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12

Ground-level ozone

A problem largely ignored in southern Europe

In view of the threat that ozone poses to health and the countries' economies in the Mediterranean region, it is surprising that the matter should have attracted so little political attention.

By
Håkan Pleijel

THE SWEDISH
NGO SECRETARIAT
ON ACID RAIN

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AIR POLLUTION AND CLIMATE SERIES

Ground-level ozone. A problem largely ignored in southern Europe

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Preface

Spain, Portugal, Italy, France, and Greece are among the countries that are most actively opposing the introduction of international measures to reduce the emissions of air pollutants in Europe. Here it is a matter both of a new protocol under the Convention on Long-Range Transboundary Air Pollution and the proposal for EU directives that would reduce the emissions of nitrogen oxides and volatile organic compounds, air pollutants that contribute to the formation of ozone. These latter include a directive for national emission ceilings and a revised version of the directive on large combustion plants.

From the present study, which has been made to show the whole extent of the problem of ground-level ozone in southern Europe, it appears that the concentrations can be very high in that region – often exceeding the levels at which harm can occur to humans as well as vegetation. They are in fact about as high as those in the most polluted parts of central Europe.

These high concentrations do not only affect crop yields and human health, but also pose a threat to the region's important tourist industry. Taking all into account, there is a distinct probability of ground-level ozone causing troublesome economic set-backs for the countries in southern Europe generally.

It must seem paradoxical that just these countries should be opposing international moves from which they would have the most to gain. We hope that this study will help to arouse opinion and so do something to bring about a change in the attitude of the South European governments in this matter.

Göteborg, March 2000

Christer Ågren

Director, The Swedish NGO Secretariat on Acid Rain

Summary

This report concerns the occurrence of ground-level ozone and the effects on vegetation and human health that can be related to ozone in southern Europe. Among the findings:

Ozone is formed under the influence of sunlight on air masses that have become polluted by nitrogen oxides and volatile organic compounds. As in Europe generally, during the last century concentrations have risen markedly in the south. There will, too, occasionally be episodes there with greatly increased and dangerous concentrations. Since the climate in southern Europe is especially favourable to the formation of ozone, strong episodes will occur relatively often during the summer months, with the result that the level of 90 parts per billion, at which EU member countries must inform their citizens of the situation, will frequently be exceeded in those parts.

In southern Europe the most serious problems occur in densely-populated areas, often near the coasts. This is especially the case in Athens and its surroundings, in parts of the coast of eastern Spain, and the Po valley in north Italy. In such places the concentrations may sometimes be as high as 150 to 200 ppb. The highest concentrations also happen to occur, by and large, in the most attractive tourist areas.

Most exposed to health risks connected with ozone are people who spend a lot of time outdoors and those engaged in considerable physical activity, such as athletes, construction workers, and children. Ozone can cause trouble in the respiratory system, with increased risk of infection. The risk is especially great for persons with a generally decreased lung function, such as asthmatics and possibly also smokers. There are moreover indications that high ozone levels can lead to increased mortality. The WHO guide value for health of 60 ppb for an 8-hour average exposure is being extensively exceeded in southern Europe.

There are reports from the Mediterranean region of visible damage, as well as other adverse effects from ozone, on several plant species. Water melon, for instance, is one that appears to be especially sensitive, although damage has also been observed on a number of other plants, such as clover. The most liable to ozone damage are crops that have been watered, which are often the most commercially valuable. The increased availability of water means that the plants also take up more ozone. The critical level that has been set in Europe for damage to agricultural crops from ozone is 3000 ppb hours AOT40, where AOT40 stands for Accumulated exposure Over a concentration Threshold of 40 ppb ozone. That threshold is now being greatly overstepped in southern Europe, with a consequent risk of a drop in production for several crops, including wheat. The southern European tree species about which there have been

the most reports of ozone damage is the Aleppo pine, although some fruit trees such as peaches are also known to be at risk.

A proposal for a new ozone directive was put forward by the EU Commission in June 1999, and a Protocol to Abate Acidification, Eutrophication and Ground-level Ozone was signed by European countries in December of that year. Aimed at reducing the emissions of ozone-forming substances, both of these documents are indicative of the great importance that is now being attached to the problem of ground-level ozone in the European effort to bring about cleaner air.

1. Aim

The aim of this report has been to describe the extent of the occurrence of ground-level ozone in southern Europe and the effects that either have been observed there or may be feared. It will also review the regulations and international agreements applying to ground-level ozone in Europe, as well as the emissions that contribute to its occurrence. Commissioned by the Swedish NGO Secretariat on Acid Rain, it has been made by the Swedish Environmental Research Institute.

2. The ozone problem in southern Europe

2.1. Rising concentrations in the troposphere

There is now general scientific consensus that ozone levels have been rising markedly during the last hundred years in the 10-kilometre high air layer, the troposphere, lying nearest the earth. This has occurred primarily in the industrialized parts of the world (Borrell et al. 1997). Ozone is formed in the troposphere through reactions between nitrogen oxides and hydrocarbons that are dependent on light. These ozone-forming substances are emitted to the air from traffic, industrial processes, and power production. The formation and occurrence of ozone in the troposphere is fully described for instance in Crutzen (1995) and more popularly in Pleijel (1999). The ozone that occurs in the air layer nearest the ground,

Units for ozone concentrations

Ozone concentrations are usually given either in ppb units (parts per billion) or as $\mu g/m^3$ (micrograms per cubic metre). In this report ppb is mainly used, but since $\mu g/m^3$ will also occur, it may be useful to know how conversions can be made.

