Modelling Photochemical Air Pollution in Hungary Using an Adaptive Grid Model

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Abstract. An adaptive grid model, describing the formation of photochemical oxidants based on triangular unstructured grids, has been developed for the Central European Region. The model automatically places a finer resolution grid in regions were higher numerical error is predicted by the comparison of 1st and 2nd order solutions. Using this method, grid resolutions of the order of 15 km could be achieved in a computationally effective way. Initial simulation of the photochemical episode of August 1998 indicate that the model captures well the spatial and temporal tendencies of ozone production.

I. Introduction

Previous EUROTRAC investigations showed that some of the highest regional ozone concentrations in Europe can be observed in Central Europe, including Hungary. During the summer ozone episodes, the ozone burden of natural and agricultural vegetation is well beyond the tolerable level. Budapest, the capital of Hungary, is one of the biggest cities in this region, emitting significant amount of ozone precursor substances. An important tool in the management of ozone problems is a computational model, which can be used to test the effect of possible emission control strategies on the ozone burden. The proper resolution of such a model is important to reduce the impact of numerical errors on the solution and to allow better comparison of the model with experimental data during validation. This paper therefore presents the development of an adaptive grid model originating at the University of Leeds, UK, [1, 2, 3] for application to photochemical air pollution in Hungary. New meteorological sub-models have been incorporated and emissions and meteorological data for Hungary included. The paper reports the first simulation results with the new model.

II. The 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. Vertical resolution of pollutants is approximated by the application of four layers representing the surface, mixing, reservoir layers and the free troposphere. The grid is adaptive, *i.e.* continuously changes in space and time to minimize numerical error. Transformation of pollutants is described within each grid cell by a detailed gas phase chemical mechanism. The wider European region is simulated using a coarse grid, while the area of interest surrounding Hungary is subject to initial refinement to a fine resolution sub-grid. This approach allows high spatial resolution for the area of interest, and also realistic boundary conditions to be simulated for this region. Transient refinement and de-refinement is then further invoked as necessary throughout the model run according to spatial errors and chosen refinement criteria.

II. 1. Model equations

The atmospheric transport-diffusion equation in two space dimensions is given by:

$$\frac{\partial c_i}{\partial t} = -\frac{\partial (uc_i)}{\partial x} - \frac{\partial (vc_i)}{\partial y} + \frac{\partial}{\partial x}(K_x \frac{\partial c_i}{\partial x}) + \frac{\partial}{\partial y}(K_y \frac{\partial c_i}{\partial y}) + R_i(c_1, c_2, ..., c_n) + \tag{1}$$

$$+ E_i - (k_{1i} + k_{2i})c_i$$

where c_i is the concentration of species i; u, v, are components of horizontal wind velocity, K_x and K_y are turbulent diffusivity coefficients and k_{1i} and k_{2i} are dry and wet deposition coefficients, respectively. E_i describes the distribution of emission sources for the ith compound and R_i is the chemical reaction term, which may contain non-linear terms in c_i. For n chemical species an n-dimensional set of partial differential equations (PDEs) is formed describing the rates of change of species concentration over time and space, where the equations are coupled through the non-linear chemical reaction terms.

II. 2. Chemical mechanisms

Two chemical schemes have been utilised. The first is a simplified mechanism introduced by Azzi and Johnson [4] and entitled the Generic Reaction Set. This mechanism represents only 7 species interacting via 7 reactions. The GRS is a small, simplistic description of tropospheric NO_x chemistry enabling fast turn around times during code development. The second scheme is the considerably larger CBM-Leeds model, which provides a more detailed representation of tropospheric chemistry. This reaction set contains 23 species and 59 reactions [5].

The rate constants have been chosen to be in agreement with those used by Derwent and Jenkin [6] and are expressed as mth order rate constants with units (molecule $cm^{-3})^{1-m}$ s^{-1}. Temperature dependent rate constants are represented by standard Arrhenius expressions. The photolysis rates were parameterised by the following function:

$$J_q = a_q \exp(-b_q \sec \Theta), \tag{2}$$

where Θ is the solar zenith angle and q is the reaction number. The solar zenith angle is calculated by

$$\cos \Theta = \cos(lha)\cos(dec)\cos(lat) + \sin(dec)\sin(lat), \tag{3}$$

where lha is the local hour angle (function of the time of day), dec is the solar declination (function of the time of year) and lat is the latitude. The concentration of H_2O was also parameterised as follows:

$$[H_2O] = 6.1078\exp(-(597.3 - 0.57(T-273.16))18.0/1.986T - 1.0/273.16)(10.0RH)/(1.38e-16T), \tag{4}$$

where RH is the relative humidity and T is the temperature. Relative humidity and temperature data were produced by meteorological model ALADIN [7], which provides data having time resolution of 6 hours and spatial resolution of 0.1×0.15 degrees.

