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## PHOTOCHEMICAL OXIDANT CONTROL STRATEGIES IN EUROPE: A 19 DAYS' CASE STUDY USING A LAGRANGIAN MODEL WITH CHEMISTRY

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<u>Abstract</u> The Norwegian Lagrangian trajectory model with atmospheric boundary layer chemistry has been applied to calculate the transport of oxidants to nine receptor points 150 km apart in southern Scandinavia during the period 26 August to 14 September 1980. Ozone and PAN measurements at several rural sites have been used for comparison, and the calculated ozone concentrations at the nine receptor points during the 19 days' period compare well with the measurements. The most important factors which control long-range transport of ozone and PAN seem to be well described. Hydrocarbon emission control in Europe is calculated to cause substantial decrease of the ozone concentrations in southern Scandinavia on days with high oxidant levels. Control of NO<sub>x</sub> only or NO<sub>x</sub> and hydrocarbons in combination are much less efficient as strategies to reduce oxidant levels.

#### 1 INTRODUCTION

It is established that oxidants alone or in combination with other pollutants, damage plants outside most metropolitan areas in the United States, Canada, Mexico and Japan (Skärby and Selldèn, 1984). There is a serious dieback of forests in many parts of Europe and North America. Exposure of forests to enhanced ozone levels is probably an important stress factor. There is a significant economical loss due to reduced yield in crops which have been exposed to oxidant pollution. In the US the loss is estimated at \$1-2 billion per year (Skärby and Selldèn, 1984). The loss in amenity due to haze and visibility reduction linked to oxidant formation is difficult to assess in economical terms.

The causes and implications of the formation of oxidants are truly of an international character. Several days may elapse between the emission of the precursors (hydrocarbons, HC, and nitrogen oxides, NO  $_{_{\rm X}}$  , the sum of NO and NO  $_{_{\rm 2}}$  ) and the formation of oxidants, notably ozone. During such a time period the air masses may have moved with the atmospheric flow over one thousand kilometers or more. Establishment of oxidant control strategies and the abatement of oxidants is therefore an international matter. Several international bodies are doing preliminary work to develop regional control strategies for photochemical oxidants and their precursors. The Environment Committee (Air Management Policy Group) within OECD is preparing a workplan for the development of regional control strategies for photochemical oxidants and their precursors in OECD member countries. The European Commission is financing a study of model approaches towards the development of oxidant control strategies. There are several bilateral agreements between countries to develop models to simulate oxidant transport and control strategies: Between Ontario Ministry of the Environment, Environment Canada and the Federal Republic of Germany (Acid deposition and oxidant model, ADOM, Misra, 1984), and between the Federal Republic of Germany and the

Netherlands (PHOXA, Interregional scale model developed to study the occurrence of photochemical oxidants in the Federal Republic of Germany and the Netherlands, and also to investigate acidification phenomena). In the latter case, a model described by Liu and Reynolds (1984) is adopted. The purpose is to apply the model to describe one oxidant episode.

The largest effort to establish a model to be used in the development of oxidant control strategies, is made in the United States by EPA. The US EPA Regional Oxidant Model (ROM) is designed to simulate hourly averaged concentrations over periods of several days on a three-dimensional grid that is  $10^3$  km in size, and with a horizontal resolution of about 18 km x 18 km. The model is intended to assist the individual states in formulating emission control plans that will bring air quality into compliance with Federal standards (120 ppbv as hourly ozone concentrations not to be exceeded more than once per year) (Lamb and Novak, 1984).

Some countries in Europe have recommended or proposed guidelines for ozone (e.g. Sweden: proposed guideline of 60 ppbv as hourly ozone concentration not to be exceeded more than once per month, Norway: 50-100 ppbv as hourly ozone concentration; Grennfelt and Schjoldager, 1984). It is essential to establish a common guideline in Europe as to the averaging time and level of ozone that is desirable to control. Ozone damage occurs both through long term (e.g. a growing season), exposure to slightly enhanced concentrations (e.g. 35-50 ppbv) and to short term (e.g. one hour) exposure to higher ozone concentrations (several hundred ppbv) (Skärby and Selldèn, 1984). Ozone guidelines in Europe should take into account both the long-term and short-term effects of exposure to enhanced ozone concentrations.

In the US it is required by law to control the maximum 1 h average ozone concentration. The EPA ROM model is planned to be applied only to one or a few severe episodes over several days of photochemical pollution over the north-eastern US. To control the long-term ozone concentration e.g. over a growing season, it would be required to run the oxidant model over at least one growing season, preferrably several, and covering a large enough area to make the results independent of the boundary conditions. US policymakers may also eventually shift to this point of view: "It is my understanding that models of long-range ozone transport are in a rather rudimentary stage of development... Of particular interest are models that can characterize growing-season concentrations over areas the size of the Eastern United States. Recent documentation of tree productivity declines throughout widespread areas of the Eastern US has accelerated the need for such tools. I am certain that ozone transport will be discussed much more widely during the next Congress than it was during the past few years' Clean Air Act reauthorization debates" (Friedman, 1984).

#### 2 CASE STUDY OF OXIDANT TRANSPORT

The Norwegian Lagrangian long-range transport model with atmospheric bondary layer chemistry was described by Eliassen et al., (1982a).

In the first application of the model, ozone formation and transport to southern Norway and south-eastern Sweden during the time period 6-14 April 1979 was studied (Eliassen et al., 1982a). Ozone measurements at Langesund, a rural, coastal site about 200 km south-west of Oslo, were used to compare with the calculated concentrations at four surrounding receptor points which were grid nodes in the  $150 \times 150 \text{ km}^2$ ,  $37 \times 39$  cell EMEP grid covering Europe and parts of the USSR eastwards to Ural. Daily sulphur dioxide and aerosol sulphate measurements taken at Rørvik at the coast just south of Gothenburg were also used for validation. The flow was primarily over eastern Europe towards southern Scandinavia.

In the second application of the model, which is reported here, the formation of oxidants during transport to southern Scandinavia during the time period 26 August to 14 September 1980 was studied (19 days). Ozone measurements were taken at Langesund, at Maridalen which is close to Oslo (Schjoldager et al., 1981) and at Rørvik (Grennfelt, Swedish Environmental

Research Institute (IVL), Gothenburg, private communication, Nielsen et al., 1981, Grennfelt et al., 1982). PAN was measured at Maridalen (Schjoldager et al., 1983) at Rørvik and occasionally at Risø (Nielsen et al., 1981, Grennfelt et al., 1982). Daily sulphur dioxide and aerosol sulphate measurements were taken at Birkenes, at Skreådalen and at Rørvik during the time period, and are used in the discussion of the model calculations (EMEP, 1981). The location of the monitoring sites is shown in Figure 1.

Nine grid points in southern Scandinavia in the 150 km grid were selected as receptor points. 850 mb, 96 h trajectories to these points were calculated four times per day (at 0000, 0600, 1200 and 1800 GMT). The flow direction was predominantly from the south-west and south during the time period (see Figure 2). Hourly ozone concentration recorded at Langesund, Maridalen, Jeløya and Rørvik is shown in Figure 3. At Rørvik, the highest concentration was found in the period 2 - 4September, at Langesund the concentration was moderate throughout the period although data are missing for the period 1-5 September, at Jeløya and Maridalen the concentration was quite high during the period 2-9 September 1980. PAN measured at Rørvik and occasionally at Risø during the time period 2-5 September is also shown in Figure 3, together with the recorded concentration in Maridalen 7-9 September 1980. The maximum hourly PAN concentration was 4-5 ppbv at all stations during the time periods shown. This is about one order of magnitude higher than what can be found in unpolluted air in the lower troposphere.

