Modelling photochemical air pollutant formation in Hungary using an adaptive grid technique

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Abstract: 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 spatial resolution. High-resolution emission inventories for Budapest and Hungary were utilised. The air pollution episode in August 1998 was modelled using a fixed coarse grid (mesh size 70 km) a fixed fine grid (17.5 km) and an adaptive, variable sized (from 17.5 to 70 km) grid. The fine and the adaptive grid models provided similar results, but the latter required 50% longer computing time. High ozone concentrations appeared downwind of Budapest and the plume extended up to about 150 km from the city at 17.00 on the simulated day. The simulation results were compared with ozone concentrations measured at the K-puszta and Hortobágy monitoring stations.

Keywords: photochemical air pollution; urban plume; adaptive gridding method; Eulerian model.

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1 Introduction

Previous EUROTRAC and EUROTRAC2 investigations have shown that ozone is increasing on both local and regional scales in the European boundary layer and that some of the highest regional ozone concentrations in Europe can be observed in Central Europe, including Hungary (Scheel et al., 1997). During summer ozone episodes, ozone concentrations can exceed legislative standards in Central Europe (see e.g. http://www.emep.int). Ozone and other photo-oxidants can cause damage to human health, natural and agricultural vegetation. Therefore, an important strategic goal is to develop reliable tools to help us to estimate these short and long-term impacts (Andrade et al., 2004; Decanini et al., 2003; Havasi and Zlatev, 2002; Palacios et al., 2002; Zlatev and Syrakov, 2004). Computational models form one set of tools that can be usefully employed to evaluate past episodes and possible future trends in photochemical oxidants. For the computational study of this phenomenon in Hungary, an Eulerian photochemical air pollution model has been developed through cooperation

among the Eötvös Loránd University, Budapest, The University of Leeds and the Hungarian Meteorological Service. A preliminary brief account of the development of this model has been published by Lagzi et al. (2001, 2002). This model fully utilises adaptive gridding methods for modelling chemical transport from multiscale sources (Ghorai et al., 2000; Hart, 1999; Hart et al., 1998; Tomlin et al., 1997, 2000). The model has been elaborated within a flexible framework where both area and point pollution sources can be taken into account with variable resolution, and the chemical transformations can be described by a mechanism of arbitrary complexity. Operational use of the model has revealed that the large cities in the region, Budapest and Vienna emit significant amounts of ozone precursors and highly influence the photo-oxidant concentrations in this region. Budapest, the capital of Hungary, is one of the major sources. This paper reports the latest developments of the model and focuses on the effects of the plume of Budapest on the concentrations of ozone in the surrounding region for a high ozone episode. Although previous studies (Delle and Stull, 2003; Havasi and Zlatev, 2002; Tilmes et al., 2002; Zlatev et al., 1993; Zlatev and Syrakov, 2004) have used an Eulerian modelling framework to study high ozone concentrations within Europe, this study is the first to describe the photochemical air pollution formation in Hungary in a detailed way, focusing on the emissions of Budapest at high resolution. An emission inventory for Budapest at 1 km resolution and an adaptive grid technique are used to predict the ozone levels in Hungary for the first time.

2 The model

The model describes the spread of reactive air pollutants in four layers of the troposphere over the Central European region. The vertical mixing of pollutants is approximated by a parameterised description of exchange between the layers. The horizontal dispersion of species is described within an unstructured triangular Eulerian grid framework. The horizontal grid is adaptive, that is, continuously changes in space and time to minimise the numerical errors. Transient refinement and de-refinement is invoked as necessary throughout the model run according to the estimated spatial errors (Tomlin et al., 1997). The modelled area is a 980 km × 920 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 the curved Earth surface onto a plane. The dispersion of species in the horizontal domain is described by the atmospheric transport reaction equation in two space dimensions:

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

where c_s is the concentration of the sth compound, u and v are horizontal wind velocities, K_x and K_y are eddy diffusion coefficients, k_s is the dry deposition rate constant, 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 change of concentrations over time and space. These equations are coupled through the non-linear chemical reaction term.

The four horizontal layers of the model are the surface layer (from surface 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. During the daytime, the height of the mixing layer is assumed to rise smoothly to the height determined by the noon radiosonde data. In the evening, it collapses to the night-time 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 of the atmosphere (Van Loon, 1996). The species exchange between the layers (i.e. the vertical transport) is modelled in two ways. Exchange between the mixing and the surface layers due to turbulent diffusion and fumigation at the top of the mixing layer are described by ordinary differential equations. These equations have been defined in our recent paper (Lagzi et al., 2004). In this way, species exchange takes place between the mixing layer and the reservoir layer or the upper layer if the reservoir layer does not exist.

The wind speed and direction, relative humidity, temperature and cloud coverage were determined by the meteorological model ALADIN (Horányi et al., 1996), which is the numerical weather forecasting model of the Hungarian Meteorological Service. The ALADIN model is a hydrostatic, spectral, limited area model using 24 layers for vertical resolution where initial and boundary conditions are determined from a large-scale weather prediction model ARPEGE. The model domain for ALADIN covers the Central European region from latitude 43.1°N to 52.0°N and from longitude 10.35°E to 25.1°E. The time resolution of data is 6 hr and the spatial resolution is 0.10 × 0.15° (approximately 10 km × 10 km). In our model, conservative interpolation methods were used to obtain data relevant to a given spatial point on the unstructured grid from the regularly gridded ALADIN meteorological data.

The dry deposition velocity was calculated using the resistance method that is based on the parameterisation of the surface resistance, the boundary layer resistance and the aerodynamic resistance. The model calculated the Monin-Obukhov length from the data of the ALADIN meteorological model.

