Ozone in the lower atmosphere causes damage to plants and affects human health. It also contributes to the greenhouse effect and damages materials. These problems are a major consideration in current European negotiations on transboundary air pollution.

Ground-level ozone is formed from nitrogen oxides and hydrocarbons under the influence of sunlight. Transport is the single most important source of these pollutants, although energy production and various types of industry also account for significant emissions. Concentrations of ground-level ozone have risen substantially in the course of the 20th century. In addition, ozone ‘episodes’ – short periods in which ozone levels are greatly increased – sometimes occur, chiefly in the spring and summer.

This report describes the mechanisms of ozone formation, current levels of ozone in Sweden, and national and international environmental objectives relating to ground-level ozone. The emphasis is on ozone’s effects on plants. On the basis of a wide range of experiments carried out in Sweden over the last 15 years, the report describes how ozone affects agricultural crops, forest trees and wild plants.
Ground-Level Ozone

– A Threat to Vegetation

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The authors assume sole responsibility for the contents of this report, which therefore cannot be cited as representing the views of the Swedish Environmental Protection Agency. The report has been submitted to external referees for review.

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PREFACE

This report presents the final results of a series of experiments investigating the effects of ground-level ozone on plants, conducted since 1984 by the Swedish Environmental Research Institute (IVL) and the Botanical Institute at Göteborg University. Between 1985 and 1989, a field chamber experiment was carried out at Rörvik, some 40 km south of Göteborg, in which Norway spruces were grown at different ozone concentrations. This project was initiated by the power and district heating and forest products industries. Here we would therefore like to emphasize the important part played by Lars Lundgren, together with the late Hans Lundberg and the late Rolf Brännland, in developing this field of research. In 1987, experiments were started at Östads säteri, roughly 40 km north-east of Göteborg, to study the effects of ozone on field-grown agricultural crops. In 1990 the experiments on forest trees were moved to Östad. A larger-scale open-top chamber experiment was set up to study the effects of ozone, drought and phosphorus deficiency on Norway spruce, and this project continued up to and including 1996. 1994 saw the launch of the EC project ESPACE-Wheat, which studied the effects of elevated carbon dioxide concentrations combined with ozone and water availability on field-grown wheat over a period of three years. Between 1994 and 1996 research was also conducted into the effects of ozone on wild plants, and for this project, too, open-top chambers at Östad were used. In addition to this work, a series of experiments on pot-grown indicator plants for ozone have been undertaken at Östad since as early as 1988, in the framework of the Convention on Long-Range Transboundary Air Pollution (CLRTAP). An experiment to study the ozone sensitivity of birch was established in 1997.

We would like to thank all the researchers, in Sweden and elsewhere, who have made important contributions to the experimental work carried on at Östad. In particular we would mention Dr D. Tingey, at the US EPA Corvallis, Prof. H. Sandemann, of GSF Munich, A. Berglen Eriksen, Senior Scientific Officer at the Phytotron, Oslo, Dr K. Ojanperä, at the Finnish Central Union of Agricultural Producers and Forest Owners, Jokioinen, and M. Werner MSc, of the Forestry Research Institute of Sweden.

Patrik Alströmer and Östads säteri played an active part in enabling the experiments to be conducted, by making available land, personnel and machinery, as well as contributing financially to the research relating to forest trees. We would like to express our special thanks to them for this. We are also greatly indebted to Cederroth International AB for the specially formulated nutrient solution used for the spruce saplings at Östad.

Finally, the authors wish to express their sincere thanks to Ulla Bertills at the Swedish Environmental Protection Agency for her enthusiastic involvement in their research, for her valuable comments and for the unstinting effort she has devoted to the preparation of this report.

GÖTEBORG, 19 APRIL 1999
THE OZONE RESEARCH GROUP
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EXECUTIVE SUMMARY

Ozone in the lower atmosphere causes damage to plants and poses a threat to human health. It also damages materials and, furthermore, acts as a 'greenhouse gas', contributing to the greenhouse effect. Ozone forms, under the influence of sunlight, in air masses polluted with nitrogen oxides and hydrocarbons. The single most important source of these ozone-forming compounds (ozone precursors) is transport, but industry and energy production also contribute. Emissions of ozone precursors have begun to fall in Europe and this trend may be expected to continue over the next ten years. Consequently, levels of ozone over the continent have probably also started to decrease. To some extent, ozone precursors disperse throughout the northern hemisphere, resulting in an elevated background concentration of ozone. Some researchers take the view that this background level is rising, while others believe that it is more or less constant.

There is now strong scientific evidence that production of sensitive agricultural crops, such as wheat and beans, is declining as a result of the ozone levels occurring across large areas of Europe. This is also the case in southern Sweden. Other plants, including various types of clover, spinach and tobacco, exhibit characteristic visible leaf injuries following ozone episodes, i.e. short periods of very high ozone concentrations. Such episodes chiefly occur in spring and summer in conjunction with high-pressure systems and sunny weather, when polluted air masses reach Sweden from more southerly areas of Europe. The main substances involved in ozone formation, which is a light-dependent process, are nitrogen oxides and certain volatile organic compounds. In southern Europe, ozone concentrations are locally greatly elevated from time to time during the warmer months of the year, and this can have very significant effects on certain crops, such as watermelon.

In various parts of the world, there is also evidence of ozone causing damage to forest trees. Here, though, the scientific case is not as compelling as in relation to agricultural crops, since most experimental studies have only involved young trees. The forests of the San Bernardino Mountains, south of Los Angeles, have suffered extensive ozone damage since the 1960s. Ponderosa (western yellow) pine especially has exhibited both significant visible injury symptoms and reduced production as a result of the very high ozone concentrations there. Ozone damage to forests has also been reported from the Mediterranean region.
In Spain, visible injury attributable to ozone has been observed in Aleppo pine, both in forest stands and in controlled experiments. In Sweden, detailed studies have been made of ozone’s effects on Norway spruce and silver birch. Following exposure to ozone, spruce needles that were more than two years old showed a decline in photosynthesis and chlorophyll concentrations. After four seasons, production was 5% lower in ozone-exposed spruce saplings than in a control involving preindustrial levels of ozone. Even cautious assumptions about the extent to which ground-level ozone interferes with the growth processes of forest stands suggest that present-day ozone concentrations will have an adverse effect of up to 10% on production in southern Swedish forests over an entire rotation. In a similar experiment on birch seedlings, ozone caused premature leaf-fall. The leaves were thus shed before their nitrogen content was returned to the stem, preventing the young trees from utilizing this nitrogen. In addition, after one season the birches exposed to ozone showed a decrease in the biomass of their roots. In the long term, high ozone concentrations probably depress production in birches. Reduced root growth may be assumed to restrict nutrient uptake, at any rate on less fertile soils, and earlier leaf-fall, combined with reduced withdrawal of nitrogen from the leaves to the stem, presumably further accentuates any nutrient deficiency.

Relatively little is known about the effects of ozone on wild plants. The enormous number of species involved is a major difficulty in this context. The experiments which have been carried out suggest that fast-growing species tend to be more susceptible to ozone than slow-growing ones. Quite a large number of wild species do not appear to be particularly sensitive to moderate increases in ozone concentrations. It should be borne in mind, though, that in the majority of ecosystems different herbs and grasses are in fierce competition with one another. This means that even fairly small differences in ozone sensitivity can result in shifts in the relative abundance of individual species if they are exposed to ozone.

The harmful effects of ground-level ozone on plants, health and materials are now an important driving force behind the international negotiations to reduce emissions of transboundary air pollutants in Europe. It is in this context that ‘critical levels’ of ozone have been formulated. These critical levels are currently based on the AOT40 concept (Accumulated exposure Over the Threshold 40 ppb ozone). AOT40 is calculated by adding together the amounts by which ozone concentrations exceed a level of 40 ppb. The purpose of the critical levels defined is to identify areas of Europe where ozone is having significant effects on vegetation. Critical ozone levels are currently exceeded in southern Sweden and, in the case of agricultural crops, to some extent in the north of the country as well.
There is now a shift in focus towards the amount of ozone taken up by plants, rather than simply the concentration in the ambient air. Sweden’s climate is favourable to ozone uptake. Long summer days and relatively high humidity mean that plants’ stomata (the tiny pores in their leaves) are open for long periods, resulting in comparatively high uptake of ozone. A given concentration may therefore have a more marked effect in this country than in an area with a drier climate and shorter days further south in Europe.

Over the next few years, research is likely to show that the exposure–response relationships that have been calculated on the basis of ozone concentrations overestimate the effects of ozone in southern Europe and underestimate its effects in the Nordic region. Preliminary estimates of ozone uptake in different parts of Europe point unequivocally in this direction.

Most of the studies presented in this report were carried out at Östads säteri. The experimental site can be seen at the bottom right of the picture. In the background, part of Lake Mjörn. Photograph: Svante Hultengren/Naturcentrum AB.

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ppb – a unit to express ozone concentrations

Concentrations of ozone in air are usually expressed in either ppb or µg/m³. In this report we use the unit ppb, which expresses what is known as a partial pressure, or the proportion of the molecules in a parcel of air which are ozone molecules. The abbreviation ppb stands for ‘parts per billion’, i.e. the number of billionths (thousand millionths) of the air molecules present which consist of ozone.

The unit µg/m³ states the mass of ozone molecules present in a cubic metre of air, in micrograms (millionths of a gram). At normal pressure and temperature, the conversion factor is more or less exactly 2, i.e. 1 ppb O₃ = 2 µg O₃/m³, but to convert precisely from one unit to the other, pressure and temperature have to be taken into account. At high altitudes, the pressure is always lower and 1 ppb O₃ corresponds to less than 2 µg/m³.
Summary

Ozone is a factor behind a number of environmental problems. In the lower atmosphere, ozone formation is currently occurring as a result of emissions of nitrogen oxides and volatile organic compounds, and here the gas is affecting plants and human health and causing corrosion of materials. Ozone also contributes to the greenhouse effect. Periods of greatly increased ozone concentrations in the lower atmosphere are known as episodes. They chiefly occur in conjunction with stable high-pressure systems in summer, especially if air masses from the continent are involved. In areas with heavy traffic, ozone levels are locally lower, since ozone reacts rapidly with the nitric oxide from vehicle emissions. The majority of the ozone in the atmosphere occurs in the ‘ozone layer’ of the stratosphere, at altitudes of about 10–40 km. Here, ozone levels are currently falling, owing to emissions of certain ozone-depleting substances, including CFCs (chlorofluorocarbons). This is a cause for serious concern, given that the stratospheric ozone layer protects life on earth from ultraviolet radiation.

1.1 Introduction

Ground-level ozone is a typical example of a regional pollutant. It forms when nitrogen oxides and volatile organic compounds undergo chemical reactions in the atmosphere under the influence of sunlight. It is no respecter of national boundaries and therefore constitutes an international problem. Ozone in the lower atmosphere has a number of different environmental effects. Apart from its effects on plants, which are dealt with in detail in this report, ozone at elevated concentrations also adversely affects human health (Bylin et al. 1996) and causes degradation of a variety of materials. Ozone is in addition a ‘greenhouse gas’, contributing to the greenhouse effect.

In the United States, and especially in southern California, the problem of ozone attracted attention in the late 1940s. Emissions from road traffic were high even at that time and, in conjunction with the special climatic conditions
of the Los Angeles basin, they were giving rise to a mix of pollutants that is generally referred to as photochemical smog, or Los Angeles smog. The most important component of this smog is ozone, although it also includes a range of other pollutants, some of them particles which radically restrict visibility (figure 1.1). In Europe, too, high ozone levels are often associated with reduced visibility.

![Figure 1.1. Photochemical smog in the San Bernardino Mountains in southern California. To the left is Paul Miller, a pioneer of research into the effects of ozone on forest trees. As early as the 1960s, he demonstrated the link between ozone and damage to ponderosa pine in the mountains south of Los Angeles. Photograph: Lena Skärby/IVL.](image)

In Europe, extensive research collaboration relating to transboundary air pollutants and their chemistry has taken place since 1988 as part of the EUROTRAC project. In this forum, ozone formation has been seen as highly relevant in terms of effects on plants, human health, materials and climate (Borrell et al. 1997). Ozone-forming compounds (ozone precursors) and ground-level ozone feature very prominently in a protocol covering several different transboundary pollutants, currently being drawn up under the ECE Convention on Long-Range Transboundary Air Pollution. It is as part of the process of establishing a scientific basis for this protocol that ‘critical levels’ for the effects of ozone on plants have been identified. These critical levels are presented in chapter 2. In parallel with this work, the European Union (EU) is currently preparing a new directive on ground-level ozone. Control strategies will be linked to both the protocol and the directive, laying down how member states are to set about reducing ozone levels.
In several countries of Europe, the ozone issue is as serious a concern as acidification, for example. In this perspective, the health effects of ozone are a key factor, alongside its effects on plants. Compared with northern Europe, the countries of southern parts of the continent have less acid-sensitive soils and often appreciably higher ozone concentrations, and the balance of environmental concerns therefore differs. Nevertheless, ground-level ozone is a major environmental threat in Sweden, too, at least in the south of the country.

There are several common misunderstandings regarding ozone, its occurrence and its environmental impacts. This is mainly because ozone forms in different ways, and has differing consequences for organisms on the earth’s surface, depending on where in the atmosphere it is found. To understand the issues involved, a historical, evolutionary approach may be helpful.

1.2 Oxygen, ozone and life on earth

When life on earth began, some 3500 million years ago, the atmosphere probably contained no oxygen, or very little (Wayne 1985). Gradually, organisms capable of photosynthesis evolved, presumably a type of blue-green algae (cyanobacteria). Oxygen gas, or molecular oxygen \( (O_2) \), is a by-product of photosynthesis. To begin with, virtually all the molecular oxygen generated by photosynthesis was consumed by inorganic processes: the earth’s surface offered a plentiful supply of substances which it could oxidize, including sulphides and reduced (ferrous) iron. Around 2000 million years ago, there was little such material left to be oxidized, and from that point on oxygen gas began to accumulate in the atmosphere (Westbroek 1991). This had several important consequences, revolutionizing life on earth.

In many respects, oxygen is a poison. The gas itself and its by-products can oxidize molecules in the cells of organisms, which are then altered and cease to function. A significant part of the metabolism of an animal or plant is concerned with providing protection against the harmful effects of oxidizing agents. A key role in this protective system is played by substances known as antioxidants. Vitamin C \( (\text{ascorbic acid}) \) is one example of such a substance. Green plants in particular need to protect themselves against oxygen and other strong oxidants, since those substances are produced by photosynthesis. In certain respects, the life of a green plant is a balancing act between efficient production based on photosynthesis and a risk of self-poisoning by the by-products of that process. Some researchers believe that life on earth could not have come about if the atmosphere had contained oxygen gas from the outset. At the same time, the high concentration of oxygen in the atmosphere is the clearest sign that the earth is a planet supporting life.
Oxygen gas, then, can be dangerous because of its oxidizing capacity, but ozone is a far more powerful oxidant. Admittedly it occurs at much lower concentrations, but it is more reactive, which is what makes it so harmful. Ozone is known to be capable of altering the chemical structures of fatty acids, modifying their chemical properties and possibly disturbing their functioning. Fatty acids are among the most important components of the membranes which surround cells and cellular organelles and divide these structures from their surroundings. Ozone can also modify the chemical structures of proteins, which is a serious matter, since proteins of different kinds play a key part in the structure and metabolism of every organism. The differing functions of individual proteins are closely linked to their specific chemical structures.

Gradually, the earth’s organisms adapted to life in a world of high concentrations of molecular oxygen. Furthermore, some organisms began to exploit the considerable energy potential which lay in using oxygen gas to oxidize the organic matter produced by photosynthesis. This process forms the basis for the supply of energy to the earth’s ecosystems at the present stage in the development of life, which began when the atmosphere started to contain large quantities of oxygen gas (Goldsmith & Owen 1992). Animals, fungi and various other organisms are dependent on the oxygen produced by green plants, and the concentration of molecular oxygen in the atmosphere thus needs to be fairly high; at present it is around 21%. It should be remembered, though, that 21% is not necessarily an optimum level for living organisms in every respect. It can be shown experimentally, for instance, that a lower oxygen concentration induces higher growth in plants, owing to a reduced oxidative stress. A higher concentration than that currently prevailing results in fires burning more vigorously, which results in oxygen being consumed. In other words, some sort of natural regulatory mechanism is in operation.

Had it not been for the change in the earth’s environment brought about by molecular oxygen, it is likely that life would have been a less prominent feature of this planet and would have consisted chiefly in mats of bacteria in the oceans. There are at least two reasons why oxygen was so important. Firstly, photosynthesis and respiration based on oxygen created conditions for far greater and more rapid conversion of energy than would otherwise have been possible. And secondly, the presence of oxygen in the atmosphere resulted in ozone beginning to form in the stratosphere. Ozone absorbs UV-B, an ultraviolet component of solar radiation which is very harmful to the majority of living organisms. Ozone formation therefore made it possible for life – previously concentrated in the protective oceans – to emerge onto the land. The ‘ozone layer’, which provides a shield against UV-B radiation, thus arose as an indirect
consequence of the accumulation of gaseous oxygen in the atmosphere. A major threat to the environment today is thinning of the stratospheric ozone layer at altitudes of around 15–40 km, so far primarily over the Antarctic, but to some extent also over other parts of the globe.

1.3 Ozone formation of ozone in the stratosphere and the troposphere

The chemical basis for the formation of ozone (O₃) is single atoms of oxygen (O). In the stratosphere, the latter can arise when short-wavelength and therefore high-energy ultraviolet rays present in sunlight break down molecules of oxygen (O₂):

\[ O_2 + h\nu \xrightarrow{\lambda < 242\text{ nm}} 2O \]  

Here, the symbol \( h\nu \) indicates that the reaction is dependent on light and the inequality above the arrow shows that the light must have a wavelength of less than 242 nm in order to drive the reaction. The shorter the wavelength, the higher the energy of the radiation. Ozone is then formed by single oxygen atoms combining with oxygen molecules:

\[ O + O_2 \rightarrow O_3 \]  

The wavelengths in sunlight which are capable of driving reaction (1) do not reach the lowest layers of the atmosphere, and this process of ozone formation is therefore of no significance for the generation of ozone there. Solar radiation with a wavelength shorter than about 300 nm is absorbed by the ozone in the stratosphere. The layer of the atmosphere below it, extending to an altitude of about 10 km, is known as the troposphere. This is where the majority of important weather phenomena occur, and here ozone forms in a different way. In the troposphere, single atoms of oxygen are freed when nitrogen dioxide (NO₂) is broken down into nitric oxide (NO) and an oxygen atom under the influence of sunlight:

\[ NO_2 + h\nu \xrightarrow{\lambda < 410\text{ nm}} NO + O \]  

Once again, the wavelengths in the sun’s radiation which drive the process are in the short-wave, ultraviolet region, but they are longer than those involved in reaction (1) and sufficient amounts of this radiation therefore reach the lower atmosphere. Reaction (2) occurs in the troposphere in the same way as in the stratosphere.
**STRATOSPHERIC OZONE FORMATION IN BROAD OUTLINE**

In the stratosphere, short-wavelength, high-energy (ultraviolet) solar radiation can break down molecular oxygen, $O_2$, into two single oxygen atoms, which are needed to form ozone, $O_3$. Ozone, too, can be broken down by sunlight. Normally, the two processes reach equilibrium at a fairly high ozone concentration.

When CFCs (chlorofluorocarbons) reach the stratosphere, they become chemically active as they are broken down by the short-wave radiation there and chlorine atoms, $Cl^*$, are released. These chlorine atoms can participate repeatedly in the cycle of reactions by which ozone and single atoms of oxygen are transformed into molecular oxygen ($O_2$). This reaction cycle reduces the ozone concentration in the stratosphere.

**TROPOSPHERIC OZONE FORMATION IN BROAD OUTLINE**

As in the stratosphere, the basis for ozone formation in the troposphere is single atoms of oxygen, $O$. Here, they are formed by longer-wavelength sunlight breaking down $NO_2$ into $NO$ and $O$.

$NO$ reacts rapidly with ozone, which is consumed in the process. These reactions alone, therefore, do not result in really high ozone levels.

