The Use of Nested Models for Air Pollution Studies: An Application of the EURAD Model to a SANA Episode

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ABSTRACT

A multiple-nesting version of the European Acid Deposition Model (EURAD) has been developed in order to increase the horizontal resolution in a region of enhanced pollution, namely the former German Democratic Republic. This new technique allows the ability to simulate large-scale features together with the development of smaller-scale structures in the nested regions. This multiple-nesting approach was applied to a case that occurred in October 1990, the so-called SANA1 episode. SANA is a German acronym that stands for "scientific program for the assessment of the air pollution situation in the former German Democratic Republic." The SANA program was established to observe the rapid change in composition of air pollutants and their concentration levels over the eastern part of Germany due to political and economical changes. Thus, within the SANA program there is a unique chance to observe and control the effect of air quality strategies.

Two nested areas are embedded in a coarse domain that covers the main parts of Europe. The second nested domain is nested within the first nested domain. For EURAD, the nesting is two-way for the meteorological part and one-way for the chemistry transport module (CTM). This means that the meteorological variables that coincide with the boundaries in the nested domain are interpolated by a monotone flux-corrected transport method from the next coarser domain and provide a feedback from the nested domain to the next coarser domain. For the CTM the inflow boundary conditions are dynamically determined by interpolation from coarse-grid results. The outflow conditions are specified by continuous advection at the boundary in order to eliminate numerical reflections. The results of the simulations indicate a strong dependence of the horizontal distribution of both meteorological quantities and atmospheric constituents on increased spatial resolution. They exhibit a much more realistic structure, especially for the simulations in the nested domain with highest resolution (8.89-km horizontal grid distance). Comparisons for the predicted SO2 concentrations with observations from selected stations clearly demonstrate that the simulations agree best with observations in the nested domain of highest horizontal resolution.

1. Introduction

One of the ongoing challenges in the simulation of transport, chemical transformation, and deposition of atmospheric pollutants is the capability to nest domains of high horizontal resolution within a regional limited-area model. There is a need to build a hierarchy of interactive models that range from a regional scale (approximately 100-km grid length) down to the local scale (lower than 10-km horizontal grid length). This demand is supported by Pleim et al. (1991) because many processes that are involved in the formation of acid deposition and photo-oxidant production occur on scales that cannot be resolved by current regional-scale models.

Conventional comprehensive air quality models either simulate on a large scale [e.g., the Acid Deposition and Oxidant Model (ADOM; Venkatram et al. 1988), the Regional Acid Deposition Model (RADM; Chang et al. 1987), the Sulfur Transport Eulerian Model (STEM; Charmichael et al. 1986)] or on a smaller scale [e.g., the Model for the Atmospheric Dispersion of Reactive Species (MARS; Moussiopoulos 1994), the three-dimensional regional transport and deposition simulation model (DRAIS; Fiedler 1987; Baer and Nester 1992), the Urban Airshed Model (UAM; Scheffe and Morris 1993)]. A coupling of both scales can be achieved by subsequently nesting domains in several steps to cover the range from continental to local scales. First developments were made by Pleim et al. (1991), who used the RADM system with horizontal grid spacings of 80 km (coarse domain) and 26.67 km (nested domain). Recently, the Regional Atmospheric Modeling System (RAMS; Pielke et al. 1992) was able to include a telescoping, interactive nested-grid capability.

In this study, the European Acid Deposition Model (EURAD) is applied in a multiple-nesting version to simulations of pollutant transport and deposition during an episode in October 1990 (SANA1). The air quality modeling system EURAD has been applied to various episodes and processes: the transport and de-
position of radionucleides after the Chernobyl accident (Hass et al. 1990), the intrusion of stratospheric ozone into the upper troposphere (Ebel et al. 1991), the simulation of a wet deposition and a photosmog episode (Hass et al. 1995), sensitivity studies of tracer fields for different spatial resolutions (Jakobs et al. 1991), and evaluation studies for a winter smog episode (Hass et al. 1993). A detailed description of the chemistry transport module is given by Hass (1991). The formulation of the European emission inventory for EURAD is described by Memmesheimer et al. (1990).

Recently, a new generation of the Pennsylvania State University–National Center of Atmospheric Research (PSU–NCAR) Mesoscale Model (MM5) became available that allows multiple nesting, that is, the nesting of up to nine subdomains within a coarse domain down to a nest level 3 (Grell et al. 1994). Thus with the model package MM5 as the meteorological driver of the EURAD model it is possible to simulate the meteorological conditions with interacting nested domains (two-way nesting technique). The PSU–NCAR model package was originally developed by Anthes and Warner (1978) and has been applied to a variety of mesoscale phenomena (Anthes 1990). Zhang et al. (1986) introduced a two-way interactive procedure in the PSU–NCAR model. This was recently improved and extended by implementing a multiple-nesting technique (Grell et al. 1994). In addition, a nonhydrostatic version of MM5 is available (Dudhia 1993).

