0 increased seedling growth of four American tree species while having no effect on seven other species[47]; another study showed increased dry matter production in seven clones of *Picea abies* (Norway spruce) subjected to pH 2.5 compared with pH 5.4 rain[59]. A recent publication[57] has tabulated the threshold pH levels for the production of visible symptoms on tree foliage, with values ranging between 2.0 and 3.2. Such symptoms are often found around hairs, stomata, or veins, where the cuticular wax is thinner and acid water may penetrate more readily into the leaf[24], although injury by artificial acid rain has recently been demonstrated on *Betula alleghaniensis* (yellow birch) leaves, in the form of collapsed epidermal cells, which was independent of cuticle thickness[61].

Most experiments with acid rain are of limited duration, as in the case of gaseous pollutants, often using extremely high levels of acidity in an attempt to obtain results. Thus their applicability to the field situation is open to question. However, there is one long-term experiment where artificial acid rain, with pH values between 2.0 and 6.0, has been applied to several tree species in the field over periods up to seven years[1]. Only in the case of *Pinus sylvestris* were any clear effects observed. Figure 2 shows that height increment of this species was stimulated by pH 2.0, 2.5 and 3.0, compared with 6.0, rain over the first two years of the investigation. Then, over the remaining five years this effect disappeared, with significant reductions being recorded at the two most acid treatments. No significant effect was found at pH 4.0.

Although this study suggests that the rainfall in rural areas of western Europe with the lowest annual mean pH (4.0–4.1) is unlikely to have deleterious effects on trees, at least with respect to direct impacts on the foliage, some recent findings suggest that there may be cause for concern. An experiment has been carried out in which *Malus hupehensis* (apple) seedlings were subjected to spray regimes with eleven different pH values between 2.25 and 7.0, with the foliage being maintained in a wet condition for nine consecutive hours every seven days for nine weeks[27]. Some rather unexpected results were obtained, in that the total dry weight was reduced between pH 2.25 and 3.0 and between pH 4.25 and 5.6, but not between pH 3.25 and 4.0, compared with 'neutral' rain at pH 7.0; the authors tentatively suggested that the lack of impact over the middle of this range was due to increased availability of sulphur and nitrogen as foliar nutrients, but provided no evidence to support this hypothesis. Further evidence for adverse impacts of rain at pH-levels within the ambient range comes from a preliminary report[62], which showed significant reductions in seedling elongation and needle differentiation in several conifer species, including *Picea rubens* (red spruce), after treatment with pH 4.6 (compared to a pH 5.6 control) artificial rain over five weeks. Relatively little attention has been paid to the potential impacts of acid rain on the sexual reproduction of trees. It has now been demonstrated that ambient pH levels can influence the germination of tree pollen[20]: the effect was determined of a pH range between 2.6 and 5.6 in liquid drop culture on the germination of the pollen of Canadian tree species; this showed LD_{50} dosages for inhibition between pH 3.63 and 3.95 for *Acer saccharum* (sugar maple), *Populus tremuloides* (quaking aspen), *Betula papyrifera* (paper-bark birch) and *Pinus strobus* (eastern white pine). These levels of acidity are common in single rain events in many areas and thus can be considered as applicable to field conditions.

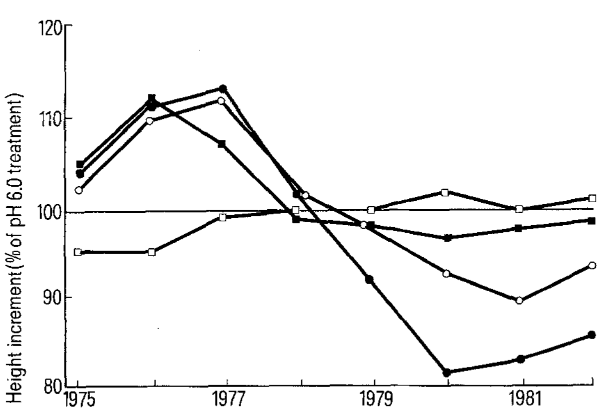

Figure 2. Height growth of *Pinus sylvestris* (c. 15 years old in 1982) exposed to artificial rain of various pH levels[1]. ● pH 2.0; ○ pH 2.5; ■ pH 3.0; □ pH 4.0. Used with the publisher's permission.