For there to be any net production of ozone, there has to be a reaction that competes with ozone for $NO$. At a certain stage in their decomposition in the atmosphere, volatile organic compounds (voc) can transform $NO$ into $NO_2$ without ozone being consumed.
Tropospheric ozone formation is complicated by two other important reactions. One is a comparatively rapid reaction between ozone and nitric oxide:

\[
\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 + \text{O}_2
\]  

(4)

Reaction (4) consumes ozone formed by reaction (2). Reactions (2), (3) and (4) form a cycle which normally does not result in high ozone concentrations. For really high levels of ozone to arise, a further step is required. This is where volatile organic compounds come in. The majority of such substances are broken down in the atmosphere by a chain of reactions, some of which are dependent on light. If, to use the chemist’s terminology, we designate any organic compound of this kind as RH (the simplest of all hydrocarbons is methane, CH₄, which in this terminology becomes CH₃-H, with CH₃ corresponding to R), the reaction steps we are most interested in here can be described as follows. In the atmosphere, hydrocarbons are primarily broken down by reactions with free radicals (usually short-lived, reactive particles). The most important of these in this context is the hydroxyl radical HO•. The dot after the chemical formula indicates that this particle has an unpaired electron, which is the main reason for its reactivity. Decomposition of hydrocarbons is initiated by the following reaction:

\[
\text{HO} • + \text{RH} \rightarrow \text{R} • + \text{H}_2\text{O}
\]  

(5)

The free hydrocarbon radical R• then reacts rapidly with atmospheric oxygen to form a peroxy radical, RO₂•. It is this radical which subsequently plays a part in ozone formation, in that it can oxidize NO to NO₂ in competition with ozone:

\[
\text{NO} + \text{RO}_2 • \rightarrow \text{NO}_2 + \text{RO} •
\]  

(6)

Reaction (6) is important because NO is converted into NO₂ without ozone being consumed as in reaction (4). Consequently, if suitable organic compounds are present in the air, nitrogen dioxide, which forms the basis for ozone formation in the troposphere, can be regenerated time and time again. The radicals RO₂• and RO• are short-lived reaction products formed during the decomposition of such organic compounds into carbon dioxide and water. (In the case of methane, the radicals concerned are CH₃O₂• and CH₃O•, respectively.) It would not be unreasonable to say that organic compounds serve as the fuel and nitrogen oxides as the catalyst for tropospheric ozone formation. The organic compounds are oxidized and thus broken down or ‘burned’, primarily producing carbon dioxide and water. The nitrogen oxides, on the other hand, can be used repeatedly to form ozone without being consumed, which tallies with the definition of a catalyst. However, a nitrogen oxide molecule cannot participate indefinitely in ozone formation, since there are competing
processes which consume nitrogen oxides in the atmosphere. One of these is deposition onto vegetation and other surfaces. Another is chemical conversion of nitrogen oxides into other nitrogen compounds which play no part in the generation of ozone, chiefly nitric acid. Such reactions are more likely to occur in heavily polluted environments. The number of ozone molecules that can form per nitrogen oxide molecule emitted is therefore larger in comparatively clean environments, although the highest ozone concentrations are still found in the areas with the largest emissions of ozone precursors. Nevertheless, the higher rate of ozone formation per emitted nitrogen oxide molecule in cleaner environments is partly responsible for the wide geographical extent of the problem of ground-level ozone.

1.4 Ozone decreasing in the stratosphere, increasing in the troposphere

More than 90% of the ozone in the atmosphere is to be found in the ‘ozone layer’ of the stratosphere. In recent decades, it has become clear that human beings have influenced the chemistry of the stratosphere in such a way that its ozone content has been depleted, a discovery that was rewarded in 1996 with the Nobel Prize for Chemistry. Certain air pollutants, above all chlorofluorocarbons (CFCs, or Freons), are able to cause changes in the stratosphere because they are highly stable, stability being one of the properties that have made such substances so useful for technical applications. As they do not degrade, they persist for a very long time in the atmosphere and are therefore eventually able to reach the stratosphere. As noted earlier, this region of the atmosphere is exposed to shorter-wavelength and thus higher-energy radiation, in the face of which CFCs are no longer chemically stable. As they break down they release chlorine atoms, which can participate in catalytic reaction cycles that consume large amounts of ozone. These reactions are particularly efficient in association with certain types of stratospheric cloud. Such clouds only develop at very low temperatures, which primarily occur in the stratosphere over the Antarctic. This is an important part of the reason why the deepest ‘ozone hole’ is to be found in the stratosphere above the South Pole. Depletion of stratospheric ozone has also been observed over the planet as a whole, but not to the same extent.

In the troposphere, the opposite problem exists. Emissions of nitrogen oxides and volatile organic compounds, above all from transport, industry and energy production, have created the basic conditions for greatly increased tropospheric ozone concentrations in much of the industrialized world. True, a certain amount of ozone was present in the troposphere even before the industrial revolution. Nitrogen oxides are formed by lightning discharges and forest fires
(Graedel & Crutzen 1993), for example, and volatile organic compounds that can play a part in ozone formation, chiefly terpenes and isoprene, are also emitted by plants (Simpson et al. 1995). However, in the course of the 20th century, background concentrations of ozone in Europe have increased by a factor of two to three from their preindustrial levels of 10–15 ppb (Borrell et al. 1997). In addition, what are known as ozone episodes occur. An episode is a relatively short period, anything from a few hours to a few days, of greatly elevated ozone concentrations, sometimes in excess of 100 ppb. In Sweden, ozone episodes usually occur when stable high-pressure systems, accompanied by strong sunlight and light winds, move in across the country from the south, i.e. from the regions with major emission sources. As was noted earlier, sunlight – which is often strong during a period of high pressure – is important in ozone formation. Since mixing of the air is restricted, ozone precursors – nitrogen oxides and volatile organic compounds – are not diluted and can reach high concentrations. In Sweden, ozone episodes occur during most springs and summers, though to a very varying extent. Warmer, sunnier weather is normally accompanied by more ozone episodes. High-pressure systems from the north do not usually produce ozone episodes, since they do not bring with them any appreciable quantities of ozone precursors.

Figure 1.3 shows how ozone levels at Rörvik, south of Göteborg, varied during the height of the summer of 1991. The values shown are 1-hour means. Fluctuations in the weather are clearly reflected in the ozone concentrations recorded: the diagram shows how spells of more or less unsettled low-pressure weather alternated with periods of high pressure. When low pressure prevailed, peak concentrations over each 24-hour period were not particularly high, perhaps reaching around 40 ppb, and levels were somewhat lower at night. At night, of course, no ozone is formed, since no light is available. High-pressure periods with more or less southerly winds were accompanied by ozone episodes. At such times, the ozone concentration rose to a diurnal maximum of almost 80 ppb, but frequently fell close to zero during the night-time. This considerable difference between day- and night-time levels is due to the fact that high-pressure systems are often associated with clear skies and little wind at night. Under such conditions, the ground surface cools down, since the thermal radiation which it emits to space is not reflected back by clouds, and a nocturnal inversion arises. The air nearest the ground ends up colder than the air above it, and vertical mixing in the lowest layer virtually ceases. All the ozone in the air closest to the ground is removed by deposition. Since mixing of the air is practically non-existent, fresh ozone is not brought in from higher layers of the atmosphere, where, even at night, concentrations are usually quite high during episodes. Lows usually entail cloudy weather and it is generally windy enough,
albeit less so at night than during the day, to cause vertical mixing. For this reason, in such weather conditions, less marked vertical gradients of ozone concentrations build up in the lowest part of the atmosphere.

To sum up, most of the ozone in the atmosphere is to be found in the stratosphere, where it ‘filters off’ UV-B radiation which would harm organisms if it reached the earth’s surface. Stratospheric ozone levels are currently falling. In the troposphere, on the other hand, ozone concentrations are rising, owing to emissions of nitrogen oxides and volatile organic compounds. These diverging trends are illustrated by figure 1.4.

A subdivision of the ozone in the troposphere is also possible, into ground-level ozone and ozone in the free troposphere. The region of the atmosphere closest to the ground is known as the boundary, or mixing, layer. This layer is strongly influenced by mechanical turbulence, caused by friction between the air and the more or less rough ground surface, and by thermal turbulence, arising from the fact that the surface and the air immediately above it are warmed by sunlight during the day. This heated air has a tendency to rise and thus to cause mixing. The depth of the boundary layer varies with the time of day and the season. Typical figures in summer may be 1000 m (about 10% of the entire troposphere) during the day and 100–200 m at night. Pollutants emitted at the ground surface disperse relatively quickly within the boundary layer. Ground-
level ozone is the ozone which occurs in this layer. Only high-altitude mountain areas reach into the free troposphere during the daytime.

There is no chemical difference between tropospheric and stratospheric ozone, but stratospheric ozone does not directly affect organisms on earth, since it is not inhaled by animals or taken up by plants. Between the troposphere and the stratosphere there is a temperature boundary which greatly restricts transfers between these air masses. Sometimes, especially in early spring, some exchange does nevertheless occur, especially at high latitudes. Since ozone concentrations are higher in the stratosphere, such an exchange involves a net input of ozone into the troposphere. The dividing line between the boundary layer and the free troposphere is not at all as distinct, and exchanges between these two air masses occur regularly.

![Figure 1.4.](image)

Concentrations of ozone (expressed as partial pressure) at different altitudes. At about 10 km, there is a clear dividing line between the troposphere and the stratosphere. The measurements were made at Hohenpeissenberg in southern Germany. An upward trend in concentrations in the lower atmosphere and a downward trend in the stratosphere are clearly discernible.

### 1.5 Ozone in environments with heavy traffic

The chemical reactions between primary air pollutants which are involved in ozone formation take some time and are regulated by sunlight, concentration changes, meteorology and other factors. This means that high levels of ozone can occur far away from emission sources and that the geographical link between sources and ozone formation is often fairly unclear. Nitrogen oxides released in one place may have an ozone-forming effect hundreds or thousands of kilometres away, by which time they will have been mixed with emissions from countless other sources. This explains the marked regional nature of the problem, and also means that emissions in other countries have a decisive impact on ozone concentrations in Sweden. The ozone level at any particular location represents the sum of many small individual contributions from a very large number of sources. On the whole, therefore, small-scale variations in ozone concentrations are limited. This is something
of a problem for anyone wanting to study the effects of ozone. Concentration gradients are very useful for such purposes, but ozone concentrations vary on such a large geographical scale that, in order to find sites with substantially different ozone levels, very appreciable differences in climate and other natural conditions will also have to be taken into account.

There is one important exception to this rule, however, and one which also illustrates the chemistry of ozone. In emissions from road traffic and combustion plants, the dominant nitrogen oxide is nitric oxide (NO). This means that the rapid reaction (4), in which ozone is consumed, is the first important reaction which comes into play when vehicle exhausts and flue-gas plumes mix with the ambient air. One consequence of this is that, in environments with heavy traffic, ozone levels are lower than in surrounding areas (Rodes & Holland 1981). This is in a sense a paradoxical state of affairs, in that transport emissions are at the same time the single most important cause of regional ozone formation, through reactions (2), (3) and (5). Because ozone concentrations are locally lower where traffic is heavy, ozone effects are in fact less marked in such areas, even though levels of several other primary pollutants emitted by vehicles are of course higher there. Figure 1.5 shows how the occurrence of visible ozone injury in an ozone-sensitive variety of clover varied with distance from the E6 motorway between Göteborg and Kungsbacka in the peak summer months of 1991 (Pleijel et al. 1994c). Noticeably less ozone damage was observed close to the road than 200 m away from it, where the degree of damage was at the level typical of rural areas of western Sweden during the period in question.

The local occurrence of lower ozone levels in the immediate vicinity of high road traffic emissions is the exception which proves the rule that concentrations of ground-level ozone do not vary appreciably, other than on a relatively large geographical scale.

![Figure 1.5. Number of leaves of subterranean clover (Trifolium subterraneum) injured by ozone, per pot, at different distances from the E6 north of Kungsbacka. The plants were exposed during the period covered by figure 1.3. From Pleijel et al. 1994c.](image-url)
INTERNATIONAL AND NATIONAL ENVIRONMENTAL OBJECTIVES

Håkan Pleijel

SUMMARY
At both the national and the international level, environmental objectives and limit values relating to ozone have been adopted. The UN agency WHO and the Swedish Institute of Environmental Medicine (IMM) have collated scientific data on levels of ozone that could be harmful to human health. WHO has proposed a level of 60 ppb for effects of this type, while the IMM wishes to set a more stringent limit of 40 ppb. In response to rising concentrations of ground-level ozone in Europe, the ECE has developed critical levels of ozone with regard to its effects on vegetation. These are being used to design effects-based control strategies to reduce emissions of ozone precursors under the ECE Convention on Long-Range Transboundary Air Pollution and within the EU. At present, critical levels are based on an exposure index known as AOT40, which refers to the accumulated exposure to ozone in excess of 40 ppb. Regarding the effects of ozone on materials, European experts have discussed using 20 ppb for the time being as a kind of critical level which should not be exceeded. In Sweden, official environmental objectives exist with regard to emissions of nitrogen oxides and hydrocarbons, the pollutants which result in ozone formation.

2.1 Introduction
As was mentioned in chapter 1, ozone is a contributory factor behind several types of environmental impact, both health effects and effects on plants and materials. As a result, various national and international bodies have formulated environmental objectives and limit values with respect to ground-level ozone and ozone precursors. The main bulk of this report is concerned with ozone’s effects on plants, and it may therefore be appropriate here also to touch on its significance for health and materials.
2.2 WHO and IMM guidelines on ozone – health effects

The World Health Organization (WHO), a specialized agency of the UN, issues guidelines on, among other things, the levels of air pollutants that are regarded as harmful to human health. As noted in chapter 1, ozone is a powerful oxidant. It has a relatively low solubility in water and is therefore drawn deep into the lungs when inhaled. After only fairly brief exposure, ozone produces inflammatory reactions in the respiratory tract. These abate when exposure ceases. In experiments, such effects have been observed at ozone levels down to around 80 ppb. Ozone impairs lung capacity and reduces resistance to bacterial and viral infections. The first of these effects is particularly serious for individuals who already have impaired lung function, such as asthma sufferers. Irritation of the eyes is another common effect of elevated, but currently occurring ozone concentrations. People who spend a lot of time outdoors and are highly active physically are particularly at risk. This category includes those who engage in sports, but generally children also have a tendency to be especially vulnerable, since they are often in the open air and move about a great deal. Epidemiological research has shown that high ozone levels co-vary with hospital admissions for lung complaints. There are also studies suggesting that ozone may increase the risk of cancer, although this risk has not been adequately assessed. On the basis of the ‘lowest observed effects’ level, WHO has formulated a guide value of 60 ppb ozone as the maximum 8-hour mean (WHO 1995). At the same time, WHO stresses that this level probably does not represent any margin of safety for the most sensitive sections of the population with regard to certain types of acute health effect. The Swedish Institute of Environmental Medicine (IMM) has recommended a low-risk level of a 1-hour mean of 40 ppb ozone, as an upper limit for human exposure (Bylin et al. 1996). This was calculated using a safety factor of 2 in relation to the lowest observed effects level.

2.3 Effects of ozone on materials – a preliminary environmental objective

Ozone has long been known to affect polymers, i.e. molecules of the type found in the majority of fibres. The polymers affected are those containing carbon atoms with double bonds, which include many natural fibre materials, such as natural rubber, cotton and cellulose. Best known is ozone’s effect on rubber, and in the 1950s this material was used as a simple means of measuring atmospheric levels of the pollutant. Ozone causes rubber to crack, and the depth of the cracks that appear over a given time is a measure of how high the ozone concentration has been. This method proved to have a good degree of preci-
One of the sectors interested in ozone levels back in the 1950s was manufacturers of car tyres, which are made from rubber. Ozone also affects a range of other materials, resulting in costs to society and damage to cultural assets. For example, it shortens the lifetimes of textiles, paints and other pigments (e.g. on museum exhibits). Unlike the effects of ozone on human health and plants, the scale of the damage it causes to materials is primarily determined by the long-term average concentration of the gas. In principle, all concentrations have some effect. Nevertheless, European experts have discussed setting a critical level for ozone with respect to effects on materials at an annual mean of 20 ppb. At present, this value is exceeded virtually throughout Europe. Current estimates of the financial cost of ozone’s effects on materials are very uncertain, but the sums involved could be considerable.

The biggest concentrations of materials that could be damaged by ozone are to be found in major towns. As indicated in chapter 1, ozone levels are usually somewhat lower there than in the surrounding countryside, since ozone is consumed locally by vehicle emissions of nitric oxide (NO). This creates a paradox: if a town manages to cut nitrogen oxide emissions locally, ozone levels will rise and with them the risk of damage to materials, or at least they will do so unless the same emission reduction is achieved over a wide geographical area. This underlines the need for large-scale strategies to reduce ozone loads in Europe.

### 2.4 International control strategies

In recent decades, various agencies have endeavoured to introduce measures to curb the effects of ozone. At the international level, the ECE Convention on Long-Range Transboundary Air Pollution has been most important. The ECE (the United Nations Economic Commission for Europe) played a significant role in promoting international dialogue in Europe during the cold war. One of its key spheres of activity was transboundary air pollution. Since this organization included the Warsaw Pact countries among its members, air pollution could provide a pretext for discussing other important political issues as well. Under the ECE Convention, several international protocols to control a range of air pollutants have been signed. At present, the final touches are being put to a protocol covering, among other things, nitrogen pollutants and volatile organic compounds (VOCs). An important driving force behind this ‘multi-pollutant/multi-effect protocol’ has been the effects of ground-level ozone, both effects on human health and damage to vegetation. ‘Critical levels’ of ozone, described in more detail in the next section, have formed an important part of the scientific basis for the protocol.
In recent years, the European Union has emerged as an increasingly powerful player in the arena of transboundary air pollution. It is currently drawing up an Acidification Strategy and an Ozone Strategy, which will guide future EU efforts in this field. A crucial consideration in the EU’s Ozone Strategy is cost-effectiveness. The emission reductions that can be achieved at low cost are already being implemented or have at least been decided on in a good number of countries, and further cuts will prove more expensive. To secure acceptance and legitimacy for the costs involved among those who will have to meet them – companies, states (taxpayers) and consumers – it is important to ensure that any action is as cost-effective as possible. In other words, it must be possible to show that the measures decided on are the ones that will yield the greatest environmental benefits per unit of currency invested. The new control strategies are therefore effects-based, which was not the case with the earliest protocols under the ECE Convention.

The EU’s new directive and strategy on ozone will be based chiefly on the ECE-defined critical levels for the effects of ozone on plants and on WHO’s guidelines with regard to health. To a large extent, the calculations underlying the cost-effective, effects-based control strategies currently being discussed within the ECE and the EU are being performed by IIASA, an international centre for systems studies near Vienna.

2.5 Critical levels of ozone

The process of establishing critical levels of ozone began in 1988, when a workshop was held in Bad Harzburg in Germany on the initiative of the ECE. At that meeting, preliminary critical concentrations with regard to effects on plants were calculated for a number of gaseous air pollutants (Guderian 1988). These efforts were guided by an older tradition in toxicology which relied on mean concentrations over a given period of time. In this early work, ozone was less prominent compared with other pollutants, chiefly sulphur dioxide and nitrogen oxides, than it is today.

To build on the results from Bad Harzburg, a similar meeting was held at Egham near London in spring 1992, and once again a range of gaseous air pollutants were dealt with (Ashmore & Wilson 1994). At that workshop, it became increasingly clear that, in quantitative terms, ozone was the dominant gaseous air pollutant in Europe with regard to effects on plants, although sulphur dioxide will probably continue to have significant effects in parts of eastern Europe for some time to come.

Another very important outcome of the Egham workshop was the launch of a new approach to describing exposure to ozone. It was demonstrated that the
observed effects of ozone showed closer agreement with the aggregate exceedance of a given threshold value than with the mean concentration figures traditionally used. This new approach was not fully developed until the next European workshop on critical levels, held in Berne in autumn 1993, which dealt only with ozone (Fuhrer & Achermann 1994). A collation of the results of European experiments on ozone exposure of field-grown wheat revealed a very high correlation with the exposure indices AOT40 and AOT30. AOT stands for ‘Accumulated exposure Over Threshold’, i.e. the aggregate exceedance of a stated ozone concentration over a given period. Assuming ozone concentrations are expressed in ppb – parts per billion (i.e. per thousand million) of the total number of air molecules – the unit used for the AOT index is ppb-hours. The main reason for choosing AOT40, i.e. for setting the ‘threshold’ at 40 ppb, was that a lower threshold would have been close to the background concentrations of ozone occurring in ambient air throughout the northern hemisphere. AOT40 is a measure which clearly reflects the extent to which ozone concentrations are elevated as a result of anthropogenic emissions of ozone precursors. It is the first step towards a dose measure indicating ozone doses harmful to plants. AOT40 does not directly reflect plant uptake of ozone, being calculated solely on the basis of concentrations in air. Efforts to develop an uptake-based exposure index for ozone are now under way, but a generally accepted method for this purpose has yet to be established.

**CALCULATING AOT40**

AOT40 is calculated as follows. Assume that the following hourly mean concentrations of ozone have been recorded over seven hours: 35, 38, 40, 41, 42, 45 and 50 ppb. The first three values do not contribute to the exposure index, since they do not exceed 40 ppb by at least 1 ppb. Only the last four values contribute to AOT40. The AOT40 value is thus: 

\[
(41-40) + (42-40) + (45-40) + (50-40) = 1 + 2 + 5 + 10 = 18 \text{ ppb-hours.}
\]

For agricultural crops, differences are aggregated over a three-month period (May–July), while for forests a six-month period (April–September) is used. Since plants chiefly take up ozone in daylight, AOT40 is calculated on the basis of values for the hours between sunrise and sunset.

