Chapter 10 Effects of sulfur dioxide on vegetation: critical levels

Sulfur dioxide (SO_2) was formerly viewed as the most important phytotoxic pollutant in Europe, and until the early 1980s was the subject of the greatest research in this field. It has since attracted less attention because of its declining concentrations in much of western and northern Europe, as emissions have been reduced, and also because nitrogen gases and ozone have been found to be of increasing significance. Nevertheless, it presents a potential threat to vegetation in many parts of Europe, in particular in the heavily industrialized regions of the Czech Republic, Poland and the eastern part of Germany.

Uptake of sulfur dioxide by plants

Sulfur dioxide penetrates into leaves primarily in gaseous form through the stomata, although there is evidence for a limited pathway via the cuticle. The aperture of the stomata is controlled largely by the prevailing environmental conditions, such as humidity, temperature and light intensity. These external factors thus influence the rate of uptake of sulfur dioxide and hence the degree of injury. When the stomata are closed, as occurs under dark or drought conditions, then resistance to gas uptake is very high and the plant has a very low degree of susceptibility to injury. However, unlike higher plants, mosses and lichens do not have a protective cuticle exposed to sulfur dioxide which is the major reason for their extreme sensitivity to this pollutant.

Effects of sulfur dioxide on plants

High concentrations of sulfur dioxide can produce acute injury in the form of foliar necrosis, even after relatively short duration exposure. However, such effects are far less important in the field than chronic injury, which results from long-term exposure to much lower concentrations of the gas and is essentially cumulative in nature, taking the form of reduced growth and yield and increased senescence, often with no clear visible symptoms or else with some degree of chlorosis. The effects of a given dose of sulfur dioxide can be modified by prevailing environmental conditions. Conversely sulfur dioxide can also modify the response of plants to other environmental stresses, both biotic and abiotic, often exacerbating their adverse impacts.

Dose-response relationships have been generated for various agricultural crops and some temperate forest tree species, but data on native herbaceous vegetation are sparse. The information used in deriving such relationships have been based on controlled fumigations, under quasi-field or defined environmental conditions, from filtration experiments and from field studies such as transects along sulfur dioxide gradients. Field and filtration studies provide data on responses under realistic conditions but are confounded by the presence of other pollutants and variable environmental conditions. Nevertheless, reasonably accurate values for no-response thresholds for adverse effects have been derived for broad categories of plants, as used in the documentation on critical levels of the United Nations Economic Commission for Europe (UNECE) *(1).*

Developments since the first edition

The chapter on sulfur dioxide (SO_2) in the first edition of these guidelines remains a contemporary account of sulfur dioxide uptake by plants and of its effects on plants and plant communities. The references selectively cover effects research up to the mid-1980s. Selectivity is based, quite appropriately, on the need to consider the smallest concentration for which adverse effects on plants have been recorded. In spite of recent shifts of concern among scientists and policy-makers away from sulfur and towards ozone and nitrogen compounds, there have been a substantial number of sulfur dioxide fumigation experiments conducted since the mid-1980s. These include the European Commission's Open Top Chamber programme *(2)*, the US National Crop Loss Assessment Network (NCLAN) *(3)*, the Hohenheim long-term experiment *(4)*, the Liphook field release experiment *(5)* and a number of trials using filtered versus unfiltered air. These recent experiments provide data against which the current guideline values can be evaluated. The current WHO guideline values for sulfur dioxide are 30 $\mu g/m^3$ as an annual average and 100 $\mu g/m^3$ as a 24-hour average.[1]

Perhaps more important than the additional effects data that have become available since 1985 are the advances made in the understanding of deposition processes, the results of new work on the impacts of acidic mist *(6)*, and work on the interactions between gaseous sulfur dioxide and other stresses and of causal sequence in forest declines *(7).* Similarly, the last ten years have seen the implementation of the EU's Large Combustion Plant Directive (1988), the signing (1985) and completion (1993) of the first sulfur protocol of the Convention on Long-range Transboundary Air Pollution and, of perhaps even greater importance, the signing of the latest sulfur protocol (June 1994).

These policy changes have recently been guided by the critical loads and critical levels approach. In terms of policy development, these approaches represent a further basis on which the WHO sulfur dioxide guideline values can be evaluated.

New experimental data

The final meeting of the European Open Top Chamber Programme was held in November 1992 *(2).* Thirteen sites using open-top chambers were employed to investigate the effects of air quality on the physiology, growth and yield of three crops: *Vicia faba, Phaseolus vulgaris* and *Triticum aestivum.* When analysed as a single data set, the results from the 13 sites gave good dose–response relationships for ozone exposure, and this programme may be of value in re-evaluating WHO guide values for ozone. At some sites, specific effects can be attributed to sulfur dioxide. In Belgium, for example, ambient pollutant concentrations did not affect the grain yield of *T. aestivum* but did decrease root growth. This effect was attributed to the combined effect of nitrogen dioxide and sulfur dioxide *(8).* Concentrations of sulfur dioxide were in the range 9–14 $\mu g/m^3$ in the presence of about 19 $\mu g/m^3$ nitrogen dioxide and 6 $\mu g/m^3$ nitric oxide and other gaseous air pollutants, including ozone. Fowler et al. *(9)* carried out similar filtered versus unfiltered air experiments over three consecutive seasons (1982/1983, 1983/1984 and 1984/1985) in Glasgow using *Hordeum vulgare* (winter barley). In two years