Effects of acid deposition on soil biota

It is well known that soil organisms are influenced by pH and thus it is possible that acid deposition might have secondary effects on plant growth through influences on microbiological processes, with respect to litter decomposition and mineral cycling. The effects of acid deposition on these processes have recently been reviewed[52,57]. Exposure of naturally acid soils to simulated rain of pH 2.5 to 6.0 over two years has shown little effect or even an increase with acid treatment on numbers of enchytraeids, spring tails, and mites[2,34]. Soil fungi are unlikely to be seriously affected by acid precipitation as they tend to replace bacteria in decomposition processes in low pH soils. There are conflicting reports on the effects on litter decomposition: application of simulated rainfall with a pH between 2.7 and 3.1 has been shown to increase the decomposition rate of pine needle litter[2,69], while a recent review[5] reported that humus decomposition rates generally decrease from pH 4.5 to 3.5. Acidification of the soil is likely to decrease nitrification, as the bacteria involved are particularly sensitive to low pH: this has been demonstrated after application of pH 4.0 rain to pH 4.4 soil[37]. The significance of this effect for plant growth is questionable in that it is likely to reduce leaching of nitrogen from the soil, but enhance its uptake by roots in the form of ammonium ions[25]. Surprisingly, very few studies have been carried out into the effects of acid deposition on mycorrhizal associations, in view of the importance of the latter in the nutrition of forest trees. It can be postulated that mycorrhizal growth would be reduced by acid deposition, as a soil pH of about 5.0 appears to be optimum for many species[54]. Some evidence for this has been provided recently: reduced mycorrhizal infection of the roots of *Quercus rubra* (red oak) occurred when the plants were subjected to simulated rain at pH 3.0 compared with 5.0[67]; in contrast, other work[35] has not shown any visible damage to the mycorrhizae of *Liquidambar styraciflua* (sweet gum) exposed to pH 3.0 rain. The effects of acid gases on soil microbiological processes are unknown, but are probably small, in view of their limited penetration into the ground. However, litter decomposition has been reduced in a mixed-grass prairie in Montana after 60 days fumigation in the open-air with 45 ppb SO_2[22].

There is a clear need for further research into the effects of acid deposition on soil microbiological processes. Very little is known about the effects on many important types of soil microorganisms and it has recently been suggested that those species which live in the top 1–2 cm may be particularly at risk e.g. protozoa, nematodes, and tardigrades[19].

The role of acid deposition in modern forest decline

During the last few years, a new type of forest damage has appeared in central Europe, which has been widely attributed to the effects of acid deposition[14]. This phenomenon often occurs in areas remote from major sources of SO_2 and NO_x and thus attention initially became focussed on the possibility of acid precipitation being the causal factor, rather than gaseous pollutants. The major exception to this pattern is in the Fichtelgebirge on the

Czechoslovakian border, where potentially phytotoxic SO_2 episodes occur as a result of transport from industrial regions to the east. In contrast, SO_2 and NO_x levels in most other forest damage sites are considerably lower than those which are known to damage higher plant species e.g. at Schauinsland, in one of the main forest damage zones in the Black Forest, where continuous measurements of a range of air pollutants are made at a so-called 'clean-air' monitoring station, annual mean SO_2 and NO_x concentrations over the last decade have been < 3 ppb[4].

The damage first appeared at high altitudes on old *Abies alba* (silver fir) individuals in the mid-1970's, with subsequently fairly similar symptoms developing on *Picea abies* (Norway spruce), the principal conifer of West Germany. Damage has now also appeared on *Pinus sylvestris* (Scots pine), *Fagus sylvatica* (beech), and some other hardwood species. Characteristic symptoms include loss of the older needles, leading to a thinning of the crown, premature growth reduction, die-back of secondary branches, intense yellow discoloration of the needles, and reduction in the fine root system[6]. The symptoms on hardwood species are not so clearly defined, but in *F. sylvatica* are manifested as premature leaf-fall and the development of serrated edges to the leaves. The most severely affected trees are generally on the edge of stands and damage was observed first on exposed west-facing slopes[63]. Over the last few years, there has been a dramatic increase in damage, with national surveys in West Germany showing an increase in the total forest area with symptoms changing from 8% in 1982 to 50% in 1984[7]. Furthermore, damage has moved from high to lower altitudes and from old to younger trees, and is now recorded in neighboring countries, including Switzerland, Austria, France, The Netherlands, and Belgium.

