LONG-TERM CALCULATIONS OF AIR POLLUTANTS WITH THE EURAD MODEL

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INTRODUCTION

Comprehensive air quality models (AQMs) have been developed during the last decades to simulate the transport, chemical transformation and deposition of air pollutants on the regional scale (Chang et al., 1987; Vogel et al., 1995; Memmesheimer et al., 1997; Ebel et al., 1997; Langmann et al., 2002; Builtjes et al., 2003). The first applications of AQMs have been performed for episodes, e.g. for the investigation of photo-oxidant formation or acid rain. In recent years the formation of secondary particles from gaseous precursors and particle dynamics has been added to the physical and chemical processes handled in AQMs (Binkowski and Shankar, 1995; Ackermann et al., 1998; Binkowski, 1999; Schell et al., 2002, Riemer et al., 2003).

The rapid development of modern information technology now allows the application of comprehensive AQMs on an annual time scale as well as short-term prediction (chemical weather forecast, Jakobs et al., 2002a,b). Long-term runs can provide data which are useful for several purposes. From the scientific point of view physical and chemical processes controlling the concentrations of air pollutants can be studied using a much larger database. This led to a better understanding of processes in the atmosphere, supports the analysis of measured data and is helpful to evaluate the parameterizations used in the modelling system for different seasons and chemical regimes.

Another main focus of interest is the application of AQMs as a tool to develop optimised air pollution abatement strategies. The results of the models allow the assessment of air quality in regions where observations are incomplete or missing. Another very important application is the study of the implications of future emission scenarios on the ambient air quality. In this paper the results of a long-term run carried out with the European Air Pollution Dispersion Model (EURAD, Jakobs et al., 2002a,b) are discussed with special emphasis on atmospheric particulate matter over Europe. Strongly polluted areas as North-Rhine-Westphalia have been considered in detail using nesting techniques. The results of the annual model run for the year 1997 have been analysed with respect to the requirements of the EU directive 96/62 on air quality control and its daughter directives, e.g. 99/30. The results shown here are partially based on an extensive report prepared for the Environmental Agency of Northrhine-Westphalia (Friese et al., 2002).

MODEL DESCRIPTION

The chemistry-transport model of EURAD (EURAD-CTM) has been used to perform the calculation of transport, chemical transformation and deposition of air pollutants. Meteorological fields are provided by the mesoscale meteorological model MM5, transport is modelled within the CTM by solving the 3-D advection and diffusion equation. Gas-Phase chemistry is handled with the RACM chemical mechanism, dry deposition is treated with a resistance model, cloud process are parameterized following Binkowski, 1999, and Friese et al., 2000. The Modal Aerosol Dynamics Model MADE has been applied with extensions to account for the formation of secondary organic aerosols (Ackermann et al., 1998; Friese et al., 2000; Schell et al., 2002). MADE provides size resolved concentrations of secondary ($\text{SO}_4^{2-}$, $\text{NO}_3^-$,
NH$_4^+$, biogenic and anthropogenic organic) and primary (EC, OC, unidentified particulate matter) aerosol species. The near-surface concentrations of PM$_{10}$ as well as gaseous air pollutants (e.g.: NO$_2$, NO$_x$, SO$_2$, CO, Ozone) relevant for the EU directive 96/62 can easily be analysed on the basis of the model output.

The calculations have been performed for the year 1997 using a one-way nesting scheme, which had been successfully applied to different regions in Europe before (Jakobs et al, 1995; Feldmann et al., 2001; Memmesheimer et al., 2001; Kessler et al., 2001; Corsmeier et al., 2002). The European Scale is covered with a horizontal grid resolution of 125 km (N0), an intermediate N1 with 25 km horizontal grid size is used for Western/Central Europe, and a resolution of 5 km has been applied to simulate the region of North-Rhine-Westphalia (N2). For episodes of particular interest an additional nest (N3) has been used. In the vertical the atmosphere is divided into 23 layers between the surface and 100 hPa (about 16 km). 15 layers are below 3000m, the lowest layer is about 40 m thick. Model runs for the N0, N1, and N2 area have been completed for the year 1997. All concentrations in the gas-phase and the aerosol phase have been stored on an hourly basis for 1997. Wet or dry deposition of gas-phase and aerosol species is also stored. Results have been compared with measurements on the European as well as on the local scale. Episodes of particular interest have been calculated with a horizontal grid resolution of 1 km for areas of specific interest.

The modelling system also provides daily short-term predictions (Jakobs et al., 2002b) which are made available to the public by internet (www.eurad.uni-koeln.de). Data and graphics from the forecast are stored in a data archive which is also available on the EURAD web site.

RESULTS AND CONCLUSIONS

Model results have been analysed with respect to the requirements of the EU directives on air quality for PM$_{10}$ and other air pollutants, compared with observations, and analysed with respect to the contribution of different sources to the total mass of the particulates. Examples from the long-term calculation for 1997 are shown in the figures below.