The ppb unit represents so-called partial pressure, i.e. the proportion of molecules in a volume of air that consists, say, of ozone. In English usage the term "mixing ratio" is often used instead of ppb. The ppb unit gives billionths in the same way as per cent gives hundredths. It has lately become ever more frequent in scientific literature to write nmole mol^{-1} (nanomole per mole) instead of ppb, in order to keep within the SI system where the unit is mole, not ppb. Mole expresses the number of molecules, while nano (10^{-9}) stands for billionths.

The unit $\mu g/m^3$ stands for the mass of ozone molecules in a cubic metre of air. At normal pressure and temperature at sea level the factor for converting ozone from ppb to $\mu g/m^3$ is almost exactly 2, i.e. 1 ppb = 2 $\mu g/m^3$ (or still more closely, 1.96). For other gases the conversion factor will be different, but it can always be calculated with the aid of the universal gas law if the molecular weight, the pressure, and temperature of the gas are known.

At high altitudes the air pressure is on an average very much lower than at sea level. In other words there will be fewer molecules per cubic metre of air. Consequently even if the proportion of molecules in the form of ozone (expressed as ppb) should be the same as at sea level, the total number of ozone molecules, and so their mass, will be markedly less. At a high altitude 1 ppb ozone will be less than 1.96 $\mu g/m^3$.

and so can harm humans, plants, and materials, is generally called ground-level ozone.

2.2. Historical studies of ozone levels in southern Europe

There are measurements of ozone levels in southern Europe dating from the late 1800s and early 1900s, using Schönbein paper or similar methods. Schönbein was the chemist who discovered ozone and devised a reaction paper that could be used to measure for instance the 24-hour levels of ozone. There is naturally some uncertainty as to whether the results of such measurements can now be used to trace the development of ozone levels over time. No living person knows anything of the time and place of the measurements, and the exact details are also often unknown. They do nevertheless show reasonable congruity, and are in any case the best means we have of tracing the course of ozone levels over the past hundred years.

Comparison with the results of modern measuring methods suggests a decided rise in ozone levels in the last 100 years

At Moncalieri in north Italy ozone levels were charted by the Schönbein method during a period of twenty years at the end of the nineteenth century. Comparison of the results with those of modern methods of measurement suggests that there has been a decided rise in ozone levels since that time, from 10 to 25 parts per billion (ppb) as 24-hour average (Anfossi et al. 1991). Taking daytime figures only, the rise would be still greater. These results are in good agreement with those from similar studies made in Paris, where measurements were also made with Schönbein paper in the 1800s (Volz & Kley 1988).

A method similar to Schönbein's, but using De James colometric papers, was employed in Athens during the first half of the last century (from 1901 to 1940). The results were compared with measurements made between 1987 and 1990 by Cartalis and Varotsos (1994). It appeared that the average summertime levels had increased from 28 to 57 ppb during the daytime and from 27 to 48 ppb at night.

Measurements made at Zagreb, Croatia, with Schönbein paper between 1889 and 1900 also give strong evidence, when compared with later series, of greatly increased concentrations – from 36 to 67 ppb during the day and 30 to 56 at night (Lisac & Grubišic 1991).

Measurements by the Schönbein method were made at a high altitude at the end of the 1800s – at the Pic du Midi observatory in the Pyrenees, more than 3000 metres above sea level. A series made towards the end of the 1900s showed that here, too, concentrations had in the meantime risen, from about 10 to 40 ppb (Marenco et al. 1994).

2.3. Episodes with a dangerous increase in exposure

The concentration figures given so far have mainly been for long-term averages. But besides the general rise in ozone levels over large areas, there now occur episodes with very greatly increased concentrations, lasting for the most part from a few hours to a few days. Although these episodes will hardly affect long-term average values, they can be a major cause of damage to health and vegetation. Prior to industrialization, such episodes probably occurred only seldom, if indeed at all. As regards dangerously high concentrations, the rise must therefore be much greater than would appear from the average figures in Section 2.2.

Although there is no generally accepted definition of the level that must be attained if the situation is to be regarded as episodical, most observers would agree that it is definitely a question of an episode when concentrations reach up to 80 ppb. Some do nevertheless consider it to be an episode at 60 ppb. In several parts of southern Europe it is quite usual to have episodes with levels above 90 ppb (see Table 1, page 9). They may even be as high as 150 to 200 ppb.

2.4. Present levels in southern Europe

There is as yet no really acceptable presentation of the occurrence of ground-level ozone in southern Europe. A number of local and even national studies have been made, although with somewhat differing aims. Information can be obtained from the EMEP computer modelling which is used to study the connection between the emissions, transformations, and loads of airborne pollutants as they move about over Europe. Figure 1 shows the extent in 1990 of exposure to ozone concentrations above 3 ppm-hours AOT40 (the critical level for crops) coming from computer modelling. AOT40 is an index of exposure to concentrations exceeding 40 ppm which is employed to relate plant damage to ozone exposure (see also Section 5). As can be seen from the chart, the overexposure is greatest in central Europe as well as in the south of the continent.

The climate of the Mediterranean region is characterized by seasonal changes between hot summers with high pressure, low winds, and strong solar radiation on the one hand and cool, mainly frost-free but rainy winters everywhere except in mountainous parts. The weather in the summer months is favourable to the formation of ozone. The long, stable periods of high pressure, with a relatively low spread of ozone-forming substances, combined with high temperatures and strong sunlight, tend to make ozone formation more of a local phenomenon there than elsewhere in Europe. The fact that the emissions of ozone-forming substances are largely confined to some densely populated parts only accentuates the local character of the ozone problem in the Mediterranean region. Those places where ozone formation is often particularly high include Athens,

Confinement of emissions to densely populated parts accentuates the local character of the problem.

