The Simulation of Photochemical Smog Episodes in Hungary and Central Europe Using Adaptive Gridding Models

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Abstract. An important tool in the management of photochemical smog episodes is a computational model which can be used to test the effect of possible emission control strategies. High spatial resolution of such a model is important to reduce the impact of numerical errors on predictions and to allow better comparison of the model with experimental data during validation. This paper therefore presents the development of an adaptive grid model for the Central European Region describing the formation of photochemical oxidants based on unstructured grids. Using adaptive methods, grid resolutions of less than 20 km can be achieved in a computationally effective way. Initial simulation of the photochemical episode of August 1998 indicates that the model captures the spatial and temporal tendencies of ozone production and demonstrates the effectiveness of adaptive methods for achieving high resolution model predictions.

1 Introduction

Previous EUROTRAC investigations have shown that some of the highest regional ozone concentrations in Europe can be observed in Central Europe, including Hungary. During summer ozone episodes, the ozone burden of natural and agricultural vegetation is often well beyond tolerable levels. Budapest is one of the biggest cities in this region, emitting significant amounts of ozone precursor substances. An important tool in the management of photochemical smog episodes is a computational model which can be used to test the effect of possible emission control strategies. High spatial resolution of such a model is important to reduce the impact of numerical errors on predictions and to allow better comparison of the model with experimental data during validation. The review paper of Peters et al.[1] highlights the importance of developing more efficient grid systems for the next generation of air pollution models in order to "capture important smaller scale atmospheric phenomena". This paper therefore presents the development of an adaptive grid model for the Central European Region describing

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the formation of photochemical oxidants based on unstructured grids. The initial base grid of the model uses a nested approach with a coarse grid covering the wider central European region and a finer resolution grid over Hungary. Further refinement or de-refinement is then invoked using indicators based on the comparison of high and low order numerical solution of the atmospheric diffusion equation. Using this method, grid resolutions of less than 20 km can be achieved in a computationally effective way.

2 Model Description

The model describes the spread of reactive air pollutants within a 2D unstructured triangular based grid representing layers within the troposphere over the Central European region including Hungary. The model describes the horizontal domain using a Cartesian coordinate system through the stereographic polar projection of a curved surface onto a flat plane. The total horizontal domain size is 1540 km × 1500 km. Vertical resolution of pollutants is approximated by the application of four layers representing the surface, mixing, reservoir layers and the free troposphere. Reactive dispersion in the horizontal domain is described by the atmospheric diffusion equation in two space dimensions:

$$\frac{\partial c_s}{\partial t} = -\frac{\partial (uc_s)}{\partial x} - \frac{\partial (wc_s)}{\partial y} + \frac{\partial}{\partial x}\left(K_x \frac{\partial c_s}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial c_s}{\partial y}\right) + R_s(c_1, c_2, ..., c_q) + E_s - (\kappa_{1s} + \kappa_{2s})c_s, \quad (1)$$

where c_s is the concentration of the s'th compound, u,w, are horizontal wind velocities, K_x and K_y are eddy diffusivity coefficients and κ_{1s} and κ_{2s} are dry and wet deposition velocities respectively. E_s describes the distribution of emission sources for the s'th compound and R_s is the chemical reaction term which may contain nonlinear terms in c_s. For n chemical species an n-dimensional set of partial differential equations (p.d.e.s) is formed describing the rates of change of species concentration over time and space, where each is coupled through the nonlinear chemical reaction terms.

The four vertical layers of the model are defined as; the surface layer extending to 50m, the mixing layer, a reservoir layer and the free troposphere (upper) layer. The mixing layer extends to a height determined by radiosonde data at 12am, but is modelled to rise smoothly to a height determined by radiosonde data at 12pm during the day[2]. The reservoir layer, if it exists above the height of the mixing layer, extends from the top of the mixing layer to an altitude of 1000 m. Vertical mixing and deposition are parameterised according to the vertical stratification presented by VanLoon[3]. Deposition velocities are assumed to be constant across the whole domain. The eddy diffusivity coefficients for the x and y directions were set to 50 m^2s^{-1} for all species.

Relative humidity and temperature data were determined by the meteorological model ALADIN with a time resolution of 6 hours and spatial resolution of 0.1 × 0.15 degrees[4]. The local wind speed and direction were obtained from the

ECMWF database ensuring conservation properties of the original data[5]. The ECMWF data has a time resolution of 6 hours and a spatial resolution of 2.5 $\times$ 2.5 degrees. These data were interpolated to obtain data relevant to a given space and time point on the unstructured grid using conservative methods as described in Ghorai et al.[6].