This new multiple-nesting capability of the meteorological driver is now being used to simulate the transport, chemical transformation, and deposition of atmospheric pollutants with the CTM (the chemistry transport module of the EURAD model). The structure of CTM was modified to also achieve the multiple-nesting approach for the prediction of the concentration fields. This enables the representation of large-scale features together with rather detailed structures by using successively finer grids, telescoping down to smaller scales in the region of interest. Some processes within the CTM operate over a wide range of temporal and spatial scales (e.g., the gas-phase chemistry). Since gas-phase concentrations are represented in an Eulerian model only as grid-box integrals, the subgrid-scale processes must be parameterized to simulate their effects on the resolvable scale. Smaller-scale variations in wind speeds, different emission resolution, different PBL height, etc., certainly change the grid-box average turbulent fluxes. Thus, covariances of subgrid-scale concentrations that result from these processes can affect grid-box average reaction rates.

Another important aspect of grid resolution is the comparison of simulated with observed quantities. Measurements of deposition and concentrations of atmospheric pollutants are usually made at fixed positions at ground level. However, in the model grids only the volume averages of the concentrations are predicted. Thus, for comparability the grid spacing should
not be larger than the length scale of the spatial variability of the concentrations.

The SANA program was established to observe the rapid change in composition of air pollutants and their concentration level over the eastern part of Germany due to political and economical changes (SANA 1993). It is expected that the conditions will alter from a "London smog"–like situation with large amount of sulfur and particulate matter emissions to a more "Los Angeles smog"–like situation, due to rapid increases in car traffic and reduction of sulfur emission. The main task of SANA is to examine the changes in concentration and composition of the trace species and their impact on sensitive ecosystems. Thus, within the SANA program there is a unique chance to observe and control the effects of air quality strategies on a comparable low timescale. Since 1990 one or two special observation periods have been performed yearly including aircraft measurements. In addition, a network of stations continuously measures the airborne concentrations of the major primary and secondary pollutants and their dry and wet deposition.

The second objective of this study is to simulate the processes relevant to air quality from local to continental scales. This task is obviously suited for a model like EURAD exploiting its new ability to telescope down from the European scale to the region of interest, namely eastern Germany, the former German Democratic Republic (GDR). In this paper, results of such an application of the EURAD model are presented focusing on the first special observation episode within SANA, the SANA1 episode, which occurred between 10 and 20 October 1990. Special emphasis was made to the transport of the sulfur compounds, which were still the main atmospheric pollutants during the first phase after the reunification of Germany.

In the next section the EURAD model system is presented and the numerical techniques related to the nesting capability is described and demonstrated. Then the results of the simulation for the SANA1 episode are presented. Finally comparisons are made between model simulations and observations, with special emphasis on the transport from eastern to western Germany.

2. Model description

The EURAD modeling system (Fig. 1) contains three major modules: the PSU–NCAR model MM5, the EURAD emission module (EEM), and the chemistry transport model (CTM). This section describes these modules, as well as describes the major dataflow in more detail.

a. The meteorological model (MM5)

The current state of the PSU–NCAR model MM5 is well described by Grell et al. (1994). By applying the preprocessors of the mesoscale meteorological model MM5, the model domain and the desired projection are selected. The NCAR global dataset of terrain height and land use with a resolution of 5 and 10 min, respectively, is taken for the interpolation to the selected grid. The meteorological initial and boundary conditions are derived from the global ECMWF (European Centre of Medium-Range Weather Forecasts) analysis dataset with a truncation of T106. Its horizontal resolution is approximately 1.68° in latitude and longitude. The data are available four times per day (0000, 0600, 1200, 1800 UTC) on 14 standard pressure levels: 1000, 850, 700, 500, 400, 300, 250, 200, 150, 100, 70, 50, 30, 10 hPa. The operationally analyzed meteorological fields are the geopotential height $Z$, tem-

<table>
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temperature $T$, the horizontal wind components $u$ and $v$, and the relative humidity RH at the mandatory pressure levels, as well as the sea level pressure SLP and the ground temperature $T_g$. After the surface pressure is determined hydrostatically from the SLP and terrain height, all three-dimensional fields are interpolated to the model $\sigma$ (normalized pressure) vertical coordinate, defined as $\sigma = (p - p_i)/p^*$, where $p$ is pressure, $p_i$ is the pressure at the top of the model, $p^* = p_s - p_i$, and $p_s$ is the surface pressure.

For all domains, the physical processes are treated as follows: a high-resolution multilayer PBL formulation (Zhang and Anthes 1982) is used to quantify the vertical transport of momentum, heat, and moisture in the lowest atmospheric layers, where surface fluxes of heat, momentum, and moisture are taken into account. The hydrological cycle is modeled by the explicit moisture scheme (Hsie 1984), in which cloud water and rainwater are explicitly predicted. It should be noted that there is almost no precipitation during the above-mentioned episode in the region of interest. For the coarse domain, a relaxation lateral boundary condition (Davies and Turner 1977) is applied. The formulation of the boundary conditions for the nested domains is discussed later. A four-dimensional data assimilation (FDDA) scheme based on Newtonian relaxation or “nudging” (Stauffer and Seaman 1990) is applied for the coarse domain to minimize the large-scale model error. This allows the model to maintain intervariable consistency and generate realistic mesoscale structures that are not resolved by the global analysis data. It should be noted that the variables $u$, $v$, and $T$ are nudged toward the gridded ECMWF analysis with a nudging coefficient of $3 \times 10^{-4}$ s$^{-1}$ and a time frequency of 6 h. For the nested domain, the FDDA procedure is not applied in order to allow the simulation to develop small-scale structures within the nested domains. All simulations with MM5 are performed in the hydrostatic mode.
FIG. 4. Mean sea level pressure (hPa), solid lines, temperature (°C), dashed lines, and wind vectors in the near surface layer for the ECMWF analysis on 1200 UTC 12 October 1990. Maximum wind speed is 29.52 m s⁻¹.