The causes of this forest decline remain a matter of considerable debate and speculation. Examination of affected trees has not revealed any pathogens, other than those which can be attributed to secondary infection of already weakened individuals, although the possibility has been raised that causal organisms may not have been identified[41]. The very widespread occurrence of damage, on many different types of soil, and in the absence of infection by pathogens, has led to the generally accepted view that some type of air pollution is involved, probably interacting with other environmental stresses, particularly climate[15].

In view of the very low concentrations of SO_2 and NO_x recorded in many of the most severely affected areas, the question must be posed as to what pollutants might be present at phytotoxic levels in such places. The only obvious candidates are O_3 and acid precipitation, both of which penetrate into rural areas remote from the sources of their precursors and occur episodically. At first attention was directed towards possible effects of acid deposition via the soil, with Ulrich[77] postulating that this was having a cumulative adverse effect on the soil's buffering capacity, leading to the release of toxic aluminium ions which destroyed the fine root system. This may be applicable in some areas, where soils are particularly susceptible to acidification, but it has now been reported[1] that culture solution experiments have failed to damage the roots of *P. abies* at concentrations which occur in soil water in damaged areas. Furthermore, damage is occurring even on strongly calcareous soils, which are very highly buffered against inputs of acidity[63]. Particular doubt must be cast upon the role of acid precipitation in view of the very clear upward trend in the pH of wet deposition occurring at Schauinsland over the period that forest damage has accelerated (fig. 3).

In contrast to the acidity of precipitation, it is now apparent that O_3 concentrations have been increasing in recent years. This is hardly unexpected, since emissions of its precursors, hydrocarbons and NO_x, have been rising markedly over the same period, largely as a result of increased motor traffic. Unfortunately, until recently, little interest in O_3 has been shown by European scientists, who have assumed that it represented a serious pollution problem only in North America. However, attention has now been drawn to East German data, which show a very marked continual increase in O_3 concentrations since 1955, at various locations, including some in rural areas[79] (fig. 3).

The possibility of O_3 being a major cause of forest damage in central Europe, has recently been examined in some depth[9]. A comparison was made of O_3 concentrations at various sites in the eastern USA, where O_3 damage has been identified on trees, with those recorded in forest damage areas of West Germany. This showed a marked similarity, especially in the case of Schauinsland, with summer mean concentrations lying between 35 and 50 ppb and annual maxima up to 155 ppb. Thus O_3 levels in German forest areas are potentially injurious to trees. Unfortunately, the species known to be injured by O_3 in the USA are generally different from those currently showing damage in central Europe. Very few fumigation experiments have so far been carried out on relevant European tree species, and most of these are rather short-term and of a somewhat preliminary nature. The results of these experiments are shown in table 3, and are some-

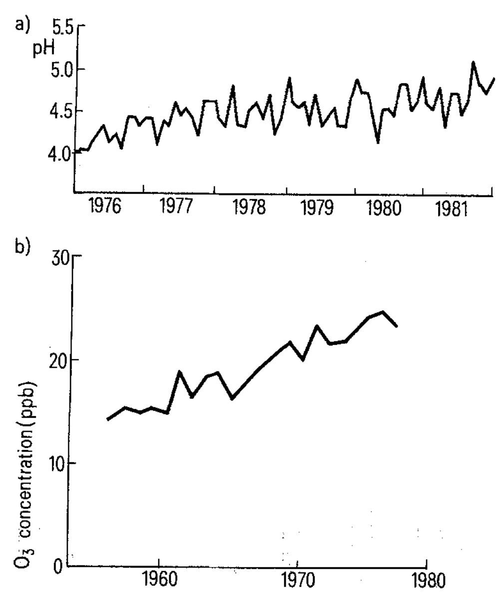

Figure 3. Trends in pollutant concentrations: *a* pH of rain at Schauinsland, Black Forest, 1976–1981[4]. *b* Mean annual ozone concentrations at a rural site in the southern Baltic, East Germany, 1956–1977[79]. Used with the publisher's permission.

what conflicting, but do not in general suggest that the main forest damage species are particularly sensitive to O_3, except possibly *P. sylvestris*. Nevertheless the absence of long-term realistic fumigation experiments on the sensitivity of these species to O_3 precludes any positive statement as to its role in the current problem.