The emission of precursor species into the domain was described by the EMEP emissions inventory for 1997 based on a 50km $\times$ 50km grid[7]. The emissions data have to be interpolated onto the unstructured mesh following each change to the mesh during refinement. This is achieved using the mass conservative method of overlapping triangles. The EMEP data is therefore split into triangular grid cells and the proportion of each EMEP triangle that overlaps each mesh triangle calculated. Point sources, although not included in the current simulations, can be included in the model by averaging them into the appropriate triangle. It follows that as the mesh resolution is improved, the description of point source emissions will also improve. During the initial simulations as described here the GRS chemical scheme was used[8] to enable fast turn around times. Photolysis rate constants have been chosen to be in agreement with those used by Derwent and Jenkin [9] and are expressed as m'th order rate constants with units (molecule $cm^{-3})^{1-m}$ s^{-1}. The photolysis rates were parameterised by the function J_q=$a_q$exp(-b_q sec θ), where θ is the solar zenith angle and q is the reaction number. The solar zenith angle is calculated from:

cos θ = cos(lha) cos(dec) cos(lat) + sin(dec) sin(lat),

where lha is the local hour angle (i.e. a function of the time of day), dec is the solar declination (i.e. a function of the time of year) and lat is the latitude. Temperature dependent rate constants are represented by standard Arrhenius expressions.

3 Solution Method

The basis of the numerical method is the space discretisation of the p.d.e.s derived from the atmospheric diffusion equation on unstructured triangular meshes using the software SPRINT2D[10,11,12]. This approach, (known as the "Method of Lines"), reduces the set of p.d.e.s in three independent variables to a system of ordinary differential equations (o.d.e.s) in one independent variable, time. The system of o.d.e.s can then be solved as an initial value problem, and a variety of powerful software tools exist for this purpose[13]. For advection dominated problems it is important to choose a discretisation scheme which preserves the physical range of the solution. A more in-depth discussion of the methods can be found in previous references[14,10,11,12].

3.1 Spatial Discretisation and Time Integration

Unstructured triangular meshes are commonly used in finite volume/element applications because of their ability to deal with general geometries. In terms of

application to multi-scale atmospheric problems, we are not dealing with complex physical geometries, but unstructured meshes provide a good method of resolving the complex structures formed by the interaction of chemistry and flow in the atmosphere and by the varying types of emission sources. The term unstructured represents the fact that each node in the mesh may be surrounded by any number of triangles whereas in a structured mesh this number would be fixed. In the present work, a flux limited, cell-centered, finite volume discretization scheme of Berzins and Ware[10,11] was chosen on an unstructured triangular mesh. This method enables accurate solutions to be determined for both smooth and discontinuous flows by making use of the local Riemann solver flux techniques (originally developed for the Euler equations) for the advective parts of the fluxes, and centered schemes for the diffusive part. The scheme used for the treatment of the advective terms is an extension to irregular triangular meshes of the nonlinear scheme described by Spekreijse[15] for regular Cartesian meshes. The scheme of Berzins and Ware has the desirable properties, see Chock[16], of preserving positivity eliminating spurious oscillations and restricting the amount of diffusion by the use of a nonlinear limiter function. The advection scheme has been shown to be of second order accuracy[11,17]. For details of the application of the method to the atmospheric diffusion equation in two-space dimensions see Tomlin et al.[12]. The diffusion terms are discretised by using a finite volume approach to reduce the integrals of second derivatives to the evaluation of first derivatives at the midpoints of edges. These first derivatives are then evaluated by differentiating a bilinear interpolant based on four mid-point values[10]. The boundary conditions are implemented by including them in the definitions of the advective and diffusive fluxes at the boundary.

A method of lines approach with the above spatial discretization scheme results in a system of o.d.e.s in time which are integrated using the code SPRINT[13] with the Theta option which is specially designed for the solution of stiff systems with moderate accuracy and automatic control of the local error in time. Operator splitting is carried out at the level of the nonlinear equations formed from the method of lines by approximating the Jacobian matrix, and the method is described fully in Tomlin et al.[12]. The approach introduces a second-order splitting error but fortunately this error only alters the rate of convergence of the iteration as the residual being reduced is still that of the full o.d.e. system. This provides significant advantages over other splitting routines such as Strang splitting.