FIG. 5. As in Fig. 4 but for 1200 UTC 15 October 1990. Maximum wind speed is 22.43 m s⁻¹.
b. The EURAD emission model (EEM)

The input emission database for the EEM used for this study are the EMEP (cooperative program for monitoring and evaluation of the long range transmission of air pollutants in Europe) inventories for NO\textsubscript{x} and SO\textsubscript{2} and for the volatile organic compounds (VOCs) for 1990. The annual emission rates for area sources are provided as low- and high-level sources for NO\textsubscript{x} and SO\textsubscript{2}. The horizontal resolution of the EMEP dataset is 150 km.

The EEM module interpolates to hourly emission rates for the chemistry transport simulations using time functions with seasonal, weekly, and hourly variations. The data are spatially interpolated to the selected grid by using a population density weighting scheme. The whole procedure is described by Memmesheimer et al. (1991). The emission rates of NH\textsubscript{3} are provided following an approach by Asman (1992). In addition, the emission rates of isoprene are taken into account (Lübker and Schöpp 1989). Since the resolution of the original EMEP dataset is very coarse (150 km × 150 km), the emission rates on the nested domains are weighted by the population density.

c. The chemistry transport model (CTM)

The chemistry transport simulation is performed with the CTM version 2 (Hass 1991; Hass et al. 1994), which uses the RADM II chemical mechanism described by Stockwell et al. (1990). For the simulations presented in this study the following algorithms are used: the subgrid-scale turbulent transport in the PBL follows a parameterization based on scaling regimes as described by Hass (1991). The calculation of the horizontal and vertical advection of the atmospheric constituents is performed using the algorithm of Smolarkiewicz (1983). For the treatment of the dry deposition, CTM uses the approach derived by Wesely (1989) to compute the deposition velocities for the atmospheric constituents. The cloud parameterization is the same as in Walcek and Taylor (1986) and Chang et al. (1987).

d. The nesting technique

As mentioned above, the meteorological model MM5 uses a two-way interactive mesh refinement scheme. The nested model domain is user specified. The only limitation is that the boundaries of the nested domain must be at least seven coarse-mesh grid points interior to the coarse domain boundary, and the nesting ratio between coarse grid and nested grid is currently set to 3. A crucial element of the refinement approach is an accurate and efficient interpolation procedure at the boundaries of the nested domain. Since shape preservation and monotonicity are important, the predicted variables are interpolated at the two grid points nearest to the nested domain boundary using the flux-corrected
transport (FCT) scheme. This scheme uses the high-order-accurate, constant-grid-flux dissipative algorithms (Smolarkiewicz and Grell 1992; Tremback et al. 1987). The feedback to the coarser grid is performed at the end of the three fine-mesh time-step integrations; that is, the variable at a coarse grid point is overwritten by the value obtained through a nine-point averaging procedure of nested-gridpoint values.

The nesting procedure in the CTM is performed only in one-way according to a procedure described by Pleim et al. (1991). The boundary conditions of the nested domain differ according to whether the flow is directed into or out of the nested domain. Under inflow conditions, all transported chemical species are interpolated with a one-dimensional linear interpolation along the boundaries of the nested domain. Since the coarse domain model output is hourly, a time interpolation is also performed. The outflow boundary conditions are specified by constant advection of the concentrations over the last two grid cell interfaces in order to eliminate reflections of outgoing waves. The concentration at the boundary will be calculated as follows. The first-order finite-difference form of advection on a staggered grid between grid cells $i$ and $i - 1$ is

$$ADV_i = \frac{u_i (c_i - c_{i-1})}{\Delta x},$$  \hspace{1cm} (1)

where $c_i$ is the concentration in grid cell $i$, and $u_i$ is the wind speed at the interface between grid cell $i - 1$ and $i$. To inhibit reflections, advection into and out of the boundary grid cell are set equal:

$$ADV_{i-1} = ADV_i.$$  \hspace{1cm} (2)

The concentration $c_i$ at the boundary is then specified as

$$c_i = c_{i-1} - \frac{u_{i-1}}{u_i} (c_{i-2} - c_{i-1}).$$  \hspace{1cm} (3)

This method minimizes flux divergence at the boundary while allowing local concentrations to evolve its response to changing conditions.

