SIMULATION OF THE EFFECT OF THE PLUME OF BUDAPEST ON THE PHOTOCHEMICAL AIR POLLUTANTS FORMATION IN HUNGARY

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The EUROTRAC investigations have shown that some of the highest regional ozone concentrations in Europe can be observed in Central Europe, including Hungary. High spatial resolution of such models is very important to reduce the impact of numerical errors on predictions. A regional air quality model has been developed that describes the transport and chemical transformation of photochemical oxidants across Central Europe using an adaptive gridding method to achieve high resolution. The basic coarse grid covers a wider Central European region. Further refinement of the unstructured triangular grid is invoked during the simulation at intermediate time-steps using spatial error estimators based on the comparison of high and low order numerical solutions of the atmospheric diffusion equation

1. INTRODUCTION

During the summer ozone episodes in Central Europe, the ozone concentration reaches very high level. For a computational study of this phenomenon, an Eulerian photochemical air pollution model was developed in a co-operation between the Eötvös University, Budapest, The University of Leeds, and the Hungarian Meteorological Service. This model describes the spread of reactive air pollutants on four layers of troposphere over the Central European region (Lagzi *et al.* 2001, 2002). The model has been developed within a flexible framework, where the pollution sources are both area and point sources and the chemical transformations can be described by a mechanism of arbitrary complexity. Operation of the model revealed, that the biggest cities, Budapest, Vienna, and Milan, emit significant amount of ozone precursors and therefore highly influence the photooxidant concentrations in this region. Budapest, the capital of Hungary, is one of the major sources and this work focuses on the effect of its plume on the surrounding region.

2. THE MODEL

Horizontal dispersion of species is described within an unstructured triangular Eulerian grid framework. The vertical mixing of pollutants is approximated by a parameterised description of mixing between four layers. The horizontal grid is adaptive, *i.e.* continuously changes in space and time to minimize the numerical errors. Transient refinement and de-refinement is invoked as necessary throughout the model run according to spatial errors. The modelled area is a 1500 km × 1540 km region of Central Europe with Hungary in the centre. The model describes the horizontal domain using a Cartesian coordinate system through the stereographic polar projection of a curved surface onto a plane. The dispersion of species in the horizontal domain are described by the atmospheric transport–reaction equation in two space dimensions:

$$\frac{\partial c_s}{\partial t} = -\frac{\partial (u c_s)}{\partial x} - \frac{\partial (v c_s)}{\partial y} + \frac{\partial}{\partial x}\left(K_x \frac{\partial c_s}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial c_s}{\partial y}\right) + R_s(c_1, c_2, \ldots, c_n) + E_s - k_s c_s,$$

where c_s is the concentration of the sth compound, u and v are horizontal wind velocities, K_x and K_y are eddy diffusivity coefficients and k_s is the dry deposition velocity. E_s describes the distribution of emission sources for the sth compound and R_s is the chemical reaction term, which may contain non-linear terms in c_s. For n chemical species, an n dimensional set of partial differential equations is formed describing the rates of change of species concentrations over time and space, and these concentrations are coupled through the non-linear chemical reaction term. The four vertical layers of the model are the surface layer (extending to 50 m), the mixing layer, the reservoir layer and the free troposphere layer. At night, the mixing layer extends to the height determined by the midnight radiosonde data. Daytime, the height of the mixing layer is assumed to rise smoothly from sunrise to the height determined by the noon radiosonde measurement. In the evening, it goes back to the night level. The reservoir layer, if it exists, 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 (Van Loon, 1996). The eddy diffusivity coefficients for the x and y directions were set to 50 m^2s^{-1} for all species. The local wind speed and direction, relative humidity, temperature and cloud coverage were determined by the meteorological model ALADIN (Horányi *et al.*, 1996), which is the limited area numerical weather forecast model of the Hungarian Meteorological Service. The time resolution of data is 6 hours and the spatial resolution is 0.10 × 0.15 degrees (approximately 8 km × 8 km) for each of the four layers. Meteorological data were interpolated to obtain data relevant to a given spatial point on the unstructured grid using conservative methods. For Budapest, the emission inventories were provided by the local authorities, which have spatial resolution 1 km × 1 km and also include the most significant 63 emission point sources for CO, NO_x and VOCs. For Hungary, the National Emission Inventory of spatial resolution 10 km × 10 km