3.2 Mesh Generation and Adaptivity

The initial unstructured meshes used in SPRINT2D are created from a geometry description using the Geompack mesh generator[18]. These meshes are then refined and coarsened by the Triad adaptivity module which uses tree like data structures to enable efficient mesh adaptation by providing the necessary connectivity. A method of refinement based on the regular subdivision of triangles has been chosen. Here an original triangle is split into four similar triangles by connecting the midpoints of the edges as shown in Fig. 1. These may

later be coalesced into the parent triangle when coarsening the mesh. This process is called local h-refinement, since the nodes of the original mesh do not move and we are simply subdividing the original elements. In order to implement the

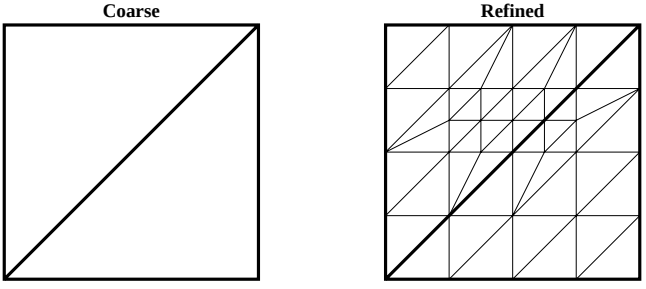

Fig. 1. Method of local refinement based on the subdivision of triangles

adaptivity module a suitable criterion must be chosen. The ideal situation would be that the decision to refine or derefine would be made on a fully automatic basis with no user input necessary. In practice a combination of an automatic technique and some knowledge of the physical properties of the system is used. The technique used in this work is based on the calculation of spatial error estimates. Low and high order solutions are obtained for each species and the difference between them gives a measure of the spatial error. The algorithm can then choose to refine in regions of high spatial error by comparison with a user defined tolerance for one or the sum of several species. For the ith p.d.e. component on the jth triangle, a local error estimate $e_{i,j}(t)$ is calculated from the difference between the solution using a first order method and that using a second order method. For time dependent p.d.e.s this estimate shows how the spatial error grows locally over a time step. A refinement indicator for the jth triangle is defined by an average scaled error ($serr_j$) measurement over all $npde$ p.d.e.s using supplied absolute and relative tolerances:

$$serr_j = \sum_{i=1}^{npde} \frac{e_{i,j}(t)}{atol_i/A_j + rtol_i \times C_{i,j}}, \tag{2}$$

where $atol$ and $rtol$ are the absolute and relative error tolerances and $C_{i,j}$ the computed concentration of species i in triangle j. This formulation for the scaled error provides a flexible way to weight the refinement towards any p.d.e. error. In these calculations a combination of errors in species NO and NO_2 were used as refinement indicators.

An integer refinement level indicator is calculated from this scaled error to give the number of times the triangle should be refined or derefined. Since the error estimate is applied at the end of a time-step it is too late to make the refinement decision. Methods are therefore used for the prediction of the growth

of the spatial error using linear or quadratic interpolants. The decision about whether to refine a triangle is based on these predictions, and the estimate made at the end of the time- step can be used to predict errors at future time-steps. Generally it is found that large spatial errors coincide with regions of steep spatial gradients. The spatial error estimate can also be used to indicate when the solution is being solved too accurately and can indicate which regions can be coarsened. The tree data structure can then be used to restore a lower level mesh which maintains the triangle quality. In order to minimise the overhead involved in calculating the spatial error and interpolating the input data and concentrations onto the new mesh, refinement indicators are not calculated at each time point but at intermediate points each 20 minutes of simulation time. The errors induced are small in comparison with the computational savings made[19]. If point sources are used it is also important to set a maximum level of refinement in order to prevent the code from adapting to too high a level in regions with concentrated emissions where high spatial gradients may persist down to very high levels of refinement.