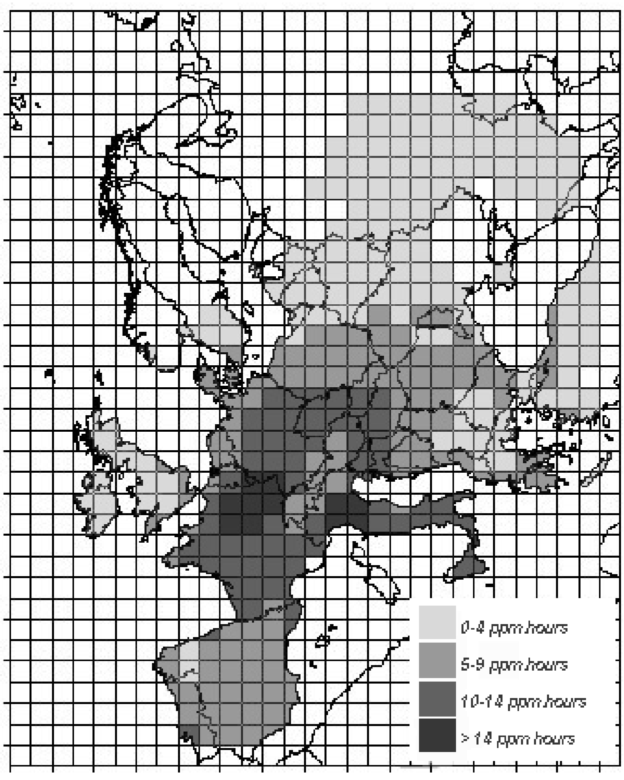

Figure 1. Total exceedings of the critical level for the effect of ozone on farm crops – 3 ppb-hours AOT40 – during the period from May to July. The underlying data is based on the emissions of nitrogen oxides and hydrocarbons in 1990, representing the average of five years' meteorological conditions taken from IIASA (Amann et al. 1999). AOT40 is the accumulated exceeding of the 40 ppb level, calculated as an hourly average (see Section 5).

Rome, Milan and its surroundings, and the coastal region of northeastern Spain, especially around Barcelona.

Intense episodes of photochemical smog similar to those occurring in California.

Detailed studies made in some of the larger urban areas in southern Europe came upon intense episodes of photochemical smog, similar to that occurring in the Los Angeles basin in California, where the problem of ozone first became evident. The ozone levels so revealed were often very high. The Athens area provides a typical example. As in Los Angeles, alternating land and sea breezes no doubt play some part is the rise of this kind of pollution in areas near the coast where emissions are high in general. Examples besides Athens are Barcelona and other parts of eastern Spain, although the meteorological conditions that led to the highest recorded concentrations may have varied somewhat from case to case (Martin et al. 1991; Borell et al. 1997).

Scientists studying the situation in Spain (Millan et al. 1999) have strongly emphasized the role of atmospheric recirculation along the coasts of the western Mediterranean. Here it is a matter of meso-scale phenomena, that is, weather systems operating over a rather limited geographical area. Sea breezes arise during the day when the land near the coast becomes warmed by strong insolation. The air above the land then tends to rise and be replaced by cooler air from the sea. At night radiation causes the surface of the land to cool off more rapidly than that of the sea, since relatively quick recirculation brings warmer sea water up to the surface, with a resulting land breeze, which is usually weaker than a sea breeze. This kind of air movement predominates when there are no other winds to disturb the pattern, as in periods of high pressure.

Weather systems give rise to a type of air circulation that favours the formation of ozone.

The high pressure systems with gentle winds that are frequent in the Mediterranean region give rise in this way to reversing land-sea breezes – a type of air circulation that favours the formation of ozone. During the night the polluted air mass over a city such as Barcelona gets carried out to sea – thus constituting a pool of pollution which is returned to land after sunrise. Fortified by fresh emissions, it can cause intense ozone formation. Depending on the strength and continuity of the sea breeze, the polluted air mass can be conveyed various distances in over land and into the mountain areas that are common behind the coast. Under steady weather conditions this type of recirculation can go on for long periods. Analyses of the chemical-meteorological processes in the air were used at Barcelona in 1992 to decide on traffic regulation at the time of the Olympic Games (Baldasano et al. 1993). Such local weather systems have been characterized by Sanz and Millan (1998) as “large natural photochemical reactors.”

Several marked episodes were noted in the course of a series of measurements carried out in and around Athens in 1984 (Güsten et al. 1988). The maximum concentration was then almost 200 ppb, a very high and harmful level. A number of hourly values over 100 ppb were recorded, especially during the summer of 1992, at Patras, in the Peloponnese, one of the largest Greek cities (Danalatos & Glavas, 1996). Observations made in 1996-97 in the mountains of the northern Peloponnese (about 1100 metres above sea level) revealed however lower concentrations, with long-term average values of 40 to 50 ppb (Glavas 1999).

Greatly raised ozone levels were also found by Bonasoni et al. (1991) around Ravenna, in the lower valley of the Po near the Adriatic. Concentrations were particularly high over the sea, peaking at 100 ppb on summer afternoons, with an afternoon average of 70 ppb. On following days the sea breeze would carry the polluted air masses back over land.

3. Effects on human health

3.1. Effects and groups at risk

Ozone is a reactive and oxidizing substance. Being only moderately soluble in water, on inhalation it can be carried down deep into the lungs. Even a short exposure may be sufficient to cause inflammation in the respiratory tract. The symptoms will however be relieved when exposure ceases. It has been shown by experiment that there can be effects at levels around 80 ppb.