#### **3 MODEL DESCRIPTION**

#### 3.1 Meteorological model

The model has been described in some detail previously (Eliassen et al., 1982a, Eliassen et al., 1982b, Hov et al., 1984). The pollutants are assumed to be completely vertically mixed throughout the boundary layer which has a variable depth along the 96 h long 850 mb trajectories. No mass transport

takes place through the top of the well-mixed layer. Lateral diffusion is neglected since the emission data are given in a 150 km grid where finer details than 150 km in the concentration fields are smoothed out.

In episode studies with short sampling times (like one hour), the rate of horizontal spread of instantaneous releases of pollutants may be an important parameter which should be considered (Eliassen, 1984). For a sampling time of many hours, like 6 h or more, the instantaneous diffusion of pollutant releases is dominated by the diffusion due to sampling time ("synoptic swinging of the trajectories", Smith, 1979). A sampling time of 24 h is used for sulphur species in EMEP, in which case the synoptic swinging of trajectories is the dominating factor for plume spread (Eliassen, 1984).

During transport, pollutants are emitted into the air parcel according to the emission maps for  $NO_x$ , HC and  $SO_2$ . Instantaneous concentrations are predicted upon arrival of a trajectory. The horizontal resolution of the concentration fields is determined by the choice of emission grid and density of trajectory arrival points. The combined effects of vertical wind shear and diffusion due to heat exchange is difficult to handle in Lagrangian models. Trajectory models are simple numerically, however, since the integration is reduced to an ordinary time-integration along certain selected trajectories.

Trajectory positions are calculated every 2 h, as described in Petterssen (1956), based on wind observations at the 850 mb level at 0000, 0600, 1200 and 1800 GMT. The observed wind data are analyzed objectively in the EMEP grid, cpr. Figure 2. In regions where wind observations are scarce, such as over sea, the wind analysis is heavily influenced by the quasi-geostrophic balanced wind produced by the Norwegian Meteorological Institute as part of its weather prediction routine.

Alternative trajectories for transport at the 925 mb level rather than 850 mb, are calculated by backing the analyzed 850 mb wind by  $10^0$  and reducing it to 90%. Radiosonde observations close to the trajectory can give an indication as to the turning and change in speed of the wind with height.

The mixing height used represents a material surface below which both old and new pollutants are mixed. The 1200 GMT mixing height is chosen. The basic data for the mixing height analysis are taken from radiosonde data (about 120 radiosonde reports are available within the grid). The estimated mixing heights are objectively analyzed to produce grid values at 1200 GMT every day. At intermediate times it is assumed that each trajectory conserves its mixing height.

Objective analysis of temperature, relative humidity and absolute humidity are carried out at 0000 and 1200 GMT in the 150 km grid, as vertical averages between the surface and the 850 mb level. The temperature is used to evaluate temperature-dependent reaction rate coefficients. The relative humidity is used as a rough indication of cloud cover, which influences the photodissociation rates (see Table 1).

# Table 1. Parameterization of cloud cover using the relative humidity.

Relative	Cloud	"Effective"
humidity	cover	albedo
> 85%	1.0	0.6
75-85%	0.5	0.3
< 75%	0.0	0.0

When the relative humidity exceeds 90%, precipitation is assumed, and a wet deposition rate coefficient of  $1 \times 10^{-4} \text{ s}^{-1}$  is applied to the concentrations of  $H_2SO_4$ ,  $HNO_3$ ,  $H_2O_2$  and  $CH_3O_2H$ . For lower relative humidities than 90%, a first order wet deposition rate coefficient of  $5 \times 10^{-6} \text{ s}^{-1}$  is applied. The individual trajectories are assigned mean values of temperature and absolute humidity at 0000 and 1200 GMT. The temperature is estimated by linear interpolation and the absolute humidity is conserved at intermediate positions.

Dry deposition velocities are given in Table 2.

Table 2. Dry deposition velocities (for references, see Eliassen et al. (1982a)).

Component	Deposition velocity	Comments
	(cm/s)	
03	0.5	Daytime over land surfaces
03	0.05	nighttime over land
03	0.0	sea surfaces
NO <sub>2</sub>	0.5	see Hov et al. (1984)
PAN	0.2	
SO <sub>2</sub>	0.8	
HNO	1.0	assumed
H2 SO4	0.1	value appropriate for
_ ,		submicron particles

#### 3.2 Chemical model

A surrogate mechanism is used to represent the hydrocarbons which are emitted into the atmosphere. Of the hydrocarbon emissions, 30% by volume (on a compound basis) are represented as  $C_{26}H_{0}$ , 20% as  $nC_{410}$ , 20% as  $C_{24}H_{0}$ , 10% as  $C_{36}H_{0}$  and 20% as m-xylene. The chemical scheme and the representation of the hydrocarbon emissions are discussed in more detail by Eliassen et al. (1982a), Hov (1983) and Hov et al. (1984). It consists of about 100 chemical reactions including photochemical reactions, and 40 different species. It is an updated version of the scheme published by Eliassen et al. (1982a).

Dissociation rate coefficients are calculated for every  $5^0$  latitude and every 15 min of the day. The total vertically integrated atmospheric ozone column is adjusted to correspond to the season and latitude in accordance with the data given by Dütsch (1978). Points along a given trajectory are allocated dissociation rate coefficients through interpolation in time and space to the appropriate latitude and local time.

The initial concentrations assigned at the starting point of the 96 h long trajectories can be important for the development along the trajectory. Ground removal is the ultimate removal mechanism for ozone, and in cases with low deposition, the lifetime of ozone is much longer than four days (Hov et al., 1978b). In such situations four days' trajectories are insufficient to trace the history of an air mass. If the weather is fair at the starting point, the air masses arriving there may have accumulated photochemically active pollution for a number of days. Therefore, in such cases, the air chemistry calculations are initiated up to four days before the start of the trajectory, depending on the length of the good weather period. The emissions are then taken as averages over  $5 \times 5 = 25$  grid squares surrounding the starting point of the trajectory. In this way the chemical development along a model trajectory is made nearly independent of the initial conditions.

The integration is started with a set of concentrations corresponding to a very slightly polluted atmosphere, with the removal processes in equilibrium with NO and NMHC emissions near the northern hemisphere average ( $2 \times 10^{10}$  molecules cm<sup>-2</sup> s<sup>-1</sup> for NO<sub>x</sub> and NMHC/NO<sub>x</sub> (volume) = 1.25). The initial concentrations of the most important species are listed in Table 3.

Table 3: Initial concentrations (ppbv)

Specie	Concentration	Specie	Concentration
NO	0.02	NMHC (C)	3.4
NO2	0.5	03	29.0
S 0 2	1.2	нноз	0.1
SO4	0.5	PAN	0.04

Natural sources of hydrocarbons are not accounted for in the model. In separate model evaluations, it is found unlikely that natural hydrocarbons contribute significantly to the forformation of oxidants on a regional scale in Europe (Derwent and Hov, 1980b, Hov et al., 1983). Natural sources of  $NO_{\chi}$  are thought to be small compared to the anthropogenic sources. Stratospheric ozone or the ozone concentrations in the free troposphere do not affect the atmospheric boundary layer chemistry as long as the upper boundary of the mixed layer is considered to be a material surface.