The nesting procedure of the modelling system is illustrated by figure 1 which shows the PM$_{10}$ concentration for all nest levels from the European scale to the local scale with a horizontal grid size of 1 km. Figure 2 show an example for an analysis of the model results with respect to the limit values given in the EU directive 96/62 and its daughter directive 99/30. Shown are the annual averaged values for NO$_x$, clearly exceeding the limit value of 30 µg/m$^3$ for the protection of vegetation, and the number of days exceeding the daily average of 50 µg/m$^3$ for PM$_{10}$ in 1997. The limit value of 35 days per year which should be reached till 2005 was clearly exceeded in NRW for 1997. Figure 3 shows a comparison of observed and measured concentrations of NO$_x$ and PM$_{10}$ in NRW. The measurements and the model results are in a reasonable agreement, except for an underestimation of NO$_x$ as well as PM$_{10}$ during summer. Extremely high NO$_x$ values also can not be simulated by model calculations. Higher spatial resolution than 5 km is needed to get a better agreement with observations for some of the measurement sites which are located near streets with high traffic density. Probably a coupling with street canyon models is necessary to get an improved description of high concentration of NO$_x$.

The results of a simple emission scenario for NRW are shown in figures 4 and 5. Within NRW all anthropogenic emissions have been set to zero in the scenario. Therefore the results of the model calculations show the importance of long-range transport for PM$_{10}$ and other air pollutants in NRW. Evidently the transport of particulate matter is important for PM$_{10}$ concentrations in NRW in the actual situation displayed in figure 4. Air loaded with particles is transported across the southern and western border of NRW and lead to quite high particle concentrations in NRW even for the emission scenario case. This can also clearly be seen in the times series for NO$_x$, ozone and PM$_{10}$ shown in figure 5 for the measurement site Köln-Chorweiler. PM$_{10}$ evidently is strongly influenced by transport whereas NO$_x$ is controlled mainly by local sources.
It can be concluded that the EURAD modelling system can be used as a tool for the optimisation of air pollution abatement strategies as well as for scientific purposes to investigate the transport of particulate matter and its formation on the regional and local scale. Future plans aim on further evaluation of the model results in particular with respect to the chemical composition (a preliminary example is shown in figure 6), development of 4DVar data assimilation schemes (Elbern and Schmidt, 2001) using information from satellites and lidar systems, and the extension of the modelling system to the hemispheric scale to account for intercontinental transport.

**Figure 1:** Example for the nesting application of the EURAD modelling system. The PM$_{10}$ mass concentration over Europe and nested areas focussed on Northrhine-Westphalia are shown for Sept.30, 1997, 06 UTC. In that case particle mass is accumulated within a stable high pressure system over Central and Western Europe (upper left). Nesting has been done with horizontal grid sizes of 25 km (N1, upper right), 5 km (N2, lower left), and 1 km (N3, lower right). The black dots show the location of larger cities. The colour scale has been changed for N3 which covers the area of Duisburg, Essen and Düsseldorf.
**Figure 2:** Example for the analysis of the annual run of the EURAD system for the year 1997 with respect to the EU directive on air quality control (96/62) and its daughter directives (e.g. 99/30). Displayed are the annual average of the NO\textsubscript{x} concentration (left) and the numbers of days exceeding a daily average of 50 µg/m\textsuperscript{3}.

**Figure 3:** Scatter diagrams. Right panel shows scatter plot for daily averages for observed and calculated NO\textsubscript{x}-concentrations; left: scatter plot of observed and calculated averages of PM\textsubscript{10} for measurements sites in North-Rhine-Westphalia. The observed values of PM\textsubscript{10} seem to be in reasonable agreement with the model calculations, except for summer where the calculated concentrations of PM\textsubscript{10} show a tendency to underestimate the observed values.
Figure 4: Emission scenario study for Northrhine-Westphalia for a selected episode in September/October 1997. All anthropogenic emissions in NRW have been set to zero, starting with September 27, 00 UTC. Shown are the model results for PM$_{10}$ for September 29, 1997. Upper left: PM$_{10}$ for the base case, Sept. 29, 06 UTC; upper right: PM$_{10}$ for the scenario, lower left: PM$_{10}$ for the base case, Sept. 29, 18 UTC; lower right: PM$_{10}$ for the scenario. Major wind direction in the morning (06 UTC) is from southeast evidently transporting PM$_{10}$ along the Rhine valley into NRW, PM$_{10}$ in the Netherlands is clearly reduced. In the afternoon wind is blowing from southwest. This brings air loaded with particles across the western border into NRW.
Figure 5: Results from the emission scenario study (no anthropogenic emissions in NRW). Shown are time series for a measurement site in Cologne for NOx, Ozone and PM$_{10}$ as calculated by the EURAD modelling system. Green line represents the results for the base case, red line for the scenario. Observations are indicated by the black line. As mentioned before wind direction was mainly from southeast till September 29 and turns to southwest during September 29. Southwest remains the prevailing wind direction till the end of September. Fresh and clean air reaches the measurement site in the beginning of October as indicated by the low concentrations of PM$_{10}$ and NOx.
Figure 6: Secondary aerosol formation due to organic gaseous precursors of anthropogenic (upper panel) and biogenic (lower panel) origin. The mass concentration of secondary organic aerosols in the aitken mode and the accumulation mode is presented for the N0 domain covering the whole of Europe. SOA formation due to anthropogenic emitted gases are dominating in central and western Europe, including the impact of oil platforms in the North Sea which can clearly be seen. In eastern and northeastern Europe biogenic precursors as pinene and limonene are dominating. For details of the modelling approach see Schell et al., 2002. Data are from an annual data set generated for the year 1997 during the ANABEL project funded by the Environmental Agency of Northrhine Westphalia (LUA; Friese et al., 2002).
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