While the mean pH of rainfall in most forest damage areas is not low enough to cause substantial vegetation injury, on the basis of the results of experiments with artificial acid rain, there remains the possibility that other processes could be important. In particular, it is known that 'occult precipitation' (i.e. fog, mist and cloud) is prevalent at many of the high altitude sites where damage first appeared in Germany[15], and that this can be considerably more acid than the rainfall at a given location[23]. This has led to speculation that acid mists may be predisposing trees to O_3 damage or vice versa. A number of European research groups are now investigating the possibility of such a pollutant interaction being involved in forest damage, but so far few results have been published. A recent experiment in which *P. abies* was exposed intermittently to 100 ppb O_3 showed a 22% increase in leaching of magnesium by an artificial acid fog which was applied subsequently, compared with plants which had previously been kept in clean air[63]. This is consistent with reports that severe Mg deficiency is highly characteristic of the needles of damaged trees in the field, a phenomenon which is rather rare in established forests[55]. However, recent work on *P. sylvestris* failed to show any interactions between applications of pH 3.0 artificial mist and O_3 fumigations up to 150 ppb over 56 days, with respect to growth, senescence, foliar injury, or Mg content of the needles[73].

Major increases in forest damage have often coincided with extreme climatic conditions, particularly severe winter weather and summer droughts. This has led to speculation that the problem is essentially the result of complex interactions between acid precipitation and/or O_3 and climatic stresses. Such hypotheses remain largely untested in the absence of experiments on the effects of climatic factors on the sensitivity of European tree species to O_3 and acid precipitation, either singly or in combination. An example of possible climate/pollutant interactions of this type is shown in figure 4. It is suggested that damage to cell membranes by O_3 increases leaching of Mg from the foliage by acid precipitation. The resulting Mg-deficiency reduces the amount of assimilate available for root growth, which in turn may predispose the tree to drought stress. A direct impact of the deficiency is a reduction in frost resistance and in stomatal control, thereby rendering the plant more susceptible to both frost and drought damage. Damage to cell membranes by O_3 will further exacerbate the frost damage, leading to increased cation leaching from the foliage. A further possible interaction arises from a reduction in Mg predisposing the photo-oxidation of chlorophyll by sunlight, which may explain the observation that yellowing occurs to the greatest extent on exposed unshaded foliage[66].

Table 3. The effects of ozone fumigations on tree species involved in current central European forest decline[9] (used with the publisher's permission)

Species	O_3 concentration (ppb)	Duration	Response	Reference
Picea abies (Norway spruce)	150	9 h/day for 35 days	None	43
	150	28 days	Foliar injury	62, 63
	150	8 h	None	21
Abies alba (Silver fir)	50	7 h/day for 42 days	No foliar injury, reduction in photosynthesis	8
	150	9 h/day for 35 days	None	43
	300	56 days	None	62
Pinus sylvestris	100	56 days	Foliar injury	72
(Scots pine)	150	56 days	Foliar injury, reduction in fine roots, increased senescence of older needles	72
	250	8 h	Foliar injury	21
Fagus sylvatica	150	42 days	Foliar injury	62
(beech)	75	42 days	Foliar injury on shaded plants	62
	50	7 h/day for 42 days	None	8

Effects of SO_2 and NO_2 on crops

There is a wealth of information available on the effects of SO_2 on agricultural crops, but much less in the case of NO_2 or the pollutant mixture. Nevertheless, even in the case of SO_2, there is considerable controversy over the minimum concentrations or doses which will reduce the growth of the most sensitive species. Thus a recent review[68] has noted that in less than 20% of experimental fumigations with SO_2 concentrations below 45 ppb have significant growth reductions been recorded. There are, in fact, numerous inconsistencies between the results of fumigation experiments, even when the same concentration of SO_2 is administered for a similar duration[11]. In