II. 3. Wind field and vertical stratification

The local wind speed and direction was considered as a function of space and time. These data were obtained from the ECMWF database [8], which ensured conservation properties. The ECMWF data has a time resolution of 6 hours and a spatial resolution of 2.5 × 2.5 degrees. These data were interpolated to obtain data relevant to a given space and time point on the model grid.

The model includes four layers: the ground layer, the mixing layer, a reservoir layer and the free troposphere (upper) layer. The surface layer extends from ground level to 50 m altitude. Above the surface layer is the mixing layer, which extends to 300 m at night, but rises to 1000 m during the day [6]. The reservoir layer, if exists, extends from the top of the mixing layer to the altitude of 1000 m. Vertical mixing and deposition are parameterised according to the vertical stratification presented by van Loon [9]. Deposition velocities are assumed to be constant across the whole domain.

II. 4. Domain, source terms and transport parameters

The model domain covers Central Europe including Hungary. The model describes the domain using a Cartesian coordinate system through the stereographic polar projection of the curved surface onto a flat plane. Global coordinates are transformed by projecting the surface of the Earth, from the opposite pole onto a flat plane located at the North Pole which is perpendicular to the Earth's axis. Due to the orientation of the projection plane this transformation places the Cartesian origin at the North Pole. To present results in a more familiar orientation the emissions inventory is rotated by 90 degrees and a false origin is placed at the south-west corner of Europe. The emission of species into the domain was described by the EMEP [10] emissions inventory for 1997. The EMEP emission inventory is transformed from latitude and longitude coordinates to the same Cartesian coordinate system. Emission data are interpolated to the triangular mesh ensuring mass conservation. The eddy diffusivity coefficients for x and y directions was set at 50 m^2s^{-1} for all species.

II. 5. Numerical discretisation

The basis of the numerical method is the space discretisation of the PDEs derived from the atmospheric diffusion equation on unstructured triangular meshes using the software SPRINT2D [11]. This approach, (known as the "Method of Lines"), reduces the set of PDEs in three independent variables to a system of ordinary differential equations (ODEs) in one independent variable, time. The system of ODEs can then be solved as an initial value problem, and a variety of powerful software tools exist for this purpose [12]. 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 references [11, 12, 13, 14, 15].

Unstructured triangular meshes are popular with finite volume/element practitioners because of their ability to deal with general two-dimensional 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. For systems of equations such as (1) it is useful to consider the advective and diffusive fluxes separately in terms of the discretisation. In the present work, a flux limited, cell-centered, finite volume discretisation scheme of Berzins and Ware [16] was chosen. 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 non-linear scheme described by Spekreijse [17] for regular Cartesian meshes. The scheme of Berzins and Ware has the desirable properties, see Chock [18], of preserving positivity eliminating spurious oscillations and restricting the amount of diffusion by the use of a non-linear limiter function. Recent surveys of methods for the advection equation [9, 18] have suggested the use of a very similar scheme for regular Cartesian meshes, preferring it to schemes such as Flux Corrected Transport.

II. 6. Adaptive gridding

The initial unstructured meshes used in SPRINT2D are created from a geometry description using the Geompack [19] mesh generator. These meshes are then refined and coarsened by the triad adaptivity module, which uses data structures to enable efficient mesh adaptation.

Since the initial mesh is unstructured, we have to be very careful in choosing the data structure which provides the necessary information for refining and derefining the mesh. When using a structured mesh it is possible to number mesh vertices or elements explicitly. This is not possible for unstructured meshes and therefore the data structure must provide the necessary connectivity. The important factor is to maintain the quality of the triangle as the mesh is refined and coarsened. This is achieved using a tree-like data structure with a method of refinement based on the regular subdivision of triangles. 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. Similar procedures are extensively used with a wide range of both finite element and volume methods for a very broad range of physical problems.