AOT40 values have been used to develop relationships between exposure and response in plants. As indicated above, the best results have been obtained with regard to effects on wheat yields. Figure 2.1, based on a large body of data from five European countries, shows what such relationships may look like. A more detailed relationship for the Nordic countries will be presented in chapter 5.
At the following ECE workshop, in Kuopio in 1996, it was agreed that, for the time being, the critical levels shown in table 2.1 were to apply (Kärenlampi & Skärby 1996).

Tabell 2.1. Critical levels of ozone defined at the Kuopio workshop in 1996. From Kärenlampi & Skärby (1996). VPD = vapour pressure deficit, a measure of how dry the air is.

<table>
<thead>
<tr>
<th>TYPE OF VEGETATION</th>
<th>AOT40 VALUE</th>
<th>PERIOD ETC.</th>
</tr>
</thead>
<tbody>
<tr>
<td>Agricultural crops</td>
<td>3 000 ppb-hours</td>
<td>May–July</td>
</tr>
<tr>
<td>Forest trees</td>
<td>10 000 ppb-hours</td>
<td>April–September</td>
</tr>
<tr>
<td>Short-term effects</td>
<td></td>
<td></td>
</tr>
<tr>
<td>– visible injury</td>
<td>500 ppb-hours</td>
<td>five days, VPD&gt;1.5 kPa</td>
</tr>
<tr>
<td></td>
<td>200 ppb-hours</td>
<td>five days, VPD&lt;1.5 kPa</td>
</tr>
<tr>
<td>Wild herbs and grasses</td>
<td>3 000 ppb-hours</td>
<td>May–July</td>
</tr>
</tbody>
</table>

Figure 2.2 shows how ozone levels, expressed as AOT40, varied across Europe in May–July over the period 1989–94 (Simpson et al. 1998). The diagram is based on modelled values, which are in close agreement with those obtained from measurements.
In scientific terms, the most certain of the critical ozone levels calculated so far is the figure for agricultural crops, which is based on yield reductions in wheat. A relatively large body of data is available, and the results are consistent. The critical level for forest trees is more uncertain. It is based on data for beech, which the information available indicated was more sensitive to ozone than spruce, for example. The AOT40 values for short-term exposure refer to the occurrence of visible injuries to leaves during ozone episodes, and are based on experiments involving clover. The abbreviation VPD in table 2.1 stands for vapour pressure deficit, which is a measure of the dryness of the air. Basically, it indicates how strongly the air tends to draw water from plants. At high VPDs, plants close their stomata, the tiny pores in their leaves which regulate their exchange of gases with the atmosphere. This reduces transpiration, i.e. the loss of water vapour to the surrounding air. Another consequence of closed stomata is that plants take up smaller amounts of gaseous air pollutants, such as ozone, and exposure therefore has to be higher to have the same effect. Broadly speaking, the two levels for short-term effects reflect the difference in climate between the warmer southern and eastern parts of Europe, with dry summers, and the damper, cooler regions of northern and western Europe. Although high VPDs do sometimes occur in Sweden in summer, the relevant AOT40 value here is by and large the lower one. The value for wild herbs and grasses is provisional. It is
based primarily on outline experiments on large numbers of species, which have indicated that the most sensitive species probably have roughly the same sensitivity as the most sensitive crops. The majority of wild plants are probably less sensitive.

All the values presented in the table above are what are known as Level I critical levels, which means that they are used to estimate the risk of production losses or visible injury in different groups of plants, regardless of all the factors which may locally modify the effects of ozone. Such factors include genetic variation within and between species, the developmental stage at which plants are exposed, the presence of pest organisms and other pollutants, and a range of climatic factors (wind, light, soil moisture, atmospheric humidity, temperature) which affect ozone uptake. Level I values are not intended as a basis for estimates of actual production losses or other effects, but simply indicate in which areas there is a risk of significant ozone damage to vegetation. The focus is now shifting towards developing Level II values, which are intended to be used to estimate the scale of effects in ecosystems. To do that, the factors mentioned have to be taken into account. Only limited progress has been made in this direction as yet, but this is likely to be a key concern of ozone research in Europe over the next few years.

2.6 Swedish environmental objectives relating to ozone and ozone precursors

The Swedish Government recently submitted a bill to Parliament entitled ‘Swedish Environmental Objectives. Environmental Policy for a Sustainable Sweden’ (Government Bill 1997/98:145). The Government’s overall goal for efforts in the environmental field is to be able to hand over to the next generation a society in which the country’s major environmental problems have been solved. In addition, at the international level, Sweden should be a driving force and pioneer of ecologically sustainable development.

To achieve these aims, the Government has proposed that a new structure for setting and implementing environmental objectives should be established. A limited number of national environmental quality objectives will be adopted by Parliament, indicating what state of the environment is to be achieved on a time-scale of one generation. The Government will have a responsibility to ensure that, where necessary to achieve these objectives, more specific goals are defined. The latter will then form the basis for the definition of aims and strategies in different sectors of society and at different levels. Since this is a new approach, the Government has set up a parliamentary advisory committee to keep the process under review, in collaboration with the government agencies
concerned. In practice, several authorities (the Environmental Protection Agency, the Board of Agriculture, the Board of Forestry etc.) have already embarked on this process. The Government’s assessment is that environmental quality objectives, together with the new Environmental Code (Government Bill 1997/98:45), offer greater scope for a decentralization of environmental protection. Opportunities for and interest in taking independent initiatives to secure a better environment will increase, not least in the business sector.

The problem of ground-level ozone is addressed under one of 15 environmental quality objectives which are proposed, namely Clean air. The objective proposed by the Government is that ‘the air should be so clean that no damage is caused to human health or to animals, plants or cultural assets’. Existing air quality problems in urban areas of Sweden are chiefly the result of Swedish emissions, with transport the most important source of all; locally, small-scale burning of wood may also be a major source. Problems relating to ground-level ozone in Sweden, however, are for the most part attributable to emissions in other countries, primarily the United Kingdom and Germany. Sweden is only the third most important of the countries contributing to the ozone levels occurring within its borders. It should not be forgotten, though, that Sweden also exports ozone precursors – nitrogen oxides and hydrocarbons – to its neighbours, inter alia from the above-mentioned urban areas.

As far as ground-level ozone is concerned, the environmental quality objective proposed means that concentrations should not exceed the limit/guide values that have been established to prevent harm to human health, animals, plants, cultural assets and materials. Emissions of nitrogen oxides need to be reduced, and under the quality objective Natural acidification only the Government has said that nitrogen oxide emissions from the transport sector in Sweden should have fallen by at least 40% by the year 2005, compared with 1995 levels. The Government’s assessment is that this objective should be supplemented with targets relating to volatile organic compounds and that ‘additional more specific targets may need to be developed’. In its Transport Policy Bill (1997/98:56), the Government put forward the view that transport emissions of VOCs in Sweden should be reduced by at least 60% by 2005, compared with 1995 levels.

It has been decided that the transport sector’s emissions of nitrogen oxides are to be cut by 40% and those of VOCs by 60% between 1995 and 2005. These reductions are relevant to the environmental quality objective Clean air. To attain this objective, action must also be taken to reduce emissions from small-scale wood burning and from industrial plants and district heating and power stations. To achieve the quality objective with respect to ground-level ozone, further emission cuts will be necessary both in Sweden and in other European
countries. Negotiations on what additional commitments different countries need to make are being conducted under the UN ECE Convention on Long-Range Transboundary Air Pollution (CLRTAP) and within the EU. Finally, further action in the transport sector is of great importance. Legislation on vehicle emissions lays down maximum permitted releases of different pollutants from road traffic. In the context of the Environmentally Sound Transport System (MaTs) project, the transport sector has set targets of a 70% reduction of VOC emissions by the year 2005 and an 85% reduction by 2020, compared with 1988.
As part of Sweden’s environmental monitoring programme, ozone concentrations are measured continuously at a number of sites around the country. These measurements show that critical levels for damage to agricultural crops and forests are exceeded in southern Sweden, but only to a very limited extent (crops) or not at all (forests) in the north of the country. By the year 2010, emissions of ozone precursors in Europe are expected to have fallen. This will result in a reduced exceedance of critical ozone levels, particularly over certain parts of the continent. In Sweden, too, the ozone load will decrease, but the critical level for crops is expected to be exceeded even after 2010.

3.1 Introduction
Ozone concentrations are measured continuously at six sites in Sweden, from Söderåsen in the south to Kiruna in the north, in the framework of a European air quality monitoring network known as EMEP (European Monitoring and Evaluation Programme). As was noted earlier, ground-level ozone is primarily a problem of a geographically very widespread nature. Table 3.1 (overleaf) shows ozone data from the six monitoring sites in Sweden, while figure 3.1 shows their location.
3.2 Variation in ozone levels over Sweden

As table 3.1 shows, long-term mean concentrations of ozone do not differ dramatically between the south and the north of Sweden. When it comes to AOT40 values and maximum hourly means, however, the differences are very pronounced. Appreciably lower values are recorded for these exposure measures at the two northern sites of Vindeln and Esrange. This is because these areas are affected by far fewer ozone episodes, and those that do affect them have usually largely abated by the time they reach this far north. On the other hand, there is a more marked spring peak in ozone concentrations at these two sites, and it occurs earlier there, normally in April. There may be various reasons for this. One possible explanation is that volatile organic compounds accumulate in the atmosphere during the polar winter, since their decomposition is a light-dependent process. When strong sunlight returns after the spring equinox, these pollutants give rise to ozone formation. Another conceivable explanation is that ozone from the stratosphere is mixed into the troposphere in the early spring.

Table 3.1. Mean ozone concentrations for April–September (24-hour, ppb), AOT40 values for the period May–July/April–September (ppb-hours), and highest recorded hourly mean concentrations of ozone (ppb) at six Swedish EMEP monitoring stations. AOT values exceeding agreed critical levels (table 2.1) are printed in bold.

<table>
<thead>
<tr>
<th></th>
<th>MEAN CONCENTRATION</th>
<th>AOT40 MAY–JULY CROPS</th>
<th>AOT40 APRIL–SEPTEMBER FOREST</th>
<th>MAXIMUM HOURLY MEAN CONCENTRATION</th>
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<tr>
<td>Vavihill</td>
<td>36</td>
<td>10 215</td>
<td>14 512</td>
<td>100</td>
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<td>39</td>
<td>13 781</td>
<td>19 758</td>
<td>98</td>
</tr>
<tr>
<td>Rörvik</td>
<td>38</td>
<td>11 113</td>
<td>17 139</td>
<td>88</td>
</tr>
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<td>4 858</td>
<td>78</td>
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<tr>
<td>Esrange</td>
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<td>3 285</td>
<td>9 053</td>
<td>80</td>
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<td>103</td>
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<tr>
<td>Norra Kvill</td>
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<td>10 062</td>
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<td>11 229</td>
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<td>2 147</td>
<td>3 195</td>
<td>64</td>
</tr>
<tr>
<td>Esrange</td>
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<td>3 843</td>
<td>7 000</td>
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<th>1996</th>
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<td>16 880</td>
<td>105</td>
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<td>Norra Kvill</td>
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<td>13 632</td>
<td>91</td>
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<tr>
<td>Rörvik</td>
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<td>4 668</td>
<td>12 026</td>
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<td>Aspvreten</td>
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<td>7 081</td>
<td>15 570</td>
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<tr>
<td>Vindeln</td>
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<td>2 477</td>
<td>5 285</td>
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<tr>
<td>Esrange</td>
<td>34</td>
<td>1 878</td>
<td>4 848</td>
<td>62</td>
</tr>
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</table>
and that this occurs to a particularly large degree in the north. Both these factors probably contribute to the pattern observed. The fairly high altitude of the Esrange measuring station is presumably also partly responsible for the comparatively high average ozone levels recorded there.

A collation of ozone data for northern and western Europe from 1989, which was a year with relatively high ozone concentrations, shows that AOT40 for the period April–September varied from about 1500 ppb-hours at Ny Ålesund in Svalbard to 20 000–40 000 ppb-hours in parts of continental Europe (Beck & Grennfelt 1994). The highest values were recorded in mountain regions.

3.3 Ozone in mountain areas

Within a given region, the highest mean concentrations of ozone are usually found at high altitudes. This has probably always been the case (Borrell et al. 1997). In addition, ozone levels follow a different diurnal pattern in mountain areas: they remain almost constant between day and night, and night-time concentrations are very high compared with those in low-lying regions. This is very much the case in the Alps, for example, and the same phenomenon has been observed on Åreskutan in the Swedish mountain range (Bazhanov & Rodhe 1996). As indicated earlier (section 1.4), monitoring sites at high altitudes in mountain regions (above approx. 1000 m) are often situated within the free troposphere, the part of the troposphere that is above the boundary layer and is therefore little affected by local emissions. The fact that they are in the free troposphere also means that ozone concentrations there are affected very little by deposition, since there are only small areas of land and vegetation at such high altitudes onto which ozone can be deposited. This is an important part of the explanation for the high ozone levels recorded at night at high-altitude stations in mountain areas. Down below, in the boundary layer, ozone is removed at the ground surface by deposition during the night. Since such a small proportion of the land mass is situated above the boundary layer, the deposition occurring there is of less significance.

3.4 Trends in ozone levels since the 19th century

Ozone was discovered in the middle of the 19th century by a chemist called Schönbein, who also developed a method of measuring the gas, known as Schönbein paper. This paper was exposed for a day and the ozone concentration could then be read off against a colour scale. Measurements performed with Schönbein paper in the 19th century – in Paris, the Pyrenees and northern Italy, for instance – suggest that background levels at that time were about 10 ppb (Volz & Kley 1988; Anfossi et al. 1991), but there are also early studies
indicating somewhat higher values. In an overall assessment of available information, carried out as part of the EUROTRAC project, researchers have concluded that preindustrial background concentrations were probably 10–15 ppb in low-lying areas, but somewhat higher in mountain regions (Borrell et al. 1997). As table 3.1 indicates, mean ozone concentrations in Sweden during the warmer half of the year are usually between 30 and 40 ppb. In other words, background levels have risen by a factor of two to three in this country. On top of this, there are ozone episodes, which have relatively little impact on mean concentrations, but contribute significantly to AOT40 (table 3.1). Such episodes probably did not occur in preindustrial times.

Regular measurements of ozone concentrations using modern methods began in Sweden in the mid-1970s, by which time concentrations were already quite high. These measurements cannot be used to assess long-term trends in ozone levels. The site in Sweden’s vicinity at which continuous ozone monitoring has been in progress for longest is Arkona on the Baltic coast of Germany, where measurements go back to the 1950s. The figures recorded at Arkona suggest an increase of more than 50% in monthly mean concentrations between 1956 and 1983 (Feister & Wrambd 1987). A similar trend probably occurred in the far south of Sweden, too.

The rise in ozone levels in northern Europe has slowed down in the last decade. Future trends will depend on what decisions on emission reductions are taken and to what extent they are actually implemented. A crucial factor is of course how emissions in Europe develop. Figure 3.2 shows modelled ozone data, based on the assumption that the most far-reaching emission reductions, including the EU’s Ozone Strategy, are implemented. As far as Sweden is concerned, the assessment is that AOT40 will fall most in absolute terms in the south of the country, while the biggest percentage decrease will be seen in the north, where critical levels are currently exceeded to only a limited extent. The largest reduction in AOT40 will occur over some western and southern areas of continental Europe. It should be borne in mind in this context that Europe is not in a position to fully control ozone levels. Background concentrations of ozone exhibit an upward trend throughout the northern hemisphere, a trend that is due to emissions over a very large area. In other words, there is a risk that the action taken in Europe to reduce ozone levels could be partly offset by a rising trend in concentrations over the northern hemisphere as a whole.
Figure 3.2. Reductions in ozone levels expressed as AOT40 (ppm-hours; 1 ppm = 1000 ppb) for crops (3 months), calculated using EMEP’s ozone model: (a) absolute reductions, (b) percentage reductions. Maps produced by David Simpson, Norwegian Meteorological Institute.
Figure 3.3 shows the AOT40 values (May–July) that are predicted on the basis of model calculations for the year 2010, assuming that planned emission reductions in Europe are achieved. Even then, the critical level for crops is expected to be exceeded in southern Sweden. However, as both figure 3.2 and a comparison with figure 2.2 make clear, ozone levels will fall over the Nordic region.

Figure 3.3. Ozone levels expressed as AOT40 (ppm-hours; 1 ppm = 1000 ppb) for crops (3 months), based on an emission scenario for 2010 calculated using EMEP’s ozone model. Map produced by David Simpson, Norwegian Meteorological Institute.
EFFECTS OF OZONE ON AGRICULTURAL CROPS

Håkan Pleijel, Helena Danielsson, Gunilla Pihl Karlsson, Johanna Gelang and Gun Selldén

SUMMARY
The problem of ozone damage to agricultural crops has been known from North America since the late 1940s. In the United States, it was estimated in the early 1980s that ground-level ozone was responsible for economic losses to farmers of between two and four billion dollars a year. Soya bean was judged to be the crop most sensitive to ozone. In Europe, research into ozone’s effects on agriculture did not begin in earnest until the middle of the 1980s. Among the crops identified as susceptible, particular mention may be made of wheat and beans. Several clovers have also been found to be very sensitive to ozone in Europe. Regarding wheat, a solid and extensive body of data is now available, and these data have among other things been used to formulate critical levels of ozone. Ozone probably has an adverse effect on wheat yields because higher exposure causes earlier ageing of leaves. This shortens the period in which photosynthesis in the leaves can supply the grains with carbohydrates, resulting in lower yields. Wheat exposed to high ozone concentrations usually produces grain with a higher protein content than wheat growing at lower levels of ozone.

4.1 Introduction
Ozone effects on agricultural crops were first documented scientifically in southern California in the late 1940s (Middleton 1950). As was noted earlier, climatic conditions there are very favourable to ozone formation and, with car use very heavy even in the 1940s, this led to significant problems of ozone and other photochemical oxidants.
Initially, studies of ozone damage to crops focused on visible leaf injury in the field and on laboratory studies of effects, often involving exposure to very high concentrations for short periods. One variety of cultivated tobacco (*Nicotiana tabacum* Bel-W$_3$) was found to be particularly sensitive to ozone (figure 4.1). It developed characteristic visible foliar injuries after periods of elevated ozone concentrations (Heggestad 1991). The symptoms had been observed on commercial tobacco farms in the eastern United States, even before the link with ozone was brought to light: previously, they had been attributed to the weather ('weather fleck disease'), which was not unreasonable, given the strong association between peak ozone levels and sunny weather and light winds (Heggestad & Middleton 1959). Subsequently, this type of tobacco was to be used as an ozone indicator for several decades, and in many different countries. In Sweden, too, studies were carried out using tobacco as a bioindicator of ozone pollution (e.g. Lång *et al*. 1980).

![Figure 4.1. The tobacco variety Bel-W$_3$ is very sensitive to ozone and has therefore been used as an indicator plant. The photograph shows one leaf that was exposed to ozone and another that was protected from exposure. Photograph: Håkan Pleijel/IVL.](image)

The real breakthrough for quantitative studies of ozone’s effects on crop yield and quality came when the open-top field chamber began to be used as an exposure system (Heagle *et al*. 1973). This is a transparent plastic cylinder, ventilated by a fan, that can be placed over plants in the field (figure 4.2). This arrangement enables concentrations of air pollutants to be controlled, while preserving a high degree of ecological realism. It can thus be used for long-term exposures to charcoal-filtered air, unfiltered air, and ozone concentrations elevated to different degrees. A charcoal filter efficiently absorbs the ozone present in the ambient air. Naturally, the microclimate of the plants is affected to some extent by the fact that they are inside a chamber. The radiation climate is differ-
ent, the temperature is somewhat higher and air flow patterns deviate from those outside the chamber (Unsworth et al. 1984; Unsworth 1991; Pleijel et al. 1994b). A particularly critical question in this context is whether the plants’ uptake of ozone is influenced by the chamber environment, and this possibility cannot be entirely ruled out. Whether ozone uptake is higher or lower in the chamber than in the open will depend on the conditions prevailing in each individual case. Nevertheless, compared with the laboratory setting, the open-top chamber technique offers a considerably more realistic alternative. In studies of agricultural crops, it is possible to place the chamber in an ordinary field (figure 4.3). It is also possible to make quantitative estimates of the chamber’s effects on such factors as pollutant uptake (Grünhage & Jäger 1994; Pleijel & Wallin 1996).

Figure 4.2. An open-top chamber is basically a transparent plastic cylinder, into which air is drawn by means of a fan. The air is then dispersed within the chamber through perforated plastic tubing and can eventually exit through the top of the chamber. Photograph: Håkan Pleijel/IVL.
In the United States, a major project involving several experimental stations, known as the NCLAN project, was carried out using the field chamber method. The final results of this research were presented in 1988, and at that time the economic losses suffered by US agriculture as a result of ground-level ozone were estimated at between two and four billion dollars a year. Soya bean was judged to be the crop most sensitive to ozone.