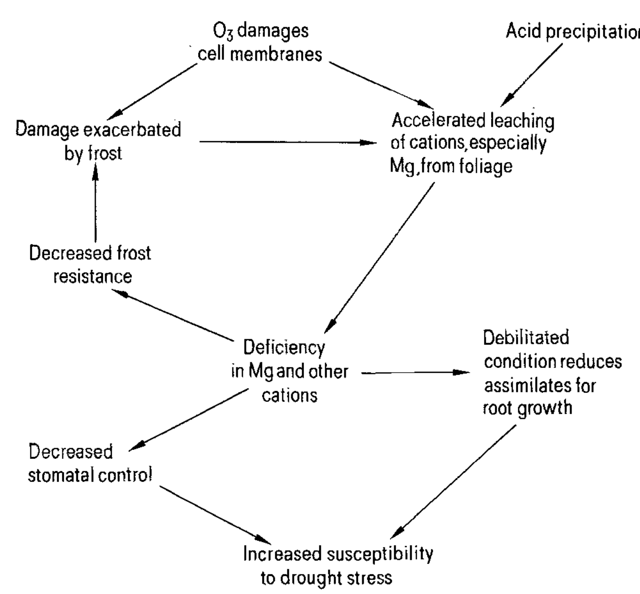

Figure 4. Possible interactions between air pollutants and climatic factors in European forest damage. (Modified with the publisher's permission from Miller[55], based on the hypotheses of Bosch et al.[17] and Rehfuess[65].)

many cases, these inconsistencies are probably explained by interactions between SO_2 and other environmental factors, as the experiments have been performed in chambers located outdoors. The lowest concentrations which have been shown to reduce crop growth are 21 ppb for 28 days for *Nicotiana tabacum* (tobacco) and *Cucumis sativus* (cucumber)[53], and 16 ppb over 173 days for *Lolium perenne* (perennial ryegrass)[13]. Various experiments have been performed in which crops are grown in chambers, ventilated with ambient polluted or charcoal-filtered air. In general these have shown much greater growth reductions for given ambient SO_2 concentrations than fumigation experiments with the same SO_2 levels[50]. It now seems very likely that this phenomenon is explained by the presence of other phytotoxic pollutants in ambient air[12], acting either on their own or in combination with SO_2, with NO_x and O_3 (summertime only) being particularly important.

It is known that synergistic interactions can occur between SO_2, NO_2 and O_3 in reducing the growth of crops or producing visible injury on their foliage[12,60,71]. However, the situation is extremely complex, with additive and antagonistic effects also having been recorded, depending upon the species concerned, and the relative and absolute concentrations of the pollutants. There are also now indications that the nature of the interactions can change markedly, according to season. This was demonstrated when *Poa pratensis* (smooth-stalked meadow grass) was fumigated continuously in outdoor chambers over eleven months with 62 ppb SO_2, NO_2, or their combination, starting in the autumn[81]. Initially SO_2 reduced growth, while NO_2 stimulated it. The pollutant mixture, however, produced a marked synergistic response, with the beneficial effects of NO_2 alone being transformed into a large growth reduction (fig. 5). As the winter progressed, the stimulatory effect of NO_2 disappeared, although SO_2 caused increasing growth reductions, and the combined impact of the pollutants became approximately additive. When more rapid growth commenced in the spring, all adverse effects progressively disappeared and by the autumn a small stimulation of growth had taken place in both treatments containing SO_2. The net impact of these effects over many seasons, is unknown, but, as in the case of trees, there is clearly a need for considerably more research in this area, before the significance of acid gases for perennial crop growth is understood. The situation is complicated even further in the case of grasses by the demonstration that the evolution of tolerance to SO_2 is a common phenomenon in species growing in polluted areas[10,82].

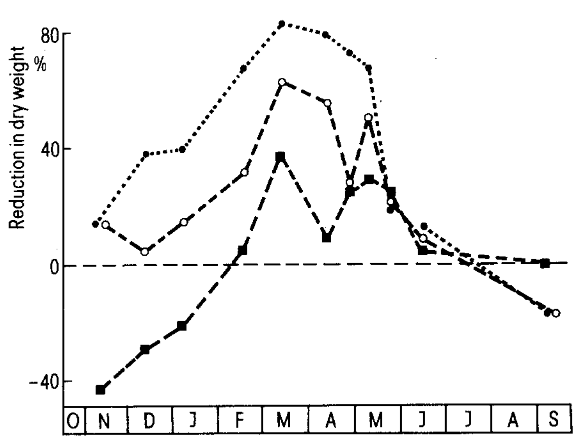

Figure 5. Percentage reduction in dry weight compared with clean air controls of *Poa pratensis* exposed to 62 ppb NO_2 (—■—), 62 ppb SO_2 (--○--), or 62 ppb NO_2 + 62 ppb SO_2 (· · ● · ·). Modified from Whitmore and Freer-Smith[81] with the publisher's permission. Data shown for total weight up to mid-May, and subsequently for shoot weight only.

