Numerical forecasting of ozone at the surface

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1. INTRODUCTION

Exposure to elevated concentrations of ozone gives rise to a health risk for the human population and impairs agricultural productivity. National air quality standards, directives or guidelines have been introduced in many countries around the world. In many countries a warning system is established to alert citizens to take precautionary steps if the concentrations exceed certain values (see Dobris, 1995). According to the European Council Directive (92/72/EEC) on air pollution by ozone, thresholds are established for population information and warning. The threshold for warning of the public is 180 ppb as an hourly concentration, while the threshold for information of the public is hourly concentrations above 90 ppb. In the summer of 1996 in Europe, the threshold for warning of the public was exceeded at three sites (two monitoring stations in Athens and one in Firenze, Italy). The threshold for information of the public was exceeded in all member states of the European Union in the summer of 1996, except Ireland. It was calculated that 46% of the population in cities with operational ozone monitors may have been exposed to ozone exceedances in the summer of 1996 (Sluyter and van Zantvoort, 1996).

The forecasting of ozone analogous to numerical weather prediction is discussed in this paper. During the summer months, some countries have introduced routine procedures to

predict ozone based on the combination of the weather forecast and the monitoring of ozone, or by running chemical models alongside the numerical weather prediction model. In this paper a coupled chemistry-transport model (CTM) is described and it is shown how an ozone forecast can be made when a modern numerical weather prediction model is run in combination with a state of the art chemistry model.

2. MODEL DESCRIPTION

The general approach used in the model is similar to that used in several other three dimensional models developed in the past decade [Liu et al., 1984; Chang et al., 1987; Carmichael et al., 1991; McKeen et al., 1991, Hass et al., 1993]. The mesoscale chemical transport (MCT) model is closely coupled to a numerical weather prediction (NWP) model with an advanced treatment of cloud physics and precipitation processes. This allows a detailed parameterization of convective transport and the calculation of photolysis rates in cloudy conditions to be made. A circumpolar grid or smaller grid can be applied (Figure 1). If a large circumpolar grid is chosen for the calculations, it is ensured that most of the conversion of anthropogenic and natural emissions of NO_x, VOC and other species over North America, Europe and Asia, take place within the model volume, and the effect of not well known upwind boundary conditions is minimized. Since the advection field has a predominant westerly component the air masses will remain inside the model domain for a substantial part of the integration period. This is further discussed in Flatøy and Hov [1996], while if a smaller grid domain is chosen (e.g. Figure 1), the boundary conditions more rapidly influence the results.

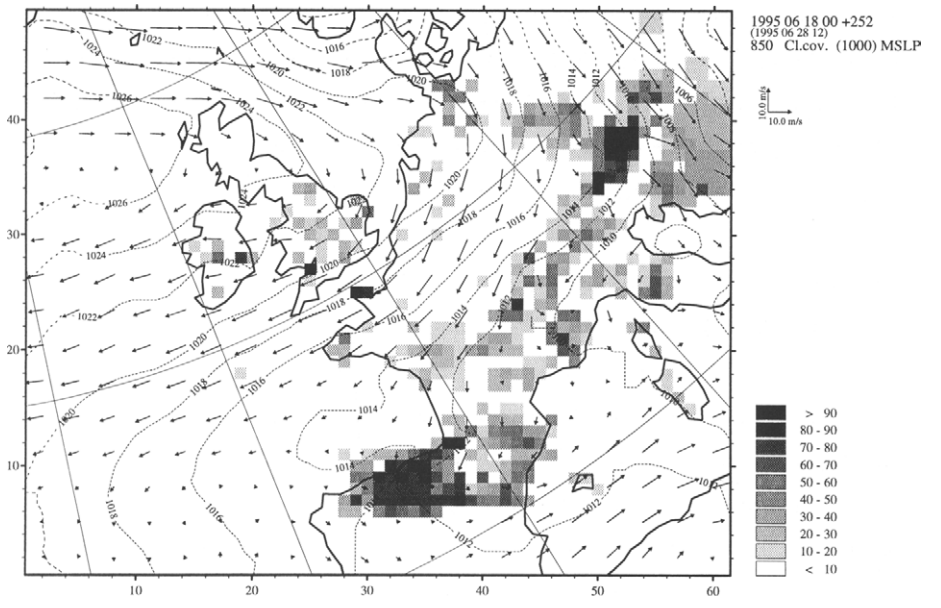

Figure 1. Mean sea level pressure, the 850 hPa cloud cover, and the 850 hPa winds at 1200 UT on June 28