3. Simulation strategy

For the SANA1 episode the EURAD model has been applied to the following five consecutive periods:

<table>
<thead>
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<th>Period</th>
<th>Start Date</th>
<th>End Date</th>
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<tr>
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<td>10 October</td>
<td>12 October 1990</td>
</tr>
<tr>
<td>P1</td>
<td>12 October</td>
<td>14 October 1990</td>
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<td>14 October</td>
<td>16 October 1990</td>
</tr>
<tr>
<td>P3</td>
<td>16 October</td>
<td>18 October 1990</td>
</tr>
<tr>
<td>P4</td>
<td>18 October</td>
<td>20 October 1990</td>
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where the period P0 is used to provide the initial conditions of the atmospheric constituents based on estimated vertical profiles of each transported species. The spatial structure and nesting configurations for the simulations with the meteorological model MM5 are shown in Fig. 2. Table 1 summarizes the design for the nested configuration. Two separate simulations have been carried out. The control simulation CNTL only runs on a coarse domain without any nesting. While in the simulation NEST, the interaction between the nested grid and the coarser grid is taken into account. The coarse domain (CO or N0), which covers nearly all of Europe, has $63 \times 48$ horizontal grid points with a grid spacing of 80 km. The center of the domain is located at 50°N, 10°E on a Lambert conformal projection.

The first nested domain (N1) covers the region of central Europe with $43 \times 37$ horizontal grid points and a horizontal grid spacing of 26.67 km (one-third of the coarse-domain grid size). The second nested domain (N2) covers the main region of interest, namely the former GDR. It has $55 \times 55$ horizontal grid points with a grid spacing of $8.89$ km (one-third of the first nested domain and one-ninth of the coarse domain). The MM5 model system is integrated over a staggered grid (type Arakawa B) with horizontal momentum, $u$, $v$, defined at the corners of the grid box, and all other variables defined at the center of the grid box. All domains have the same vertical
structure, that is, 15 unequally distributed vertical layers, defined at the following \( \sigma \) levels: 0.050, 0.150, 0.250, 0.350, 0.450, 0.550, 0.650, 0.740, 0.810, 0.865, 0.910, 0.945, 0.970, 0.985, 0.995. The top of the model \( (p_f) \) is 100 hPa.

The CTM uses a slightly different nesting configuration. In order to avoid boundary problems for the nested simulations with CTM, some MM5 grid points at the lateral boundaries are neglected. The selected configuration for CTM is also demonstrated in Table 1. CTM is running on an Arakawa C-type staggered grid, but using the same vertical structure as MM5.

The effect of increasing the horizontal resolution is demonstrated by looking at the terrain height of the different domains compared to the coarse domain. As seen in Fig. 3, the mountainous regions in the nested domain N2 (8.89-km horizontal grid spacing) are well represented (e.g., the Harz Mountains, the Erzgebirge) but are not indicated in the coarse domain (80-km horizontal grid spacing).

4. The SANA1 episode

As mentioned above, the SANA special observation episodes take place once or twice a year, preferably in early spring or late summer, starting in the year 1990. All episodes exhibit similar meteorological conditions: weak pressure gradients together with relatively low wind speeds and almost no precipitation in the region of interest, namely eastern Germany. During the SANA1 episode (10–20 October 1990) aircraft measurements were made on October 14 covering the area around the severe polluted region of Leipzig–Halle–Bitterfeld. First results of these measurements were presented by Schaller et al. (1992). In addition, ground-based measurements of concentration and deposition were made by a SANA network (SANA 1993).

The meteorological situation of the SANA1 episode is characterized by weak pressure gradients over central Europe: at the beginning of the episode (1200 UTC 12 October 1990) a ridge is situated over eastern Europe.
together with a trough over the northeastern Atlantic. Ahead of this trough warm air from the Mediterranean area flows toward central Europe (Fig. 4). Germany is located between the two pressure systems, so that a shift of the ridge leads to a change of the wind direction over eastern Germany from west to southeast. At later times the ridge moves slowly eastward (Fig. 5). A weak frontal system in the west of Ireland follows and reaches Germany on 15 October 1990 (Fig. 5), but no major precipitation occurred in the area of eastern Germany during the whole simulated episode. Wind speeds are relatively low except in frontal zones. At the beginning, in central Europe westerly winds are prevailing. Due to a shift of a high pressure system over eastern Europe in the consecutive time, the wind direction changes several times to a northwesterly direction (13, 15, and 17 October) with intermediate eastward flow.

5. Results

a. Meteorology

Figures 5 and 6 show that the large-scale features of the meteorological fields are well represented by the coarse-domain simulations. The main reason for the good agreement is the use of the four-dimensional data assimilation (FDDA) during the simulation. With regard to the effects of grid resolution and the interaction of the nested domains with the coarse domain, remarkable differences occur, especially in the flow fields. Figure 7 demonstrates this effect, showing the wind vectors in the near surface layer for the simulation CNTL and the simulation NEST in the coarse domain on 1200 UTC 15 October 1990. The flow field without any interaction or feedback from a nested grid exhibits a flow that is directed more to the west than for the simulation with interacting domains in the region of the nested domain N2. Thus, the feedback of the nested-grid wind field is large enough to dominate the forcing due to FDDA on the coarse domain. As will be shown later, this finding will have strong consequences for the transport of the atmospheric pollutants, especially when comparing the simulations to the observations.