4 Simulation of Photochemical Smog Episode

The model was tested via the simulation of a photochemical oxidant episode that took place in Hungary in August, 1998. During almost the whole month wind speeds were low limiting dispersion, and strong sunshine resulted in high photo-oxidant levels over most of Europe. The simulation period was from midnight 1 August 1998 to midnight 5th August 1998. Both fixed and adaptive mesh simulations were carried out in order to investigate the influence of mesh resolution on predicted concentrations. The coarse level 0 grid covered Central Europe as seen in Fig. 2 and is defined by a maximum edge length of 228 km. For

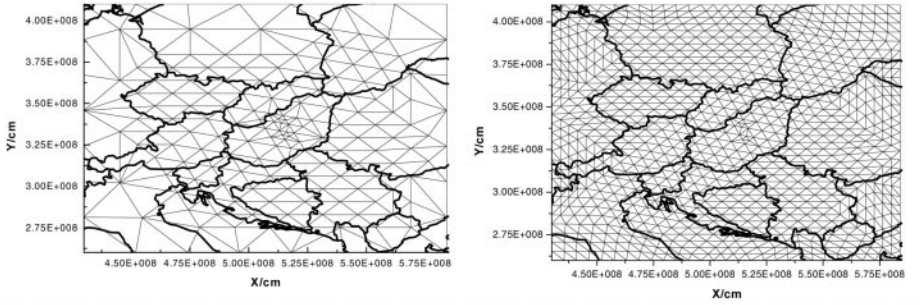

Fig. 2. The structure of level 0 and level 2 base meshes

comparison, simulations where also carried out using a level 2 refined base grid as shown in Fig. 2 with a maximum edge length of 56km. For both simulations extra mesh refinement to 3 levels (edge length 28km) was carried out around the K-puszta monitoring station of the Hungarian Meteorological Service, located 70 km south-southeast from Budapest, in order to carry out comparisons of measured and simulated ozone concentrations. The initial conditions for the most important species were as a follows: 1.9 ppb for NO_2, 3 ppb for NO, ppb 20.3 O_3, 4 ppb for VOC and were constant across the domain. A certain amount of spin up time was therefore required for the model in order to eliminate the impact of the initial conditions. Fig. 3 shows the simulated ozone concentrations after 4 days of simulation on 5th August, 1998 at 15.00 for the two base grids where no transient refinement has taken place. The impact of mesh resolution can be seen and the higher resolution simulation predicts higher peak concentrations over the region South of Hungary.

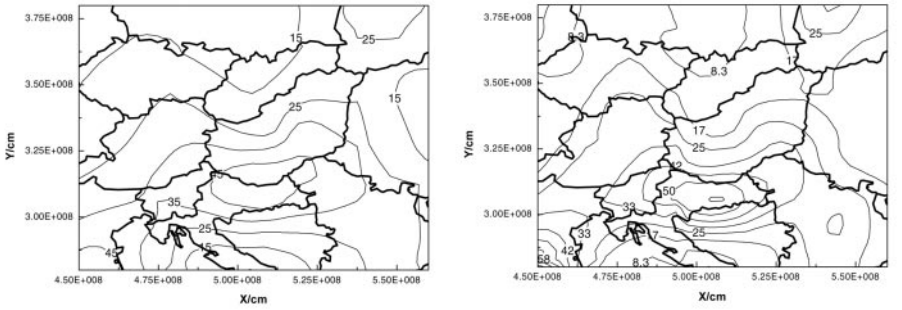

Fig. 3. Calculated ozone concentration after 4 days of simulation on 5th August, 1998 at 15.00, level 0 vs level 2 results

An adaptive simulation was also carried out where transient refinement was limited to 4 levels below the base mesh, in this case leading to a minimum edge length of approximately 20 km. Fig. 4 shows the grid at 18.00 on the third simulation day where refinement has effectively taken place around regions of high gradients in NO and NO_2 concentrations. The refinement region is seen to cover a large part of Hungary and the surrounding region where steep concentration gradients occur. The simulated ozone concentrations for a level 2 base mesh and the level 4 adaptive mesh shown in Fig. 4 at the simulated time of 18.00 on 5th August, 1998 are shown in Fig. 5. The high ozone concentration area is found to be in the Western region of Hungary where the grid is also dense for the adaptive solution, and to Southwest of Hungary. There are significant differences in the predicted peak ozone concentrations for the base mesh and the adaptive simu-

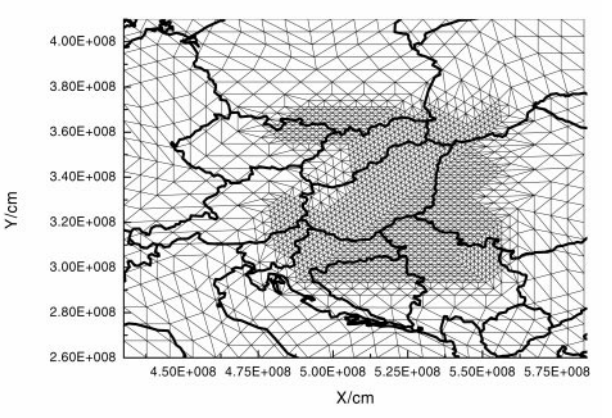

Fig. 4. The adaptive mesh at the simulated time of 18.00 on 5th August, 1998

lation with the peak not rising above 50ppb for the coarse mesh and reaching 90ppb in the South West region of the mesh for the adaptive solution.