The NWP model is based on the limited area model NORLAM from the Norwegian Meteorological Institute, described in Grønås et al. [1987], Nordeng [1986], and Kvamstø [1992]. Sigma coordinate primitive equations which are integrated on a stereographic map projection are used. The horizontal domain and the size of the grid elements are the same in the NWP and MCT models. Meteorological fields from the NWP model are in the present setup stored once every hour. The horizontal grid resolution of the input and the results is 50 kilometer at 60°N and there are 18 unequally spaced vertical layers extending up to 100 hPa in the version used here. With this resolution no interpolation in time of the meteorological data is thought necessary before use by the MCT model. The NWP model also supplies data for land use, topography, vegetation and ground albedo. Analysis every 6 hours from the ECMWF are used as initial and boundary conditions for a set of 18-hour prognosis with the NWP model. The first 6 hours of each prognosis are considered as spin up and are skipped, while the following 12 hours are used. The meteorological input to the MCT model then consists

of a chain of independent 12 hour prognosis segments. In a forecast mode, a prognosis would have to be generated based on the most recent analysis which is available.

The transport part of the MCT model is described by Flatøy et al. [1995], and Flatøy [1993] and the chemistry part by Flatøy et al. [1995]. The chemistry scheme includes more than 40 chemical species and more than 120 chemical reactions for the gas phase formation of photooxidants. The emissions of volatile organic compounds (VOC) are represented by six nonmethane hydrocarbons (NMHC) and several oxygenated VOC's, in addition to isoprene. Nighttime chemistry is assumed to occur through nitrate radical attack on VOC's. Both peroxy radical + NO and peroxy radical + peroxy radical reactions are included to assure validity of the reaction scheme over a wide range of NO concentrations. Aerosol and liquid phase chemistry is parameterized. The chemistry scheme is documented by Strand and Hov [1994], who applied it in a study of global ozone. The choice of initial values and boundary conditions is discussed in Flatøy et al. [1995].

EMEP emissions' inventory has been utilized for NO_x, SO_2 and anthropogenic NMHC emissions. The natural VOC emissions represented by isoprene are derived by an emission-temperature relationship from Lübkert and Schöpp [1989] using the forest cover and the variable surface temperature from the result of the NWP model calculations. The formula gives low isoprene emissions at night, following the diurnal cycle in the surface temperature. The temperature is here also a surrogate for the effect of the variation in the photosynthetically active radiation (PAR), which strongly affects the isoprene emissions. Empirical relationships for the dependence of the isoprene emissions on both temperature and PAR have been reported in the literature [Guenther et al., 1993]. Aircraft NO_x emissions are taken from the global ANCAT/ECAC Emission inventory database group [1995] and the lightning NO_x emissions are from Køhler et al. [1995].

3. FORECAST OF OZONE

The period of calculation to be discussed here is June 24 to 29, 1995 where a high pressure center north west of Scotland gave rise to a westward air flow off the coast of

South England and France. During part of the period a low pressure center was found over the Iberian peninsula which increased and narrowed the air flow and gave a flow situation with a continental plume from Europe flowing into the North Atlantic. We have performed several 18 days' simulations (18 June-5 July 1995) with different grid resolutions to investigate large and small scale features of this situation. The same period was also used in a study of the importance of lightning emissions of NO_x and how results from simulation models are influenced by uncertainties in this source. Lightning emissions are connected with convective activity. A set of simulations were made on a circumpolar grid covering the northern hemisphere from the pole to 35°N with a horizontal resolution of 150 km and 10 unequally spaced layers. The simulations were run with different types and localizations of emissions to study the combined effect of North American and European emissions on the atmospheric chemistry over the North Atlantic. Part of this work is presented in Flatøy et al. [1996]. It was shown how the transport of pollutants in the free troposphere across the Atlantic only takes in certain atmospheric flow situations.