### 4.2 Experiments in Sweden

One of the first important results of open-top chamber studies of ozone in Sweden was the clarification of ozone’s role as a cause of visible injury and production losses in spinach crops in Skåne in the south of the country (Skärby & Jönsson 1985; figure 4.4). This study also covered peas, French beans and potatoes. In 1987, larger-scale research on commercially important species was launched as part of a European network of chamber studies exploring the effects of ozone on farm crops. This network was EC-based, but Switzerland and Sweden also took part. Experiments were conducted over a period of four years, studying wheat (two years), barley (one year) and oats (one year).

In 1992 and 1993, a Swedish open-top chamber experiment was performed to investigate the effects of ozone on a ley consisting of red clover (*Trifolium pratense*), timothy (*Phleum pratense*) and meadow fescue (*Festuca pratensis*). Over the period 1994–96, an EU-funded research programme, ESPACE, was carried out using the chamber exposure system. Sweden’s contribution to this
programme was a series of experiments studying the effects on wheat of ozone, elevated carbon dioxide concentrations and water availability. All the chamber experiments on agricultural crops, apart from the one in Skåne mentioned first, were carried out at Östads säteri, north-east of Göteborg. Figure 4.3 shows the type of chamber used in the Swedish crop experiments.

4.3 Effects on cereals

Some plants develop characteristic visible injury symptoms when exposed to elevated ozone concentrations, usually consisting of spots or flecks on the leaves. To begin with, the flecks are pale: the leaf tissue is more or less intact, but chlorophyll and other pigments are broken down, hence the pale colour. This type of injury is known as chlorosis. Fairly soon, the damage develops a stage further, as the tissue dies and the flecks turn brown. This phenomenon is called necrosis. Spinach, clover and tobacco are examples of plants which suffer these types of injury, first chlorotic and later necrotic, following exposure to ozone, which is why they make good indicator plants for ozone.

Other plants do not respond with characteristic visible damage when exposed to realistic levels of ozone. In cereals, for example, the most important effect of ozone is to shorten the life of the leaves. They age more rapidly at higher ozone levels, and this curtails the period in which the leaves produce carbohydrates that can be supplied to the ear, adversely affecting the yield (Pleijel et al. 1997a). On the other hand, there are no typical, easily recognizable visible symptoms, beyond what can be interpreted as natural ageing (senescence), un-
less extremely high ozone concentrations are used. Figure 4.5 shows the percentage of green leaves on wheat roughly two weeks before harvest, following different ozone treatments.

The effect on leaf longevity which is described quantitatively in figure 4.5 was also visible to the naked eye. Figures 4.6a and 4.6b show what the plants in the charcoal-filtered and unfiltered treatments in the 1987 wheat experiment looked like when the chambers were removed prior to harvest, on completion of the season’s exposure.
The observations made at the macroscopic level are supported by biochemical and ultrastructural studies, which also suggest that ozone’s effects on wheat can to a large extent be interpreted as premature ageing (Grandjean & Fuhrer 1989; Ojanperä et al. 1992).

The flag leaf of a cereal is the leaf situated immediately below the ear. This leaf is considered to be of most importance in filling the grains of the ear with carbohydrates from photosynthesis. After a time it begins to age, and then it soon ceases to produce carbohydrates that can be supplied to the ear. Figure 4.7, based on microscopic studies, shows how the size of chloroplasts developed in wheat exposed to different ozone concentrations. The higher the ozone exposure, the more rapid the ageing process. A horizontal line has been added to the diagram to show at what point the chloroplast area had decreased to about 5 µm², roughly half the maximum size. The number of days between the flowering of the ear, when grain filling begins, and the time at which chloroplast size has decreased to around 5 µm² can serve as a measure of the effective longevity of the leaf. There are data in the scientific literature which support the assumption that this corresponds roughly to the period in which the flag leaves function as effective carbohydrate producers. When the yield was plotted against

![Figure 4.7](image-url)
flag leaf longevity, thus defined, a strong linear relationship was found, as shown in figure 4.8. Exposure to ozone after flowering is probably of far more significance for yield losses than exposure before flowering (Pleijel et al. 1998), since the ageing process is more easily induced at the post-flowering stage.

Since the late 1980s, a succession of open-top chamber studies of ozone’s effects on wheat production have been carried out, a relatively large proportion of them in the Nordic countries. Data from these Nordic experiments are presented in figure 4.9, in which yields relative to those obtained at zero exposure are plotted against the exposure indices AOT30 and AOT40, calculated according to the principles developed as a basis for critical ozone levels within the ECE. AOT is calculated by adding together, hour by hour, the amounts by which ozone concentrations exceed a certain level, in this case 30 and 40 ppb (see chapter 2). Linear relationships with a high correlation between yield and ozone exposure are obtained with both exposure indices, somewhat better in the case of AOT30. Other threshold levels of ozone result in poorer correlations, and most of the evidence now suggests that ozone only has an appreciable negative impact on wheat yields at levels above approx. 30–40 ppb.

As well as the size of the harvest, it is important to establish whether its quality is affected. One of the most important quality criteria is protein content, which in fact tends to be higher, the greater the adverse effect of ozone on the yield. This can be seen as a dilution effect. The supply of nitrogen available in the soil is the limiting factor for the total amount of protein the plants can produce. If production of carbohydrates is depressed, e.g. because of ozone exposure, there will be more protein in proportion to carbohydrate in the wheat grains, while the converse will be true if carbohydrate production is stimulated, e.g. by a higher concentration of carbon dioxide. However, this has to be quali-
fied somewhat. Total production of protein per hectare is lower when the overall yield of grain is reduced, but higher when the yield is enhanced by an elevated carbon dioxide concentration, for example. The conclusion, therefore, is that higher ozone levels result in a lower total production of protein, but since the total production of carbohydrate is reduced even more, the net result of ozone stress is a higher proportion of protein in the grains produced.

The effects of ozone on cereals other than wheat are less well researched. A number of ozone experiments on barley have been carried out, and the overall picture they provide is that this cereal is less sensitive than wheat (Pleijel et al. 1992). Recent analyses suggest that the difference in ozone sensitivity between wheat and barley is not all that great (Fuhrer 1996). Oats have been tested in a Swedish study, in which filtration of the air produced no effect at all (Pleijel et al. 1994a). Rye has not yet been tested for ozone susceptibility in any chamber experiment from which results have been published.

Figure 4.9. Relationships between relative yield and ozone exposure, expressed as AOT30 and AOT40, for open-top chamber experiments on wheat carried out in the Nordic countries. From Pleijel et al. 1997b.
4.4 Effects on legumes

Beans have been widely studied with respect to ozone damage in Europe and the United States, but only on a relatively small scale in Sweden. This is due to the greater commercial importance of beans in other countries. A collation of the European studies carried out (Fuhrer 1994) showed that French (common) bean (*Phaseolus vulgaris*) was possibly, on average, somewhat more sensitive than wheat. On the other hand, the results of different experiments varied much more widely in the case of French bean than in studies of wheat, making assessments of the effects of ozone more uncertain. This variation was probably due in part to the fact that there are many varieties of this species, with differing genetic properties.

Exposure of French bean to elevated ozone concentrations results in characteristic visible injury to the leaves, in the form of a spotted pattern which is initially pale, but soon turns a brownish colour. In south-west France, where open-top chamber experiments on this species have been carried out, the same type of damage was also observed in the open during the years with the highest ozone concentrations.

Another legume that is highly sensitive to ozone is lucerne (*Medicago sativa*), also known as alfalfa. This crop, which is not grown on a particularly large scale in Sweden, rapidly develops extensive, whitish injury symptoms when exposed to moderately elevated concentrations of ozone. In North American studies of alfalfa, production losses due to ozone have also been observed (Heck *et al.* 1988). The visible symptoms are illustrated in figure 4.10. Other important legumes which are ozone-sensitive include various species of clover. These are mainly used as forage crops, and are therefore dealt with in section 4.7.

Figure 4.10. Ozone injury to lucerne (alfalfa). Note that the younger leaves are symptom-free. Young leaves often contain higher levels of antioxidants and are generally less susceptible to ozone than older ones. Photograph: Håkan Pleijel/IVL.
4.5 Effects on potato and tomato

Tomato is the plant species with regard to which symptoms of ozone damage elicit most enquiries from growers and plant health experts. In the spring and early summer, quite extensive visible injuries sometimes arise in tomato plants grown outdoors or under glass, in the form of necrotic (dead) areas on the leaves, which later assume a brownish to bronze colour. Very little research has been done into ozone’s effects on tomato. The symptoms observed by growers in Sweden, and which cannot be attributed to known pests, tally with the accounts of ozone damage to tomato found in the scientific literature. For a site in eastern Småland (south-east Sweden), a comparison was made between the times such injuries occurred and periods of high ozone concentrations. It emerged that the symptoms always arose a few days after ozone episodes, but that they were not observed after every episode. There is a great deal to suggest that some of the damage observed is due to ozone, but it has to be borne in mind when interpreting these results that ozone episodes coincide with special, often warm and sunny weather conditions. To clarify the relationships between these factors in more detail and with greater certainty, more in-depth experimental studies would need to be carried out.

With respect to another important crop too, potato, the significance of ground-level ozone has not been adequately investigated, but there are indications that current ozone concentrations could cause production losses and other changes. The study carried out in Skåne in the early 1980s also included potato (Bintje). A moderate increase in ozone levels resulted in both reduced production and visible leaf injury (Skärby & Jönsson 1988). In potato, the visible symptom of ozone damage consists of a mottled, purple-brown pattern on the leaves (figure 4.11).

Figure 4.11. Early stage of ozone injury to potato. Photograph: Håkan Pleijel/IVL.
4.6 Effects on oilseeds
With regard to oilseeds, even less is known about the effects of ozone than in the case of potato and tomato. German studies of oilseed rape suggest that exposure to realistic levels of ozone has a clearly detrimental effect on yields, but that rape is nevertheless less sensitive than spring wheat, for example (Adaros et al. 1991; Hertstein et al. 1995). The current state of knowledge concerning ozone effects on commercially important oilseeds must be described as very inadequate.

4.7 Effects on forage crops
An open-top chamber study of field-grown forage crops has been carried out in Sweden. A grass–clover ley was exposed to four different ozone concentrations and harvested three times a year over two growing seasons. At all six harvests, the two highest ozone treatments resulted in a lower yield than the two lowest exposures. The effect, summarized in figure 4.12, was smaller in proportion to the exposure than in spring wheat, for example. The red clover and the two grasses included, meadow fescue and timothy, appeared to be roughly equally sensitive to ozone, with all three species exhibiting visible ozone injury in the two highest ozone treatments. A contributory reason why ozone had less effect on this ley than on wheat is probably that the biomass of forage crops is harvested before it has started to wither. In the case of wheat, the harvest consists of the grains, i.e. the mature seeds, and it is only gathered in when the plants have withered. Since plants which are beginning to age are usually more susceptible to ozone than plants at an earlier stage of development, it is reasonable that forage crops should be less sensitive. The lower sensitivity of young leaves is believed to be due to, among other things, higher levels of antioxidants, which provide protection against ozone.

Figure 4.12.
Relative yield of a grass–clover mixture in relation to ozone exposure, expressed as AOT40. From Pleijel et al. 1996.
Many clovers are sensitive to ozone, but there are considerable differences between genotypes. In a Finnish study of ozone effects on forage crops, very similar to the Swedish one, a larger yield reduction was observed, and it affected clover to a greater extent. In addition, the latter succumbed more rapidly to competition from grasses, the higher the ozone exposure was. A corresponding Swiss study (Fuhrer et al. 1994) found no appreciable effect on the overall yield of a grass–clover pasture, but showed that the clover was far more quickly displaced by the grasses at higher ozone concentrations.

4.8 Exposure–effect relationships
Several factors can influence the effect of a given ozone exposure on a plant. These factors can be divided into two main groups:

1) factors affecting the uptake or dose of ozone, and
2) factors affecting the scale of the response to a given dose.

Typical response-modifying factors, which have already been touched on briefly, are the development stage of the plant and the strength of its antioxidant defences. As mentioned in chapter 1, antioxidants are substances present in plants (and animals) which react with strong oxidizing agents and thus render them harmless. Dose-modifying factors are connected, on the one hand, with micrometeorological conditions around the plant and, on the other, with the plant’s own mechanisms for controlling its gaseous exchange with the surrounding air. These different elements are discussed below.

4.8.1 Transport of ozone in the atmosphere
Let us assume that we know the ozone concentration at a given point, e.g. 5 m above a wheat field. To get inside a wheat plant, where it can do harm, the ozone first has to be transported down to the level of the field. This transfer is governed by atmospheric movements which are dependent on wind strength, thermal stratification of the air above the plants and the roughness of the surface below. That the last-mentioned factor should be of significance is perhaps somewhat surprising, but it is in fact very important. It explains why vertical gradients of ozone concentrations, for example, arise far more easily over a field of wheat than over and in a spruce forest. The spruce forest is considerably more uneven and therefore generates greater mechanical turbulence for a given wind speed. The decisive factor here is how much friction there is between the moving air and the surface over which it passes. The smoother the surface, the lower the friction, and hence the less mixing of the air there is and the slower the transfer of ozone (for example) from one height above the ground to another.
Another way of expressing this is to say that the aerodynamic resistance ($R_a$ in figure 4.15) to vertical transport of ozone is greater over a comparatively smooth surface. If straw shorteners are used in a field, its roughness will be reduced. The crop will be lower and the surface therefore smoother, and consequently the aerodynamic resistance will increase. This means that the ozone dose to the field will be smaller than if straw shorteners had not been applied.

Figure 4.13 shows how marked the gradients of ozone concentration and AOT40 were found to be over a field of barley. The most pronounced gradients occur at night, owing to the fact that winds are lighter then and the air is often stably stratified (see chapter 1). This stability is due to the ground surface cooling down at night as a result of outward radiation not being offset by incoming sunlight. If the air closest to the ground is colder than the air above it – i.e. an inversion has arisen – vertical transfer of air between different levels is impeded. The gradient for AOT40 is much more marked than the gradient for the mean concentration, as AOT40 is based on a concentration threshold (40 ppb). The probability of that threshold being exceeded decreases rapidly with a falling mean concentration. Any attempt to estimate yield losses on the basis of concentrations recorded, say, at monitoring stations must take the concentration gradient into account. Otherwise there is a danger that losses will be greatly overestimated.

![Figure 4.13](image-url)

Figure 4.13. Average ozone concentration and AOT40 over a barley field, during the day and at night, in relation to the height above the ground at which measurements were made. The height of the crop was about 1 m. From Pleijel et al. 1995b.

### 4.8.2 Transfer of Ozone into the Plant

Once aerodynamic resistance has been taken into account – it can be calculated by micrometeorological methods – we know the concentration of ozone at the plant level. There are then two more barriers which this ozone has to overcome.
to reach a plant’s internal tissues. The first is the laminar boundary layer immediately surrounding the leaf, where the air is so still that molecules can only be transferred by diffusion. How thick this layer is depends on the wind strength and the shape of the leaf. Large, broad leaves have a larger boundary layer resistance ($R_b$ in figure 4.15) than small, segmented ones. When this barrier has been penetrated, ozone can enter the plant through its stomata. This also occurs by diffusion. The strength of the resistance at this point to a gas entering a plant depends on the various factors affecting the degree of stomatal opening. Of these, light is of dominant importance (figure 4.14). It is through the stomata that the carbon dioxide needed for photosynthesis diffuses into the plant.

Since at night there is no light and photosynthesis therefore cannot occur, plants normally do not have their stomata open then. A price they inevitably have to pay for opening them is a loss of water vapour to the atmosphere. In other words, by closing their stomata at night, plants save water at a time when photosynthesis is in any case not possible. Furthermore, the great importance of water conservation for plants explains why dry soil, and also dry air, can result in the stomata closing even during the daytime. All the factors which affect stomatal opening also influence plants’ uptake of ozone.

It may be appropriate in this context to take into account the effects of the open-top chamber on plant ozone uptake. The rate of air mixing in a chamber is constant and fairly high. This means that the boundary layer resistance ($R_b$) is probably usually somewhat lower in open-top chambers than outside them, which would promote greater ozone uptake. On the other hand, light levels are somewhat lower, roughly 20% lower in the case of the chambers used for farm
crops in Sweden, which has the opposite effect. The soil can easily become drier inside a chamber than outside, but this is normally compensated for by irrigation. Usually, atmospheric humidity is roughly the same inside and outside the chamber, but in very warm, dry weather it can end up considerably lower in the chamber, which counteracts the uptake of ozone.

![Figure 4.15. Schematic diagram illustrating the factors affecting the representativeness of ozone measurements, and also the chamber effect on plants' uptake of ozone. $R_a =$ aerodynamic resistance to transfer of ozone between different layers of air, $R_b =$ boundary layer resistance, $R_c =$ plant surface resistance to ozone uptake, with the stomata the dominant factor involved. Both $R_b$ and $R_c$ can differ between the environment inside an open-top chamber (OTC) and outside it (AA).](image)

4.9 The most ozone-sensitive crops

Of the commercially important crops, wheat is the one that has been studied in greatest depth in Europe and for which there is the strongest scientific evidence of ozone sensitivity. Some varieties of French bean may be even more sensitive, but ozone susceptibility varies greatly from one type to another. Subterranean clover is a very ozone-sensitive species. It is very useful as a bioindicator for this
pollutant (Pihl Karlsson et al. 1995a) and is the plant which most rapidly produces visible injury symptoms even at moderately elevated ozone concentrations.

Figure 4.16. Harvesting ozone-exposed wheat. Photograph: Håkan Pleijel/IVL.

4.10 Important gaps in current knowledge

In the case of wheat, barley, forage crops, French bean and a number of less important crops, we now have a relatively good body of knowledge regarding ozone damage in the Nordic countries. One of the key tasks for the future is to further refine methods of estimating yield losses, which should be done by studying the relationships between the scale of effects and ozone uptake by plants. Good quantitative estimates of yield reductions and of the economic consequences of ozone for the agricultural sector are dependent on the development of such relationships. Existing AOT40-based models are too crude and static to permit sufficiently accurate estimates of yield losses. Development work in this area is now in progress in Europe as part of the Level II approach to critical levels of ozone. Researchers are moving on from the cruder, simpler Level I method, which has primarily relied on AOT40 to identify areas of Europe at risk of ozone damage, to a more sophisticated level, at which dose- and
response-modifying factors are taken into account. Only by means of a Level II approach will it be possible to quantify the effects of ozone.

One important response-modifying factor that needs to be studied more closely is the biochemical defence against ozone provided by antioxidants. Variations in antioxidant defences between species, between developmental stages and between geographical regions are an area of research which must be given the highest priority.

Crops for which there are indications of major effects occurring, but about which our knowledge in this area is virtually non-existent, include potato and tomato. Studies of these species are urgently needed. Oilseeds and rye are other crops about which we need to know more. Over the next few years, rye and potato are to be studied, potato as part of a major EU-funded project involving several groups in different countries.
SUMMARY

We know that ground-level ozone can adversely affect young trees, and there is a great deal to suggest that mature trees are equally sensitive to ozone. On a time-scale of one to a few years, deciduous species are more sensitive than conifers. Viewed over an entire forest rotation, however, conifers are at least as sensitive as deciduous trees. In a large-scale, long-term experiment, elevated concentrations of ozone were found to reduce growth and the chlorophyll content of older needles in young Norway spruces. After four years exposure to ozone, the trees’ biomass was around 5% lower compared with a control, corresponding to a reduction of 2% in the relative growth rate. A simple model simulation showed that critical levels of ozone (10 000 ppb-hours) could cause a volume reduction of 3–11% in spruce trees of harvestable age. Another Swedish experiment showed young birches to be affected by elevated ozone concentrations after exposure for only 3 months.

5.1 Introduction

Most European and US studies of the effects of ozone on forest trees have been carried out on young plants in laboratories (climate chambers) or in outdoor chamber systems. A general problem with these studies has been that they have covered very short periods, at most perhaps 5% of the life cycle of the trees concerned. In Europe, the species studied most closely are beech (Pearson & Mansfield 1994; Braun & Flückiger 1995; Mortensen et al. 1996; Steingröver et al. 1995), birches (Günthardt-Goerg et al. 1993; Pääkkönen et al. 1997a; Mortensen & Skre 1990) and spruces (Ogner 1993; Skärby et al. 1995; Holland et al. 1995; Lucas et al. 1988). The effects demonstrated include everything from visible foliar injury and premature ageing (senescence) of foliage to reduced chlorophyll content, depressed photosynthesis, altered allocation of carbon and
slower growth. In poplars, for example, the effects of ozone include both accelerated ageing of leaves and reduced growth (Mooi 1980; Keller 1988; Matyssek et al. 1993). In the case of conifers, statistically significant effects on growth have only been demonstrated in a few cases (Skärby et al. 1998). In addition, intraspecific variation in ozone sensitivity is known to exist, for example in silver birch (Pääkkönen et al. 1995) and Norway spruce (Payer et al. 1990; Karlsson et al. 1997). Extensive studies of oaks in the United States show that this genus, too, is susceptible to ozone damage (Samuelson & Kelly 1996 and 1997; Samuelson & Edwards 1993; Samuelson et al. 1996). However, we still lack a sound knowledge base concerning the effects of ozone on European forests from the practical forestry point of view (Fuhrer et al. 1997; Bussotti & Ferretti 1998).