Ozone can cause lowered lung capacity and reduced resistance to infection from bacteria and viruses. Reduction due to exposure can be serious for persons whose lung function is already reduced, as in the case of asthmatics (Bates 1996). Smokers can reasonably be assumed to be sensitive to ozone exposure. Since the efficiency of their lungs will already be affected by smoking, the least extra effect will be more serious than it would be for non-smokers. The high concentrations of ozone occurring nowadays can also cause eye irritation.

Especially at risk are persons who spend much time out-of-doors and are physically very active, such as those engaged in some types of construction work, and athletes, although children must be included in this category, too, since they are also very active and spend a lot of time in the open air. Their high basic metabolism and incompletely developed immune systems may, too, make children extra sensitive to ozone. Short-term exposure to concentrations of 60 to 120 ppb has been shown to effect their lung functioning.

Another group of persons that should not be forgotten are tourists, who often are also much out in the open air. Many of the most popular tourist resorts in southern Europe are just in those densely populated areas near the coast where ozone concentrations are highest.

Epidemiological studies have shown a parallel between admissions to hospital for lung trouble and high levels of ozone in the air. One such study, named APHEA, even found a statistical connection between ozone exposure and mortality, with an average increase in mortality of 2.3 per cent for each increase of 25 ppb in the highest daily ozone value (Touloumi et al. 1997). There are also studies indicating that ozone can increase the risk of cancer, although that has been insufficiently investigated.

3.2. Guide values

Among the guidelines issued by the World Health Organization are some for the levels of air pollutants at which there may be a risk to health. Taking the lowest sure level at which there may be effects, it has arrived at a guide value for ozone of 60 ppb as the maximum average during eight hours (WHO, 1995), while at the same time emphasizing that this does not represent a safe margin for some types of acute effects on the most sensitive persons.

The Swedish Institute for Environmental Medicine recommends a one-hour value of 40 ppb as the upmost limit for human exposure to ozone (Bylin et al. 1996), using a safety factor of 2 in relation to the lowest scientifically determined level for known effect. Many experts in the field of environmental medicine are of the opinion that the lowest threshold for effect, in the form of a risk-free level for ozone, has yet to be found.

3.3. Overstepping of the WHO guide values

The charts in Figures 2 and 3 represent two ways of showing the extent of ozone exposure. Figure 2 shows the number of days per year on which the WHO guide value of 60 ppb was exceeded as an 8-hour average in various parts of Europe. The chart is built on the results of IIASA computer modelling (Amann et al. 1999). Expressed in this way the exposure levels turn out to be very high in some parts of the Mediterranean region.

The chart in Figure 3 illustrates the extent to which AOT60, the total of oversteppings of the 60 ppb level, varied in Europe according to calculations based on the emissions of air pollutants in 1990. In this case it is not a matter of the threshold being exceeded during an 8-hour period, but is simply a summing up of all the 1-hour values when the 60 ppb threshold was crossed. Since it tends not to bring out the big episodes, the effects on southern Europe appear less when mapped in this way.

3.4. EU levels for informing the public

There is a directive, 92/72/EEC, setting the level for ozone at which the public in the EU countries has to be informed of the situation. This is when concentrations exceed 90 ppb as a 1-hour average. There is also a so-called warning level for ozone, requiring the authorities in each country to take action to warn people of the possible health risks. This has been set at 180 ppb, a value that is however seldom exceeded. How often the information level was exceeded during one year in each country can be seen from Table 1, showing it to have been most frequent in southern Europe – where the highest 1-hour average values were recorded, being greatest in Spain and Italy, although Greece and France also had high

Level at which public must be informed is exceeded most frequently in southern Europe.

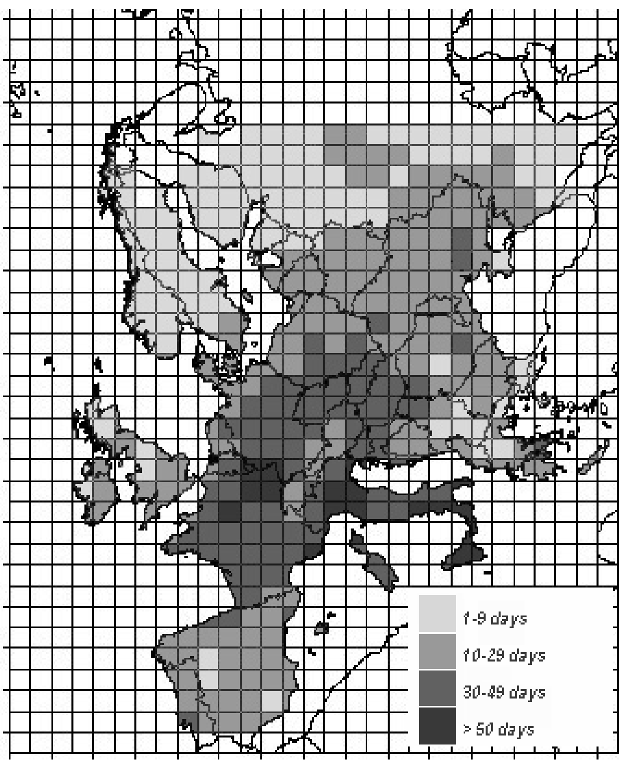

Figure 2. Number of days on which ozone concentrations exceeded 60 ppb as an 8-hour floating value. The underlying data, based on the emissions of air pollutants in 1990, represents floating average values from five years of meteorological observations.