#### 3.3 Emissions

As a basis for the model calculations, emission data for  $NO_{\chi}$ , SO<sub>2</sub> and HC were needed in a grid covering Europe. The uncertainties in these data are necessarily high. However, the degree of consistency obtained between the calculations and the measurements in the case study reported by Eliassen et al. (1982a), suggests that the emissions are reasonably well estimated.

An inventory of European sulphur emissions has been prepared in connection with EMEP (Dovland and Saltbones, 1979). This inventory gives the estimated annual (1978) emission in 150 km grid squares.

The estimated total national emission figures are listed in Table 4. The uncertainty is estimated to be 10-15% at best, and considerably larger for many of the countries.

The estimates of national emissions of NO<sub>x</sub> in OECD Europe (i.e. Austria, Belgium, Denmark, Finland, France, Federal Republic of Germany, Greece, Iceland, Ireland, Italy, Luxembourg, The Netherlands, Norway, Portugal, Spain, Sweden, Switzerland, Turkey and the United Kingdom (UK)), are based on information obtained from OECD. For the UK the emission given by Apling et al. (1979) is used. For the remaining European countries, the emission estimates are taken from Semb (1979).

The national emission figures estimated for  $NO_{\chi}$  are listed Table 4. Chemically,  $NO_{\chi}$  is assumed to be emitted as NO. Uncertainties are likely to be larger than for the  $SO_2$  emissions.

As a first approximation,  $NO_{\chi}$  emission data in the 150 km grid have been generated from the  $SO_2$  emission inventory by assuming that for each country, the distribution of  $NO_{\chi}$  emissions on grid elements is identical to that of  $SO_2$ . In certain grid elements where the sulphur emissions are thought to be anomalously high relative to the energy consumption, lower  $NO_{\chi}$  emissions have been assumed.

The estimates of non-methane hydrocarbons (NMHC) are based on information obtained from OECD (OECD, 1982), with the exception of UK, where the emission data of Apling et al. are used. According to these data, the ratio between national NMHC

TABLE 4: Assumed annual emissions of sulphur dioxide (SO<sub>2</sub>), oxides of nitrogen (NO<sub>x</sub>) and non-methane hydrocarbons (NMHC) for countries in Europe. Units:  $10^3$  tonnes, SO<sub>2</sub> measured as S, NO<sub>x</sub> as NO<sub>2</sub>, and NMHC by their total mass. For remarks about the uncertainties, see text.

	S	502-S	NO <sub>3</sub> -NO	)2	1	NMHC
Albania		50	10			10
Austria	1	215	275			280
Belgium		380	410			390
Bulgaria	•	500	240			240
Czechoslovakia		1500	600			600
Denmark		228	240			220
Finland		270	200			200
France		1800	1650			2000
German Dem. Rep.		2000	680			680
Germany, Fed. Rep.		1815	3350			2450
Greece		352	500			260
Hungary		750	220			220
Iceland		6	10			15
Ireland		87	90			105
Italy	1	2200	1550			1750
Luxembourg		24	50			30
The Netherlands	-	240	700			600
Norway		75	110			170
Poland		1500	1000			1000
Portugai		84	110			200
Romania		1000	460			460
Spain		1000	850			1050
Sweden		275	260			380
Switzerland	-	58	.160		28	260
Turkey		483	600			600
USSR (within grid)	1	8100	5000			5000
United Kingdom		2490	1730			1158
Yugoslavia		1475	210			210
Remaining area						
within grid		256	50			50

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and NO<sub>x</sub> emissions in OECD-Europe varies between 0.5 and 1.82 (NMHC measured by their total mass and NO<sub>x</sub> as NO<sub>2</sub>). For countries in non-OECD Europe, the NMHC emissions were estimated very roughly to be equal to the NO<sub>y</sub> emissions.

The resulting NMHC emission estimates are listed in Table 4. The uncertainties are thought to be considerably larger than for  $SO_2$  and may approach a factor of 2, in particular for countries in non-OECD Europe.

Emission grid data for NMHC have been generated by distributing the national emissions according to the sulphur emission inventory. In areas with many oil refineries and petrochemical industry, increased NMHC emissions are assumed.

#### 3.4 Mathematical formulation

The mass conservation equation determining the mass concentration  $c_i$  of species i can be written as

$$\frac{Dc_i}{dt} = -\left(\frac{v_d}{h} + k_w\right)c_i + \frac{E_i}{h} + S_i$$

The notation is

D/dt	Lagrangian (total) time derivative along a
	trajectory
v <sub>d</sub> (x,y,t)	dry deposition velocity
h(x,y,t)	mixing height
k <sub>w</sub> (x,y,t)	wet deposition rate
$E_{i}(x,y)$	direct emission of pollutant
S <sub>i</sub>	chemical sources or sinks.

In the integration procedure the appropriate back trajectories are first calculated from the analyzed wind fields. Then the quantities  $v_d$ , h,  $k_w$ , etc. originally given as Eulerian fields, are converted into Lagrangian information, i.e. as a function of transport time along the trajectories. These operations transform the mass conservation equation into an ordinary differential equation in time. Lastly, this equation is integrated to obtain calculated instantaneous concentrations at the receptor points.

The integration of the mass conservation equation has been done with a version of a quasi-steady-state approximation method (QSSA), described in detail by Hesstvedt et al. (1978). This method is explicit and applies a fixed time step. The method has been shown to give accurate predictions in a wide range of model calculations of atmospheric chemistry when compared with Gear-type methods with automatic error control (Hesstvedt et al., 1978; Derwent and Hov, 1979). The upper limit for the computational error is estimated to be 5%.

#### 4 THE CASE STUDY

The measurements of ozone at Jeløya during the 19 days' period 26 August - 14 September 1980 are used for comparison with the model calculations. The measurements at Rørvik, Jeløya and Maridalen (Figure 3) were quite similar with distinct peaks around 100 ppbv in ozone during the first ten days in September.

At Langesund, data are missing for the period 1-5 September, and the concentration was somewhat lower than at the other measuring sites during the rest of the period.

A reference model calculation was made calculating the chemical composition of air arriving at 9 receptor points 150 km apart in the EMEP grid in southern Scandinavia (Figure 2). The ozone concentration at each of the nine receptor points is shown in Figure 4, together with the mean and the standard deviation. In Table 5 the calculated mean ozone concentration during the 19 days' period for each individual receptor point is given. The concentration is calculated to decline for the receptor points over land northwards (the concentration at receptor point 1,  $c_1$ , is less than  $c_4$  which is less than  $c_7$ , similarly  $c_2 < c_5 < c_8$  and  $c_3 < c_6 < c_9$ ). Also, the concentration is calculated to increase to the east ( $c_1 < c_2 < c_3$ ,  $c_4$  $\langle c_{5} \langle c_{6}, c_{7} \langle c_{8} \rangle \langle c_{9} \rangle$ . The decrease in concentration northwards is partly a reflection of the efficient ground removal of ozone over land, while the increase eastwards should partly reflect that the transport is predominantly from the southwest, indicating an increase in travel time from the pollution sources eastwards. Gradients in the emission field could also contribute to the calculated spatial pattern of the ozone concentration at the receptor points.