[1] Sulfur dioxide: 1 $\mu g/m^3$ = 0.35 ppb; 1 ppb = 2.86 $\mu g/m^3$ at 20 °C and 1013 hPa.
Nitrogen dioxide (NO_2): 1 $\mu g/m^3$ = 0.532 ppb; 1 ppb = 1.88 $\mu g/m^3$ at 20 °C and 1013 hPa.
Nitric oxide (NO): 1 $\mu g/m^3$ = 0.813 ppb; 1 ppb = 1.23 $\mu g/m^3$ at 20 °C and 1013 hPa.
Ozone (O_3): 1 $\mu g/m^3$ = 0.5 ppb; 1 ppb = 2 $\mu g/m^3$ at 20 °C and 1013 hPa.

(1982/1983 & 1984/1985) the plants in unfiltered air (polluted with about 26 μg/m^3 sulfur dioxide and about 23 μg/m^3 nitrogen dioxide) gave larger grain yields by some 8–9%, and above-ground dry matter was also increased. In 1983/1984, effects were in the opposite direction for similar average sulfur dioxide and nitrogen dioxide concentrations, with grain yields lower by 13%. The summer of 1984 had a notable dry period in May and June, giving rise to the interpretation that interactions in the effects of sulfur dioxide plus nitrogen dioxide and drought, together with ozone exposure, may have accounted for the contrasting response in this season.

Weigel et al. *(10)* grew winter and spring barley in open-top chambers at Braunschweig in Germany. Sulfur dioxide concentrations in ambient air ranged between 34 μg/m^3 and 127 μg/m^3 as monthly mean values between November 1995 and August 1986, and between 16 μg/m^3 and 26 μg/m^3 between April and August. Compared with filtered controls, ambient air increased plant weight in winter barley but reduced leaf weight at some harvests. Ambient air did not affect grain yield or leaf sulfur content. In further work by the same group, a significant reduction in top dry weight and 1000-seed dry weight of *Ph. vulgaris* and *Brassica napus* was demonstrated after fumigation with 51 μg/m^3 sulfur dioxide for 3 months in summer. In a similar study, Adaros et al. *(11)* found a significant reduction in 1000-seed dry weight in *H. vulgare* after fumigation with 63 μg/m^3 sulfur dioxide, and a reduction in ear, 1000-seed, grain and whole plant dry weights of *T. aestivum* after fumigation with 56 μg/m^3 sulfur dioxide. Thus, the effects of sulfur dioxide at concentrations between 26 μg/m^3 and 127 μg/m^3 are variable and, even where present along with nitrogen oxides and ozone, impacts are often difficult to detect or contrast in direction between years.

Colls et al. *(12)* used a field release system to expose winter barley to sulfur dioxide for 1 day at 500 μg/m^3, 3 days at 168 μg/m^3 or 6 days at 77 μg/m^3 on 24 occasions over the growing season. Even at these large concentrations, imposed along with other pollutants in ambient air, there were no responses of shoot dry weight or grain yield to sulfur dioxide. This effect contrasts with earlier work of the same group *(13)* with continuous exposure to sulfur dioxide at similar concentrations. Colls et al. *(12)* concluded that for sulfur dioxide in these concentration ranges, plant response is determined by cumulated dose (e.g. ppb·days) or average concentration rather than by intermittent peak exposures.

The NCLAN programme also identified the importance of pollutant mixtures and of interactions with abiotic and biotic factors in producing crop losses. Although effects of sulfur dioxide on North American prairie grass have been shown to be significant in field release experiments *(14)*, the NCLAN programme placed the main emphasis on ozone and on interactions between ozone and water deficit *(3)*. A comprehensive review of crop response data from the Netherlands *(15)* came to similar conclusions. National average concentrations of ozone (at 89 μg/m^3) were reported to give some 3% reductions in crop volume, while the average sulfur dioxide concentration at 24 μg/m^3 apparently resulted in a volume loss of some 1%. These authors concluded that air pollution decreased crop production by about 5% at that time, and that of this loss some 70% was caused by ozone. Both their evaluation and the NCLAN studies showed ozone to be of greatest importance to crop loss, but "other pollutants such as sulfur dioxide and HF appear more important to the Dutch situation than in the USA".

A study by Dueck et al. *(16)* in the Netherlands involved fumigation of heathland bryophyte and higher plant species with 32, 66, 130, 260 and 520 μg/m^3 sulfur dioxide for 42 days. Dose–response relationships for reduced root or shoot growth, changes in root:shoot ratio and visible injury were generated and thresholds established. The most sensitive higher plant species was *Hieracium pilosella,* with a leaf injury threshold of 50 µg/m^3, followed by *Antennaria dioica* with a growth reduction threshold of 75 µg/m^3. In contrast, the most sensitive bryophytes were *Dicranum polysetum* and *Hypnum cupressiforme*, with thresholds of 19 µg/m^3 and 21 µg/m^3 sulfur dioxide for visible injury and growth reduction, respectively. Dueck et al. *(16)* adopted a probabilistic approach, on which basis they established an effect threshold of 8 µg/m^3 sulfur dioxide at which 95% of the species in a heathland community would be protected over a 42-day period.