Two long-term (4–5 years) and several short-term studies of how ozone affects Scots pine and Norway spruce were carried out in Sweden over the period 1979–97. In addition, an experiment has been conducted in which birches have been exposed to ozone for two years.

5.2 Effects on Scots pine

A study of ozone uptake and its effects on transpiration, net photosynthesis and dark respiration in Scots pine was carried out on two 20-year-old pines at the Jädraås Research Station in Gästrikland in 1979 (Skärby et al. 1987). Current-year shoots were exposed to gradually increasing ozone concentrations (60–200 ppb) in the course of a month in the late summer. The study showed that ozone uptake co-varied with the diurnal variation in stomatal opening, increasing during the daytime when the stomata were open and decreasing at night when they were closed. When exposure exceeded 125 ppb, the degree of stomatal opening during the night increased. By the end of the experiment, dark respiration had increased by 60%, while net photosynthesis was unchanged. Similarly, a study by Wingsle et al. (1992) has shown that short-term exposure to high levels of ozone can affect Scots pine.

5.3 Effects on Norway spruce

In the early 1980s, forest decline was identified in European public debate as an important threat to the environment. Elevated ozone concentrations were suggested as one possible cause of the problem. In view of the pre-eminent importance of Norway spruce to the Swedish forestry sector, a series of long-term experiments was launched in 1985 to study how the ozone levels occurring in Sweden affect young trees of this species. The trees were exposed in open-top chambers, a technique originally developed for agricultural crops (Heagle et al. 1973; see chapter 4).
5.3.1 RÖRVIK 1985–1989 ♦ PHOTOSYNTHESIS AFFECTED

The first chamber experiment was started in 1985 at the Rörvik field station on the Onsala peninsula, south of Göteborg. Cloned Norway spruce saplings were exposed to three different ozone concentrations in 12 open-top chambers over five growing seasons (Skärby et al. 1995). The aim of the study was to investigate how ozone influences physiological processes within young trees which are of significance for their growth, e.g. photosynthesis.

This experiment showed that elevated ozone concentrations (30 ppb above the ambient concentration, daytime) can reduce the chlorophyll content and alter the ultrastructure of needles, in particular the structure of the chloroplasts (Wallin et al. 1990; Sutinen et al. 1990). Similar changes occurred in trees exposed to unfiltered air (which has roughly the same concentration of ozone as the ambient air), though later than in the elevated ozone treatment. The rate of photosynthesis was also affected: it increased somewhat in young shoots, but decreased substantially in shoots that were two years or older (figure 5.1). The study was not designed in such a way as to permit a direct investigation of how growth was affected. Detailed studies of photosynthesis showed that the effects of ozone were above all linked to a lower carboxylation efficiency. This meant that rubisco, an extremely important enzyme which fixes atmospheric carbon dioxide in the ‘dark reactions’ of photosynthesis, was affected. The light
reactions involved in photosynthesis also responded to ozone exposure, but this effect was only of significance at low light intensities (Wallin 1990; Wallin et al. 1992a, b).

The results presented in figure 5.1 were used to estimate the effects of ozone on the total photosynthesis of a tree, by means of a model simulation (Skärby et al. 1995). This simulation showed that ozone affects total crown photosynthesis to a far greater extent in older trees (89 years) than in young trees (8 years). This is because needles that are at least two years old make up roughly 70% of the needle mass of an old tree (Schulze et al. 1977), while the proportion in younger trees is considerably smaller.

5.3.2 **The Ozone–Spruce Project at Östad, 1992–96**

*Growth Affected*

On the basis of the Rörvik results, a new long-term experiment on Norway spruce was launched. A new field station was established at Östads säteri, 50 km north-east of Göteborg.

![Figure 5.2. Norway spruces in open-top chambers at Östad. Photograph: Per Erik Karlsson/IVL.](image)

The main purpose of this study was to quantify the effects of ozone on growth in young Norway spruce trees. Another aim was to investigate how climatic and nutrient stresses influence the ozone effect. Drought stress was chosen as a climatic factor on the basis that earlier experiments had suggested that ozone disturbed regulation of the stomata (Wallin & Skärby 1992). Low phosphorus availability was chosen as a nutrient stress, since there is a risk of phosphorus deficiency or an imbalance between phosphorus and nitrogen in acidified forest soils in southern Sweden.
The design of the experiment is presented in detail in the appendix. A total of 828 Norway spruce saplings from a clone, propagated in 1989, were exposed to different ozone concentrations in 42 open-top chambers over four growing seasons, from 1992 to 1995 (figure A1, appendix). In addition, 12 experimental plots without chambers were used to assess the effects of the chamber technique. An optimum nutrient supply to each pot ensured that the plants had the best possible conditions for growth. When plants are cultivated in pots, they eventually outgrow their containers, with the result that their growth is restricted. To investigate whether the pots (120 l) did in fact restrict the trees’ growth during the experiment, a treatment without pots was also used. Here the saplings were planted together in a sand-filled basin, spaced at the same intervals as those in pots. The pots were not found to have any significant negative effects on growth; on the contrary, initially they appeared to favour growth.

Measurements of SO$_2$, NO$_x$ and soot, both in the open air and in chambers with the charcoal filter treatment (CF), showed that concentrations were very low even in the ambient air, with seasonal means of between 0 and 5 µg m$^{-3}$. In the chambers supplied with filtered air, levels of SO$_2$ and NO$_x$ were reduced by another 50% approximately. SO$_2$ and NO$_x$ thus did not cause any problems in the experiment, since their levels were so low. Ambient concentrations of soot were around the detection limit of 1 µg m$^{-3}$. Climatic measurements were made inside and outside the chambers. Air temperature and humidity were monitored continuously for at least three plots subjected to each of the ozone treatments. The measurements showed that the presence of the chambers had an appreciable effect on temperature, corresponding to an increase of 1°C (24-hour means calculated for days with a mean temperature >5°C). It could be said that the chambers transposed the entire experiment a few degrees of latitude south. On the other hand, there were no significant temperature differences between chambers receiving different treatments (detection level approx. 0.1°C; calculated for days with a mean temperature >5°C).

The spruces were exposed in open-top chambers to three different ozone levels (table 5.1, and tables A5 and A6, appendix): (1) charcoal-filtered air (CF) with an ozone concentration around 25% of that of the ambient air (corresponding to the preindustrial atmosphere); (2) non-filtered air (NF) with an ozone level corresponding to the ambient concentration; and (3) non-filtered air with added ozone bringing the concentration to 1.5 times the ambient level (NF+). The current critical level for forest trees (10 000 ppb-hours, see chapter 3) was exceeded every year in the NF+ treatment. The ozone concentrations used in this treatment must be regarded as entirely realistic, in comparison with those occurring in southern Sweden in recent years (cf. table 5.1 and chapter 3).
The study was designed as two separate experiments, each of which was sub-
jected to a separate statistical analysis (table A1, appendix): (1) the main ex-
periment (Ozone-m), involving three ozone treatments (CF, NF, NF+) com-
bined with optimum nutrient and water supplies, and two ozone treatments
(CF/PD, NF+/PD) combined with reduced availability of phosphorus; and (2)
the drought stress experiment (Ozone-d), comprising two ozone treatments
(CF/W, NF+/W) with optimum nutrient and water supplies and two ozone
treatments (CF/D, NF+/D) with a reduced supply of water.

The statistical analysis (analysis of variance) of the total biomass of the spruces
in the main experiment demonstrated an effect due to harvest date and treatment, but
no effect attributable to either the position of the plot within the experimental site
or the position of the plant within the plot (in terms of compass direction). This
meant that direction and plot did not need to be taken into account in the statisti-
cal analysis. Subsequently, a two-way analysis of variance was used, with ozone and
low phosphorus or drought as independent variables, or alternatively a three-way
analysis, with time also included as an independent variable.

5.3.2.1 Effects on needles
A common effect of ozone exposure is reduced chlorophyll concentrations in
needles or leaves. In the Rörvik study, the chlorophyll content of the spruces’
needles gradually declined. Significant effects of the ozone treatment could be
demonstrated in 2- to 4-year-old needles (Skärby et al. 1995). Similar results
were obtained in the experiment at Östad. Needle chlorophyll content was meas-
ured in December each year (1992–96). Figure 5.3 shows how ozone affected
the chlorophyll content of needles that developed in 1992: the levels in needles
from the different ozone treatments are presented as percentages of the con-
centrations in needles from the corresponding controls. The results of the
statistical analysis (two-way analysis of variance), which did not include NF, are
presented in table 5.2.
All the trees treated with ozone, apart from those exposed to ozone in combination with a low phosphorus supply, exhibited the same pattern over time. The chlorophyll content of 1-year-old needles, which had been exposed to ozone for two seasons, was higher than in the corresponding controls. The increase was significant in the case of NF+/W (18%) and NF+/D (11%). Levels then gradually decreased with increasing needle age, and in 3-year-old needles they had fallen by 13–24% compared with the CF controls. The decrease in chlorophyll content was significant in 2- and 3-year-old needles in all the ozone treatments. A similar trend was observed for NF, but with one year’s delay compared with NF+ (figure 5.3).

When trees were exposed to ozone combined with low phosphorus availability, a different pattern emerged. In this case, the chlorophyll content fell steadily from one year to the next (figure 5.3). A significant interaction between ozone and phosphorus was only found after the first season with a low phosphorus supply (1993). No interaction between ozone and drought could be demonstrated.

Table 5.2. Statistical analysis (two-way analysis of variance) of the treatments’ effects on needle chlorophyll content. P-values less than 0.05 (bold type) indicate significant differences between treatments or significant interactions between ozone and phosphorus or water availability. M = main experiment, D = drought stress experiment.

<table>
<thead>
<tr>
<th>YEAR</th>
<th>NEEDLE AGE</th>
<th>OZONE-M</th>
<th>OZONE*LOW PHOSPHORUS</th>
<th>OZONE-D</th>
<th>OZONE*DROUGHT</th>
</tr>
</thead>
<tbody>
<tr>
<td>1992</td>
<td>Current-year</td>
<td>0.808</td>
<td>0.863</td>
<td>0.911</td>
<td>0.494</td>
</tr>
<tr>
<td>1993</td>
<td>1 year</td>
<td>0.545</td>
<td>0.010</td>
<td>0.034</td>
<td>0.543</td>
</tr>
<tr>
<td>1994</td>
<td>2 years</td>
<td>&lt;0.001</td>
<td>0.190</td>
<td>0.009</td>
<td>0.123</td>
</tr>
<tr>
<td>1995</td>
<td>3 years</td>
<td>&lt;0.001</td>
<td>0.303</td>
<td>&lt;0.001</td>
<td>0.267</td>
</tr>
</tbody>
</table>

Figure 5.3. Change over time in the chlorophyll content (% of the corresponding CF control) of needles from 1992 shoots. NF = non-filtered air; NF+ = added ozone; NF+/W = added ozone and well watered; NF+/D = added ozone and drought stress; NF+/PD = added ozone and low phosphorus level.
The reduction in the chlorophyll content of older needles suggests that ozone could cause abnormal ageing of needles. This represents an interesting, but as yet unproved, link with the increasingly widespread crown thinning observed in Europe, in both deciduous trees (oak and beech) and conifers (chiefly spruce, but also pine), over the last 10 years. Ozone may be a contributory cause of premature leaf- and needle-fall.

5.3.2.2 Effects on growth

The biomass growth of the spruces (needles, branches, stems and roots) was measured regularly throughout the experiment. Figure 5.4 shows total biomass production at the last six harvests in the main experiment, with the three different ozone treatments and a balanced nutrient dosage. No significant differences were found between the treatments at any individual harvesting date, but the control (CF) produced the largest amount of biomass, while the highest ozone treatment (NF+) yielded the smallest amount.

To obtain as many replications as possible, an analysis of variance was performed which included the effects of time, ozone, and drought or low phosphorus. Figure 5.5 shows the effects of the highest ozone treatment (NF+, 1.5 x ambient concentration) on the biomass harvested during the three periods in the main (Ozone-m) and drought stress (Ozone-d) experiments, as a percentage of the biomass in the relevant control (CF). Period 1 comprised 1992–93, period 2 1994 and period 3 1995–96. All the data (excluding NF) for each group of treatments were included. Data for the high ozone treatments with both balanced and low phosphorus doses, on the one hand, and with both normal watering and drought stress, on the other, could thus be combined and compared with the corresponding controls, providing a larger set of data for testing. The effect of

![Figure 5.4. Effects of different ozone treatments on growth (total biomass) in the main experiment, between April 1995 and May 1996.](image-url)
ozone was considerably smaller than the effect of drought or low phosphorus dose. In the main experiment (Ozone-m), the negative effect of ozone on biomass increased gradually from year to year. In the drought stress experiment (Ozone-d), too, ozone reduced the biomass, but the effect was more complex. Overall, i.e. for the whole-tree biomass and over a period longer than one year, the effect of ozone in combination with drought stress or low phosphorus supply does not appear to be either synergistic or antagonistic.

The effect of elevated ozone was not significantly negative until during period 3, i.e. after four seasons of exposure. At that point, the high ozone (NF+) treatment caused a 5% decrease in biomass in the main experiment and a 4% decrease in the drought stress study, compared with CF (the control with a low ozone level). The effect of the drought treatment was already significantly negative during period 2: biomass was 19% lower after three seasons and 31% lower after four seasons compared with spruces that had been watered regularly. The low phosphorus dose resulted in 23% lower biomass in period 3, compared with a normal dose.

The relative growth rate over the entire life of the experiment was calculated by plotting logarithms of all the harvest results against time. Linear regression analysis showed a very high correlation coefficient for all the treatments. Elevated ozone concentrations (NF+) caused a decrease in the relative growth rate of about 2%, in both the main and the drought stress experiment, compared with the control (CF). Table 5.3 presents the estimated orders of magnitude of the treatment effects. If the growth rate was instead based on measurements of stem volume, the same reduction due to ozone was obtained, i.e. about 2%.
These results could be used to test different scenarios in model simulations and to estimate production losses in Norway spruce in southern Sweden attributable to ozone. However, important questions still need to be answered (see 5.8 for further discussion):

- Can the clone studied here be regarded as representative of the Norway spruces growing in Sweden with regard to their response to ozone?
- Are trees able to develop ozone tolerance after a certain time or at a certain age, and can they recover following exposure to high concentrations?
- How does the ozone sensitivity of mature trees compare with that of saplings?

The average annual AOT40 value for the NF+ treatment during the 4 years of the experiment was about 20 000 ppb-hours, which is twice the critical ozone level of 10 000 ppb-hours. The associated average reduction in growth was 2%. If we assume that growth reduction is directly proportional to ozone dose, then the present critical level of ozone, 10 000 ppb-hours, would give rise to half that reduction in the growth rate in Norway spruce, i.e. 1%.

To illustrate the possible consequences of a 1% reduction in the relative growth rate, we carried out a simple calculation. The basic data consisted of a model simulation of stem volume growth in a Norway spruce on a high-productivity site in southern Sweden over 110 years, performed at the Swedish University of Agricultural Sciences (Ulf Söderbergh, personal communication). This simulation was used as a control. The control was then modified by introducing a 1% growth rate reduction during three different periods in the tree’s life cycle: 0–5, 0–20 and 0–110 years. The aim was to simulate the development of a Norway spruce assuming that ozone adversely affects stem volume growth (1) only in young plants, (2) only in young trees during their intense growth period, or (3) throughout the life cycle of trees until they are harvested. The results are presented in figure 5.6. If ozone only affected the growth of the young plant, the stem volume of the spruce at harvestable age would be reduced by roughly 3%.

<table>
<thead>
<tr>
<th>OZONE-M</th>
<th>OZONE-D</th>
<th>DROUGHT</th>
<th>LOW PHOSPHORUS</th>
</tr>
</thead>
<tbody>
<tr>
<td>-2</td>
<td>-2</td>
<td>-12</td>
<td>-8</td>
</tr>
</tbody>
</table>

Table 5.3. Estimated reductions (%) in the relative growth rate (total biomass) caused by ozone (1.5 x ambient concentration), drought and low phosphorus availability. The ozone effect is compared with a charcoal-filtered control, the drought stress effect with a well-watered control, and the effect of low phosphorus with a control given a balanced nutrient dose.
If on the other hand ozone acted throughout the phase of intense growth, 0–20 years, the reduction would be about 8%. Finally, if ozone depressed the growth rate over the entire growing period of the spruce, its stem volume at harvestable age would be reduced by around 11%.

This simple calculation can give some indication as to what scenarios may be relevant: (1) The influence of ozone on the sapling over a 5-year period means that the growth of the tree is retarded and therefore has a major impact on its final volume, or alternatively on the date at which it can be harvested. (2) If ozone affects the tree’s growth rate throughout its life cycle in the same way as it affects that of the young tree, very sizeable production reductions could follow. This is assuming that the tree’s capacity to recover is limited.

For a forest owner in southern Sweden, the volume reduction caused by current ozone concentrations could entail losses in earnings of the order of SEK 10 000 per hectare of felled forest, depending on whether the harvested trees are sold for timber (table 5.4).
In the Ozone–Spruce study, then, it was found:

- that ozone resulted in an increase in the chlorophyll content of 1-year-old needles and a subsequent decrease in needles that were 2 years or older. In 3-year-old needles, the chlorophyll content was found to have decreased by 13–24%;
- that ozone at an AOT40 level averaging 20,000 ppb-hours per season (twice the critical level for forests) caused a reduction of around 2% in the growth rate over the entire period of the experiment. Estimates suggest that current ozone levels, compared with those occurring 100 years ago, reduce the biomass of Norway spruce of harvestable age in southern Sweden by between 3% and 10%;
- that low phosphorus availability (a moderate deficiency) reduced the relative growth rate by about 8% over the period of the experiment;
- that recurrent periods of drought reduced the relative growth rate by around 12% over the period covered by the experiment.

### Table 5.4. Estimated economic losses due to the effects of ozone, arising in conjunction with the felling of Norway spruce on a high-productivity site in southern Sweden. Two different levels of ozone effects have been used for the estimates, corresponding to a 3% and an 11% volume reduction (figure 5.6), and separate estimates are shown for different end uses of the trees harvested (saw timber or pulpwood). The growing stock at harvestable age is assumed to be 300 m³ ha⁻¹, the price of saw timber SEK 450 m⁻³ and the price of pulpwood SEK 250 m⁻³.

<table>
<thead>
<tr>
<th>REDUCTION OF STEM VOLUME DUE TO EFFECTS OF OZONE</th>
<th>ECONOMIC LOSS (SEK ha⁻¹)</th>
</tr>
</thead>
<tbody>
<tr>
<td>3 %</td>
<td>4,000</td>
</tr>
<tr>
<td>11 %</td>
<td>14,800</td>
</tr>
</tbody>
</table>

In the Ozone–Spruce study, then, it was found:

- that ozone resulted in an increase in the chlorophyll content of 1-year-old needles and a subsequent decrease in needles that were 2 years or older. In 3-year-old needles, the chlorophyll content was found to have decreased by 13–24%;
- that ozone at an AOT40 level averaging 20,000 ppb-hours per season (twice the critical level for forests) caused a reduction of around 2% in the growth rate over the entire period of the experiment. Estimates suggest that current ozone levels, compared with those occurring 100 years ago, reduce the biomass of Norway spruce of harvestable age in southern Sweden by between 3% and 10%;
- that low phosphorus availability (a moderate deficiency) reduced the relative growth rate by about 8% over the period of the experiment;
- that recurrent periods of drought reduced the relative growth rate by around 12% over the period covered by the experiment.

### 5.4 Östad Birch, 1997–99

After spruce and pine, the most important type of tree for the Swedish forest industry is birch. In Sweden, and also in Finland, there is a growing interest in deciduous species, on account of their ability to improve long-term soil productivity.

Several experiments suggest that young deciduous trees are affected more rapidly by high ozone concentrations than conifers. In different birch species, ozone has been shown to affect growth (Mortensen & Skree 1990; Matyssek et al. 1992; Pääkkönen et al. 1996; Pääkkönen et al. 1997a, b), ageing of leaves (Matyssek et al. 1992; Günthardt-Goerg et al. 1993), stomatal density (Pääkkönen et al. 1993), chloroplasts (Pääkkönen et al. 1995) and photosynthesis (Pääkkönen et al. 1996). There is thus ample evidence that ozone has effects on birch. A broader range of data, above all including additional information on growth,
will provide a sound basis for determining a critical level for the effects of ozone on growth in birch species.