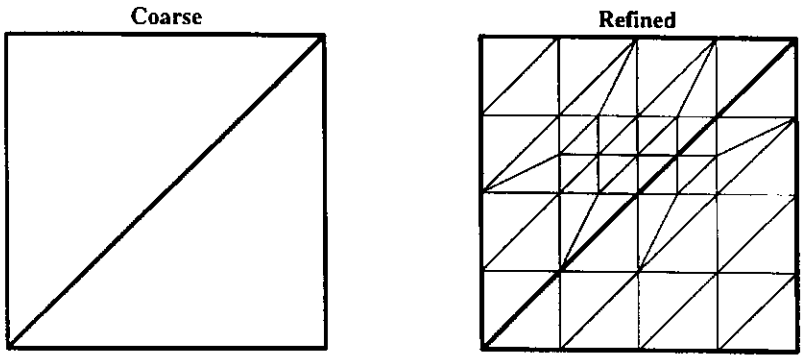

Fig. 1. Method of local refinement based on the subdivision of triangles.

Once a method of refinement and derefinement has been implemented, it remains to decide on a suitable criterion for the application of the adaptivity. 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 and the difference between them gives the spatial error. The algorithm can then choose to refine in regions of high spatial error by comparison with a user defined tolerance.

For the *i*th PDE component on the *j*th 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 PDEs this estimate shows how the spatial error grows locally over a time step. A refinement indicator for the *j*th triangle is defined by an average scaled error $serr_j$ measurement over all *npde* PDEs using supplied absolute and relative tolerances:

$$serr_j = \sum_{i=1}^{npde} \frac{e_{i,j}(t)}{atol_i / A_j + rtol_i \cdot c_{i,j}} \tag{5}$$

where *atol* and *rtol* are the absolute and relative error tolerances, $e_{i,j}(t)$ is local error estimate of species i over element j, $c_{i,j}$ is the concentration of species i over triangle j and A_j is the area of *j*th triangle. This formulation for the scaled error provides a flexible way to weight the refinement towards any PDE 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.

III. Results

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 and strong sunshine resulted in high photo-oxidant levels over most of Europe. High ozone levels were also measured at the K-puszta monitoring station of the Hungarian Meteorological Service, which is located 70 km south-southeast from Budapest.

The initial, rather coarse, grid covered Central Europe as seen in Fig. 2. The resolution of this coarse grid is defined by an edge length of 125 km. The grid was initially manually refined around K-puszta to an edge length of 30.2 km in order to allow comparisons with measured concentrations in this region even under circumstances where spatial errors are low. Transient refinement/derefinement took place every 20 minutes of simulation time throughout the model run. This methodology is used rather than allowing grid adaption every time-step to avoid excessive overhead in interpolating data to the new mesh. Initial test simulations have shown that the method does not lead to greater errors than refining every time-step where often only one or two grid cells are changed. Refinement was limited to 1 levels below the base mesh in this case leading to a minimum edge length of 14.7 km. Fig. 3 shows the grid after 4 simulation days 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. The high ozone concentration area is found to be northwest of Hungary and in this region the grid is also dense. The GRS scheme was used and the initial conditions for the most important species were as a follows: 0.04 ppb for NO_2, 1 ppb for NO, 20.3 ppb O_3, 4 ppb for VOC.

Fig. 4 shows the calculated ozone concentrations on 3rd August, 1998. The highest concentration region is in eastern Austria. The calculations showed that the high ozone levels there originate from the pollutants emitted in the southern Polish industrial areas. Another high ozone concentration area is the industrially developed North-Italy. Also, somewhat higher ozone concentrations can be found downwind from Budapest.

The calculated ozone levels are lower than the ozone levels measured at K-puszta. Improved agreement between the measured and calculated data is expected from further developed version of the model, which will use a more sophisticated method for the calculation of the height of mixing layer and a more detailed emission inventory for Hungary.