The Hohenheim long-term experiment *(4)* was an investigation of the effects of sulfur dioxide, ozone and simulated acidic precipitation, using large open-top chambers and *Picea abies* (Norway spruce), *Abies alba* (silver fir) and *Fagus sylvatica* (beech). Sulfur dioxide treatments were 10 µg/m^3 or 120 µg/m^3 (weekly mean values) and ozone treatments were 20 µg/m^3 and 180 µg/m^3 (also weekly concentrations). Sulfate concentration in the simulated acid rain was 4.3 mg/1. These large concentrations of sulfur dioxide and of sulfur dioxide plus ozone resulted in increased peroxidase activity associated with detoxification of free radicals, and increased glutamate dehydrogenase and glutamine synthetase activity. There were, nevertheless, increases in foliar sulfur content. Sulfur dioxide also resulted in decreased total chlorophyll and total xanthophyll contents of the conifer needles. Sulfur dioxide at 120 µg/m^3 resulted in decreased photosynthesis and transpiration in *A. alba* but not in *P. abies.* These biochemical effects might be regarded as the classic responses to moderate sulfur dioxide concentrations, and by the fourth growing season of the experiment (1987) they were resulting in decreased growth rates. In later reports on this investigation it was shown that 50–60 µg/m^3 sulfur dioxide in summer produced reductions in fine roots of *F. sylvatica* and *A. alba (17)* and in net photosynthesis and transpiration of *A. alba (18).*

Steubing et al. *(19)* reported changes occurring in the species composition of beech forest floor flora after fumigation with an apparently low level of sulfur dioxide. Further details of the same investigation are given by Steubing et al. *(20)*, who reported a reduction in leaf area index of *Milium effusum* and *Arum maculatum* after subjecting them to a mean sulfur dioxide level of 15–39 µg/m^3 over four growing seasons. These results should be interpreted with caution, however, as they involved intermittent fumigations with 300 µg/m^3 sulfur dioxide for 4 hours in open-top chambers placed over the natural vegetation.

A major forestry study reported since 1985 is the forest stand work undertaken by Schulze et al. *(7)* in the Fichtelgebirge, north-east Bavaria. The potential role of sulfur dioxide at the two Fichtelgebirge sites (Oberwarmensteinach and Wulfersreuth) was not ignored; indeed the Fichtelgebirge was considered to be an area of Germany in which sulfur dioxide concentrations might have their greatest direct effect. However, the causal sequence identified for these two sites placed the main emphasis on the indirect, soil-mediated effects of sulfur and nitrogen depositions. Thus the results of this major study will be of relevance mainly in setting sulfur critical load guideline values or guideline values for total acidity.

A major forestry study of more direct value in setting sulfur dioxide guideline values was the Liphook sulfur dioxide and ozone field (or rather forest) release experiment. This was a major study with over 4000 trees examined for various responses, and a number of papers have been published. Fortunately an overview, which includes a discussion of data use in setting critical loads and levels, has been published by McLeod & Skeffington *(5)*. *Pinus sylvestris* (Scots pine), *Picea abies* (Norway spruce) and *Picea sitchensis* (Sitka spruce) were grown in the forest soil (a humoferric podzol) between May 1987 and December 1990. Treatments were ambient air (sulfur dioxide annual mean concentration of about 12 μg/m^3) and two levels of sulfur dioxide (continuous values of 32 μg/m^3 or 53 μg/m^3) in factorial combination with one level of ozone (1.3 × ambient). The ozone fumigations did not affect tree growth, but in contrast these sulfur dioxide exposures gave a range of responses. For *P. sylvestris* there were no effects of sulfur dioxide on growth; in *P. sitchensis* extension growth was significantly increased with both sulfur dioxide concentrations by the final year of the experiment. There were no significant effects on root growth. The increased extension growth was associated with the co-deposition of sulfur dioxide and ammonia *(21).* Indeed the establishment of the importance of co-deposition in this context was a major achievement of this experiment.

In contrast to *P. sitchensis, P. abies* was poorly suited to this site and showed a decrease of stem diameter increment with both sulfur dioxide concentrations. McLeod & Skeffington *(5)* suggest that this finding supports the hypothesis that trees may be more likely to respond to pollution stress if suffering other stresses. An alternative interpretation would be that *P. abies* is more sensitive to sulfur dioxide than *P. sitchensis.* Only *P. sylvestris* showed leaf injury from sulfur dioxide and this was restricted to certain individuals, implying a genetic predisposition. Such symptoms were unexpected, since such injury had been identified previously at 665 μg/m^3 sulfur dioxide *(22).* Detailed analysis showed that the visible injury related to sulfur dioxide exposure in a 20-day period immediately after budburst. The threshold concentration for injury was as low as 16–26.6 μg/m^3 over this period in two years and between 22.3 μg/m^3 and 47.9 μg/m^3 in the third. In general, the growth of the individuals affected in this way was not decreased. A late frost in the spring of 1989 led to damage on about 10% of trees of the two spruce species in all plots, which increased to 20% in the trees exposed to 53 μg/m^3 sulfur dioxide. Interacting effects of sulfur dioxide and frost were therefore not a strong feature of the experiment, not being recorded on other occasions.