To study the ozone sensitivity of birch in southern Sweden in relation to the critical level, a two-year chamber experiment was started at Östad in spring 1997. Silver birches were raised from seed in January of that year and planted in open-top chambers at the beginning of June, when they were about 50 cm high. Figure A1 in the appendix shows a plan of the experimental site. The birch experiment involved 4 treatments, with 4 replications per treatment, which means that 16 of the 56 plots were used. The seedlings were exposed to four different ozone levels up to the end of November 1997 and were subjected to further exposure in 1998 (table 5.5). Since ambient ozone concentrations were relatively low in 1997, ozone doses during that season ended up below the current critical level of 10 000 ppb-hours, with the exception of the highest ozone treatment (NF++), in which the ozone dose was roughly three times the critical level.

Table 5.5. Ozone concentrations (means) and ozone doses during daylight hours in the different treatments used in the open-top chamber experiment on birch at the Östad field station, 4 June–30 September 1997.

<table>
<thead>
<tr>
<th>Treatment</th>
<th>Ozone concentration (ppb)</th>
<th>AOT40 (ppb-hours)</th>
</tr>
</thead>
<tbody>
<tr>
<td>CF</td>
<td>Filtered air (control)</td>
<td>6</td>
</tr>
<tr>
<td>NF</td>
<td>Non-filtered air</td>
<td>30</td>
</tr>
<tr>
<td>NF+</td>
<td>Non-filtered air+20 ppb extra ozone</td>
<td>40</td>
</tr>
<tr>
<td>NF++</td>
<td>Non-filtered air+80 ppb extra ozone</td>
<td>65</td>
</tr>
</tbody>
</table>

The highest level of ozone (NF++) very rapidly affected the birches. After just three months’ exposure, accelerated leaf-fall could be observed (figure 5.7). In addition, it was noted that around 10% of the leaves were not ageing normally.

Figure 5.7. Accumulated leaf-fall during 1997 in the Östad Birch experiment, as a percentage of the total number of leaves.
(figure 5.8). The cells died before their nitrogen had been withdrawn into the stem (figure 5.9).

![CF and NF++ leaves](image)

**Figure 5.8.** Examples of birch leaves exposed to filtered air and the highest ozone level. Leaves from the filtered air treatment (CF) showed normal yellowing. Ozone-exposed leaves (NF++) were bronze-coloured.

![Graph](image)

**Figure 5.9.** Nitrogen concentrations in birch leaves collected in October 1997 in the open-top chamber experiment at Östad.

Root growth, in particular, was adversely affected by the NF++ treatment, showing a 25% reduction compared with CF. As the ozone dose increased, the distribution of biomass between shoots and roots was gradually affected, to the disadvantage of the roots (figure 5.10). A statistically significant increase in the shoot/root ratio was obtained even with the NF treatment.
The effects of ozone on the biomass of the entire trees were more complex (figure 5.11). After one season, growth was significantly higher in the NF and NF+ treatments than in CF, but lower in the NF++ treatment, although not significantly lower than in CF. Ozone’s effects on whole-tree growth might be attributable to a combination of two different effects. On the one hand, ozone causes a redistribution of biomass to above-ground plant parts. In the short term, this favours growth, since more biomass (an increased leaf area) becomes available for photosynthesis. On the other hand, high concentrations of ozone have an adverse effect on growth, since the leaves are shed sooner and are thus productive for a shorter period. At very high ozone levels, the latter effect is so marked that it overshadows the first, beneficial effect. What is more, in the longer term, the first effect will be detrimental to tree growth, since a reduced root biomass can result in decreased uptake of nutrients and water. The earlier leaf-fall, combined with reduced withdrawal of nitrogen from the leaves to the stem, may be expected to further accentuate any nutrient imbalance.

Figure 5.10. Effect of ozone on the shoot-to-root ratio (dry weight) in birches harvested in November 1997 in the chamber experiment at Østad.

Figure 5.11. Total biomass (dry weight), including leaves, of birches harvested in November 1997 in the chamber experiment at Østad.
The most important findings after one year’s exposure of birches to ozone are:

• that ozone accelerates leaf-fall. The trees exposed to the highest ozone levels did not have time to withdraw all the nitrogen from their leaves to their woody parts before the leaves were shed;

• that ozone at the levels occurring in ambient air affects the distribution of biomass between shoots and roots, to the disadvantage of the roots.

### 5.5 Diagnosing the effects of ozone

How can the effects of ozone be distinguished from those of other stresses, such as nutrient deficiencies, drought, high light intensity, or other pollutants? There is an urgent need for reliable methods that could be used routinely, or at least on a large scale, to ascertain to what extent different stress factors are influencing the vigour of forest trees. Such methods would have a very important part to play in confirming that experimentally based conclusions about the scale of the problems are truly applicable to the wider ecosystems concerned. A simple and specific methodology to distinguish the impacts of ozone from those of other stresses would provide a good basis for a survey of ozone’s significance for forest trees in Sweden.

Trees often react to stress by shedding needles or leaves, and estimates of crown density are the commonest means of classifying tree vigour. As a diagnostic method, though, they are less suitable, since needle or leaf loss is a fairly non-specific response to a variety of stresses, including ozone stress. In addition, as has already been noted, needle- or leaf-fall is the last in a series of changes. By means of light microscopy and electron microscopy, responses can be detected at an earlier stage.

Electron microscopic studies of spruce and pine needles and birch leaves show that ozone-related changes first appear in the chloroplasts, the parts of cells where photosynthesis occurs. The chloroplasts become smaller, and this process begins in the layers of cells beneath the epidermis, in the part of the needle (leaf) facing the sky (Sutinen 1987; Sutinen et al. 1990; Pääkkönen et al. 1995; Sutinen et al. 1998b). Other cell layers remain unharmed. As time passes, however, these changes penetrate deeper into the needle. The basic substance of the chloroplast, the stroma, darkens and becomes granular, and it becomes difficult to discern its membranes, or thylakoids, under the microscope. Other cell organelles only exhibit changes at a later stage. The mitochondria, for example, show no structural changes until the cytoplasm of the cell has been completely destroyed. Older needles show more numerous and more serious changes than young ones.
In the Rörvik study, when trees were exposed to around 60 ppb of ozone (24-hour mean) for two seasons (1985 and 1986), all the chloroplasts in their needles were found in April 1987 to have been affected, having decreased in size by about 25%. In July 1988, changes also began to appear in needles from trees exposed to non-filtered ambient air, compared with those growing in filtered air. At the time, these needles were about 14 months old. In needles that had developed in charcoal-filtered air, on the other hand, changes of this type were never observed (Sutinen et al. 1990). Spruces from the open-top chamber study at Östad were also examined, and the same pattern of damage was found as in the Rörvik experiment.

If ultrastructural changes are to be used to diagnose ozone effects, they must be specific to ozone, i.e. they must be of a kind which cannot be produced by any other stress. Pollutants such as SO₂ and NO₂ (Rantanen et al. 1994), acid rain (Holopainen & Nygren 1989) and fluoride (Soikkeli & Tuovinen 1979) have been found to induce different types of changes in needles, and these changes in turn differed in appearance from those caused by ozone. Deficiencies of nutrients, e.g. magnesium, potassium and calcium, also give rise to structural changes distinct from those arising from ozone exposure (Fink 1989). The effects of a phosphorus deficiency were studied in the Östad experiment, in which this factor was found to induce changes in the chloroplasts, mitochondria and cytoplasm (Sutinen et al. 1998a). Unlike those caused by ozone, these changes (which included increased chloroplast size) first occurred in current-year needles.

Most studies of how ozone affects coniferous and deciduous species have been carried out on trees exposed to ozone by means of various exposure systems. The response to ozone has been found to be similar, regardless of the system used. In a field study of Scots pine in Finland, many trees displayed the same symptoms of ozone damage (Sutinen et al. 1998b), indicating that ultrastructural methods could in future be developed to diagnose ozone effects in the field. However, it has still to be demonstrated that these methods are entirely ozone-specific and can be used on a large scale.

### 5.6 Ozone combined with drought stress or phosphorus deficiency

Plants take up carbon dioxide through their stomata, at the cost of a loss of water. Under drought stress, plants react by closing their stomata, thereby reducing their uptake of ozone. In other words, drought would appear to reduce rather than increase the risk of ozone damage. However, a number of studies have shown that trees exposed first to ozone and later to a period of drought lose more water than trees not exposed to ozone (Norway spruce: Wallin &
A hypothesis frequently put forward in this context is that ozone disturbs stomatal function. Other results point in a direction which contradicts the hypothesis that ozone harms the stomata (Karlsson et al. 1997) and suggest that the combination of drought and ozone in fact provides protection against adverse ozone effects on growth (Temple et al. 1993). According to Mansfield (1998), however, there are strong indications that ozone disturbs the water relations of many plants by affecting the stomatal opening mechanism. He refers to a ‘sluggish stomatal response’, which can result in a greater uptake of ozone in a drought stress situation. Pääkkönen et al. (1998) showed that birch plants subjected to severe drought stress were protected from ozone damage, whereas milder drought was accompanied by greater ozone damage. This suggests that the effect of ozone is dependent on the severity of the drought. Another ozone effect of great relevance to a plant’s ability to maintain satisfactory water status is the promotion of shoot growth at the expense of root growth. Scientific consensus regarding the effects of ozone combined with drought stress has yet to emerge.

The results of the Östad study (see above) offer no clear picture of how ozone stress interacts with drought stress or phosphorus deficiency. As shown in figure 5.3, chlorophyll levels initially increased in spruce needles exposed to ozone, and then gradually decreased again. The same pattern occurred in needles exposed to both drought and ozone, but here the response was more muted in both directions. The interaction between ozone and drought was not statistically significant (table 5.2); whether this reflects some form of protection against ozone damage is open to speculation. Phosphorus deficiency interfered with ozone with regard to the effects on chlorophyll content, which in this case fell continuously with time and needle age. This could be taken to mean that the combination of ozone and insufficient phosphorus initially made needles more sensitive to ozone, but this effect was only significant in one-year-old needles.

There were also differences in the pattern of ozone’s effects on biomass, depending on whether or not the ozone stress was combined with drought stress or a phosphorus deficiency. Overall, i.e. in terms of the whole-tree biomass and over a period of more than a year, the effect of ozone combined with drought or phosphorus deficiency appeared to be neither synergistic nor antagonistic.
5.7 Effects on ozone on older trees

We have a poor understanding of how ozone affects older trees, and the results that do exist are partly contradictory. In red oak (*Quercus rubra*), for instance, ozone sensitivity increases with age (Samuelson & Edwards 1993; Edwards *et al.* 1994), whereas it decreases with age in giant sequoia (*Sequoiadendron giganteum*; Grulke & Miller 1994) or remains unchanged, as in Douglas fir (*Pseudotsuga menziesii*; Smeulders *et al.* 1995).

For practical and economic reasons, it has proved difficult to conduct controlled experiments on mature trees over extended periods of time. In the United States, however, a few studies have been carried out on red oak, comparing the effects of ozone between seedlings and 30-year-old trees, for example (Samuelson *et al.* 1996). The 30-year-olds turned out to be more sensitive: exposure to ozone resulted in retention of carbon in the form of starch in their leaves, and in reduced fine root growth (Samuelson & Kelly 1996). This was not the case with the seedlings. Ozone uptake by leaves and needles also appears to be crucial in determining whether older or younger trees are more sensitive (Samuelson & Kelly 1997). Various types of models have begun to be used to simulate the effects of ozone at the tree and stand levels. Depending on the climatic and soil conditions involved, very sizeable variations in growth and in ozone effects emerge. According to Chappelka and Samuelson (1998), more research is needed if the effects of ozone are to be scaled up from small to large trees, and from physiological processes to growth.

A simulation of the impact of ozone on photosynthesis in the crowns of Norway spruces of different ages (8 and 89 years) was carried out using data from the Rörvik study (Skärby *et al.* 1995). The ozone effect was much greater in the case of the 89-year-old spruce: crown photosynthesis decreased by up to 36%, compared with just 7% in the young tree. Shading effects in older trees were not taken into account, since measurements performed on an 89-year-old spruce (Schulze *et al.* 1977) have shown that needles aged two years or older account for 67% of a tree’s total uptake of carbon in the course of a year, irrespective of light conditions.

The forests of the San Bernardino Mountains, south of Los Angeles in California, are the best-documented example of a coniferous forest ecosystem affected by ozone (Miller *et al.* 1997). Here researchers have been able to show how ozone combined with various biotic and abiotic factors has affected growth and species composition. Both old and young trees have been affected, the most sensitive species being ponderosa (western yellow) pine (*Pinus ponderosa*).

At the Skogaby experimental site in Halland (south-west Sweden) and elsewhere, research is in progress to establish whether, and if so how, the results of
experiments on young saplings can be extrapolated and validated for older trees and perhaps entire stands. At Skogaby, ozone fumigation of 35-year-old Norway spruces is being carried out using branch chambers, located at a height of 12 m in the stand, which is about 15–16 m high (see figure 5.12). Annual measurements are being made of photosynthesis, transpiration, ultrastructure and lipid composition. The results will subsequently be used in process-oriented growth models and to evaluate epidemiological studies.

Figure 5.12. Branch chamber measurements of ozone sensitivity in mature trees were started in 1998 at the Skogaby experimental forest in southern Halland. Photograph: Göran Wallin.
5.8 Differing sensitivity to ozone

A rough-and-ready way of classifying Swedish forest trees (deciduous and coniferous) in terms of their ozone sensitivity is to divide them into fast- and slow-growing groups. Rapidly growing trees generally have a higher rate of gaseous exchange through their stomata and take up more carbon dioxide per unit of time than those that grow slowly. Since ozone enters a plant via its stomata, their degree of opening is of great significance in determining how much ozone reaches the plant’s leaf tissues. Trees subjected to drought grow more slowly and normally have a low exchange of gases through their stomata.

Depending on the time-scale applied and also on the parameter considered (e.g. visible injury to needles or leaves, premature shedding of needles/leaves, sensitivity to other stresses, stem growth/production), the answer to the question which tree species is most sensitive can vary. Table 5.6 is an unranked listing of the tree species occurring in Europe which have been identified as being ozone-sensitive.

Table 5.6. List (unranked) of tree species grown in Europe that have shown sensitivity to ozone.

<table>
<thead>
<tr>
<th>English Name</th>
<th>Latin Name</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Hybrid poplar</td>
<td><em>Populus euroamericana</em></td>
<td>Matyssek et al. 1993</td>
</tr>
<tr>
<td>Beech</td>
<td><em>Fagus sylvatica</em></td>
<td>Braun &amp; Flückiger 1995</td>
</tr>
<tr>
<td>Silver birch</td>
<td><em>Betula pendula</em></td>
<td>Pääkkönen et al. 1997a,b</td>
</tr>
<tr>
<td>Norway spruce</td>
<td><em>Picea abies</em></td>
<td>Wallin et al. 1990</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Karlsson et al. 1995</td>
</tr>
<tr>
<td>Sitka spruce</td>
<td><em>Picea sitchensis</em></td>
<td>Lucas 1989</td>
</tr>
<tr>
<td>Scots pine</td>
<td><em>Pinus sylvestris</em></td>
<td>Kellomäki &amp; Wang 1998</td>
</tr>
<tr>
<td>Aleppo pine</td>
<td><em>Pinus halepensis</em></td>
<td>Gimeno et al. 1992</td>
</tr>
<tr>
<td>Eucalyptus</td>
<td><em>Eucalyptus spp.</em></td>
<td>Monk &amp; Murray 1995</td>
</tr>
</tbody>
</table>

In terms of the effects of exposure over as short a period as one or two seasons, the deciduous species beech, birch and poplar are more sensitive than spruce and pine. This is why European researchers involved in determining critical ozone levels for forest trees have agreed to choose beech as representing the most ozone-susceptible receptors (see also chapter 2).

Over a longer time-frame, such as an entire forest rotation, the answer to the question of the most ozone-sensitive species may be that conifers are at least as sensitive as deciduous trees. The reason for this is that, over the life cycle of a
spruce or pine, the ozone effect can gradually increase, since – in spruce at least – an increasingly large proportion of the needle mass consists of older needles. This in turn means that the adverse effect of ozone on biomass accumulation is amplified as total photosynthesis in the crown decreases. In deciduous trees, the foliage is replaced every season. Consequently, each individual season is probably of roughly equal importance, provided that there are no lingering effects which lead to a reduced quantity of leaves with rising age or an altered shoot/root ratio following ozone exposure.

5.9 Gaps in current knowledge
Sweden and Europe need more, and more certain, knowledge concerning a number of questions relating to the effects of ozone on forest trees:

5.9.1 How much ozone is taken up in different climates?
Ozone has to be taken up by leaves/needles before it can have an effect. In summer, the climate of the Nordic countries, compared with that of continental and southern Europe, is favourable to ozone uptake, owing to such factors as long hours of daylight and relatively high humidity. At present, a concentration-based measure of exposure forms the basis for critical levels of ozone. Data on ozone concentrations are collected from different measuring sites in Europe and converted into a dose which describes the amount of ozone plants have been exposed to over a given period (AOT40; see chapter 2). However, work is also under way to develop an uptake-based exposure index, taking into account the degree of stomatal opening over time. The aim is to get closer to a real, effects-related ozone dose, rather than the more administrative dose which AOT40 represents. There is a great deal to suggest that the new, uptake-based index will show that the effects of ozone in the Nordic region are significant, and possibly more far-reaching than has previously been assumed in comparison with the rest of Europe (Emberson et al. 1998).

5.9.2 How ozone-sensitive are pine, oak, aspen and goat willow?
With the exception of spruce, beech and birch, our knowledge of how different Swedish tree species respond to ozone is still limited. We know relatively little about Scots pine and a range of deciduous trees (poplar, oak, aspen, ash and goat willow). Scots pine, oak, goat willow and aspen should be studied more closely. These trees may be at least as important as the better-understood species in the years to come, as sources of raw materials for the paper pulp, joinery and bioenergy sectors.
5.9.3 Sensitivity of seedlings, older trees and entire forests

It should in future be possible to use models to translate experimental data obtained from ozone exposure of seedlings or saplings into ozone effects on older trees. Models should also be able to extrapolate the ozone impact to realistic field conditions over large areas. This is a new and extensive area of research, and one in which work has in fact only just begun. Nevertheless, a number of models do already exist, and they should be tested with regard to the effects of ozone on trees of different ages. The Ozone–Spruce study at Östad has generated a very large database which can be used in various applications involving modelling of growth and biomass production. The results of the branch chamber study at Skogaby will be of use in verifying these modelling studies.

5.9.4 Is it possible to develop diagnostic methods for field use?

It is still difficult to extrapolate knowledge gained from controlled ozone experiments to conditions in the field. The question is whether we can find biochemical and/or ultrastructural markers that could be used both to identify and to quantify ozone effects on trees growing in situ in forests. This is not a new area of research, but its methodology is still little developed. It is not entirely clear yet whether it is possible to find methods of diagnosing ozone damage in the field. It has to be admitted that the many attempts made around the world to establish such methods have so far proved unsuccessful. One exception is the use of ultrastructural approaches, which have proved capable of distinguishing specific ozone effects, but it is too early to say whether they could be used in general environmental monitoring. Further analysis of results from the Ozone–Spruce study, among others, should shed light on how useful such methods could be on a large scale.

5.10 Economic estimates of production losses

With discussions under way in Europe about ways of reducing emissions of ozone precursors, it has been asked what gains may be expected to accrue from lower air pollutant emissions. At present, we know that ozone can have a detrimental effect on biomass production in young trees, but our limited understanding of how it affects older trees makes any estimate of production losses in forestry uncertain. However, the problems are not insurmountable. It would be possible even now to develop a model simulating the effects of ozone at a regional level. This would enable scenarios to be drawn up indicating the different costs associated with better or poorer air quality in terms of ozone. A proposal for a step-by-step procedure for calculating such costs is presented in figure 5.13.
Figure 5.13. Scaling up the effects of ozone on trees, from experiment to economic risk assessment. Vertical arrows represent scaling up between different levels of complexity. Horizontal arrows indicate the knowledge needed at each level of complexity. An attempt has been made to evaluate the uncertainties in the knowledge needed within the different levels or for each scaling step: +++ = present knowledge is sufficient; ++ = present knowledge is not sufficient, but enough to give a reasonably good estimate; + = present knowledge is poor; ? = state of knowledge is not clear. From Karlsson et al. 1999.
EFFECTS OF OZONE
ON WILD PLANTS

Helena Danielsson and Håkan Pleijel

SUMMARY
The current state of knowledge regarding the effects of ozone on wild plants is relatively poor, poorer than in the case of agricultural crops and certain trees. A small number of studies have been carried out in Sweden and Norway, judging from which fast-growing species generally appear to be more sensitive to ozone than those which are slow-growing. According to a recently published European survey, the most susceptible wild plants are adversely affected by ozone at roughly the same exposure levels as the most sensitive crops, but many species of wild plants seem to tolerate quite high ozone concentrations. Ozone exposure can alter the competitive balance between species with differing susceptibility to ozone. The most ozone-sensitive species in the Nordic countries include alpine cat’s-tail, timothy, and white and red clover.