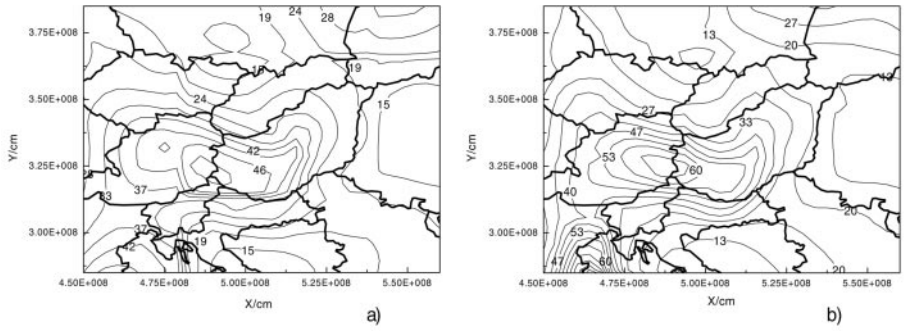

Fig. 5. Comparison of ozone predictions , at the simulated time of 18.00 on 5th August, 1998 using a) a level 2 base mesh and b) a level 4 adaptive mesh

Fig. 6 shows the simulated and measured ozone concentrations at the K-puszta station as a function of time during the simulation. As expected the initial comparisons are poor during the model spin up period reflecting the need for more accurate initial conditions. During the latter part of the simulation the simulated ozone levels increase but are still considerably lower than the ozone levels measured at K-puszta. There are several possible reasons for this. The meteorological data used for the simulations was quite coarse and therefore the region of predicted high ozone levels may be spatially inaccurate as a result. High ozone concentrations are predicted in the Southwest region of the mesh but lower concentrations in the area of K-puszta. It is also clear that the assumption

of constant initial conditions across the mesh is not accurate and affects the early part of the simulation. The amount of ozone generated over the 5 days is quite well represented by the model, again leading to the requirement for better initial conditions. The emissions data is also based on a fairly coarse grid. Future improvements to the model will therefore be to include higher spatial resolution meteorological data and emissions data for the Budapest region. The incorporation of measured concentrations for initial conditions where possible would also improve the simulations.

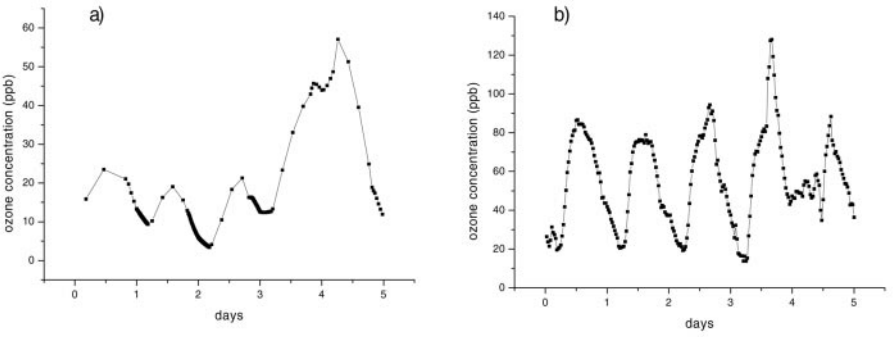

Fig. 6. Comparison of a) Model predictions with b) measured ozone concentrations at the K-puszta station

5 Conclusions

An adaptive grid model describing the formation and transformation of photochemical oxidants based on triangular unstructured grids has been developed for the Central European Region, which includes Hungary. The model automatically places a finer resolution grid in regions characterised by high concentration gradients and therefore by higher numerical error. Using an adaptive method it is therefore possible to achieve grid resolutions of the order of 20 km without excessive computational effort. Initial simulations of the photochemical episode of August 1998 indicate that the model under predicts ozone concentrations, but captures the spatial and temporal tendencies. The comparison of level 0 and level 2 base mesh simulations with a level 4 adaptive simulation demonstrates the impact of mesh refinement on predicted ozone concentrations and indicates that using a coarse mesh may lead to underpredictions of peak values.

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