Point no.	1	2	3	4	5	6	7	8	9	Mean	Initial value
0 <sub>3</sub> (ppbv)	34.9	39.7	43.2	38.2	42.3	45.8	38.8	44.5	49.1	41.8	29.0

Table 5: Calculated mean ozone concentration at each of 9 receptor points for the time period 26 August-14 September 1980.

There is a considerable spread in the calculated ozone concentration at the nine receptor points. At all points except No. 1, a maximum concentration is predicted for 28 August, which is not detected in the measurements. Calculated values at points 1 and 4 fail to predict the observed maximum around 4 September, while the maximum 8-9 September is calculated quite correctly at all receptor points. The calculated mean value fit well with observations during the whole period, except for 28 August where about 100 ppbv of ozone is predicted and 50 ppbv observed. In the following, all the model results discussed will be averages for the nine receptor points shown in Figure 2.

The prediction of the reference model will be compared with other calculations where physical and chemical processes have been assigned different values in order to see the impact of slightly different interpretation of the meteorological situation, solar radiation, initial conditions, deposition and in particular the emissions. The sensitivity study is summarized in Table 6. Table 6a: Parameters altered in sensitivity studies.

Parameter	Description
Backing of trajectories	$10^0$ and $30^0$ backing, reduction of wind to 90%
Smoothing of emissions	Moving average of 9 grid cells along the trajectories
Initialization	Four days' initial calculation at the trajectory starting points with emissions equal to 5x5 grid cell average.
Solar radiation	Zero cloud cover or full cloud cover along all trajectories.
Ozone ground deposition	v <sub>d</sub> = 0.6 cm/s always, or v <sub>d</sub> = 0.0 cm/s throughout

Table 6b: Emission control scenarios

	f all emissions by 50% all emissions
-	f all emissions to 10%
Reduction of Reduction of	f all NO <sub>x</sub> emissions by 50% f all HC emissions by 50%
Reduction o	f all SO, emissions by 50%
	f UK emissions to 10%
Reduction of	f non-Scandinavian emissions to 10%

The effect of backing the trajectories and reducing the wind speed to 90% can be seen in Figure 5. Backing by  $30^{\circ}$  causes the trajectories to pass over important pollution sources in the UK, with an average ozone concentration of 54.9 ppbv during the 19 days' period, compared to 47.4 ppbv in the  $10^{\circ}$  backing case and 41.8 ppbv in the reference model calculation. By backing and reducing the wind speed, a lower transport height is assumed than 850 mb.

Averaging the emissions along the trajectory over 9 grid cells does not change the average results over the 19 days' period very much, as can be seen from Table 7. There is an increase of 2.0 ppbv (4.8%) in the average ozone concentration for the 9 points for the 19 days' period, while the 19 days' average for each of the 9 cells goes up from 0.9 to 2.9 ppbv. On days with significant ozone formation, however, there are marked changes in the results. This reflects the gradients in the emission fields.

Table 7: Ozone concentrations (ppbv) for standard calculation and calculation where the emissions are averaged over 9 grid cells along the trajectories. The receptor point numbers refer to the information given in Figure 2.

Receptor point	1	2	3	4	5	6	7	8	9	Average
Sept. 4, 0600 h										
Standard, Averaged	40.6	104.5	86.3	49.0	104.8	91.6	42.8	126.6	98.8	82.8
emissions	48.6	80.1	108.2	69.1	91.7	113.6	67.6	88.6	100.9	85.4
Sept. 8, 1800 h										
Standard, Averaged	93.7	103.7	117.8	89.7	117.1	104.7	37.1	95.5	92.8	94.7
emissions	104.0	110.0	100.9	101.1	115.9	112.0	47.7	104.8	112.5	101.0
Average, 19 days' period										
Standard, Averaged	34.9	39.6	43.0	38.1	42.2	45.6	38.7	44.3	48.9	41.7
emissions	37.0	41.6	45.5	39.1	43.1	48.5	40.9	46.3	51.1	43.7

Four days of initialization at the start of the trajectory makes less difference to the results than averaging the emissions over 9 grid cells along the trajectories. This is due to the meteorological situation during the period considered, where the 96 h trajectories mainly originate in the Atlantic far away from important emissions (Figure 2). In the case study reported by Eliassen et al. (1982a), the effect of extending the calculations beyond 4 days was important because the 96 h trajectories started over industrial areas in East Europe.

The extent of cloud cover is an important parameter as can be seen from Figure 6 where zero and 100% cloud cover is assumed, respectively. The calculated peak in ozone on 28 August almost vanished when 100% cloud cover was assumed. Indeed, inspection of the surface weather maps on 28 August and the preceeding days, indicates overcast weather and frontal passage over the UK and the North Sea towards southern Scandinavia. It seems as if the meteorological data assigned to the trajectory positions for 28 August contain values of

the relative humidity which give too little cloud cover in the model calculation.

Further support for the hypothesis that the trajectories calculated for 28 August were about right while the meteorological data for cloud cover were wrong, can be found in Table 8 where calculated diurnal mean SO<sub>2</sub> and aerosol sulphate concentrations for the nine receptor points are given together with the measured daily average SO<sub>2</sub> and aerosol sulphate for EMEP sites in southern Scandinavia (Skreådalen, Birkenes and Rørvik; EMEP, 1981). The sulphur concentration was high on 28 August, indicating air mass passage over significant pollution sources with the potential of ozone formation if the sun had been out.

Table 8: Calculated diurnal mean concentration of SO<sub>2</sub> and aerosol sulphate averaged for the nine receptor points, together with measured values at Skreådalen, Birkenes and Rørvik. In µgS/m<sup>3</sup>.

		Measured
Date	Calculation Skreådaler SO <sub>2</sub> SO <sub>4</sub> SO <sub>2</sub> SO <sub>4</sub>	n Birkenes Rørvik SO <sub>2</sub> SO <sub>4</sub> SO <sub>2</sub> SO <sub>4</sub>
26 August 1980 27 28 29 30 31 1 September 1980 2 3 4 5 6 7 8 9 10 11 12 13	$\begin{array}{c ccccccccccccccccccccccccccccccccccc$	$ \begin{array}{ c c c c c c c c c c c c c c c c c c c$
11	4.5 0.4 1.7 3.32	0.2

The days with maximum diurnal mean SO<sub>2</sub> and sulphate aerosol during the 19 days' period as measured at Skreådalen, Birkenes and Rørvik coincide well in time with the calculations. The absolute levels do not always agree well, but it should be remembered that the model includes gas phase chemistry only, and the airborne sulphur at the receptor points is only a small fraction of the total emissions along the trajectories.

As concluded also by Eliassen et al. (1982a), ozone ground removal is a dominant factor when the boundary layer concentration is calculated. There is not very much difference between the zero- $v_d$  case and the reference case, indicating that much of the transport during the 19 days' period occurred over sea.

The calculations of the ozone concentrations at the nine receptor points shown in Figure 2 during the period 26 August to 14 September 1980 and comparison with measurements at Jeløya, Maridalen and Rørvik show good agreement. The essential controlling factors for ozone are well described. The model seems sufficiently verified to be used to indicate the efficiency of ozone control strategies.

calculated mean PAN concentration with standard The deviation for the nine receptor points is shown in Figure 7. There are three days with distinct peaks: 28 August, 3-4 September and 8 September. PAN measurements at Rørvik and Risø from 2-5 September and Maridalen 7-9 September are shown in Figure 3. Peak concentration of 4 ppbv was recorded at Risø in good agreement with the calculations. About 5 ppbv was recorded in Maridalen on 8 September, again the calculations agree well. PAN is shown to be a sensitive indicator of oxidant pollution, with a large difference in the mean PAN concentration for the nine receptor points during the 19 days' period (0.9 ppbv) and the maximum calculated concentration of about 5 ppbv. In Table 9 the nine point, 19 days' average of PAN is shown for the different sensitivity computations.