A neglected experimental approach is exposure of plants to ambient pollutants at different concentrations under field conditions. Two such studies relating to sulfur dioxide have been reported. Ashmore et al. *(23)* carried out a study on the effects of ambient air pollution on *Trifolium pratense*, along a transect 38 km westwards from central London. Plants were exposed as standard cultures along the transect, which represented a gentle gradient of sulfur dioxide and nitrogen dioxide concentrations declining in a westerly direction. Sulfur dioxide, nitrogen dioxide and ozone concentrations were measured at each site by simple physicochemical or biomonitoring techniques, and meteorological parameters were also evaluated. A multiple regression analysis showed a negative correlation between sulfur dioxide concentration and top and root dry weights and flower numbers. Unfortunately, the method of measuring sulfur dioxide only gave a deposition rate, but it was suggested that concentrations ranged between about 7 μg/m^3 and about 40 μg/m^3.

Another study was carried out around the industrial area of Yokkaichi in Japan by Matsushima et al. *(24).* They suggested that an April–November sulfur dioxide concentration of 19 μg/m^3 over two years reduced the dry weight per needle of Pinus *thumbergii* when potted plants were exposed in the field. These results should be interpreted with extreme caution, however, as the sulfur dioxide concentrations of 5 μg/m^3 at the control site appear to be based on some form of extrapolation, and the authors recognized that oxidants were also present.

Effects on lichens and bryophytes

The first edition of these guidelines considered the effects of sulfur dioxide only on higher plants. Thus they did not include the abundant evidence that many species of lichen and bryophyte are considerably more sensitive than the most susceptible species of higher plant. As mentioned previously, these types of plant lack the protection of a cuticle and of stomata that can close and prevent the uptake of pollutants. In other words, they are effectively exposed to pollutants continuously, at least when they are in a moist condition and metabolically active. These characteristics render both lichens and bryophytes particularly sensitive to sulfur dioxide, which is highly soluble in the surface water film.

There is little doubt that sulfur dioxide has been the main pollutant responsible for the dramatic decline in bryophytes and lichens in urban and industrialized areas over the past 100 years or so *(25).* However, in urban areas sulfur dioxide is often accompanied by nitrogen oxides and other pollutants *(26).* Most field investigations have failed to distinguish between the effects of individual pollutants when they occur in mixtures. In considering the impact of sulfur dioxide on bryophytes and lichens, there is uncertainty about whether direct effects on metabolism or indirect effects on the acidity and chemistry of the substratum have greater importance. The direct toxic effects of sulfur dioxide occur at much lower concentrations than those causing injury to vascular plants, and thus bryophytes and lichens will often determine the critical level set for sulfur dioxide in natural environments.

Direct effects of sulfur dioxide on the metabolism of bryophytes and lichens have been summarized by Winner *(27)*, Fields *(28)* and Richardson *(29).* Among some commonly measured indicators of metabolic vitality, the sequence of events on exposure to sulfur dioxide is: declining nitrogen fixation (only in lichens with a cyanobacterial photobiont), increased membrane leakiness, reductions in photosynthesis and respiration and, least sensitive, destruction of photosynthetic pigments. Adenosine triphosphate levels in lichen thalli also appear to be a sensitive indicator of pollution stress. The transfer of carbohydrate from alga to fungus within lichens, a critical process in the symbiosis, is mediated by a carrier protein that appears to be more sensitive to sulfur dioxide than the less accessible photosynthetic apparatus of the alga *(30).*

The pH of water films in contact with metabolically active bryophytes and lichens and their substrata may significantly modify the toxicity of sulfur dioxide molecules as they dissolve and enter the individual cells. Where the substratum is basic, the predominant ionic form is the relatively harmless sulfite ion, whereas on acid surfaces the considerably more toxic bisulfite ion and sulfurous acid are formed. Calcium ions in basic environments also appear to have a direct ameliorative action that counters sulfur dioxide toxicity in lichens (31). Exposure of poorly buffered surfaces such as bark to sulfur dioxide over long periods has caused a reduction in pH through the leaching effects of sulfurous acid *(32).* This will tend to exacerbate the effects of sulfur dioxide on bryophytes and lichens. It is notable that some of the most sulfur dioxide tolerant lichens (e.g. *Lecanora conizaeoides,* Scoliosporum *chlorococcum, Hypogymnia physodes*) and mosses (e.g. *Dicranoweisia cirrata, Ceratodon purpureus)* are also those that best tolerate acid conditions.