Effects of acid rain on crops

The effects of acid precipitation on agricultural crops has recently been reviewed in considerable depth by Irving[38], who examined the results of a vast number of experiments with artificial rain. The results of these experiments are extremely difficult to interpret, in view of a considerable range in both the size and direction of effects for any given pH value; a selection of these is shown in table 4. In contrast to work with trees, artificial rain of pH 4.0 has been shown to reduce the marketable yield of various species, compared with pH 5.6 controls (e.g. *Daucus carota* (carrot), *Brassica japonica* (mustard green)). However, a further decrease in pH does not necessarily result in further yield reductions. There are also many

Table 4. Effects of simulated acid rain on crops compared with pH 5.6 control rain. (Selected from Irving[38] with the publisher's permission)

Species	Rate of application ($cm \cdot h^{-1}$)	Duration and number of applications	pH	Effect	Reference
Phaseolus vulgaris (bean)	1.64	18 × 0.67 h	4.0	No effect on yield; increased senescence	40
Daucus carota (carrot)	0.67	44 × 1.5 h	4.0	− 27% market yield	47
			3.5	− 45% market yield	
			3.0	− 44% market yield	
Zea mays (maize)	0.67	20 × 1.5 h	4.0	No effect	47
			3.5	No effect	
			3.0	+ 13% market yield	
Capsicum annuum (green pepper)	0.67	38 × 1.5 h	4.0	No effect	47
			3.5	+ 20% market yield	
			3.0	No effect	
Fragaria chiloensis (strawberry)	0.67	80 × 1.5 h	4.0	+ 51% market yield	47
			3.5	+ 72% market yield	
			3.0	+ 72% market yield	
Medicago sativa (alfalfa)	0.67	56 × 1.5 h	4.0	No effect on market yield	47
			3.5	+ 31% market yield	
			3.0	No effect on market yield	
Brassica japonica (mustard green)	0.67	14 × 1.5 h	4.0	− 14% market yield	47
			3.5	No effect	
			3.0	− 31% market yield	

examples of acid rain increasing yield, even at pH 3.0. In the case of *Capsicum annuum* (green pepper) and *Medicago sativa* (alfalfa), stimulations in yield occurred at pH 3.5, but with no effect at pH 4.0 or pH 3.0, which reflects to some extent the pattern observed in apple seedlings[27]. It was concluded tentatively 'that the net response of a crop to acidic deposition is the result of the interaction between the positive effects of sulphur and nitrogen fertilization, the negative effects of acidity, and the interaction between these factors and other environmental conditions such as soil type and the presence of other pollutants. Available experimental results appear to indicate that the effects of acidic precipitation on crops are minimal and that when a response occurs it may be positive or negative'[38].

Interactions between acid precipitation and other air pollutants, have scarcely been studied. *Glycine max* (soybean) has been subjected to 214 ppb SO_2 and/or artificial rain at pH 3.1 simultaneously[39]: SO_2 reduced seed yield, while acid rain stimulated it, but the pollutants in combination gave an additive effect, with the positive and negative effects apparently balancing out. Similarly, no interactions were found on growth of *G. max* with exposures to pH 4.0 acid rain and 1125 ppb SO_2[72]. On the other hand, a recent report[76] showed that reductions in *G. max* caused by ambient O_3 were increased as the pH of artificial rain was reduced between pH 4.0 and 2.8. A study on *G. max* with a mixture of 200 ppb SO_2 and 100 ppb O_3, in combination with a range of simulated acid rain exposures, between pH 2.6 and 5.6, produced no evidence of interactions between the gases and rain treatments[58]. On the basis of current information, it is impossible to make any definitive statement concerning the interactive impacts on crops of gaseous pollutants and acid precipitation. However, this is clearly a matter requiring further and substantial investigation, using a range of major crop species, with combinations of the pollutants simulating conditions prevailing in the field, with respect to concentration, duration, and coincidence of occurrence.

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