Table 9: Nine receptor points, 19 days' average PAN concentration in ppbv (The initial concentration was 0.04 ppbv).

Zero cloud cover always : 0.9 100% cloud cover always : 0.6 Reference run : 0.9 Moving average of emissions over 9 cells : 1.0 Four days initialization : 1.0 Backing of wind 10<sup>0</sup> : 1.3 Backing of wind 30<sup>0</sup> : 1.8 Zero cloud cover always : 0.9 100% cloud cover always : 0.6

## Ozone ground removal 0.6 cm/s : 0.7 Ozone ground removal 0.0 cm/s : 0.9

#### 5 EMISSION CONTROL STRATEGIES

#### 5.1 <u>Ozone</u>

The model was applied to evaluate the effect of the emission changes specified in Table 6b on the concentration of oxidants at the 9 receptor points in southern Scandinavia during the period 26 August to 14 September 1980. The results for ozone are shown in Figure 8 and Table 10. In the left hand panel of Table 10, the average ozone concentration calculated for only those receptor points where the concentration exceeded 100 ppbv on 4 and 8 September 1980, is shown. The effect on ozone by doubling and reducing by 50% all emissions are less than proportional, even when it is taken into account that 29.0 ppbv of ozone was present at the starting point of each trajectory. Hydrocarbon emission control is much more efficient than NO control or a combined HC-NO control. A 50% HC emission control is calculated to reduce ozone much more than reducing both HC, NO  $_{\rm x}$  and SO  $_2$  by 50%. SO  $_2$  control alone can be seen to influence ozone very slightly, demonstrating that with the chemical scheme adopted, the sulphur chemistry has little impact on the HC-NO chemistry.

Even though there is a predominant southwesterly flow with passage over The British Isles for most of the trajectories during the period 26 August-14 September 1980, emissions on continental Europe play an important role in the high ozone cases. As can be seen from Table 10, only 12.8 ppbv of ozone is generated (in excess of the initial concentration) averaged over the receptor points where the ozone concentration is calculated to exceed 100 ppbv on 4 and 8 September when all non-Scandinavian emissions are down by a factor 10, while this number is 59.9 ppbv when only the UK emissions are cut by 90%, and 80.7 ppbv in the reference case.

In the middle panel of Table 10, average ozone concentrations are calculated where data for all 9 receptor points are included if one or more of the receptor points had ozone concentrations exceeding 100 ppbv on 4 or 8 September 1980. The effect of changing all emissions is less pronounced, while the high efficiency of HC-control alone versus NO<sub>x</sub> control or combined NO<sub>x</sub>-HC control is as striking as in the lefthand panel.

Averaging over all receptor points throughout the 19 days' period gives a different picture. In this case, all the days with very little ozone generation along the trajectories contribute strongly to the average concentration. It can be seen that 12.7 ppbv of ozone is generated as an average in excess of the initial concentration, and the UK emissions have contributed about 7.1 ppbv (41.7-34.6 ppbv) or 56%, while Scandinavian emissions have contributed 0.7 ppbv or 5.5% and emissions on continental Europe 4.9 ppbv or 38.5%. In the high ozone cases (left hand panel in Table 10), UK emissions have contributed about 26%, Scandinavian emissions 16% and emissions on continental Europe 58% of the amount of ozone generated above the initial concentration. UK emissions thus have contributed the major part of the precursors for ozone during the days with low ozone at the receptor points, while the dominant source of precursors on high ozone days was continental Europe during the 26 August - 14 September 1980 time period. Or said differently; the weather conditions were more favourable for oxidant formation for the cases with transport over continental Europe towards Scandinavia than during the cases with direct transport over The British Isles and the North Sea.

Scandinavian emissions were not important for the calculated ozone concentrations at the 9 receptor points.

611071	Average ozone concentration at all 9 receptor points during the calculation period 26 August - 14 September 1980	relative to reference	41.7 1.00		35.9 .86	49.0 1.18	28.4 .68	37.9 .91 37.3 .89		34.6 .83		29.7 7.71		
סוורבוורז מ		0												
	centration points whe them, ozon 0 exceed 8 Sept, 19	relative to reference	1.00		. 75	1.35	64.	. 69	.98	.82		44.		_
- ATT THE CONTRACTOR CONTRACT AND THE CHARGE OF CALCULATACED OFOLG CONCENT ALTONS	Average ozone concentration at all 9 receptor points when at one or more of them, ozone was calculatead to exceed 100 ppbv on 4 and 8 Sept. 1980		19.8		59.9	107.6	34.2	71.4	78.6	65.1		35.1		
	e concentration points calculated 0 ppbv on 4 and 1980 {ppbv}	relative to reference	1.00		11.	1.42	.36	. 63	.99	. 81		.38		
1012000	C 0	Standard relative deviation reference	(2.2)		(1.6)	(2.5)	(6.0)	(2.0)	(2.1)	(6.2)		(1.1)		
	Average ozon at receptor to exceed 1 8 September		109.7		78.7	155.9	39.7	92.3	108.2	88.9		41.8		29.0
· · · · · · · · · · · · · · · · · · ·			Reference Run	Emission	changes: Down factor 2	Up factor 2	Down factor 10	NOx down factor 2 HC down factor 2	SO, down factor 2	UK <sup>c</sup> down factor 10	Non-Scandinavia down	factor 10	Concentration at the start of	the trajectory

Table 10: Emission control and the effect on calculated ozone concentrations

#### 5.2 Peroxyacetyl nitrate (PAN)

Table 11: Nine receptor points, 19 days' average PAN concentration in ppbv. (The initial concentration was 0.04 ppbv).

> Emissions reduced factor 2 : 0.5 Emissions increased factor 2 : 1.6 NO<sub>x</sub> emissions reduced factor 2: 0.7 HC emissions reduced factor 2 : 0.4 SO emissions reduced factor 2: 0.9 UK<sup>2</sup> emissions reduced factor 10: 0.5 Non-Scandinavian emissions reduced factor 10 : 0.2

The effect of emission controls on the concentration of PAN can be seen in Table 11. It can be concluded that the efficiency of HC control over  $NO_{\chi}$ -control is marked. Even when averaging is done only over the receptor points with the highest PAN concentrations on 4 and 8 September 1980, HC control is much more efficient than  $NO_{\chi}$  control. This can be understood by noting that the low PAN days (Figure 7) contribute much less to the average concentration than in the case with ozone. There is typically a factor 2 difference in ozone on days with high and low oxidant pollution, while the difference can be at least a factor 10 for PAN.