was applied and it included both area and point sources. Outside Hungary, the emission inventory of EMEP for CO, NO_x and VOCs was used, having spatial resolution of 50 km × 50 km. The emission data had to be interpolated onto the unstructured mesh following each change to the mesh during refinement. This was achieved using the mass conservative method of overlapping triangles. In the present simulations, the GRS chemical scheme (Azzi and Johnson, 1992) was used, although the model allows the utilization of any other reaction schemes. Photolysis rate constants are calculated as described by Derwent and Jenkin (1990) and are expressed as *m*th order rate constants with units (molecule cm^3) $^{m-1}s^{-1}$. Temperature dependent rate constants are represented by standard Arrhenius expressions.

The basis of the numerical method is the spatial discretisation of the partial differential equations derived from the atmospheric transport–reaction equation on unstructured triangular meshes. This approach, (known as the 'method of lines'), reduces the set of partial differential equations in three independent variables (*x, y, t*) to a system of ordinary differential equations in one independent variable, time. The model uses the flux limited, cell centred finite volume scheme of Berzins and Ware (1995). This is achieved by the integration of the atmospheric diffusion equation over each finite volume, the use of the divergence theorem and the evaluation of the line integral along the boundary of each volume using the midpoint quadrature rule. Unstructured triangular meshes are commonly used in finite volume and finite element applications because of their ability to deal with general complex geometries. They have advantages for air quality models in that they are capable of achieving high levels of mesh refinement in regions where concentration gradients are steep. In regional modelling of air pollution, this is very advantageous describing urban areas with mixed point and area sources, and plumes coming from single point sources. The system of ordinary differential equations is integrated by code SPRINT2D. The Theta method of SPRINT2D has been invoked since it is specially designed for the solution of stiff systems with moderate accuracy and controls the local error automatically in time (Berzins *et al.*, 1992). Operator splitting is carried out at the level of the non-linear equations by approximating the Jacobian matrix.

The initial unstructured meshes used in SPRINT2D are created from a geometry description using the Geompack mesh generator (Joe and Simpson, 1991). These meshes are then refined and coarsened by the Triad adaptivity module. Low and high order solutions are obtained for each species and the difference between them gives a measure of the spatial error (Tomlin *et al.* 1997). The algorithm can choose to refine in regions of high error by comparison with a user-defined tolerance for one or the sum of several species. An original triangle is split into four similar triangles by connecting the midpoints of the edges. These may later be coalesced into the parent triangle when coarsening the mesh. The choice of tolerances will therefore reflect to a certain extent, a balance between desired accuracy and available computational resources, since tighter tolerances usually lead to a higher number of grid cells. It is also possible within the code for the user to control the maximum number of levels of adaptivity, thus limiting the minimum grid size in regions of very steep gradients *i.e.* close to the point source.

3. RESULTS AND DISCUSSION

The model simulated a photochemical oxidant episode that took place in Hungary and Central-Europe in August, 1998. During almost the whole month, wind speed was low and strong sunshine resulted in high photooxidant levels over most of Central-Europe. Figure 1 shows the calculated ozone concentrations in ppb