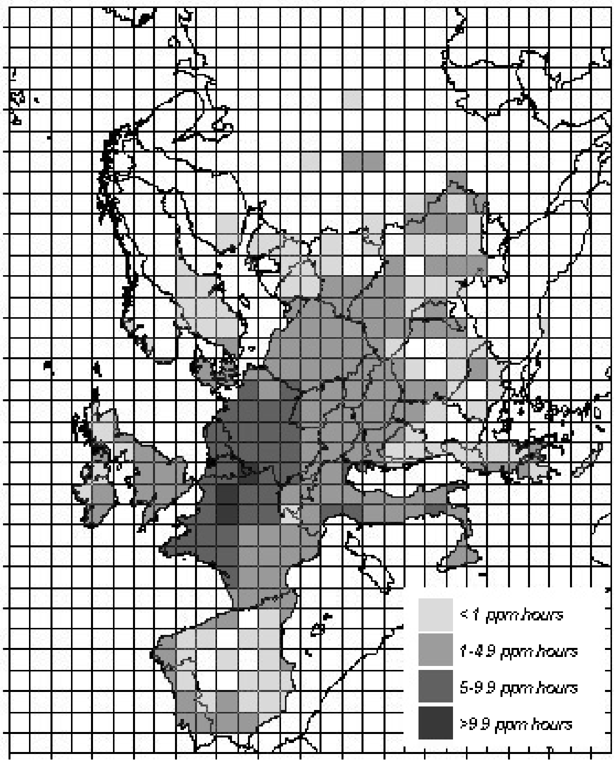

Figure 3. A0T60, the accumulated exceeding of an ozone level of 60 ppb based on 1-hour values and 1990 emissions. The map shows the next-highest values from five years' meteorological observations, as a means of illustrating a high-level situation that is still not extreme as a result of values having been affected by unusual circumstances.

figures. It should be noted that the accuracy in these cases may depend on the number of observation points in any country, and also on where they were situated. The figures should nevertheless give a fairly true picture of the occurrence of ground-level ozone in the EU during the summer of 1999.

Table 1. Exceeding of the ozone level of 90 ppb as a 1-hour average within the EU during the summer of 1999. Preliminary data from Sluyter and Camu (1999).

Country	Number of days with more than 90 ppb at any observation point	Maximum concentration (ppb), 1-hour average
Austria	8	112
Belgium	11	115
Denmark	0	<90
Finland	0	<90
France	42	138
Germany	19	110
Greece	53	152
Ireland	0	<90
Italy	68	170
Luxembourg	2	102
Netherlands	7	126
Portugal	10	121
Spain	40	170
Sweden	0	<90
United Kingdom	9	114
EU-15		170

4. Effects on vegetation, especially as concern southern Europe

4.1. Southern European peculiarities

It is well proven that ozone in the concentrations occurring in the industrialized parts of the world causes damage to vegetation. This appears in the form of visible effects on the leaves, for example of tobacco, beans, various species of clover, water melon, and spinach. The leaves may also age prematurely as a result of exposure to ozone, as in the case of wheat and other species of grain. There may also be a physiological effect, manifesting itself in reduced photosynthesis without any outward signs of damage.

Southern Europe differs in several ways from the rest of the continent in respect of ozone. This is mainly because:

❒ Ozone concentrations tend to be very high in some parts of the region.

❒ During the summer months the climate in southern Europe is very dry.

❒ Some crops, as well as other kinds of plant, are largely confined to the region.

In a dry climate the plants' uptake of ozone is counteracted by the low dampness of the soil and air (expressed as VPD, vapour pressure deficit) spurring the stomata to close. Stomata are small pores in the leaves, which allow gas exchange – the intake of carbon dioxide and transpiration of water vapour. At night the stomata are normally closed, since for lack of light no photosynthesis can then take place. There will be no intake of carbon dioxide, and to minimize the loss of water vapour the stomata are kept closed. They will be closed, too, if conditions are dry. Dry soil makes it impossible for the plant to compensate for the loss of water vapour through the leaves if the stomata should be open. The cost to the plant through water lost, compared with carbon dioxide taken in, would therefore be heavy, and the plant might be forced to react and reduce gas exchange by closing the stomata.

Ozone enters the plant essentially through the stomata, so the uptake will be less if the stomata are closed. Although concentrations are often high in the Mediterranean region, on account of the dry climate the plant's uptake of ozone may not be so great as it would have been in more humid conditions. An important factor in this connection is however that crops in southern Europe often have to be watered, so that the ozone uptake will be much greater than it would have been without irrigation. It is most likely that the worst damage from ozone in Europe occurs in irrigated crops in the south. It can happen that whole fields become more or less knocked out there as a result of severe ozone episodes – something that is hardly ever heard of in other parts of Europe.

Velissariou (1999), who has studied the extensive crop damage that sometimes occurs around Athens as well as in other part of Greece in connection with big ozone episodes, has come to the conclusion that watered crops are much more sensitive than others. Watering is in fact very frequent in the most important agricultural districts of southern Europe. From a study made in Catalonia, northeastern Spain, using tobacco plants with varying degrees of sensitivity as bioindicators, it appeared that ozone damage was greatest in the coastal region, and that air humidity had much to do with the amount of damage that occurred at any given concentration (Gimeno et al. 1995). Similarly Peñuelas et al. (1999) found that the damage to indicator plants was less in Catalonia when the meteorological conditions were such as to favour the closing of the stomata.