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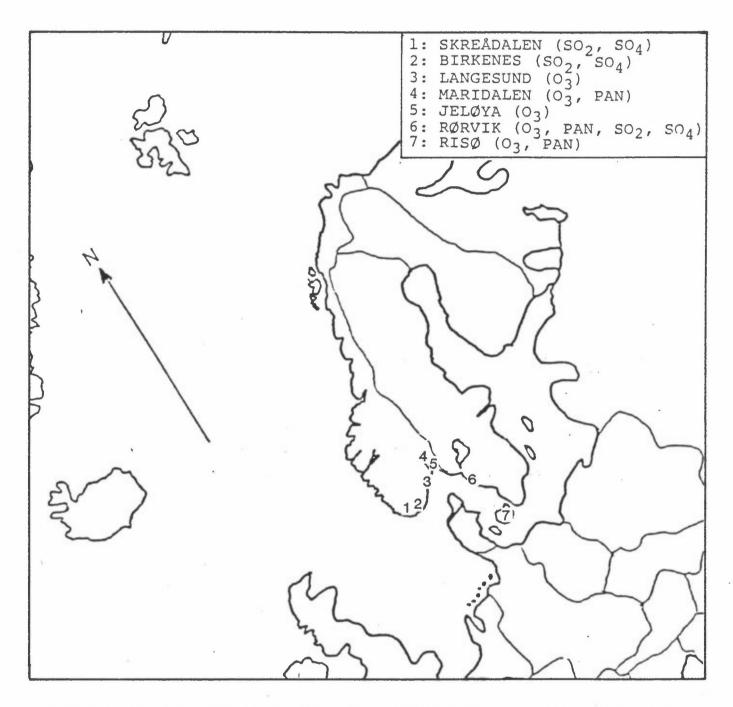


Figure 1 Location of monitoring sites: Rørvik (0, PAN, S0, S0), Jeløya (0), Maridalen (0, PAN), Risø (0, PAN), Langesund<sup>2</sup> (0), Birkenes and Skreådalen (S0<sup>3</sup>, S0).

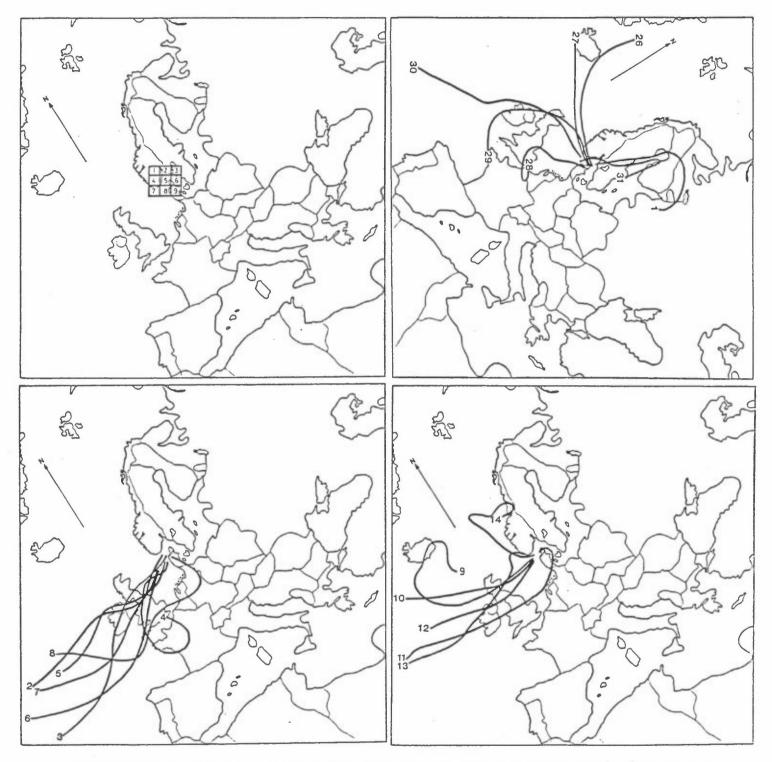
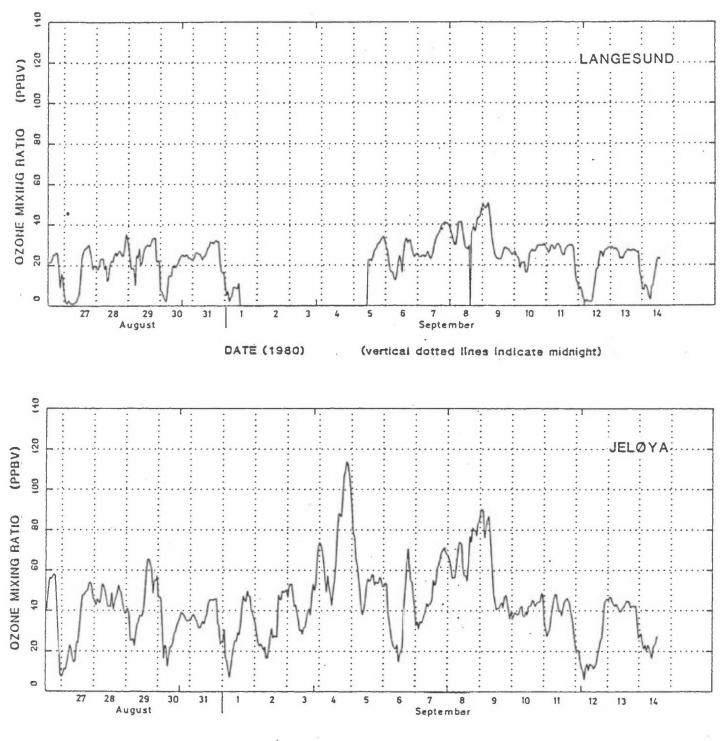


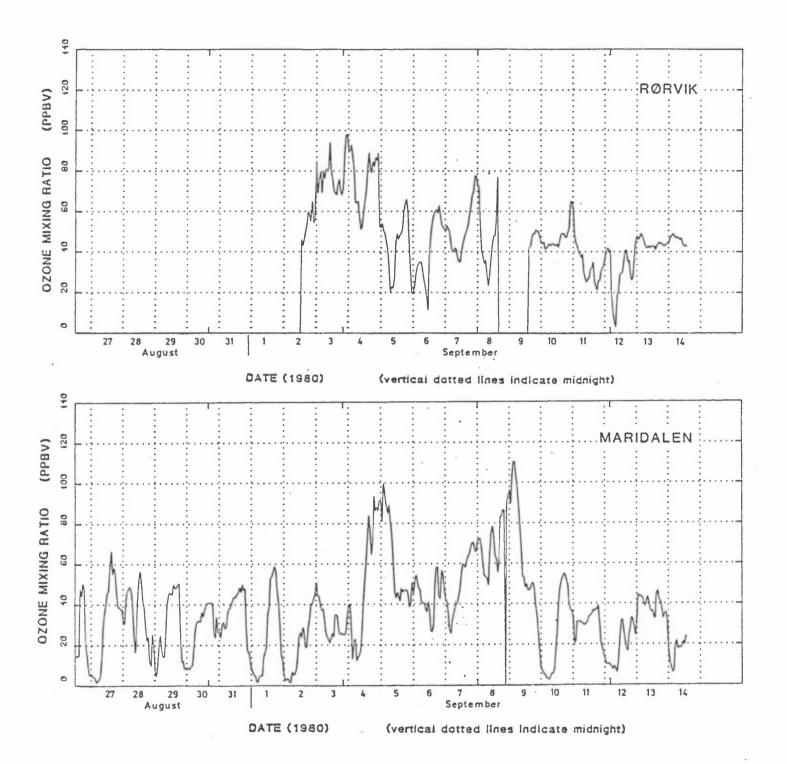
Figure 2 Map of the EMEP grid. The nine receptor points are indicated and numbered. 1200 GMT, 96 h, 850 mb back trajectories to receptor point no. 5 are indicated for the period 26 August - 14 September 1980.