Here the focus will be on a simulation where the horizontal scale was 50 km and with 18 layers in the vertical and covering a small part of the northern hemisphere (see Figure 1). This gives a possibility to study the small scale meteorological and chemical structures which a plume of continental emissions contains. Especially the effect of topography, better resolved convection and land-sea interactions are important for the meteorology and more detailed inventories of surface emissions and improved convective transport are important for the chemistry computations.

Figure 1 shows the mean sea level pressure, the 850 hPa cloud cover, and the 850 hPa winds at 1200 UT on June 28 1995 from the simulation with the 50 km grid. Before reaching the ocean the air masses passed over large sources of oxides of nitrogen, volatile organic compounds and other pollutants in the United Kingdom and in Central Europe. This gave rise to a well defined plume of polluted continental air downwind of the sources. The period when the plume developed and existed was characterized by fair weather and the plume was not disturbed by frontal motions and only to a limited degree by convective activity.

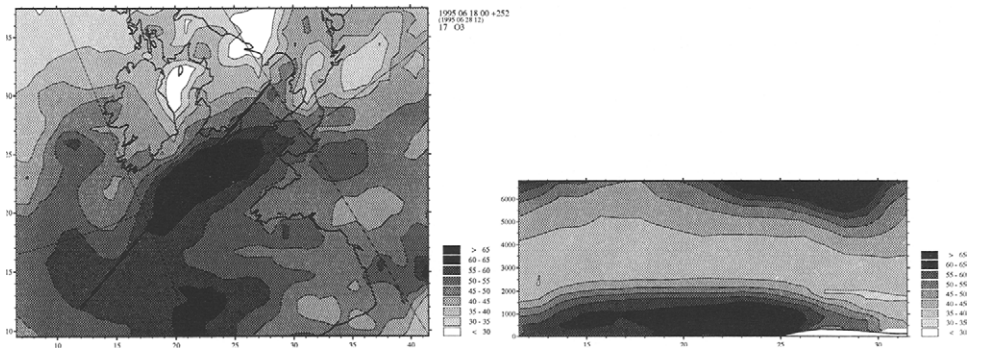

Figure 2. Ozone concentration at 1200 UT on June 28 for model layer 17, ~250 m a.s.l. (left panel) and vertical cross section of ozone concentration (right panel).

Figure 2 shows the O_3 plume at 1200 UT on June 28 1995 for model layer 17 (~250 m a.s.l.). Air with increased ozone concentration is transported with the large scale winds westward off the United Kingdom and the European continent. Ozone production (not shown) takes place close to the source regions, the largest ozone production is not found where the NO_x concentration has its maximum but in areas where NO_x is from 1-3 ppb.

Figure 3 shows scatter plots from June 28 1995 for model layer 17 of the nett ozone production by chemistry between 1200 UT and 1500 UT versus the NO_x concentration (left panel) and versus the NMHC concentration (right panel). The most favorable conditions for ozone production is seen to occur for NO_x concentrations around 2 ppb and NMHC concentrations around 20 ppbC.

Lightning emissions of NO_x can have an important influence on the chemical ozone production in the troposphere. To investigate if this can be properly handled by simulation models we have made a simulation for the above period where NO_x from lightning is distributed according to the convection modelled in the employed numerical weather prediction model, and compared this to the base simulation when a constant

lightning inventory is used. The results indicate that when a realistic distribution of the lightning sources of NO_x in time and space is calculated, the upper tropospheric fields calculated for ozone and nitrogen oxides, differ significantly from the results of a standard run where the lightning sources of NO_x are distributed according to some monthly or seasonal climatological average. The differences are larger than e.g. the effect calculated from aircraft emissions of NO_x. This work is further described in Flatøy and Hov [1997].