In addition, the meteorological variables show much more detailed structure in the nested areas N1 and N2. This effect is demonstrated in Fig. 8 for the vertically integrated cloud water on 1200 UTC 16 October 1990 when a weak front is crossing central Germany. With increasing horizontal resolution there is more pronounced horizontal structure with much greater maxima. Clearly, 80- and 26.67-km grid resolution are insufficient to resolve convective cloud systems that start to become evident on a 8.89-km grid. Grid resolution also greatly affects the simulated wind fields. With increasing horizontal resolution the wind field exhibits a more realistic structure: the flow around the Harz Mountains and the channeling of the wind field in the valley between Erzgebirge and Riesengebirge (see Fig. 10) is enhanced in the simulation on domain N2 but of course cannot be resolved by the coarser-domain simulations.

b. Chemistry and transport

As mentioned above, for the discussion of the results of the chemistry transport module, emphasis is put on the transport of SO2, which is the dominant atmospheric pollutant during that episode. Figure 9 shows the annual sulfur emissions for 1990 based on the EMEP emission inventory and interpolated to the coarse domain used in this study. It should be noted that these emissions represent both high- and low-level sources. The total amount of sulfur emitted in 1990 was 19 400 kilotons (kT). Main maxima are located in the central United Kingdom and in the so-called Black Triangle (the southern part of eastern Germany, northwestern Czechoslovakia, and western Poland), where the grid box with the highest emission is located (387 kT, mainly due to high-level sources). This plot clearly demonstrates the high sulfur emission situation in eastern Germany and the need to study its effects with multiple nesting, especially with respect to the fluxes to the surrounding areas.

The distribution of the atmospheric pollutants is characterized by accumulation over the major high- and low-level emission sources in the southern part of eastern Germany and western Poland during stable nighttime conditions and accompanying low wind speeds. In the early morning hours the wind speed increases and the plumes move out of the emission area and are mixed within the planetary boundary layer.
Figure 10 shows the horizontal distributions of the SO$_2$ concentrations in the surface layer (approximately 35 m above ground level) together with the flow field on 1200 UTC 15 October 1990 for the different domains. For orientation purposes there is still a border line plotted in Germany, although the political reunification was effective at 3 October 1990. As demonstrated in the simulation of the coarse domain (Fig. 10a), transport from the region of greatest emissions (eastern Germany) to the western part of Germany is predominant that time. It is also seen that the pollution in western Germany reaches very high values (greater than 140 ppbv), which is unusually high for western European conditions. The results for the area of the nested domain N1 exhibit a more detailed structure of the near surface SO$_2$ field (Fig. 10b). The center of maximum values over eastern Germany is split into two parts. When looking at the results of the simulation in the nested domain N2 (Fig. 10c), the overall pattern of the SO$_2$ field is quite similar to the simulation on the nested domain N1, but the simulation on domain N2 certainly shows more detail: the main plume over central eastern Germany due to high-level sources is elongated now to parts of northwest Germany, again
accompanied with a splitting of the maximum. Furthermore, the plumes of the emission centers in southern parts of eastern Germany are evident, but they are not resolved in the coarse-domain simulations of domain N0 and are only slightly visible in the simulation of domain N1. The plumes follow more or less the predicted wind fields. It should be noted that the wind field represents the current flow at 1200 UTC 15 October 1990, but the concentration field is a result of the emission scenario and the transport prior to the selected date. The elongation effect of the main plume is due to the fact that the wind speeds in the nested domain N2 are larger than those in the coarser domain (see also Fig. 7). For example, at the point (34, 28) in the coarse domain (the center of the domain N2) the horizontal wind speeds are 5.3, 6.3, and 7.2 m s⁻¹, respectively, in the simulations for the domains N0, N1, and N2.

Figure 11 shows the same field for the simulated domains 1 day later (1200 UTC 16 October 1990). Now a westerly flow is predominant ahead of a weak front that is located over the border between the former
tions in the nested domain N2. Only stations in western Germany are used, because only they are available with a hourly resolution for the SANA1 episode. Stations from eastern Germany so far have only daily averaged values for SO₂. The choice of these stations allows analysis of the effect of transport from the area of highest emissions (source region) to an area of greatest impact (receptor region) in a relatively SO₂-clean atmosphere in western Germany. In this study two stations are selected for a detailed comparison between simulations and observations (the stations are encircled in Fig. 12): Waldhof (WA) is located in the northwest of the domain, and Witzenhausen is located in the southwest of the nested domain N2.

a. Meteorological fields

Figure 13 shows the comparison of the measured and simulated temperature (Fig. 13a), wind direction (Fig. 13b), and horizontal wind speed (Fig. 13c) at Waldhof from 13 to 19 October 1990. Table 2 provides a summary of the measures of performance. The measures are defined as follows: the correlation coefficient is

\[
\text{COR} = \frac{1}{N \sigma_P \sigma_O} \sum_{i=1}^{N} [(P_i - \bar{P})(O_i - \bar{O})];
\]

the root-mean-square error is

\[
\text{rmse} = \left[ \frac{1}{N} \sum_{i=1}^{N} (P_i - O_i)^2 \right]^{1/2};
\]

and the bias is

\[
\text{bias} = \frac{1}{N} \sum_{i=1}^{N} (P_i - O_i).
\]

Terms \(P_i\) and \(O_i\) are the predicted and observed values, respectively, at time \(i\), \(N\) is the total number of observed values at the selected station, and \(\bar{P}\) and \(\bar{O}\) are the average of the predicted and observed values over \(N\). Here \(\sigma_P\) and \(\sigma_O\) are the standard deviation of the predicted and observed values, respectively.