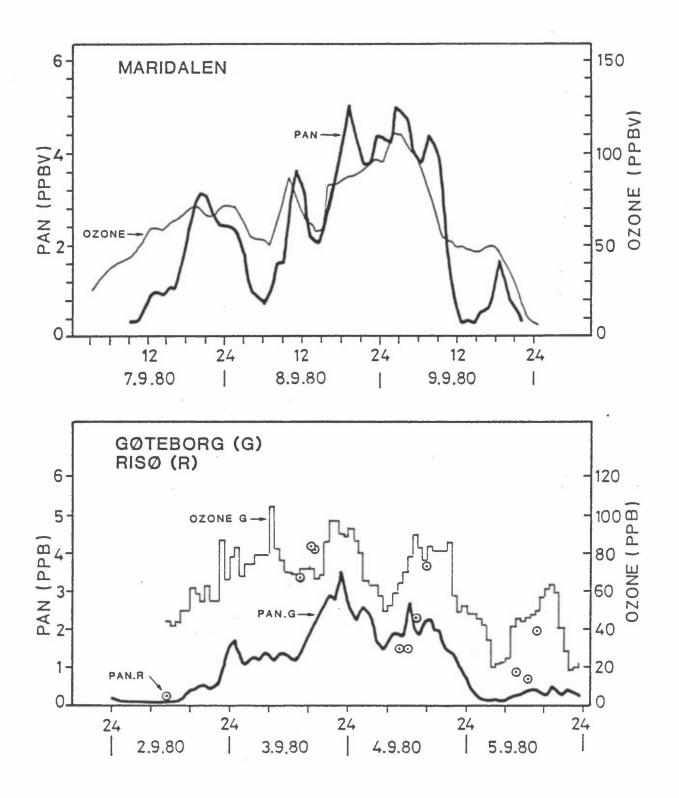


DATE (1980)

(vertical dotted lines indicate midnight)

Langesund, Figure 3 Measured hourly ozone concentrations at Jeløya, Maridalen and Rørvik throughout the period 26 August- 14 September (Schjoldager et al., 1981; Grennfelt, 1980 with some exceptions private communications). PAN at Rørvik, Risø and Maridalen for a part of the period is also shown (Nielsen et al., 1981, Schjoldager et al., 1983.





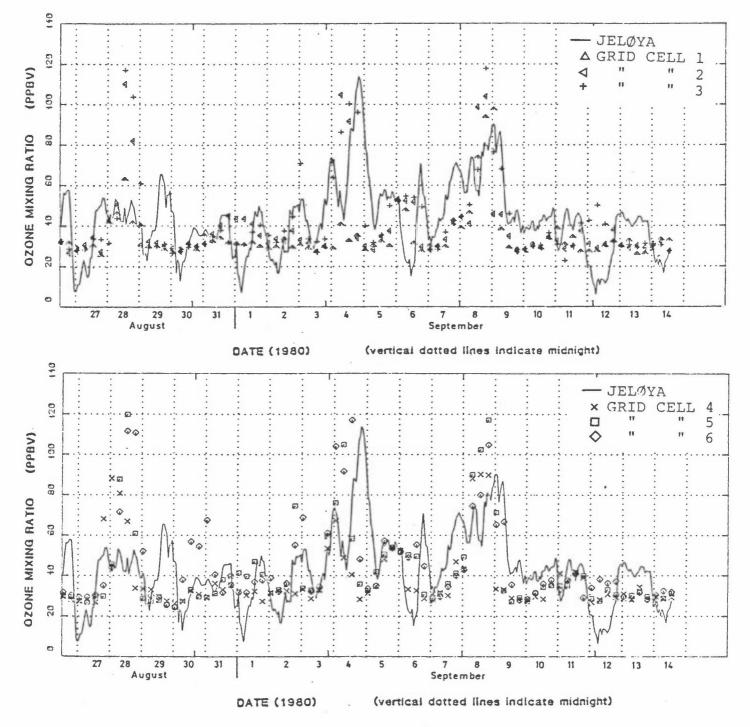
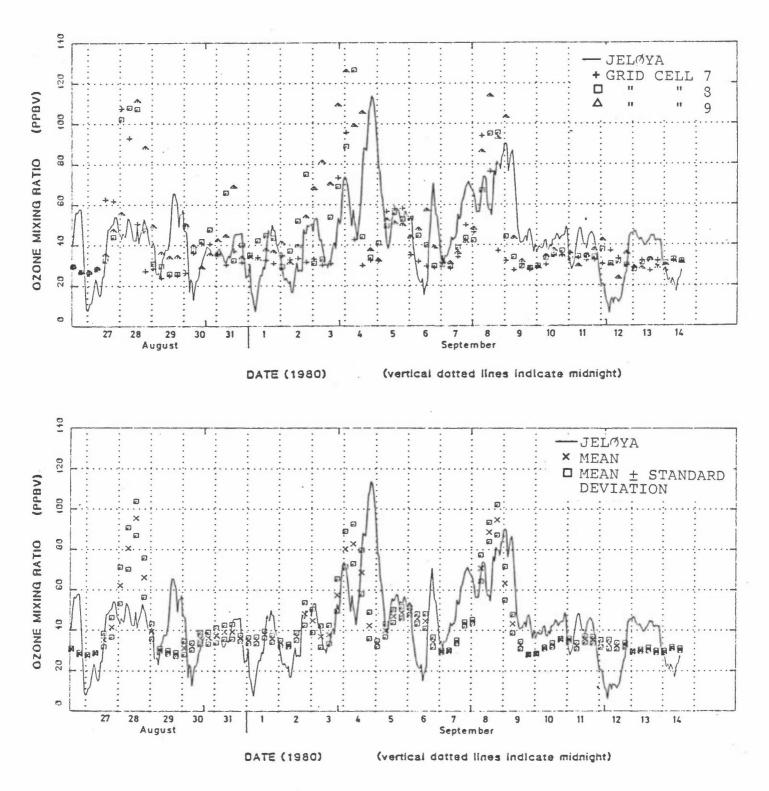
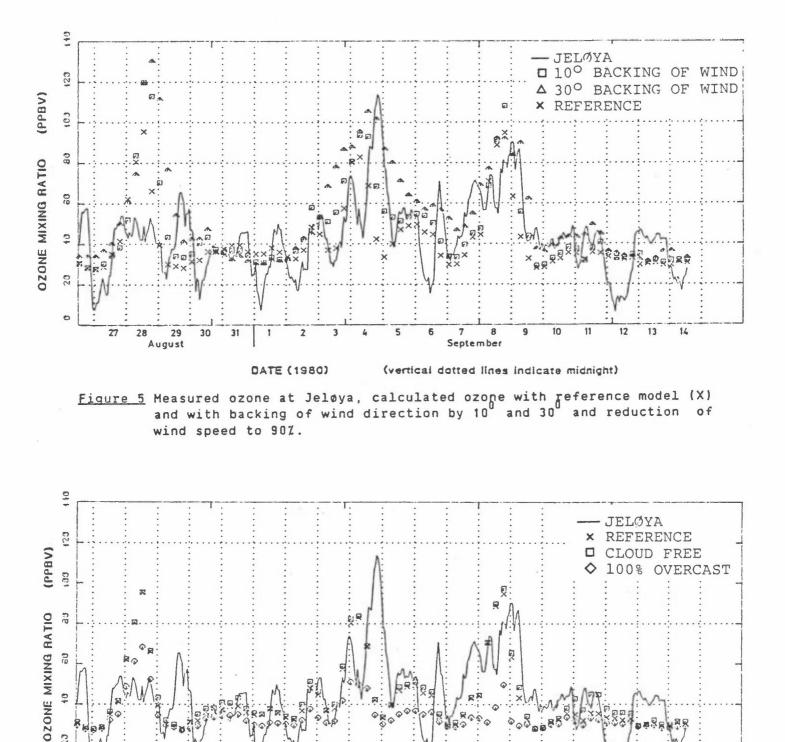
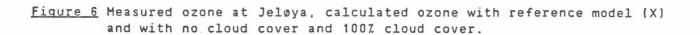


Figure 4 Measured ozone at Jeløya and calculated ozone to receptor points 1-9, in addition to  $0_3$  mean and standard deviation for the nine points.