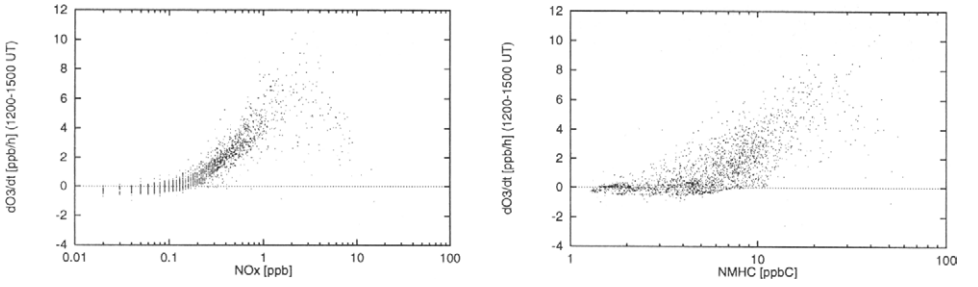

Figure 3. Scatter plots from June 28 for model layer 17 of the ozone production by chemistry between 1200 UT and 1500 UT versus the NO_x concentration (left panel) and versus the NMHC concentration (right panel).

The employed chemistry transport model gives a possibility to make estimates of the oxidation capacity of the atmosphere and the role of VOC and NO_x in the chemical ozone formation. This is of importance when measurement campaigns are planned and also when measured data is analyzed. An other important aspect of the model is the possibility to separate the different physical and chemical processes that modify the concentration distribution and thereby increase the understanding of the mechanisms behind observed concentration, see examples in Flatøy et al., (1995, 1996), Flatøy and Hov (1996, 1997) and Hov and Flatøy (1997).

The coupled NWP and chemistry model can be run with sufficient computational efficiency to be used in a forecast mode, provided that an appropriate weather analysis is available to initialize the numerical weather prediction model. The initial and boundary

conditions of the chemical compounds are of particular importance, however, in particular of those species which have a chemical decay time comparable to the transport time through the model domain. Such compounds are ozone, CO, oxides of nitrogen, and a long range of hydrocarbons and oxygenated species. In particular the boundary conditions for ozone aloft and on the lateral boundaries are essential for the absolute levels of ozone calculated in the free troposphere. Run in a forecast mode, i.e. covering a few days only, the ozone distribution in the free troposphere is to a large extent determined by dynamical processes, and consequently the aloft and lateral boundary conditions are important.

4. MODEL VALIDATION AND DISCUSSION

The calculations shown are examples of historic forecasts of ozone on a regional or continental scale, based on meteorological forecast by a numerical weather prediction model and based on precursor emissions estimated from annual values. The model has been compared with surface measurements, ozone sondes and aircraft measurements. In Flatøy et al. [1995] comparison were made of the calculated distribution of ozone over Europe and surface ozone as well as ozone sonde measurements. It was found that the upper boundary condition chosen for ozone (at 100 hPa) is quite important for the absolute level of ozone in the troposphere, while the relative changes with height and with time are caused mainly by dynamical/physical processes when a time period of a few weeks is studied.

The model results have also been compared with aircraft measurements made in international projects like the EU sponsored OCTA and POLINAT in 1993 and 1995 over parts of the North Atlantic, and where also CO, NO_x and NO_y were measured in addition to ozone. More details on the model validation are given in Flatøy et al. [1996] and Flatøy and Hov [1996].

A comprehensive field campaign to measure the transformation of European precursor emissions into secondary products like ozone and PAN is being undertaken in the summers of 1996 and 1997. In the EU-funded project Testing Atmospheric Chemistry in

Anticyclones (TACIA) the British Meteorological Research Flight aircraft C130 is instrumented to measure ozone, NO, NO_x, NO_y, HNO_3, PAN, CO, carbonyls, peroxides, photolysis rate coefficients and perhaps peroxy radicals on a continuous basis and individual nonmethane hydrocarbons on an offline basis, in air masses flowing off the European continent over the North Atlantic in anticyclonic conditions. These measurements will serve as a comprehensive basis for model validation and an assessment of the prognostic capability of the CTM.

ACKNOWLEDGEMENT

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