4.2. Agricultural crops

One plant that has especially drawn attention on account of the effects of ozone in southern Europe is the water melon, which has been seen to be suffering extensive damage in the Ebro delta in northeastern Spain since as far back as 1976. Experiments have shown there to be a clear relation between air pollution and effects (Reinert et al. 1992; Fernandez-Bayon et al. 1993). Visible damage from ozone, with possible reduction of harvests, has been found generally in Catalonia (Ribas & Peñuelas 1999).

A survey of recorded visible damage to plants in the Mediterranean region was made by Velissariou et al. (1996). It has excellent colour photographs showing the effects of ozone on a number of crops that are of importance in those parts, as well as a map marking the areas where there was clear evidence of visible damage to various types of plant in that area. See Figure 4. The authors lament the lack of dose-response functions for ozone in respect of a number of the region's important crops – a serious lacuna in view of the fact that concentrations are often high in the best growing areas.

Many kinds of clover have been found to be highly sensitive to ozone, developing characteristic visible damage to the leaves after no more than moderate exposure. As in the case of many other species, the damage ap-

Figure 5. Characteristic ozone blight on burrowing clover (*Trifolium subterraneum*), a species much used as a bioindicator in European work on air quality under the LRTAP Convention. Photo Håkan Pleijel.

[PHOTO]

Burrowing clover is an especially sensitive species that is used for monitoring effects.

pears in the form of spots or discolouration between the leaf veins, where the cells first lose their colour and then die and become brownish. An especially sensitive species is burrowing clover (*Trifolium subterraneum*), which quickly shows typical damage (Fig. 5) and so has been much used for monitoring the effects of ozone. Another type, which is also highly sensitive but largely confined to the Mediterranean region, is Egyptian clover (*Trifolium alexandrinum*). It has been proposed as a suitable indicator for those parts (Velissariou & Kyriazi, 1996). In Italy some kinds of wheat have found to suffer from exposure to ozone (Badiani et al. 1996b), just as they have in other parts of Europe.

There are strong indications that the ozone is also having effects on crops in North Africa, on the southern side of the Mediterranean – as shown for instance by experiments with radishes and turnips in Egypt, near Alexandria (Hassan et al. 1995). There high concentrations were recorded, reaching up to 67 ppb as a 6-hour average value.

4.3. Trees

A species that is seen to have suffered damage from ozone, both in Greece (Velissariou et al. 1992b) and Spain (Elvira & Gimeno 1996; Elvira et al. 1998; Sanz et al. 1999), and in Israel (Naveh et al. 1980) is the Aleppo pine (*Pinus halepensis*). This is the most widespread species of pine in the Mediterranean region. The damage appears chiefly as visible symptoms on the needles, but also as lower levels of chlorophyll (in other words, bleaching).

The effects of ozone have also been seen on several kinds of fruit trees that are common in southern Europe, such as the peach, which seems to be relatively sensitive to this pollutant (Badiani et al. 1996a). Here a special biological mechanism is involved. It has been proved that even a moderate rise in the ozone level will result in increased attacks of a troublesome rust fungus (Badiani et al. 1993).

4.4. The effect of breeding on crops' sensitivity to ozone

An important study of the trend of crops' sensitivity to ozone during the last century has been made with a number of Greek wheat species. It involved exposing several varieties that had been brought onto the market between 1932 and 1980 (Velissariou et al.1992a; Barnes et al. 1990). It turned out that the effects of ozone became greater as new varieties were successively introduced. Increased yield was accompanied by increased sensitivity. Although the harvest per hectare may be greater, it is probable that the adverse effects of ozone are now also greater, in terms of a percentage reduction of growth. The increase in yield is nevertheless much greater than any drop due to the effects of ozone. It seems the plant breeders have unintentionally increased the sensitivity to ozone, probably because the increase in yield is accompanied by an increase in gas exchange, which in turn will lead to a greater uptake of ozone. Good growth, no matter whether it is due to improved varieties or to favourable conditions in the way of the availability of nutrients and water, tends in general to bring an increase in plants' sensitivity to ozone.

5. Critical levels for ozone

Critical levels for ozone have been developed in Europe through cooperation between experts working under the Convention on Long-Range Transboundary Air Pollution. The levels now agreed upon are the result of an international conference of scientists in Kuopio, Finland, in 1996 (Kärenlampi & Skärby, 1996). They have since been reviewed in the scientific press (Fuhrer et al. 1997) and are set forth in Table 2. In this case the exposure index for ozone is AOT40 (Accumulated exposure Over a concentration Threshold of 40 ppb). For the way AOT40 is calculated, see box.

Table 2. Critical levels for ozone emanating from the conference held in Kuopio, Finland, in 1996. VDP, vapour pressure deficit, is a measure of the drying effect of the air on plants. From Kärenlampi & Skärby (1996).

Type of vegetation	AOT40 value	Time period, etc.
Crops	3 000 ppb hours	May-July
Forest trees	10 000 ppb hours	April-September
Acute effects, visible damage	500 ppb hours 200 ppb hours	five days, VPD >1.5 kPa five days, VPD <1.5 kPa
Semi-natural vegetation (mainly wild herbs and grasses)	3 000 ppb hours	May-July

Calculation of AOT40

This is done as follows. Assume average hourly values for ozone of 35, 38, 40, 41, 42, 45, 50 ppb. The first three will not affect the exposure index because they do not exceed the 40 ppb level by at least 1 ppb. Only the last four contribute to A0T40. The value will be (41-40) + (42-40) + (45-40) + (50-40) = 1+2+5+l0 = 18 ppb hours. For agricultural crops the summing-up period is three months from May to July, and for forest trees six months from April to September. Since plants take up ozone mainly during daylight hours, when the stomata are open, AOT40 is calculated only for the hours between sunrise and sunset.