Individual bryophyte and lichen species differ widely in their sensitivities to sulfur dioxide as a result of habitat, growth-form, morphological and physiological differences. Thus epiphytes, which form species-rich communities on exposed tree trunks and branches, often appear to be more sensitive to sulfur dioxide than species that occupy more sheltered niches, such as on soil in forests. This differential sensitivity has given rise to striking zonation patterns caused by progressive impoverishment on approaching towns and point sources of sulfur dioxide. Gilbert *(33,34)* and Hawksworth & Rose *(35,36)* made practical use of this phenomenon in the "zone scale" biomonitoring approach. Zone scales allow rough prediction of the average sulfur dioxide concentration of the atmosphere from the assemblages of species present within a standard habitat. They are based entirely on field observations, and their effectiveness depends ultimately on the soundness of the correlation of the various lichen or bryophyte "zones" with average concentrations of sulfur dioxide worked out in test areas.

Most of the studies suggesting acute sensitivity of cryptogams to sulfur dioxide have involved correlation of field distributions of species with ambient concentrations. They indicate that winter or annual means of 30 $\mu g/m^3$ are sufficient to eradicate the most sensitive taxa. Community changes were observed at average concentrations below 10 $\mu g/m^3$ in one careful study around a newly established rural point source *(37).* Critical levels of 10 $\mu g/m^3$ annual mean have been proposed, but these levels may have to be further reduced as detailed information becomes available for the most sensitive species.

Interaction between sulfur dioxide and other pollutants

While fumigation studies have been performed for many years on mixtures of pollutants, the great majority of these have used concentrations substantially greater than the critical levels, for at least one of the pollutants in each combination. In this section, the literature will be reviewed on combinations where both or all of the pollutants are reasonably close to the UNECE critical levels *(1).* The requirement for controlled studies of this type is obvious in view of indications from filtration experiments of their potential importance.

Adaros et al. *(11)*, working with open-top chambers, showed that an 8-hour mean of about 47 $\mu g/m^3$ ozone (maximum 85 $\mu g/m^3$, which is not far above the critical level) added to a mean of 56 $\mu g/m^3$ sulfur dioxide (maximum concentration in 24 hours of 91 $\mu g/m^3$), again not far above the critical level, produced a synergistic interaction on *Triticum aestivum,* increasing growth reductions. In contrast, applying the same mixture of sulfur dioxide and ozone showed that the addition of sulfur dioxide stimulated yield, overcoming ozone-induced growth reduction. The same group working with *Brassica napus* showed a number of significant ozone × sulfur dioxide interactions *(38).* Thus 46 $\mu g/m^3$ sulfur dioxide stimulated pod dry weight, but this was antagonized by the addition of 70 $\mu g/m^3$ ozone as a maximum 8-hour mean, these again being close to the critical levels. In another experiment, a reduction in 1000-seed dry weight caused by 56 $\mu g/m^3$ sulfur dioxide was eliminated by a maximum 8-hour mean ozone level of 85 $\mu g/m^3$. A substantial impact of sulfur dioxide below the critical level for crops was shown by Ashmore et al. *(39)*; when added to the ambient air in a rural English location, 21 $\mu g/m^3$ sulfur dioxide produced a substantial reduction in the yield of peas in air containing more than 120 $\mu g/m^3$ ozone on 21 days during the growing season; in contrast, no effect was seen when the same concentration of sulfur dioxide was added to air from which the ozone had been removed by filtration.

Ashmore (unpublished data) showed a marked synergistic interaction of sulfur dioxide and nitrogen dioxide well below the critical levels for crops (21 $\mu g/m^3$ and 11 $\mu g/m^3$, respectively) on *P. sativum*, whereby stimulation of vegetative and pod dry weights was transformed to reductions below the control when present in combination. In an experiment using *Spinacia* *oleracea* with the same fumigation conditions, a synergistic interaction occurred with respect to dry weight reduction. Adaros et. al. *(11)* found an antagonistic effect of adding 39 µg/m^3 nitrogen dioxide to 56 µg/m^3 sulfur dioxide in reducing the yield of *T. vulgare.* The same group *(39)*, working with the same pollutant regimes and *B. napus*, found an antagonistic interaction in the form of reduced growth stimulation compared with the single pollutants.

Mooi *(40)* demonstrated a remarkable effect on *Populus* × *interamericana* of adding an ozone concentration (12-hour mean 58 µg/m^3), which appears to be around the critical level, to 60 µg/m^3 sulfur dioxide plus 57 or 23 µg/m^3 nitrogen dioxide: leaf fall was almost doubled compared with sulfur dioxide plus nitrogen dioxide, which had increased this by 430% over the untreated controls. Adaros et. al. *(11,38)* showed strong synergistic impacts of the three pollutant mixtures on 1000-grain dry weight of *T. aestivum* and pod length of *B. napus.*

The interaction between sulfur dioxide and ammonia was also examined by Van Hove et. al. *(41)* for *Populus* × *euramericana* cv. Flevo. During a 7-week fumigation, it was shown that the addition of 46 µg/m^3 sulfur dioxide counteracted the stimulation in photosynthesis produced by 64 µg/m^3 ammonia.

It is very difficult to make any meaningful generalization concerning guidelines for pollutant mixtures, in view of conflicting reports of the effects of low concentrations and the absence of any dose–response data within this range. It is apparent, however, that there is evidence that the addition of ozone and/or nitrogen dioxide to sulfur dioxide at around the critical levels of all three pollutants can under some circumstances produce markedly increased adverse effects. On present evidence it is impossible to make any more firm conclusions than a plea for more research on this matter, and for a cautious approach to be adopted in view of the not infrequent occurrence of elevated concentrations of all three pollutants in the field.