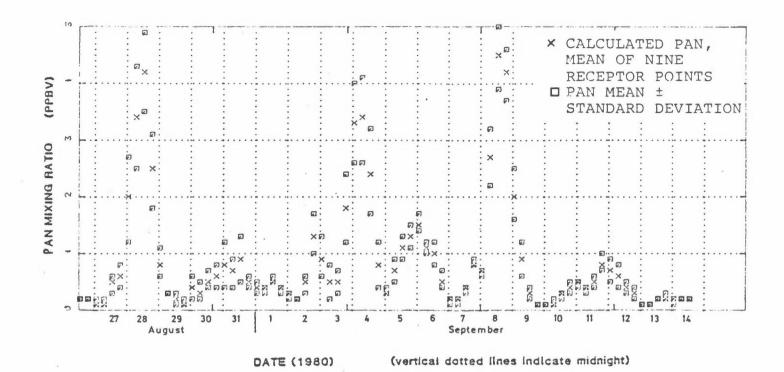


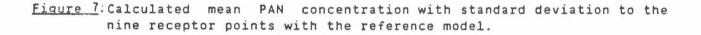
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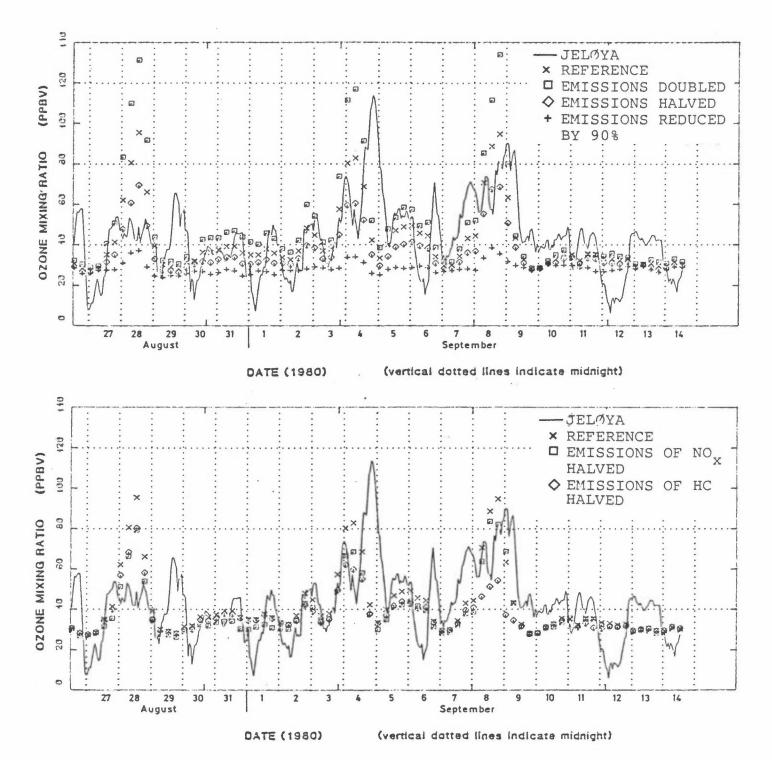
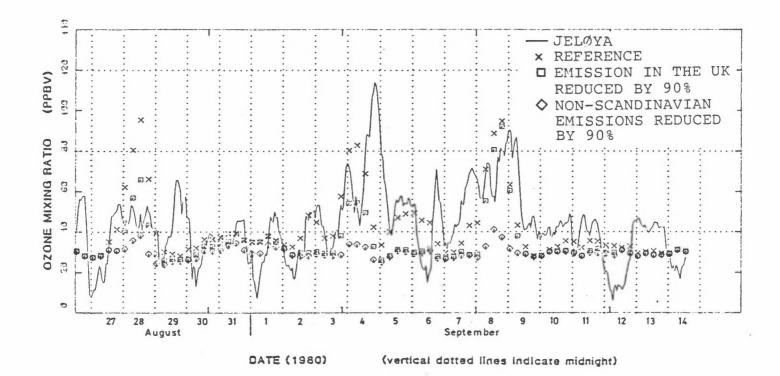


Figure 8 Measured ozone at Jeløya, calculated ozone with reference model (X) and for different emission control scenarios: All emissions up a factor of 2, down a factor of 2, down a factor of 10, NO<sub>x</sub> emissions down a factor of 2, hydrocarbon emissions down a factor of 2, UK emissions reduced by 90% and non-Scandinavian emissions reduced by 90%.



#### NORSK INSTITUTT FOR LUFTFORSKNING (NILU) NORWEGIAN INSTITUTE FOR AIR RESEARCH

#### (NORGES TEKNISK-NATURVITENSKAPELIGE FORSKNINGSRÅD)

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TITTEL Photochemical oxidant control strategies in Europe: A 19 days' case study		PROSJEKTLEDER Ø. Hov
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FORFATTER(E) Ø. Hov, F. Stordal og A. Eliassen		TILGJENGELIGHET A
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OPPDRAGSGIVER (NAVN OG ADRESSE) Statens forurensningstilsyn (SFT) Miljøverndepartementet (MD) Norges Teknisk-Naturvitenskapelige Forskningsråd (NTNF)		
3 STIKKORD (à maks. 20 anslag) kontrollstrategi ozon modell		
REFERAT (maks. 300 anslag, 7 linjer). Trajektoriemodellen med atmosfærekjemi utviklet ved NILU, UiO og MI (EMEP) er brukt til å beregne oksidantkonsentrasjonen i 9 ankomstpunkter i sør-Skandinavia i perioden 26/8-14/9 1980. Beregningene viser at ozonkonsentra- sjoner over 100 ppbv i ankomstpunktene effektivt reduseres hvis de europeiske utslipp av hydrokarboner reduseres. Kontroll av ut- slippene av nitrogenoksider eller både hydrokarboner og nitrogenok- sider er mye mindre effektivt som kontrollstrategi for ozon.		
TITLE Kontrollstrategier for fotokjemiske oksidanter i Europa. En 19 dagers modellberegning.		
ABSTRACT (max. 300 characters, 7 lines). The trajectory model with atmospheric chemistry developed by NILU, University of Oslo and DNMI (EMEP) has been applied to calculate the oxidant concentration at 9 receptor points in South Scandinavia in the period 26 August to 14 September 1980. The calculations show ozone concentrations exceeding 100 ppbv at the receptor points are efficiently reduced through re- duction of European HC emissions, while reduction of NOx or NOx and HC in combination is much less efficient to control ozone.		
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