The work on critical levels is now characterized by an attempt to estimate the actual uptake of ozone. The AOT40 approach rests only on concentrations in the air surrounding the plant. From what has been said above it must be evident that a change will be needed, to an exposure index related to ozone uptake. Moves in that direction have barely begun; some practical result can however be expected within the next 3 or 4 years (Fuhrer & Achermann, 1999).

6. Proposal for a new ozone directive

In June 1999 the EU Commission put forward a proposal for a new directive on ground-level ozone entitled *Proposal for a Directive relating to ozone in ambient air* (COM(1999)125 final), thereby bringing the EU more actively into the process of attacking the problem in Europe. The Commission's proposal takes account of the effects of ozone both on health and vegetation, with long-term aims as shown in Table 3.

Table 3. Long-term objectives and target values for 2010 in the proposed special directive for ozone (CEC, 1999).

To protect	Long-term objectives	Target value 2010
Human health[1]	60 ppb as an average during 8 hours	The average of 60 ppb during 8 hours may not be exceeded on more than 20 days of the year
Vegetation[2]	AOT40 = 3 000 ppb hours	AOT40 = 8 500 ppb hours

[1] Average for 3 years. [2] Average for 5 years.

The critical level for crops will also protect forest trees as well as other types of vegetation.

Those aims are based on values put forward by the World Health Organization, which are meant to apply throughout the European Union. As regards plants, WHO has selected the value for long-term effects on crops emanating from the work on critical loads that has been done within the Convention on Long-Range Transboundary Air Pollution. The critical level for crops is taken to be the strictest, since it will also protect forest trees as well as other kinds of non-agricultural vegetation. Actually the Commission was of the opinion that the proposed aims could not be attained in the medium term. It has therefore not set any definite date for their fulfillment. They are however intended to serve as benchmarks on which to evaluate progress, and to emphasize high intentions in the long run.

In the strategy for ozone that it put forward simultaneously with its proposal for a new directive, the Commission named the following as partial aims to be attained by 2010:

❒ For the protection of human health. A reduction by at least two-thirds from the extent by which the proposed long-term figures has been exceeded in 1990.

❒ For the protection of vegetation. A reduction by at least one-third.

❒ Uniform ceilings of 2.9 ppm hours for the health-related AOT60 level and 10 ppm hours for AOT40 above the critical level of 3 ppm hours for vegetation.

These aims are proposed to be reviewed at the end of 2004. They have been converted to the values (target values) in Table 3 by means of computer modelling and scenario analyses carried out by IIASA. By "target values" is meant the quality of air that should as far as possible be attained within a certain time.

Simultaneously with the presentation of its proposals for an ozone strategy and a new directive for ozone, the Commission put forward a proposal in the form of a directive on national ceilings for ozone-forming and acidifying air pollutants (CEC, 1999). The reductions that would be involved, which on the whole go beyond those recently agreed upon under the LRTAP Convention (see Section 7), are also proposed to be achieved by 2010. If the EU member countries should bring down their emissions under the proposed ceilings, the interim targets for environmental quality would be attained, as well as the partial aims for reducing acidification in the EU. The environmental improvement would be considerable. Both proposals are now going through their mill, a process that usually takes about two years.

7. Another international move

In December 1999 twenty-five European countries signed, together with the United States and Canada, a new protocol under the Convention on Long-Range Transboundary Air Pollution. Entitled *The Protocol to Abate Acidification, Eutrophication and Ground-level Ozone*, it covers four pollutants: sulphur dioxide, nitrogen oxides, volatile organic compounds,

Table 4. Emissions of the ozone-forming air pollutants nitrogen oxides (NOx) and volatile organic compounds (VOCs) in 1990 and according to the reference scenario (REF) and the new LRTAP protocol (PRO) by 2010. Figures are from Amann et al. (1999) and the protocol itself (UN ECE 1999). Kilotons.

	Nitrogen oxides			Volatile Organic Compounds		
Country	**1990**	**REF**	**PRO**	**1990**	**REF**	**PRO**
Austria	192	103	107	352	205	159
Belgium	351	191	181	374	193	144
Denmark	274	128	127	182	85	85
Finland	276	152	170	213	110	130
France	1867	858	860	2382	1223	1100
Germany	2662	1184	1081	3122	1137	995
Greece	345	344	344	336	267	261
Ireland	113	70	65	110	55	55
Italy	2 037	1130	1000	2055	1159	1159
Luxembourg	22	10	11	19	7	9
Netherlands	542	280	266	490	233	191
Portugal	208	177	260	212	144	202
Spain	1 162	847	847	1008	669	669
Sweden	338	190	148	511	290	241
U.K.	2839	1186	1181	2667	1351	1200
EU-15	13226	6849	6671	14031	7128	6600

and ammonia. Besides their effects in the way of acidification and eutrophication, the risks of ground-level ozone for plants and human health have also been taken into account in this protocol. How it is expected to perform, country by country, in respect of the emissions of ozone-forming substances is shown in Table 4, which also gives comparisons with emissions as they were in 1990 and are expected to be as a result of measures either planned or already taken in each country. (REF is the reference scenario.)

8. Biogenic emissions of ozone-forming substances

It may be worth noting that there are also natural emissions of ozone-forming substances. These come mainly from organisms in the soil, from growing vegetation and forest fires (Simpson et al. 1999). Although natural, they are partly a consequence of human activity, being dependent on the crops that are cultivated, the kinds of tree that are planted, fertilization, and the dampness of the soil. How much they amount to is uncertain. It seems the proportion of natural emissions of nitrogen oxides is small (about 10 per cent) in comparison with those of the anthropogenic kind, which are mainly a result of combustion. It is on the other hand a known fact that some kinds of plants give off relatively large amounts of a substance called isoprene (Simpson et al. 1999). Since emissions are stimulated by hot weather, they can be considerable in southern Europe.