Interactions with other stresses

Some indications of interactions between sulfur dioxide and frost in the Liphook experiment are reported earlier in this chapter. There is, however, evidence from field studies that ambient sulfur dioxide at low concentrations can interact markedly with low temperature stress and lead to increased damage. The most extensive study on this subject is the work of Materna and his colleagues in the polluted Ore Mountains in the Czech Republic. They examined the health of spruce stands near which sulfur dioxide monitoring stations had been established, over a relatively small area. Nitrogen oxide and hydrogen fluoride levels appear to have been low, although other adverse impacts such as soil acidification and heavy metal toxicity cannot be precluded. Annual mean concentrations showed a highly significant correlation with the 97.5 percentile of high concentrations, which indicates the value of long-term means in setting guidelines for sulfur dioxide. On the basis of this study, it was determined that the duration of exposure until the spruce stands began to disintegrate depended on a combination of sulfur dioxide concentration and altitude, the latter being negatively correlated with time (Table 1). Makela et al. *(42)* developed a model by calculating the effective temperature sum (ETS) (i.e. the annual sum of daily temperatures exceeding a threshold concentration), this being negatively correlated with altitude and being a meaningful parameter with respect to changes in the sensitivity of trees to sulfur dioxide with increasing altitude. Taking mean annual sulfur dioxide concentrations and ETS values in Europe, the authors were able to map different degrees of risk to forests, under a range of emission scenarios. The lowest concentrations that apparently damaged spruce under the most severe climatic conditions were in the range 20–30 µg/m^3.

However, the study was limited by altitude and it is possible that under even more severe conditions, as in northern Europe, that damage could have occurred at even lower concentrations.

Table 1. Time (in years) between the beginning of air pollution incidence and the disintegration of *Picea abies* stands in Czechoslovakia

Annual mean sulfur dioxide concentration (μg/m^3)	Elevation above sea level (metres)			
	0–600	**600–900**	**900–1050**	**>1050**
<20				
20-30			30–40	20
30-50	50–60	20–30	20	
50-70	40–50	20	10	
70-90	30–40	10–15		
>90	20–30	<10		

Source: Makela et al. *(42).*

It has been known for a long time that air pollution apparently modifies outbreaks of pests and pathogens in the field, although it is only more recently that causal relationships have been established in controlled experiments. Familiar examples are changes in invertebrate herbivore populations around point sources of pollution in Poland, increased outbreaks of bark beetles in oxidant-damaged forests in California, and the former absence of *Diplocarpon rosae* (the black spot fungus of roses) in urban areas of the United Kingdom with high sulfur dioxide levels.

Most of the controlled fumigation or filtration experiments that have been used to elucidate and quantify the effects of sulfur dioxide on pathogens and pests have involved extremely high concentrations. Thus their relevance to the field is questionable, although they may provide valuable indications as to processes and mechanisms by which pollutants influence pathogen and pest performance in the field. A further problem is that only very few experiments have translated the effects of pollutants on pathogens and pests into effects on the growth and yield of their host plants, and thus their significance for vegetation is unclear (although in many cases this may be predicted on a theoretical basis). Another complication is that there are many cases of pollutants reducing the performance of pests and pathogens, thereby producing a potential beneficial impact on plant growth. This applies particularly in the case of obligate fungal pathogens, where until recently it was believed that air pollution always reduced performance due to interference with the host plant's metabolism. This can no longer be seen as a universal phenomenon, however, in view of the recent demonstration by Mansfield *(43)* and Mansfield et. al. *(44)* that the performance of the obligate pathogen *Erysiphe graminis* was stimulated when infecting barley fumigated with sulfur dioxide concentrations of 53 μg/m^3 for 24 hours or 38 μg/m^3 over the growing season.

In the case of pests, ambient air containing a mean of 25 μg/m^3 sulfur dioxide plus 25 μg/m^3 ozone over 3 days resulted in a significant stimulation of the mean relative growth rate of rose aphids feeding on roses *(45)*. A study along a gradient of sulfur dioxide and nitrogen dioxide declining outwards from central London showed that sulfur dioxide concentrations (which probably ranged between about 7 μg/m^3 and about 40 μg/m^3) were positively correlated with the performance of cereal aphids on barley, although a similar gradient of nitrogen dioxide was also present *(46)*.

Thus there is evidence that low concentrations of sulfur dioxide can influence biotic stresses, but it is not possible with present knowledge to determine thresholds that could be used in the formulation of air quality guidelines.

Other air quality standards for sulfur dioxide

In spite of developments in understanding of the role of pollutant deposition in forest decline, the International Union of Forest Research Organizations (IUFRO) has not revised its recommended sulfur dioxide values for the protection of trees (Table 2). It can be seen that the 24-hour mean agreed by IUFRO is the same value as the current WHO guideline value (100 μg/m^3), except that the IUFRO value can be exceeded 12 times in a 6-month period. The IUFRO annual value, at 50 μg/m^3, exceeds the current WHO guideline value. Similarly the EU limit values set less rigorous standards than the current WHO guideline values. The median (50th percentile) of daily mean values taken throughout the year should not exceed 80 μg/m^3, and values taken throughout the winter should not exceed 130 μg/m^3 (Box 1).