The daily temperature variations are well reproduced by all simulations (see Fig. 13a and the correlation coefficients in Table 2); only at 13 October do the NEST simulations predict too-high temperatures at noon. There is an overall tendency that the simulation CNTL predicts relatively too low temperatures (negative bias) compared to simulation NEST. In general, the NEST simulations exhibit the smallest forecast error. This figure also demonstrates that all simulations predict well the weak cold front passage at 17 October 1990.

An important aspect for transport calculations is the right prediction of the wind direction. As seen in Fig. 13b, the wind direction is also well predicted for all simulations compared to observa-

East and West Germany. This front was well reproduced in all simulations, as seen from strong gradients in the SO₂ concentrations associated with strong reduction of SO₂. Note that the maximum values of SO₂ reach only 50 ppbv. This figure again demonstrates the effect of telescoping the simulations from a coarse domain down to the nested domain N2. The plumes again exhibit a more detailed structure with increasing horizontal resolution. This effect is well demonstrated in the simulation of the nested domain N2 with the plumes of the major cities (emission centers) in southern East Germany. But at that time the area of greatest impact is the western part of Poland.

As mentioned earlier in this paper, main emphasis was put on the transport of the sulfur compounds. It should be noted that the CTM of EURAD predicts not only SO₂ but also higher reactive gases such as ozone for this episode. A detailed description of the effect of multiple nesting on these gases and on the deposition of the trace gases will be a subject of a further investigation.

6. Comparison with observations

In order to evaluate the results of the simulations with the EURAD model, measurement data were taken from the German Federal Environmental Agency (Umweltbundesamt, UBA) and from regional networks of the German federal states, namely Hessen and Schleswig-Holstein. Available data are SO₂, NOₓ, and ozone. Figure 12 shows the location of these sta-
The control simulation CNTL (with a coarse domain only) obtains best performance for both wind speed and wind direction. This figure indicates the episodes of high-pollution events in western Germany with predominant easterly flow on 13, 15, and 17 October 1990. (The wind direction is less than 180°.) These events are clearly exhibited in Fig. 14a.

The comparison of predicted wind speeds with observation does not provide the same good performance as for temperature and wind direction. The simulation CNTL has the lowest rmse but predicts too-low wind speeds compared to the NEST simulations and observation (negative bias). There is a negative correlation for all simulation under stable nighttime conditions at 13–14, 16–17, and 18–19 October 1990. The best correlation is seen for the NEST simulations at 15 and 17 October 1990, when Waldhof is affected by severe pollution with associated easterly winds (Figs. 13b and 14a).
b. $SO_2$

Figure 14a shows the comparison of measured with simulated concentrations of $SO_2$ at the station Waldhof from 13 to 19 October 1990. As already seen in Fig. 13b, the data indicate three major periods of easterly flow from the high emission regions that coincide with periods of high pollution: during these periods, at 13, 15, and 17 October, very high concentrations (almost 200 ppbv) of $SO_2$ are observed. Between these events the concentrations are reduced to the local background values. The event at 15 October exhibits a double-peak structure. The simulated concentrations of $SO_2$ for the control simulation CNTL, in general, exhibit too high values especially at times with low measured concentrations. The observed double-peak structure at 15 October could not be resolved. This pattern is probably due to the relatively coarse resolution of emissions and the effect of numerical diffusion. The predicted concentrations of $SO_2$ for the simulations with interaction (NEST) all show nearly the same pattern for measured background concentrations lower than 50 ppbv (13–14, 16, and after 18 October), but they show great differences at the time of maximum observed concentrations (15 October). The simulation NEST on domain N0 gives a strong reduction of the $SO_2$ concentrations compared to the control simulation. An indication of a double-peak structure is seen for the simulation on domain N1, which is strengthened for the simulation on domain N2, where the simulation agrees best with observations. This is a remarkable finding considering the relatively narrow plume of $SO_2$ concentrations transported from eastern Germany to Waldhof (see Fig. 10c). Thus, the simulation with highest horizontal resolution fits best with observation during high pollution events.

The same comparison was made for the station Witzenhausen, which was affected by the transport from eastern Germany mainly at 17 October 1990 (see Fig. 14b). Maximum measured values (approximately 120 ppbv) are reached at that date as a result of a flow to southwest from the main emission centers in the southern part of East Germany. The predicted relatively high $SO_2$ concentrations for the control simulation CNTL, as compared with observations, are evident. The same effect of the simulations with interactive nested domains is seen for this station: the best agreement with observations occur in the simulation with highest horizontal resolution (for nested domain N2). This comparison again demonstrates the importance of using high-resolution nested domains where the flow fields of the nested domain interacts with the coarser domain. A simulation with a conventional regional atmospheric transport modeling system, which is running with a grid spacing of around 80 km, would have significant errors in predicting the concentrations of the main transported species as $SO_2$ in the near range of the emission sources. The simulations presented in this paper demonstrate very well that an increase of the horizontal resolution in a certain area of interest can reduce these errors significantly.