6.1 Introduction
The current state of knowledge concerning ozone’s effects on wild herbs and grasses is appreciably poorer than in the case of farm crops and certain trees. The economic importance of agriculture and forestry has provided an impetus for relatively extensive research on species relevant to those sectors. The flowers of forests and meadows are of less direct economic significance and there have therefore been fewer studies of ozone damage to such plants. In recent years, following increasingly intense discussion about the conservation of biological diversity, interest in the effects of air pollutants on wild plants has grown. One difficulty is that there are many more species of wild plants than of agricultural crops or forest trees, and the studies carried out so far must therefore be regarded as random tests of the ozone sensitivity of our flora.

Most of the investigations of ozone’s effects on wild plants undertaken in Europe have related to vascular plants. A survey of the findings that have so far emerged from these studies concludes that the most sensitive wild species are adversely affected by ozone at roughly the same levels as the most sensitive crops (Ashmore & Davison 1996). At the same time, there appear to be many wild
species which are affected little or not at all by realistic concentrations of ozone. This picture is broadly applicable to the Nordic region, too, where studies have primarily been carried out in Sweden and Norway. Bryophytes (mosses etc.) and lichens are less well studied with respect to ozone sensitivity than vascular plants. It is difficult, therefore, to draw any general conclusions at present as to the extent to which these groups of organisms are harmed by current levels of ozone, although there are indications that they too are affected.

An interesting question from a biological point of view is whether any genetically determined variation in ozone sensitivity occurs within individual species, and whether elevated ozone concentrations therefore constitute a selection pressure favouring the forms with the greatest resistance to ozone. Such variability could, for example, result from certain genotypes taking up less ozone or having stronger antioxidant defences. Antioxidants are substances which are present in living organisms and which react with strong oxidizing agents, such as ozone, reducing them and rendering them harmless. There is much to suggest that selection for less ozone-sensitive genotypes is occurring, at least in certain annual species, whose short life cycle makes it possible for natural selection to rapidly favour the best adapted forms. An example of a species in which selection of this kind has taken place is greater plantain (Plantago major). British and Greek researchers compared the ozone susceptibility of populations of this species from Athens, where ozone concentrations are very high, with that of populations from rural areas of northern England, where they are lower. It was found that the Athenian populations were not particularly sensitive to ozone, whereas the English ones responded to ozone treatment with a marked reduction in growth (Reiling & Davison 1992a). The study was developed to include greater plantain populations from different parts of Britain, and the least ozone-sensitive ones were found in the areas which had experienced the highest ozone levels (Reiling & Davison 1992b).

Ozone concentrations have been appreciably elevated for just a few decades. In species with a much longer life cycle than greater plantain, genetic adaptation is a slower process. What is more, it is not certain that every species has the potential to adapt to an ozone-polluted environment within the foreseeable future, and where such adaptation does occur it may be at the cost of other characteristics being lost.

Since ozone sensitivity appears to vary widely between species, there is reason to suspect that the competitive balance between species in the natural environment may shift as a result of ozone pollution. There are clear indications in the international literature that this is indeed the case. In an experiment conducted outside London, a plant community consisting of grasses and herbs was exposed to filtered and unfiltered air in field chambers. Filtration of the air
resulted in increased growth of the grasses, with the consequence that the herbs became less abundant (Evans & Ashmore 1992).

6.2 Experiments in the Nordic countries

In Sweden, four studies of ozone sensitivity in wild plants were carried out over the period 1994–96 at Östads säteri, north-east of Göteborg. In 1994, an experiment was performed on 27 species of herbs and grasses, which were exposed in open-top chambers to three different concentrations of ozone (CF = filtered air, NF = ambient air, NF+ = ambient air with added ozone) (Pleijel & Danielsson 1997; figure 6.1).

This was followed in 1995 by an experiment to test the effects of the same ozone treatments on competition, on the one hand between red and white clover (Trifolium pratense and T. repens), on the other between three grass species: annual meadow-grass (Poa annua), sheep’s fescue (Festuca ovina) and cocksfoot (Dactylis glomerata). The plants were grown in large pots, and both combinations of species were subjected to cutting (simulated grazing or haymaking) at two different frequencies during the growing season. In addition, for the grasses, two different levels of nitrogen availability were used, in combination with the ozone treatment. Overall, the effects of ozone exposure were considerably smaller than the effects of differing nitrogen supplies and cutting intensities. In 1996 a test was made of the ozone sensitivity of various genotypes of timothy (Phleum pratense) and alpine cat’s-tail (P. alpinum) deriving from different parts of the Nordic region, from Denmark in the south to the Norwegian Arctic Ocean coast.
in the north. The seeds were obtained from the Nordic Gene Bank. The same year, the ozone susceptibility of a few lichen species was also investigated.

In Norway, a series of ozone sensitivity studies of a large number of wild plant species have been carried out (Mortensen 1992; Mortensen & Nilsen 1992; Mortensen 1993; Mortensen 1994). Since the Norwegian flora is broadly similar to that of Sweden and the two countries have similar climates, the results of these studies are also taken into account here, broadening the basis for an assessment of ozone’s effects on wild plants in north-western Europe.

6.3 Effects on grasses and herbs

6.3.1 Visible injury and effects on growth

Many earlier studies of the effects of ozone on wild plants focused on the occurrence of visible injury symptoms on leaves (Threshow & Anderson 1989). Clearly, such injuries are an indication that a plant is subject to a detrimental influence. They also mean that certain plants, which exhibit characteristic symptoms when exposed to only moderately elevated ozone concentrations, can be used as indicators of harmful levels of ozone. Potential indicator plants include several clover species. Figure 6.2 shows a few examples of foliar injuries resulting from ozone. However, the correlation between leaf symptoms and other, perhaps more important effects, such as reduced production or effects on flowering and seed set, does not appear to be very strong. The occurrence of visible injury is sometimes, but need not be, linked to depressed growth (Reiling & Davison 1992a; Mortensen 1993). What is more, an adverse effect on growth may occur without any characteristic visible injury.

6.3.2 Ozone sensitivity in relation to systematics and growth strategy

Early reports on ozone’s effects on wild plants often sorted species according to the groupings of botanical systematics, and species belonging to a given plant family, such as legumes (the pea family) or grasses, were considered to be more sensitive than those belonging to another. This dimension may need to be taken into account, but many plant families, especially the large ones, include species with widely differing ecological adaptations, growth patterns and life strategies. In many cases, annual weeds, woody plants and long-lived herbs all form part of the same family. There is good reason to assume that species which adapt ecologically in roughly the same ways also resemble one another in terms of their sensitivity to ozone, even if they belong to different families. We have not been able to find a clear link between plant family and susceptibility to ozone, and have instead tried to relate the ozone sensitivity of the wild herbs and grasses
studied in the Swedish and Norwegian experiments to three different indices reflecting the plants’ responses to nitrogen availability and their inherent growth potential. The hypothesis is that species with a high maximum growth rate and which rapidly translate available nitrogen into growth are more susceptible to ozone than slow-growing, stress-tolerant species. Such an idea finds support in the scientific literature (Reiling & Davison 1992a; Selldén & Pleijel 1995). A fast-growing species has a higher gaseous exchange with its surroundings, to provide carbon dioxide for photosynthesis. A greater exchange of gases through the stomata is accompanied by higher uptake of ozone. What is more, the normally rapid growth of species of this type means that any detrimental effect on photosynthesis will quickly be manifested in reduced growth. Slow-growing species rarely or never utilize their maximum potential for photosynthesis, and consequently a relatively small reduction in their photosynthetic capacity does not feed through into as large an effect on growth. Such species husband their resources and have a strategy which gives survival priority over growth.

Figure 6.2. Visible ozone injuries to (a) subterranean clover (Trifolium subterraneum), (b) white clover and (c) red clover. Photographs: Håkan Pleijel/IVL.
The first index used in figure 6.3 is Ellenberg’s ‘nitrogen number’ (Ellenberg 1979). The second is a similar nitrogen index developed in Sweden for assessments of the nutrient status of meadows and pastures (Ekstam & Forshed 1992). Both these indices reflect the extent to which a species is favoured by a good supply of nitrogen. Ellenberg’s index is on a scale from 1 to 9, Ekstam’s on a scale from 1 to 3. At Sheffield University in England, a system has been developed to classify plants according to their growth strategies (Grime 1979; Grime et al. 1990). Plants are divided into ruderals, competitors and stress-tolerators. Both ruderals and competitors are fast-growing, but the first group are short-lived, while the second are long-lived. Stress-tolerators are to be found in low-productivity environments and have a low maximum potential growth rate, but can withstand conditions unfavourable to growth better than plants which employ one of the other two strategies. Grime defines all manner of variants intermediate to the three main strategies. We have created a scale from 1 to 4, with the most stress-tolerant species at one end and the ruderals and competitors at the other. Only species included in all of these index/strategy systems were taken into account in this analysis, and they were divided into (1) those
found to be adversely affected by ozone even in a comparison between filtered air (CF) and ambient air with current levels of pollution (NF), (2) those only harmed by ozone at above-ambient concentrations (NF+) and (3) those not damaged at all by ozone in the studies. Both reduced growth and visible injury were used as effect parameters.

As figures 6.3a and 6.3b show, a majority of the plants were not affected at all, by either ambient or above-ambient concentrations of ozone. Elevated concentrations were needed to produce visible injury symptoms in seven species and reduced biomass growth in eight, while ambient levels of ozone were sufficient to induce visible injury in seven species and reduced growth in four. The plants affected even by ambient concentrations included a higher proportion of species with high Ellenberg, Ekstam and Grime indices than the other groups. In the case of biomass growth, this difference was statistically significant with respect to the Ellenberg index. The elevated ozone concentrations and increased nitrogen deposition rates now prevailing affect the high-index species in different directions. If the negative effect of ozone were reduced, these species would possibly become more competitive, at the expense of less fast-growing species. The species which are currently declining in the natural environment as a result of various environmental changes, such as reduced management of hay meadows and pastures, are often of the latter, slow-growing and stress-tolerant type. Many red-listed and other rare species exhibit relatively slow growth and have a low Ellenberg index (Ellenberg 1988). It should be pointed out that our understanding of how ozone and other gaseous pollutants affect the composition of the flora is still relatively limited, and that more information is needed to be able to draw definite conclusions. In addition, it needs to be emphasized that changing land-use patterns are no doubt much more far-reaching in their effects on the floristic composition of most ecosystems than it would appear from existing knowledge that ozone is.

When sensitivity to ozone, as reflected in biomass losses, was compared between different genotypes of timothy from throughout the Nordic region in the 1996 experiment, a relationship was found between growth rate and ozone sensitivity (Danielsson et al. 1999). The genotypes with the slowest growth at low or moderate ozone concentrations were less sensitive to ozone than those that grew more rapidly (figure 6.4). By contrast, no link could be identified between ozone susceptibility and geographical origin. One might have expected that genotypes from southern areas, where ozone levels are highest, would have been subject to selection that would have resulted in genes for ozone resistance spreading within these populations. However, the most sensitive genotypes did not originate in any particular part of the Nordic region. Timothy appears to be a species with a widely varying rate of growth. Ozone sensitivity seems to be more
strongly correlated to growth rate than to concentrations of ozone at the sites from which the timothy genotypes were obtained.

The numbering of the points (1–9) refers to the different sites, from south to north through the Nordic region, from which the seeds were collected.

6.3.3 THE IMPORTANCE OF COMPETITION

As mentioned at the beginning of this chapter, the competitive balance between different species may shift if they are exposed to ozone and they have differing sensitivities to this pollutant. Such an effect has in fact been observed in experiments on forage crops, in which ozone-sensitive species were displaced by less susceptible ones at higher ozone levels (chapter 4; Fuhrer et al. 1994). In one of the experiments carried out in 1995, white and red clover growing together in pots were exposed to different ozone concentrations. The plants were cut three times during the growing season. The results of this experiment are presented in figure 6.5. At a higher ozone exposure, the red clover more effectively outcompeted the white. This reflects the difference in ozone susceptibility existing between these species, and found by us in earlier experiments in which they were grown separately (Pihl Karlsson et al. 1995b). Several other examples of this type of effect can be found in the scientific literature. Ashmore and co-workers (1995), in a study of calcareous grassland communities in England, found that realistic ozone concentrations gave rise to an altered species composition. Slow-growing species were favoured by the ozone treatment, at the ex-
pense of fast-growing, nitrophilous species. This lends further support to the hypothesis that species with a high maximum growth rate suffer more marked negative effects as a result of ozone than those that are slow-growing.

6.4 Effects on lichens and bryophytes

There are indications that both lichens and bryophytes (mosses, liverworts etc.) can be adversely affected by realistic concentrations of ozone. Eversman and Sigal (1987), for example, found detrimental effects on photosynthesis and ultrastructural changes in the lichens *Flavoparmelia caperata* and *Umbilicaria mammulata* (a rock tripe) after five days’ exposure to ozone levels known to occur in the industrialized world during the warmer months of the year. In other studies, by contrast, not even very high concentrations of ozone produced measurable effects in lichens. As for bryophytes, even less is known than regarding lichens. However, there are indications that this plant group may also be sensitive to ozone. Potter and co-workers (1996) exposed hair moss (*Polytrichum commune*) and the bog moss *Sphagnum recurvum* to 70–80 ppb ozone for 6–9 weeks. The growth of the mosses was adversely affected by this treatment, more so in the case of the bog moss than in the hair moss.

In 1996 an experiment was carried out in Sweden in which lichens were grown in cuvettes placed in open-top chambers and exposed to three different ozone levels. *Lobaria scrobiculata*, a species believed to have declined appreciably in southern Sweden in recent decades, grew rapidly and exhibited positive net photosynthesis in the experimental system. Judging from the results of this experiment, ozone is not a major cause of this species’ decline, since growth and photosynthesis were somewhat higher in the ozone-treated specimens (figure 6.6).
Filtering the air appears to have had a somewhat unfavourable impact on the species. Since lichens obtain most of their nutrients by absorbing substances which end up on their surfaces, the lower rate of growth in filtered air compared with unfiltered air may have been due to the slightly lower levels of nitrogen compounds in the filtered air. *Sticta sylvatica*, a lichen whose decline in Sweden is even more dramatic than that of *Lobaria scrobiculata*, showed a tendency to be negatively affected by ozone. It therefore cannot be ruled out that regional levels of ozone have contributed to the retreat of this species, but since it exhibited very little net growth in the experiment such a conclusion is uncertain.

![Figure 6.6](image_url)

**Figure 6.6.** Net photosynthesis and net growth in the lichen *Lobaria scrobiculata*, grown at different ozone concentrations: CF = filtered air, NF = ambient air, CF+ = filtered air with added ozone, NF+ = ambient air with added ozone. The treatments CF+ and NF+ involved roughly the same ozone exposure. ns = no significant differences; different lower case letters above two columns indicate a significant difference.
6.5 The most sensitive species

As will have emerged from this chapter, we still know relatively little about how ozone affects our flora. Certain species can nevertheless be identified as definitely sensitive to ozone. They include timothy, alpine cat’s-tail and white clover. Red clover is also highly susceptible to ozone, although, as has already been pointed out, less so than white clover. The effects of ozone on the growth of timothy and alpine cat’s-tail in the Swedish experiment in 1996 are shown in figure 6.7. Moderately elevated ozone concentrations appreciably affected the growth of both species, which also exhibited substantial visible injury.

![Figure 6.7. Biomass production in timothy and alpine cat’s-tail grown in open-top chambers at different ozone concentrations: CF = filtered air, NF = ambient air, CF+ = filtered air with added ozone, NF+ = ambient air with added ozone.](image)

The treatments CF+ and NF+ involved roughly the same ozone exposure. AA refers to a control with no chamber. Different letters above columns indicate statistically significant differences (p<0.05).

6.6 Important gaps in current knowledge

There are major gaps in our knowledge concerning the effects of ozone on wild plants. Relatively few species have been tested. Furthermore, many studies have been conducted using individual plants grown in pots under virtually optimum conditions. More needs to be known about how ozone affects plants in the natural environment, where competition and other interactions, both with herbivores and with factors such as nitrogen availability, have to be taken into account. There is a growing interest in Europe in how the wild flora is influenced by ozone. At the last meeting on critical ozone levels, in Kuopio in 1996, it was agreed that the most significant gaps in our understanding of ozone’s effects on wild plants related to the structure and dynamics of populations, variations in sensitivity between species and between genotypes, the signifi-
cance of nitrogen status and nitrogen deposition, developmental stage, and the impact of various climatic factors and parasites on ozone sensitivity.

Figure 6.8. Clones of timothy and alpine cat’s-tail exposed to (a) unfiltered air and (b) unfiltered air with added ozone in open-top chambers at Östad in 1996. Photograph: Håkan Pleijel/IVL.
7.1 Effects of ozone on plants – current state of knowledge and research needs

A large number of studies show that current concentrations of ground-level ozone are causing damage to plants across much of Europe. Best understood are the effects of ozone on agricultural crops. This is chiefly because such plants are easy to cultivate, normally grow in what are more or less monocultures, with no interspecific competition, and are usually short-lived. In many experiments around Europe, including Sweden, filtration of ozone from the air has been found to significantly increase growth in wheat, an effect that can be attributed to a longer period of growth for the plants exposed to filtered air. It may be concluded that current ozone levels entail an appreciable cost to the agricultural sector as a result of reduced productivity. An earlier, relatively rough estimate of ozone’s impact on Swedish agriculture pointed to losses of the order of SEK 1 billion a year. With the knowledge now available, it should be possible to determine the cost of the yield reductions caused by ozone in the crops studied most closely.

Short periods of greatly elevated ozone concentrations, ‘ozone episodes’, are known to produce visible injury symptoms in a number of different plants. Plants that have been used particularly frequently to study such injuries include clovers, spinach and certain varieties of tobacco, and some of them are therefore used as bioindicators of ground-level ozone. Visible ozone damage has also been observed in a range of other plants in Europe and North America, many of them wild species which only occur in natural or semi-natural ecosystems.

In general, we know very little about the effects of ozone on wild herbs and grasses. Only a small number of species have been studied closely. The most sensitive wild plants are probably as susceptible to ozone as certain crops, such as wheat. Fast-growing species appear to be most sensitive, but too few species have been studied to allow any definite, general conclusions to be drawn. Important research topics in this area are the significance of competition between species with differing ozone sensitivity, and whether there is a trend towards higher genetic tolerance of ozone in wild plants. If the latter is the case, an important follow-up question is whether the price paid for this greater tolerance is the loss of other characteristics.
Concerning the effects of ground-level ozone on forest trees, two factors complicate efforts to establish definite relationships between ozone exposure and effects. First, the longevity of trees poses a problem. Most studies have been carried out on young specimens (< 10 years), and there is much to suggest that they are not generally representative of older trees. Some studies suggest that mature trees may be more sensitive to ozone than younger ones, but this conclusion is based on assumptions for which better empirical support is needed. It is therefore important to test the ozone sensitivity of mature trees by a variety of methods and to compare the results with data on their younger relatives.

Secondly, assessments of ozone's effects on forest trees are made more difficult by the fairly complex dynamics of a forest stand over the course of a rotation. Competition within and between species means that it is not possible to establish simple relationships between experiments on small trees in a controlled environment, on the one hand, and forest stands, on the other. As has been made clear in this report, partly differing conclusions will be reached, depending on the assumptions made. However, even cautious assumptions about the extent to which ground-level ozone interferes with the growth processes of forest stands suggest that present-day ozone levels will have an adverse effect of up to about 10% on production in southern Swedish forests over an entire forest rotation. Adapting existing relationships between ozone exposure and tree growth response to forest stands is one of the most important challenges for research in this area over the next few years, and in this context modelling will be an important element. Furthermore, in the framework of international cooperation on long-range transport of air pollutants in Europe, there is a need for knowledge concerning the economic consequences of current and future ozone levels for production in the forest sector.

An important question in the European perspective is how different types of forest damage – chiefly in the shape of crown thinning, but also to some extent actual dieback of trees – are to be explained. This is particularly true as regards oak forests, which are currently regarded as showing the most rapid increase in forest damage in Europe. At present, it is difficult to assess what part ozone is playing in forest decline. In the case of oak especially, it would make sense to investigate whether ozone could be a factor underlying the damage symptoms observed. Although various types of disease and parasitic attack are often the visible causes of trees suffering damage or dying, ozone may act as an additional stress, weakening trees and finally leaving them susceptible to parasites or diseases. For forest owners in southern Sweden, this is a question of major concern.