Table 2. Current IUFRO air quality standards for sulfur dioxide (1978, confirmed in 1980)

Period	Maximum level of sulfur dioxide (μg/ m^3) that allows:	
	full production in most sites	full production and environmental protection[a]
Annual average	50	25
24-hour average	100[b]	50
97.5 percentile of 30-minute values in growing season	150	75

[a] Environmental protection against erosion and avalanches and to ensure full production in higher regions of mountains, boreal zones, extreme sites, etc.

[b] The 24-hour average may be exceeded 12 times in a period of 6 months.

Source: Wentzel *(47)*.

Box 1. EU statutory standards for sulfur dioxide

Median (50th percentile) of daily values taken throughout the year

120 µg/m^3 if smoke is < 34 µg/m^3
80 µg/m^3 if smoke is > 35 µg/m^3

Median of daily mean values taken throughout the winter (1 October to 31 March)

180 µg/m^3 if smoke is < 51 µg/m^3
130 µg/m^3 if smoke is > 51 µg/m^3

98th percentile of all daily mean values taken throughout the year

350 µg/m^3 if smoke is <128 µg/m^3
250 µg/m^3 if smoke is >128 µg/m^3

Guideline values for year

Arithmetic mean of daily values taken throughout the year: 40–60 µg/m^3

Guideline values for 24 hours

Daily mean value: 100–150 µg/m^3

Note: Limit values depend on presence and concentration of smoke. The smoke values are quoted in British Standard Units: EC units × 0.85.

Source: European Commission *(48)*.

This is not the situation for UNECE critical levels for sulfur dioxide, which are presented in Box 2 *(1)*. The use of a 24-hour guideline value for sulfur dioxide has been abandoned by UNECE. The annual means of 30 µg/m^3 sulfur dioxide for the protection of agricultural crops and 20 µg/m^3 for the protection of forests and natural vegetation were confirmed in 1992, and should now be applied as winter means (October to March) for relevant vegetation. This is an effective lowering of the targets, and lower values have also been agreed for lichens (10 µg/m^3) and where the accumulated temperature sum above 5 °C is less than 1000 degree-days per year (15 µg/m^3 for natural vegetation and forests). Much of the experimental work referred to in the first edition of the WHO guidelines was also taken into account at the Bad Harzburg meeting *(1)* where these critical levels were first set. Similarly, much of the more recent experimental work described briefly above was presented at Egham in 1992 *(49)*.

Box 2. UNECE critical levels for sulfur dioxide

As set at Bad Harzburg (1)

24-hour mean:	70 μg/m^3
Annual mean:	30 μg/m^3 to protect agricultural crops
	20 μg/m^3 to protect forests and natural vegetation

As set at Egham (49)

24-hour mean abandoned.

Annual means to protect agricultural crops (30 μg/m^3) and forests and natural vegetation (20 μg/m^3) were confirmed. However, these critical level values should be applied in winter (October to March) for relevant vegetation types, such as coniferous trees.

A critical level of 15 μg/m^3 as annual and winter means has been set for natural vegetation and forests in areas of low temperatures. These areas are defined as where the accumulated temperature sum above + 5 °C is < 1000 °C·days per year.

Annual mean to protect certain lichen species: 10 μg/m^3

Critical loads and levels

The critical levels approach has been developed over the last few years in order to define the thresholds of concentrations or dose of gaseous pollutants above which adverse effects occur on plants or other receptors.

The critical loads concept has been developed in parallel with the critical levels approach in order to determine the input values of total sulfur, total nitrogen or total acidity that should not be exceeded if specific receptors are not to be damaged. Gaseous sulfur dioxide entering plant foliage via the stomata may be contributing to an exceedence of critical levels and, since the dry deposition of sulfur dioxide is included in total inputs, such sulfur dioxide will also be included as a component of total deposition for calculation of critical load exceedence.

Like gaseous sulfur dioxide, sulfate ions in mist and in rain are potentially damaging to vegetation, and thus critical levels for the sulfate-sulfur content in mist and rain could be set. As for critical levels of gaseous sulfur dioxide, it is not of practical importance that sulfur inputs in mist and rain may also contribute to indirect effects on vegetation (i.e. soil-mediated) and thus may also be included in critical load calculations. This is because, in order to protect receptors, neither critical levels in air, mist or rainfall nor critical loads of total deposition should be exceeded. Although the major impact of wet-deposited sulfur on terrestrial ecosystems is likely to occur by indirect damage via the soil, it would be possible to extend WHO guideline values to sulfur concentrations or total acidity values (pH) in mist, rain or both, and to total sulfur deposition (critical loads).

Acid mists (aerosols)

The experimental data on trees exposed to acid mist have been examined, and a method proposed for defining critical levels of sulfur for the protection of forests *(50).* Since mist and cloud occur more frequently at higher altitudes, and forests intercept greater amounts of mist, these are likely to be the most sensitive receptors for these inputs.