To further support this finding and to demonstrate that this fact is not restricted to the two presented stations, Fig. 14c shows the time series of the average over all 13 sites shown in Fig. 12, both measured and simulated. In this plot, the extreme overprediction of the control simulation is obvious. It demonstrates that for the simulations with interactive nested grids, an ex-
Fig. 14. Observed and simulated concentrations of SO₂ (ppbv) for the time 13–19 October 1990: (a) station Waldhof and (b) station Witzenhausen, and (c) the average for all 13 sites as shown in Fig. 12. The lines correspond to the simulations as shown within the figure.

tremely good agreement between observation and simulation is found for low SO₂ concentration, with the best reproduction of the measurements from 13 to 14 October and less correlation but also relative good agreement from 18 to 20 October. The first severe pollution event on 15 October is observed and simulated mainly at the northern stations, whereas the second peak on 17 October is only seen at the southern stations. The simulation on the nested domain N2 best reproduces the first event at the northern sites. The same is true for the southern sites at 17 October, but the arrival time of that plume is about six hours too early. The reason for that shift may be due to the incorrect locations of the emission sources, which are only interpolated from a 150 km × 150 km grid with population density weighting. A more correct place-
Fig. 15. Distribution of the bias (calculated minus observed) for SO$_2$ (ppbv). The lines correspond to the simulations as shown in the figure.

ment of the point sources due to power plants that are remote from the big cities will probably improve the results.

In Fig. 15 the distribution of the bias [predicted-minus-observed SO$_2$ concentrations, see Eq. (6)] for all mentioned sites and hours of the different simulations is presented. Positive bias means overprediction, negative values correspond to underprediction. This diagram demonstrates the effect of overprediction in the control simulation exhibited by the asymmetric structure of the distribution. This structure is also seen for the simulation on the coarse domain with nested grid interaction, but there is already a tendency toward a more symmetric pattern. With increasing horizontal resolution, the distribution of the bias becomes more and more symmetric with the best pattern for the simulation on the nested domain N2.

The performance of the model simulation is also demonstrated in a frequency distribution of the SO$_2$ concentration for different concentration classes (Fig. 16). Nearly 50% of the measured values for that episode are less than 10 ppbv, but the control simulation CNTL has only 22% below that background value of 10 ppbv. For this class a significant tendency is seen that a better representation of the background concentrations is reached with increased horizontal resolution. The same tendency is seen regarding higher concentration classes, but a reverse effect is evident. For example, about 1.2% of the measured values range between 60 and 70 ppbv, but the control simulation predicts about 4% in this range. With increasing horizontal resolution, a decrease of the percentage close to the observation is pronounced.

Table 3 summarizes these previous results in calculating the maximum, the average, and the median of the different simulations for the SO$_2$ concentrations at all 13 sites. The calculations are separated for northern and southern sites because the maximum concentrations correspond to different events: the northern sites at 15 October and the southern sites at 17 October (see Fig. 14). The tendency due to the nesting effect is well demonstrated for all measures, especially for the median, which can be interpreted as a measure for the background concentrations. The predicted maximum concentration increases from 90 (N0) to 160 ppbv (N2), compared to measured 196 ppbv for the northern sites. The median decreases with increasing horizontal resolution (13 ppbv for domain N0 to 9 ppbv for domain N2) and nearly reaches the observed value of 11 ppbv. Both effects are typical for Eulerian models: due to the large volume of the coarse grid boxes, the high and low end of the concentration distribution are biased. The same pattern is more evident for the statistical measures at the southern sites. Thus, the nesting facility enhances the performance of the model to reproduce the near surface fluxes in a distance of 100–500 km away from the main source region of SO$_2$. It should be noted here that the coarse-domain simulation is still needed for the prediction of the synoptic-scale variations.

When comparing the simulated with the observed quantities, the question of representativeness of simulated concentrations should be addressed. Measure-
Table 3. Statistical measures for the SO$_2$ concentrations.

<table>
<thead>
<tr>
<th>Experiment name</th>
<th>Domain name</th>
<th>Grid spacing (km)</th>
<th>Maximum (ppbv)</th>
<th>Median (ppbv)</th>
<th>Average (ppbv)</th>
</tr>
</thead>
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<tr>
<td>Northern stations</td>
<td></td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CNTL</td>
<td>CO</td>
<td>80.00</td>
<td>143</td>
<td>21</td>
<td>37</td>
</tr>
<tr>
<td>NEST</td>
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<td>80.00</td>
<td>90</td>
<td>13</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>N1</td>
<td>26.67</td>
<td>135</td>
<td>12</td>
<td>22</td>
</tr>
<tr>
<td></td>
<td>N2</td>
<td>8.89</td>
<td>160</td>
<td>9</td>
<td>21</td>
</tr>
<tr>
<td>Observation</td>
<td></td>
<td></td>
<td>196</td>
<td>11</td>
<td>18</td>
</tr>
<tr>
<td>Southern stations</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
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<td></td>
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<td></td>
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<td>10</td>
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</tbody>
</table>