Plants' emissions of volatile organic compounds can also contribute to ozone formation.

Furthermore, other kinds of plants give off other volatile organic compounds, including terpenes, the overall emissions of which are probably of the same order as those of isoprene. Both of these substances can in principle contribute to the formation of ozone. Terpenes take part however in somewhat different types of chemical and physical process in the atmosphere. Unlike isoprene they can consume ozone to a considerable extent, and also form light aerosols (a kind of particle), thus no longer contributing to ozone formation. A general conclusion must therefore be that isoprene is of greater importance to ozone forming than terpenes are. A sensitivity analysis to ascertain the importance of isoprene in anti-ozone strategies have shown that its emissions do not affect the load to any great extent in respect of long-term average values (Simpson, 1995). The same study suggested on the other hand that the maximum ozone levels would be more likely to be affected by isoprene. It was found that in Spain, for instance, the assumption of very high isoprene emissions would mean a great reduction of the relative effect of anthropogenic emissions on the highest ozone concentrations.

9. Conclusions

There is a great amount of data indicating that ozone is a serious problem for southern Europe, and that concentrations have risen greatly in the past hundred years. Episodes with concentrations high enough to be definitely harmful to health often occur in built-up areas near the coasts. The irrigated crops in some parts of the Mediterranean region may well be the worst affected of any type of vegetation in Europe, although there are parts of central Europe where exposure to ozone is also very high. Recently a number of studies have shown ground-level ozone to be a particular problem for Spain, Italy, and Greece.

Levels sufficient to harm health occur regularly in several parts of the Mediterranean region, and especially in some thickly populated parts of Spain (east coast) and northern Italy (around Milan and in the valley of the Po), as well as in Athens and its surroundings. In these places the environmental situation would be improved, both for the local population and visiting tourists, if the emissions of ozone-forming substances from traffic and industries were reduced.

Emissions will also have to be reduced in order to save vegetation. Studies have revealed considerable damage to it in southern Europe, and although the risks have yet to be fully evaluated, there are indications that the effects may be even greater than is now believed. Continued research will thus be needed for a proper estimation of the size of the problem. As yet the dose-response connection is lacking for a number of important crops in the Mediterranean region. It is just this kind of information that will also be needed for making an economic evaluation of the situation – an evaluation that is especially called for in view of the extent of the damage, both potential and already documented.

It would also have been valuable to have had analyses in depth of the total situation in respect of ozone in southern Europe – one in which air-quality data and the outcome of research were put together in a collective south-European perspective, with reconsideration then of the conditions peculiar to the region. Studies of this kind have previously leant towards central and northern Europe, despite the problems of ground-level ozone being just as great, if not greater, in southern Europe. Of late, important information in this respect has nevertheless been forthcoming.

In view of all that has now been said, it must seem surprising that reluctance to institute measures for the reduction of the emissions of ozone-forming air pollutants should be greater in southern Europe than it is further north.

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This study is the first to present a comprehensive view of the situation as regards ground-level ozone in the Mediterranean region, showing that the concentrations often exceed the levels at which harm can occur both to humans and vegetation.

Despite the threat posed by ozone to agriculture and tourism, which are so important to the economies of the region, governments there are stubbornly opposing international efforts to bring about reductions of the emissions of air pollutants that lead to the formation of ozone.

Håkan Pleijel, Ph.D. in plant physiology, has long been engaged in ozone research, and has been associated with the international work for determining critical concentrations of ozone in the atmosphere. He has also been acting as representative of the European Environmental Bureau in the group of experts appointed to develop a new ozone directive for the European Commission.

The Swedish NGO Secretariat on Acid Rain

The essential aim of the Swedish NGO Secretariat on Acid Rain is to promote awareness of the problems associated with air pollution, and thus, in part as a result of public pressure, to bring about the required reduction of the emissions of air pollutants. The eventual aim is to have those emissions brought down to levels – the so-called critical loads – that the environment can tolerate without suffering damage.

In furtherance of these aims, the secretariat operates as follows, by

- Keeping under observation political trends and scientific developments.
- Acting as an information centre, primarily for European environmentalist organizations, but also for the media, authorities, and researchers.
- Publishing a magazine, Acid News, which is issued four to five times a year and is distributed free of charge.
- Producing and distributing information material.
- Supporting environmentalist bodies in other countries by various means, both financial and other, in their work towards common ends.
- Acting as coordinator of the international activities, including lobbying, of European environmentalist organizations, as for instance in connection with the meetings of the bodies responsible for international conventions, such as the Convention on Long Range Transboundary Air Pollution.
- Acting as an observer at the proceedings involving international agreements for reducing the emissions of greenhouse gases.

The work of the secretariat is largely directed on the one hand towards eastern Europe, especially Poland, the Baltic States, Russia, and the Czech Republic, and on the other towards the European Union and its member countries. By emitting large amounts of sulphur and nitrogen compounds, all these countries add significantly to acid depositions over Sweden.

As regards the eastern European countries, activity mostly takes the form of supporting and cooperating with the local environmentalist movements. Since 1988, for instance, financial support has been given towards maintaining information centres on energy, transport, and air pollution. All are run by local environmentalist organizations.

To date, four European conferences on strategy for environmental NGOs have been arranged by the secretariat, where common objectives and cooperative projects were developed. An important outcome has also been the agreement on the demands, based on scientific data concerning critical loads.