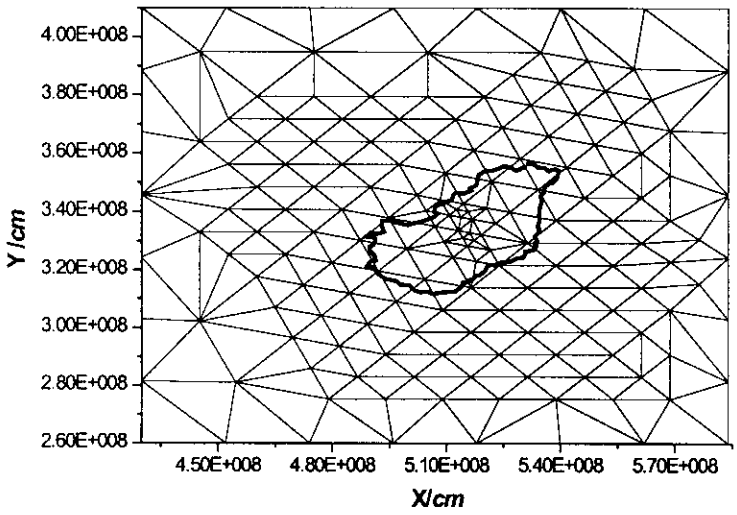

Fig. 2. Initial coarse grid at the beginning of calculations.

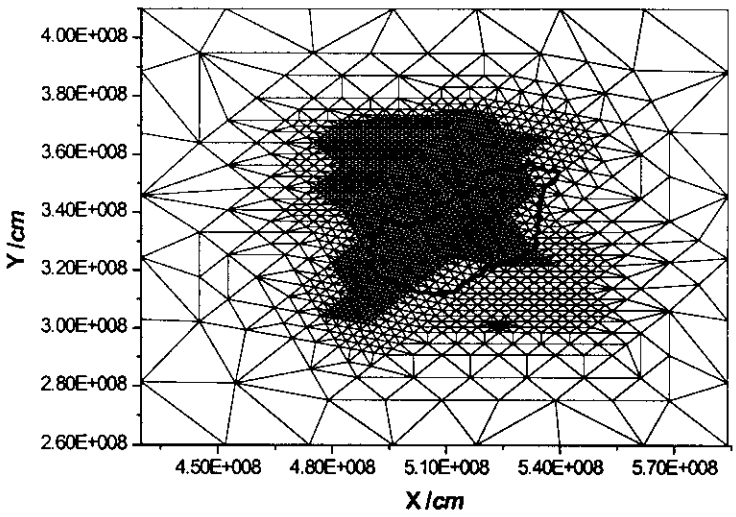

Fig. 3. Grid after 96 hours of simulation.

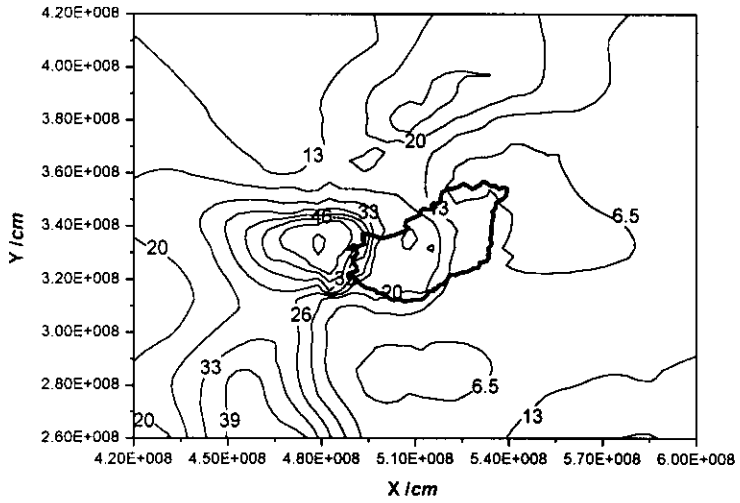

Fig. 4. Calculated ozone concentrations after 3 days of simulations, on 3rd August, 1998 at 16.00

IV. Discussion and 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 15 km without excessive computational effort. Initial simulations of the photochemical episode of August 1998 indicate that the model under predicts ozone concentrations, but captures well the spatial and temporal tendencies. In the near future the model will be developed further by applying higher resolution emission inventory and more sophisticated calculation of the height of the mixing layer.

Acknowledgement: The authors acknowledge the support of OTKA grant T025875 and UK-Hungarian cooperation grant GB50/98.

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