ments of deposition and concentrations of atmospheric pollutants are usually made at fixed positions at ground level, but in the grids of the EURAD model only the volume averages of the concentrations are predicted. Using the multiple-nesting approach one grid box in the coarse domain with 80 km x 80 km spacing contains nine grid boxes in the nested domain N1 and 81 grid boxes in the nested domain N2. Thus, for comparison of simulated with measured concentrations, it is obvious that with increasing grid resolution the concentrations in these grid boxes are more comparable with observations at fixed locations. This effect is demonstrated in Fig. 17. Figure 17a shows the simulated SO$_2$ concentrations in the coarse grid box (32, 28) where Waldhof is located for the simulation in the 81 grid boxes of nested domain N2 at 0800 UTC 15 October 1990 when the maximum SO$_2$ concentration is observed. The coarse-grid value for this box is 61.99 ppbv, but the simulation in the nested domain N2 predicts 109 ppbv, compared to observed 196 ppbv. The average value for the nested domain N2 in the coarse grid box is 95.43 ppbv with a relatively high variability (standard deviation is 19.8 ppbv). In Fig. 17b the simulated SO$_2$ concentrations for the nested domain N1 is presented at nine grid boxes within the coarse-domain grid box (32, 28). The average and the variability is less than for the simulation in the nested domain N2 (82.06 ppbv and the standard deviation of 14.55 ppbv). The value corresponding to the station Waldhof is only 84 ppbv. This figure also shows that the simulation in the nested domain N2 can produce a relatively narrow plume with a width of approximately 20 km, which is not seen in the simulation of nested domain N1 and of course not resolvable in the coarse-domain grid box.

7. Summary and conclusions

A multiple-nesting version of the EURAD model has been developed in order to increase the horizontal resolution in a region of enhanced pollution, namely the former German Democratic Republic (GDR). This new technique allows the ability to simulate large-scale features and at the same time to study the development of smaller-scale structures in the nested regions. Two nested areas have been embedded in a coarse domain that covers the main parts of Europe, where the second nested domain is embedded in the first nested domain. The nesting is two-way for the meteorological part and one-way for the chemistry transport module of the EURAD model. That means that the meteorological variables at the boundaries in the nested domain are interpolated by a monotone flux-corrected transport method from the next coarse domain and create a feedback for the coarser domain. For the CTM the inflow boundary conditions are dynamically determined by interpolation from coarse-grid results, the outflow conditions are specified by continuous advection at the boundary in order to eliminate numerical reflections.

This multiple-nesting approach was applied for a case study in October 1990, the so-called SANA1 episode. This episode is dominated by weak pressure gradients over central Europe, associated with low wind speeds and little precipitation. The results of the simulations indicate a strong dependence of the horizontal structure of the fields; both meteorological and atmospheric constituents exhibit the most realistic structure on increased horizontal resolution. The simulations in the nested domain with highest resolution (domain N2 with 8.89-km horizontal grid distance) exhibit the most realistic structure. At the highest resolution the meteorological model MM5 predicted the temperature the best compared to measurements but showed less agreement with observation for wind direction and wind speed. Comparisons for the SO$_2$ concentrations with observations from selected stations gave a strong indication that the simulations agree best with observations in the nested domain N2, especially for the background concentrations. This finding is supported by statistical analysis for all 13 sites that are available and common for all domains. It was also found that the interaction of the meteorological variables, especially the wind fields between each domain (two-way nesting approach) has a strong influence for the prediction of the
atmospheric pollutants. Without interaction or feedback from the nested domain variables, the different prediction of the coarse-domain meteorological variables, especially the wind field, will lead to different prediction of the trace gas concentrations. This leads to the conclusion that the resolution of emissions and the numerical diffusion are probably more important. This finding is only valid when comparing the simulations with station observation in the near range of the main emission sources. Evaluation studies show that the EURAD model, when running on a domain with an approximate 80-km grid size, is well suitable to simulate the transport, chemical transformation, and deposition in the far range (Hass et al. 1990; Hass et al. 1993).

Although the simulations with highest horizontal resolution agree very well with observations, there are still some discrepancies between predicted and observed chemical species, for example, the arrival time of the species SO₂ is not always in accordance with observations for the major pollution events. This is possibly due to the fact that the meteorological model predicts the wind direction very well, but the wind speeds do not have the same accordance with observation, especially under stable nighttime conditions. Another reason for this effect could be the inappropriate emission inventory used for this study. The EMEP emission inventory based on a 150 km × 150 km resolution is interpolated to the selected grid by using a population density weighting. Thus, the major cities are the main emission centers in the high-pollutant area of eastern Germany. A more sophisticated emission inventory with the inclusion of the real point sources (power plants with high SO₂ emissions are usually remote from the cities) will probably lead to a more realistic distribution of the atmospheric constituents, especially for SO₂. Within the SANA project it is planned to create a much more detailed emission inventory for the region of eastern Germany. Thus, additional simulations are necessary to study the sensitivity of the transport of atmospheric constituents due to a choice of improved emission scenario.

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