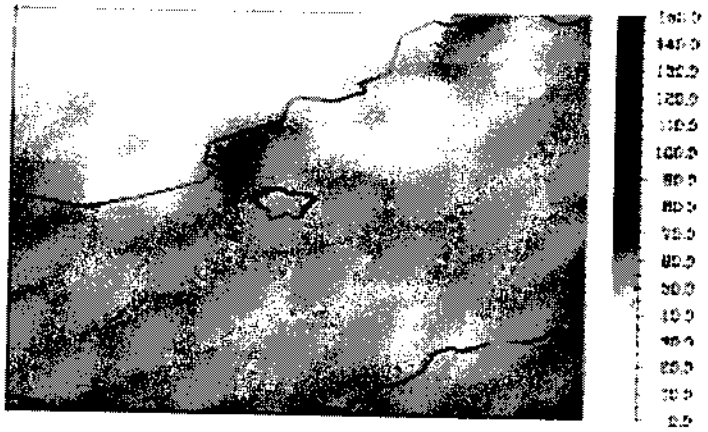

Figure 1. Calculated ozone concentration at 16.00 on 2nd August, 1998

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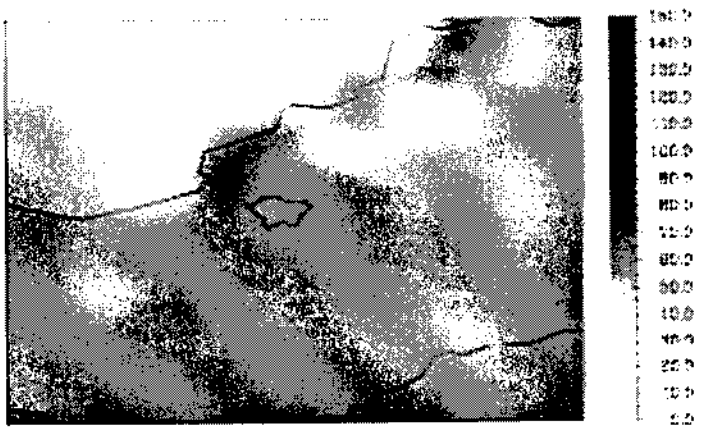

Figure 1. Calculated ozone concentration at 16.00 on 2nd August, 1998

at 16.00 on 2nd August, 1998 using grey scale. The dark territories correspond to high ozone level. The high ozone concentrations can be observed in north-west direction from the Budapest. The figure shows that the south-east wind transports the ozone precursors from Budapest, which generate high ozone concentration north-west from the city. Figure 2 shows the calculated ozone concentrations at 16.00 on 3rd August, 1998. There is a high ozone concentration in a wide region around Budapest, but in the city the ozone concentration is much lower due to the high local NO emission.

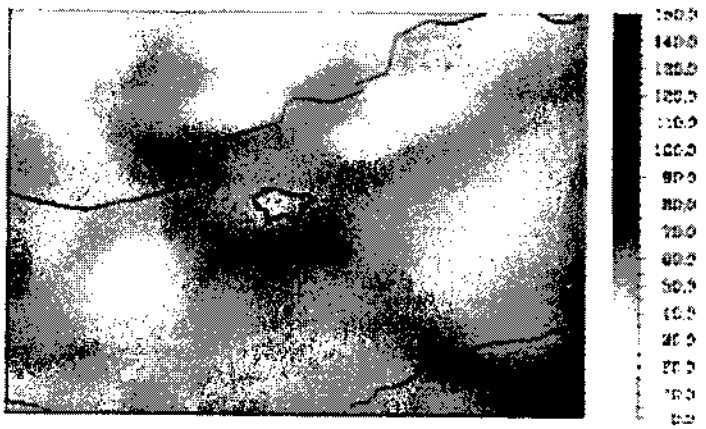

Figure 2. Calculated ozone concentration at 16.00 on 3rd August, 1998

4. CONCLUSIONS

An adaptive grid model that describes the formation and transformation of photochemical oxidants, based on triangular unstructured grids has been developed to study the photochemical air pollution in the Central-European region. The model was applied here to investigate the influence of the emission of Budapest for the ozone concentration around the city. The two typical patterns are that (i) the high ozone precursor emission of Budapest causes a plume-like formation of ozone within about 100 km downwind even if no regional photochemical air pollution episode is present; (ii) in case of a regional zone episode, the large amount of NO emitted in Budapest significantly decreases the ozone concentration in the city. This latter influence is limited to a narrow region of Budapest. The model can be used for the elaboration of integrated ozone concentration maps for each year, which will allow a more comprehensive study of the emission of Budapest.

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