Just as the dose of a pollutant gas experienced by a plant can be described as the product of concentration and stomatal conductance *(51)* or the product of concentration and duration of exposure, so the dose of pollutants carried in mist can be defined as the product of concentration and contact time *(7).* One problem is that the concentrations of solutes change after rain or mist has been deposited on to leaf surfaces, as evaporation of water occurs *(52).* Experiments have also shown that intermittent exposure to acid mist, as will occur for vegetation growing at cloud base, causes greater effects than continuous exposure at the same chemical composition *(53).* Quantification of effective dose is clearly difficult.

Direct foliar injury can result from polluted, acidic rain but such damage is unusual, occurring only when rainfall pH is less than 3 *(54)*, and ion concentrations in rain are seldom, if ever, sufficient to cause such injury. Direct events are so unusual that it is not necessary to set guideline values for rainwater quality. This is not so for mist (fog and cloud), which can have solute concentrations up to 10 times those in rain. Cape *(7)* collated United States and European measurements of fog and cloud water. These show that minimum pH values in mist range between 2.1 and 2.5 for more than half of the 21 sites at which measurements were made. Four major pollutants, hydrogen, ammonium, nitrate and sulfate ions, make up 80% of the ion composition of most hill cloud, and the four ions are usually present in approximately equal concentrations. The largest ion concentrations in mist occur at cloud base and they decrease thereafter with altitude; the cloud base regions will also experience the most frequent wet–dry cycles. Acid rain and mist may also contain significant concentrations of magnesium and calcium ions. In contrast, the major component of urban fogs is nitric acid *(55).* In regions exposed to hill mist or advected coastal fog, the major land uses are likely to be grass, moorland or forest. Measurement of cloud droplet capture have shown that depositions are as much as four times greater to forest than to shorter vegetation *(56).* Cloud water can contribute to up to half the wet deposition of solutes to upland forest *(57).*

Cape *(7)* undertook a very thorough review of experimental work using simulated acid mist. He concluded that the threshold concentration for visible injury to leaves caused by long-term exposures “may be taken as pH 3, or 500 mmol/litre sulfate”. For red spruce *(Picea rubens)* there is good enough data for a dose–response relationship between acid mist (hydrogen and sulfate ion concentrations) and visible injury. Sulfuric acid has much greater effects than nitric acid for the same pH *(50).* There is also an extensive literature covering acid mist effects on leaf surfaces (wax structure and wettability to water droplets). Here also, sulfuric acid appears more damaging than nitric acid *(58).* Thresholds for response (about 150 mmol/1itre sulfate) are generally lower than for visible injury. It is difficult to evaluate the consequences of leaf surface injury (non-visible) for plant growth and condition. This is also the case for acid mist effects on stomatal function; effects have been reported for mist at pH 3.0 *(59)* but their long-term consequences are not clear. Exposure to acid mist may also render conifer foliage less tolerant of winter cold by decreasing frost hardiness.

The idea that foliar leaching of base cations occurs, resulting from exposure to acid mist and perhaps acid rain, was developed by Krause et al. *(60).* It is now clear that low nutrient status (magnesium, calcium or potassium) is a major and widespread feature of forest declines, but such problems are primarily associated with soil-mediated problems of poor supply rather than with accelerated foliar leaching. In many of the relevant studies, the impacts of these two mechanisms cannot be distinguished. Guide values for total deposition would thus be the appropriate policy to ensure protection of ecosystems from effects resulting from enhanced foliar leaching. For hill land, because duration of cloud cover increases with altitude but ion concentration in cloud decreases with altitude, the product of concentration and exposure duration may be approximately constant with altitude. The pattern of exposure may, therefore, be characterized by concentration × time of exposure.

Cape has suggested using sulfate content rather than acidity to set critical levels for the protection of trees from acid mist. This proposal is based on the dominance of sulfate in the ion composition of mist, and the experimental evidence that sulfate, rather than nitrate, is the main ion responsible for observed effects. Cape *(6)* presents the relationship between atmospheric particulate sulfate content and cloud sulfate concentrations, and suggests that critical levels for mist should be defined by setting threshold values for sulfate particle concentration in air. Such values are currently measured, and modelled sulfate particle concentrations of 1.0–3.3 $\mu g/m^3$ would correspond to concentrations of sulfate in mist of 150–500 mmol/litre. The experimental data suggest that these values are the sulfate concentrations in mist that would protect trees from effects on leaf surface structure and visible foliar lesions, respectively. Cape suggests that these concentration thresholds are appropriate where cloud cover occurs for more than 10% of the time. In essence, the difficulties of measuring cloud water composition can be overcome by measuring or modelling particulate sulfate concentrations in the atmosphere and combining these with data on cloud occurrence. Cape suggests that critical levels of cloud water for forest trees should correspond to non-marine aerosol sulfate concentrations in the range 1.0–3.3 $\mu g/m^3$, depending on the level of protection required. However, these values only apply when calcium and magnesium concentrations in cloud do not exceed hydrogen and ammonium ion concentrations, because no data exist to establish responses under these conditions, such as occur in the Mediterranean, eastern Europe and the Alps.

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