Another important field of research is the development of relationships between plant uptake and effects of ozone. The indices of ozone exposure cur-
rently most widely used, including AOT40, are based on ozone concentrations in air and therefore take no account of the factors affecting how much ozone is taken up by plants, i.e. the dose they actually receive. Research in recent years has shown that uptake is the key factor. If allowance is made for this, it has to be concluded that the AOT40 concept overestimates the effects of ozone in southern and central Europe and underestimates its effects in the Nordic region. This is a question of vital importance to Sweden. Our climate is comparatively favourable to plant uptake of ozone, which increases the risk of adverse effects from a given level of ozone. Long summer days and relatively high humidity are among the climatic factors promoting high uptake in Sweden. In southern and to some extent in central Europe, too, the climate is such that ozone is taken up more slowly than in the Nordic countries. Ozone uptake by plants in more southerly parts of Europe therefore need not be as markedly higher than further north as current concentrations of the pollutant might suggest. Perhaps in some cases ozone uptake is in fact higher in our region.

In addition, the factors affecting how plants respond to the ozone dose which they take up need to be studied under realistic conditions. The importance of phenology (the developmental stage of plants) and the strength of plants’ antioxidant defences against the harmful effects of ozone are key issues to be addressed by research over the coming decade.

7.2 Problems and prospects

Ground-level ozone is regarded as an issue of major relevance to international air quality cooperation in Europe. In several countries, especially in southern Europe, the ozone problem ranks higher than acidification and eutrophication. In this context, health concerns probably carry somewhat greater weight than ozone’s effects on plants, but both types of damage are now important considerations in the ECE’s and the EU’s activities to tackle transboundary air pollution.

As far as health effects are concerned, maximum ozone concentrations and hence ozone episodes are of decisive importance, and to a relatively large extent these parameters can be influenced by action taken in Europe. Control strategies to tackle maximum ozone levels usually involve larger emission reductions for volatile organic compounds than for nitrogen oxides. Such measures are regarded as relatively effective in cutting the highest ozone peaks in densely populated parts of southern and central Europe. On the outer fringe of Europe, including in Sweden, on the other hand, nitrogen oxide emissions are the most important factor in ozone formation.
In relation to effects on plants, attention is currently focused on ozone levels in excess of 40 ppb, whereas with regard to health levels exceeding 60 ppb are the main concern. Ozone episodes are of much greater significance when it comes to exceedance of 60 ppb, compared with 40 ppb. Many researchers studying the links between ozone and health are suggesting that the criterion of 60 ppb should be lowered, but this is the level currently guiding practical efforts to reduce air pollution. The regional background concentration of ozone, which averages just over 30 ppb for Europe as a whole, has a greater impact as regards exceedance of 40 ppb than in relation to exceedance of 60 ppb. Background levels are not only affected by emissions in Europe itself. They are the result of aggregate emissions throughout the northern hemisphere, where emissions of ozone precursors are rising in some areas, even though they have begun to fall in Europe.

Ozone’s effects on materials are even more strongly linked to elevated background concentrations than its effects on plants. In principle, effects on materials are proportional to the concentration of ozone; there is no level below which they do not occur. The quite significant contribution which ozone makes to the greenhouse effect (5–10%) also has a direct link with the current elevated background levels of this gas. To address both these categories of effects, there is a need for international air quality cooperation extending beyond Europe. It should be emphasized, however, that Europe’s own emissions of ozone precursors are responsible for the great majority of the ozone damage occurring on this continent.

Many studies point to a sharp rise in background ozone concentrations in the northern hemisphere between the 1950s and the 1980s. Certain results, moreover, indicate that these background levels are still rising. However, the picture is somewhat inconsistent, and it is often difficult to distinguish regional and local contributions to ozone levels from truly large-scale contributions. It is important to note that a rising background level could counteract the effects of measures introduced in Europe under protocols to the ECE Convention on Long-Range Transboundary Air Pollution and the EU’s Ozone Strategy. How great the improvement will be will depend on which category of effects is considered. The higher the effects threshold in terms of ozone concentrations which we take as our starting point, the greater the chance of achieving an improvement by the measures implemented in Europe. These measures may therefore be expected to have the greatest beneficial impact in terms of health, somewhat less of an impact in relation to plants, and somewhat less again with regard to damage to materials and ozone’s contribution to the greenhouse effect.
We would like to take this opportunity to thank all the individuals, companies and organizations that have supported our research into the effects of ground-level ozone in Sweden.

As noted in the preface, the Swedish Environmental Research Institute (IVL) and the Botanical Institute at Göteborg University have conducted a series of experiments since 1984 relating to the effects of ground-level ozone on plants. Over the period 1985–89, an open-top field chamber experiment was carried out at Rörvik, some 40 km south of Göteborg, in which Norway spruces were grown at different ozone concentrations. This project received financial support from the Swedish Environmental Protection Agency, the Nils and Dorthi Troëdsson Research Foundation, the Swedish Council for Forestry and Agricultural Research (SJFR), the Foundation for the Swedish Environmental Research Institute, and industry (the Association of Power and District Heating Producers (KVM), the OK Environmental Foundation, Saab, SSVL and Volvo).

In 1987, experiments were started at Östads säteri, roughly 40 km north-east of Göteborg, to study the effects of ozone on field-grown agricultural crops. This work was funded by the Environmental Protection Agency, the Swedish Farmers’ Foundation for Agricultural Research and SJFR from 1987 to 1990, and by the latter two bodies only between 1991 and 1993. In 1990 the experiments on forest trees were moved to Östad. A larger-scale open-top chamber experiment was set up to study the effects of ozone, drought and phosphorus deficiency on Norway spruce. This project continued up to and including 1996 and was supported financially by the Environmental Protection Agency, Swedish industry (the Association of Swedish Automobile Manufacturers and Wholesalers, Boliden Mineral, Borealis, Elforsk, Hydro Polymers AB, the Swedish Steel Producers’ Association, the Association of Swedish Chemical Industries, MoDo, Norrköpings Energi, Ovako Steel, Saab, SSVL, the Ångpanneföreningen Foundation for Research and Development, Stora Skog, Svensk Bilprovning, Södra Skogsägarna, Vattenfall and Volvo), the Nils and Dorthi Troëdsson Research Foundation, SJFR, the Foundation for the Swedish Environmental Research Institute, the National Road Administration and the National Board of Forestry. 1994 saw the launch of the EC project ESPACE-Wheat (funded in Sweden through the Environmental Protection Agency and SJFR). Between 1994 and 1996 the Environmental Protection Agency also supported research into the effects of ozone on wild plants. In addition to this work, a series of experiments on pot-grown indicator plants for ozone have been undertaken at Östad
since as early as 1987, in the framework of the Convention on Long-Range Transboundary Air Pollution (CLRTAP). These studies were made possible chiefly by grants from the Environmental Protection Agency, but also received some support from the Swedish Farmers’ Foundation for Agricultural Research (1991).

An experiment to study the ozone sensitivity of birch was established in 1997 with funding from the Environmental Protection Agency, the Swedish Foundation for Strategic Environmental Research (MISTRA), the Nils and Dorthi Troëdsson Research Foundation, the National Road Administration, the National Board of Forestry, the Östad Foundation and industry (the Association of Swedish Automobile Manufacturers and Wholesalers, Borealis, Elforsk, the Ångpanneföreningen Foundation for Research and Development, SSVL, Södra Skogsägarna and Volvo). The main source of funding for the experiments carried out in Sweden has been the Environmental Protection Agency, in recent years within the project area Acidifying Substances and Tropospheric Ozone. This project area is also responsible for the publication of the present report.

Finally, special thanks are due to the Nils and Dorthi Troëdsson Research Foundation for its generous grants towards the development of measuring instruments and for its funding of Gun Selldén’s research professorship.
REFERENCES


in relation to ambient levels of ozone in Finland. *Environmental Pollution* 96, 117–127.


Experimental design for the Ozone–Spruce Project at Östad, 1992–96

Experimental plots and chambers

A total of 56 plots were established, 42 of them with open-top chambers. Figure A1 shows a plan of the site.

Figure A1. Plan of the experimental site at the Östad field station. The plots enclosed in a box in the top portion of the plan (1–12, 17) were used for the drought stress experiment (Ozone-d). The other plots were used for the main experiment (Ozone-m). The treatments are explained in table A1. S = reserve plot.
Figure A2 shows the layout of a single plot in the main experiment (A) and the drought stress experiment (B), and a cross-section of a main experiment plot (C).

Figure A2. Plot layout in the main experiment (A) and the drought stress experiment (B), and cross-section of a main experiment plot (C). There were 18 pots per plot in the main experiment, 24 in the drought stress experiment. One Norway spruce was grown in each pot, and at each harvest one tree per plot and treatment was harvested. The harvests were divided into 3 blocks, shown by different shading in A and B. (a) subsoil water tube, Ø 32 mm, (b) polyethylene pot, 120 l, (c) pipe, Ø 16 mm, (d) sampling tube, Ø 45 mm, (e) sand, 0–6 mm, (f) fibre mat, (g) PVC sheet, (h) crushed stone, 10–18/6–12 mm, (i) filter for drainage water, (j) crushed stone, 10–18 mm, (k) main drainage pipe, (l) container to collect drainage water, 30 l, (m) surrounding soil.
PLANT MATERIAL
The plant material consisted of a clone (C77-0068 Minsk) of Norway spruce, *Picea abies* (L.) Karst., from Hilleshög AB. This clone is of Belarussian origin, and has been widely used in nursery and field experiments. It was one of the fastest-growing within its origin, and has been sold commercially by Hilleshög AB. Two-year-old spruces were planted in sand, one per pot, in June 1991 (figure A2).

Table A1. Overview of treatments in the Ozone–Spruce experiment. Treatments 1–7 constituted the main experiment, 1–3 being used to estimate the effects of the chambers and the pots, and 4–7 to estimate the effects of ozone with a low or optimum supply of phosphorus. Treatments 8–11 constituted the drought stress experiment and were used to estimate the effects of ozone with or without periods of drought.

AA = ambient air; UP = unpotted; P = potted; NF = non-filtered air; NF+ = non-filtered air with added ozone; CF = charcoal-filtered air; PD = phosphorus deficiency; D = drought; W = well watered; N = normal; L = low; H = high.

<table>
<thead>
<tr>
<th>TREATMENT</th>
<th>OZONE</th>
<th>WATER</th>
<th>PHOSPHORUS</th>
<th>CHAMBER</th>
<th>POTS</th>
</tr>
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<tbody>
<tr>
<td>1</td>
<td>AA/UP</td>
<td>N</td>
<td>N</td>
<td>N</td>
<td>-</td>
</tr>
<tr>
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<td>AA/P</td>
<td>N</td>
<td>N</td>
<td>N</td>
<td>+</td>
</tr>
<tr>
<td>3</td>
<td>NF</td>
<td>N</td>
<td>N</td>
<td>N</td>
<td>+</td>
</tr>
<tr>
<td>4</td>
<td>CF</td>
<td>L</td>
<td>N</td>
<td>N</td>
<td>+</td>
</tr>
<tr>
<td>5</td>
<td>NF+</td>
<td>H</td>
<td>N</td>
<td>N</td>
<td>+</td>
</tr>
<tr>
<td>6</td>
<td>CF/PD</td>
<td>L</td>
<td>N</td>
<td>L</td>
<td>+</td>
</tr>
<tr>
<td>7</td>
<td>NF+/PD</td>
<td>H</td>
<td>N</td>
<td>L</td>
<td>+</td>
</tr>
</tbody>
</table>

| 8         | CF/W  | L     | N          | N       | +    |
| 9         | NF+/W | H     | N          | N       | +    |
| 10        | CF/D  | L     | L          | N       | +    |
| 11        | NF+/D | H     | L          | N       | +    |

EXPERIMENTAL DESIGN
The study was designed as two separate experiments, which were subjected to separate statistical analyses (table A1): (1) the main experiment, with a focus on ozone and phosphorus (Ozone-m), and (2) the drought stress experiment, with a focus on ozone and water (Ozone-d). The biomass growth of the spruces (needles, branches, stems and roots) was measured regularly throughout the experiment. The main experiment ran from May 1992 to May 1996 and comprised 18 harvests, divided into 3 blocks (figure A2 and table A2).

Each block consisted of six harvests over a specific period, with block 1 comprising the first six harvests (1992–93), block 2 the next six (1994) and block 3 the last six (1995–96). The drought stress experiment ran from May 1992 to September 1995 and involved 6 harvests, divided into 3 blocks, i.e. trees were
harvested twice a year, in April and September 1993–95. Within each block, the experiments were designed according to a Latin square, enabling the variables time (harvest date), treatment, direction (compass direction; shading effects) and plot (e.g. differences in drainage and exposure to pest attack) to be analysed statistically, while minimizing the number of spruces harvested. There were 6 replications of each treatment.

Table A2. Schedule of harvests from all the plots in the main experiment. Each block consisted of six harvests. ◆ block 1; ■ block 2; ● block 3.

<table>
<thead>
<tr>
<th></th>
<th>Jan</th>
<th>Feb</th>
<th>Mar</th>
<th>Apr</th>
<th>May</th>
<th>June</th>
<th>July</th>
<th>Aug</th>
<th>Sep</th>
<th>Oct</th>
<th>Nov</th>
<th>Dec</th>
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<td>1992</td>
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<td>◆</td>
<td></td>
<td>◆</td>
<td></td>
<td>◆</td>
<td></td>
<td>◆</td>
<td></td>
<td>◆</td>
</tr>
<tr>
<td>1993</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
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<td></td>
<td></td>
<td></td>
<td>◆</td>
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<tr>
<td>1994</td>
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<td></td>
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<td></td>
<td></td>
<td></td>
<td>■</td>
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<tr>
<td>1995</td>
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<td></td>
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<td></td>
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</tbody>
</table>

Table A3 shows key events during the Ozone–Spruce study, from planting to the end of the experiment, including dates of ozone exposure, watering, drought periods and nutrient inputs over the six years of the experiment.

Table A3. Dates (day/month) of important events during the Ozone–Spruce study.

<table>
<thead>
<tr>
<th>Spruces planted</th>
<th>Chambers installed, filtration started</th>
<th>Ozone exposure started</th>
<th>Ozone exposure stopped</th>
<th>Chambers dismantled, filtration stopped</th>
<th>Automatic watering started</th>
<th>Watering stopped</th>
<th>Nutrient supply started</th>
<th>Nutrient supply stopped</th>
<th>Phosphorus deficiency started</th>
<th>Drought period started</th>
<th>Drought period stopped</th>
<th>Experiment ended</th>
</tr>
</thead>
<tbody>
<tr>
<td>5/6</td>
<td>25/6 15/4 29/4 12/4 3/4</td>
<td>6/7 16/4 21/4 13/4</td>
<td>23/10 30/10 2/11 14/11</td>
<td>2/11 3/11 2/11 14/11 22/5</td>
<td>15/6 15/5 26/4 28/4 15/5 30/4</td>
<td>20/10 6/10 13/10 17/10 12/10 22/5</td>
<td>18/7 16/5 19/5 12/5 1/6</td>
<td>31/8 25/8 30/7 18/7 31/7</td>
<td>19/5</td>
<td>24/8 30/7 23/7 4/8</td>
<td>21/9 10/9 15/9 19/9</td>
<td>22/5</td>
</tr>
</tbody>
</table>

Water and nutrient supplies were controlled using a computerized irrigation system. The trees were watered daily throughout the season. Nutrients were supplied daily in May–July, with maximum doses in mid-June. Annual nutrient doses were calculated on the basis of tree weight and expected growth, with the
aim of achieving optimum growth and a needle nitrogen concentration of 18–20 mg g\(^{-1}\) (dry weight).

Half the spruces in each plot in the Ozone-d experiment were subjected to a drought period of 7–8 weeks during each of the seasons 1993, 1994 and 1995, while the other half were kept well watered at all times. During the drought periods, regular watering was suspended and the drought-stressed trees were given water on just a few occasions, depending on how rapidly they dried out. Drought stress was quantified by measuring the water potential early in the morning. Water potential was measured as the negative pressure arising in the trees’ conducting tissues when water was in short supply. The greater the negative pressure, the more marked the drought stress. The lowest water potential values reached were -1.9, -3.2 and -1.7 MPa during the drought periods in 1993, 1994 and 1995, respectively. Mean water potentials for the same periods were -1.1, -1.0 and -0.8 MPa. Water potentials did not differ between the two ozone treatments in the drought stress experiment.

The phosphorus concentrations recorded in current-year needles in the main experiment are presented in table A4. Critical levels of phosphorus in spruce needles are considered to be 1.1–1.3 mg P g\(^{-1}\) and 0.10–0.12 expressed as a P:N ratio. Typical values in southern Swedish forests are 0.9–1.5 mg P g\(^{-1}\) and a P:N ratio of 0.09–0.16. The lowest phosphorus level recorded in the main experiment was 1.3 mg P g\(^{-1}\), a moderate phosphorus deficiency compared with what can be found in the field.

Table A4. Phosphorus concentrations (P, mg g\(^{-1}\) dry weight) and phosphorus:nitrogen (P:N) ratios in current-year needles from the main experiment treatments involving normal and low phosphorus doses.

<table>
<thead>
<tr>
<th></th>
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<th></th>
<th></th>
<th></th>
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</thead>
<tbody>
<tr>
<td>Normal P</td>
<td>P conc.</td>
<td>2.2</td>
<td>2.0</td>
<td>1.9</td>
<td>1.9</td>
</tr>
<tr>
<td></td>
<td>P:N ratio</td>
<td>0.10</td>
<td>0.10</td>
<td>0.08</td>
<td>0.12</td>
</tr>
<tr>
<td>Low P</td>
<td>P conc.</td>
<td>2.2</td>
<td>1.9</td>
<td>1.3</td>
<td>1.5</td>
</tr>
<tr>
<td></td>
<td>P:N ratio</td>
<td>0.10</td>
<td>0.09</td>
<td>0.05</td>
<td>0.09</td>
</tr>
</tbody>
</table>

OZONE CONCENTRATIONS

Table A5 shows the ozone concentrations that prevailed in the different treatments, as 24-hour means over the season and as means during daylight hours. Ozone levels were lowest during the first two years and highest during the third year, 1994. The corresponding ozone doses, expressed as AOT40, are shown in table A6. The current critical level for forest trees (10 ppm-hours, see chapter 3) was exceeded every year in the NF+ treatments.
Table A5. Seasonal mean concentrations of ozone (24-hour and daylight hours, ppb) between 1 April and 30 September in chambers and ambient air. AA/5m = concentration in ambient air 5 m above the ground; other abbreviations, see table A1. *Note that data for 1996 only cover the period 1 April–22 May.

<table>
<thead>
<tr>
<th>TREATMENT</th>
<th>24-HOUR MEAN</th>
<th>MEAN, DAYLIGHT HOURS</th>
</tr>
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<tr>
<td>AA/5m</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>AA</td>
<td>23</td>
<td>24</td>
</tr>
<tr>
<td>NF</td>
<td>22</td>
<td>23</td>
</tr>
<tr>
<td>CF</td>
<td>4</td>
<td>6</td>
</tr>
<tr>
<td>NF+</td>
<td>29</td>
<td>34</td>
</tr>
<tr>
<td>CF/PD</td>
<td>5</td>
<td>7</td>
</tr>
<tr>
<td>NF+/PD</td>
<td>30</td>
<td>34</td>
</tr>
<tr>
<td>CF/W, D</td>
<td>5</td>
<td>6</td>
</tr>
<tr>
<td>NF+/W, D</td>
<td>30</td>
<td>34</td>
</tr>
</tbody>
</table>

Table A6. AOT40 expressed as ppm-hours (1 April–30 September), aggregated over 24-hour periods and daylight hours. Abbreviations: see table A1. *Note that data for 1996 only cover the period 1 April–22 May.

<table>
<thead>
<tr>
<th>TREATMENT</th>
<th>24 HOURS</th>
<th>DAYLIGHT HOURS</th>
</tr>
</thead>
<tbody>
<tr>
<td>AA</td>
<td>6.9</td>
<td>5.3</td>
</tr>
<tr>
<td>CF</td>
<td>5.8</td>
<td>0.3</td>
</tr>
<tr>
<td>NF</td>
<td>6.6</td>
<td>4.2</td>
</tr>
<tr>
<td>NF+</td>
<td>13.8</td>
<td>22.5</td>
</tr>
</tbody>
</table>
Ozone in the lower atmosphere causes damage to plants and affects human health. It also contributes to the greenhouse effect and damages materials. These problems are a major consideration in current European negotiations on transboundary air pollution.

Ground-level ozone is formed from nitrogen oxides and hydrocarbons under the influence of sunlight. Transport is the single most important source of these pollutants, although energy production and various types of industry also account for significant emissions. Concentrations of ground-level ozone have risen substantially in the course of the 20th century. In addition, ozone ‘episodes’ – short periods in which ozone levels are greatly increased – sometimes occur, chiefly in the spring and summer.

This report describes the mechanisms of ozone formation, current levels of ozone in Sweden, and national and international environmental objectives relating to ground-level ozone. The emphasis is on ozone’s effects on plants. On the basis of a wide range of experiments carried out in Sweden over the last 15 years, the report describes how ozone affects agricultural crops, forest trees and wild plants.