For Budapest, the emission inventories for CO, NO_x and VOCs were provided by the local authorities with a spatial resolution of 1 km × 1 km and also include the most significant 63 emission point sources. For Hungary, the National Emission Inventory of spatial resolution 20 km × 20 km was applied which included both area and point sources. Figure 1 shows the emission inventories of NO_x for Budapest and Hungary. Outside Hungary, the emission inventory of EMEP for CO, NO_x and VOCs was used, having a spatial resolution of 50 km × 50 km. The emissions data had to be interpolated onto the unstructured grid following each change to the mesh during refinement. This was achieved using the mass conservative method of overlapping triangles. Point sources are averaged into the appropriate grid cell for their location and hence when the grid is refined the definition of point sources improves.

In the present simulations, the GRS chemical scheme (Azzi et al., 1992) was used, although the model allows the utilisation of any other reaction scheme. The GRS scheme is a reduced mechanism that was created using a semi-empirical approach; it contains seven reactions of seven species (see Table 1). The GRS scheme was developed by comparison with smog chamber data and has been evaluated by comparison with smog chamber data and predictions from more detailed chemical schemes (Cope et al., 2005). Previous studies have shown that the scheme performs well for the prediction of ozone in polluted conditions although it can overpredict ozone concentrations in rural locations.

The scheme has been selected in the current application for its computational efficiency and because its accuracy can be assumed to be reasonable in the region of interest that is, down wind of major NO_x sources. The rate constants were calculated as described by Derwent and Jenkin (1990) and were expressed as *m*th order rate constants with units (molecule $cm^3)^{m-1}$ s^{-1}. The photolysis rates were parameterised by the following function:

$$J_q = (1 - 0.75N^{3.4})a_q \exp\left(-b_q \sec\Theta\right)$$

where Θ is the solar zenith angle, N is the cloud coverage and a_q and b_q are the rate parameters of reaction q. Temperature dependent rate constants were represented by standard Arrhenius expressions.

Figure 1 The emission inventories of NO_x in 1998: (a) for Budapest with resolution 1 km × 1 km and (b) for Hungary with resolution 20 km × 20 km

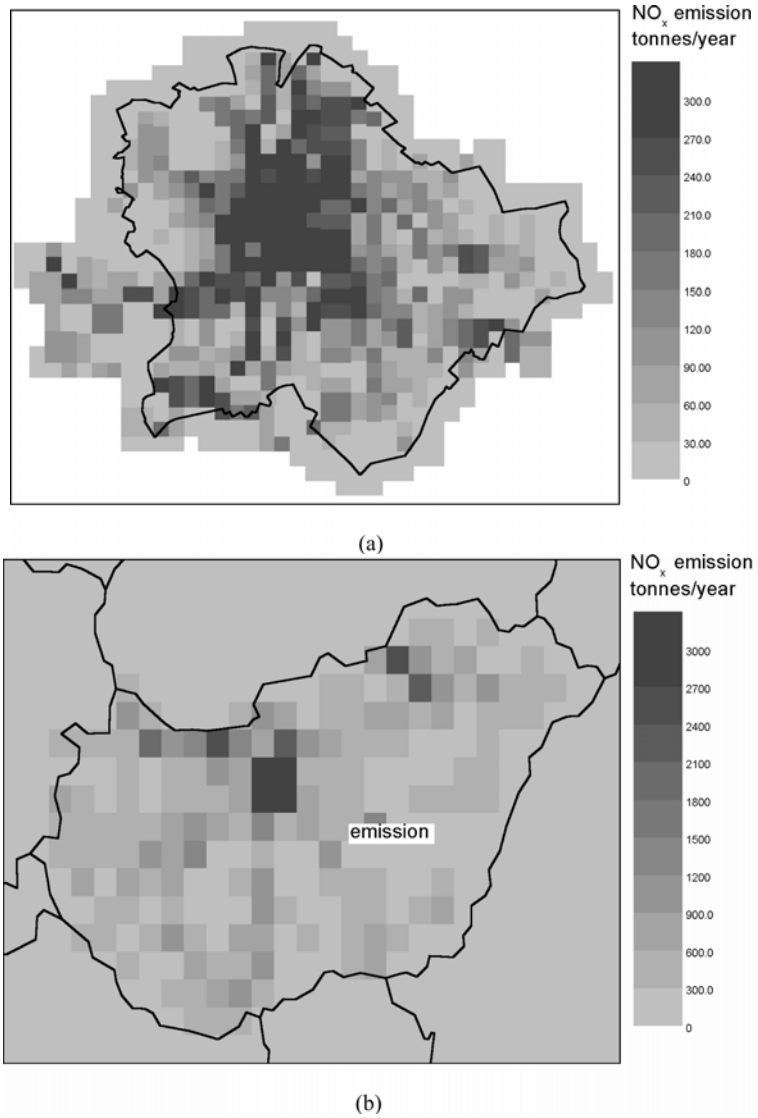


Table 1 The GRS mechanism

Reactions	*Reaction rate constants*	
ROC + $h\nu \rightarrow$ RP + ROC	$k_1 = 1000 \exp(-4710/T)J_3$	[R1]
RP + NO $\rightarrow$ NO_2	$k_2 = 3.7098 \times 10^{-12}\exp(242/T)$	[R2]
$NO_2 + h\nu \rightarrow$ NO + O_3	$J_3 = 1.45 \times 10^{-2}\exp(-0.4 \sec \Theta)$	[R3]
NO + $O_3 \rightarrow$ NO_2	$k_4 = 1.7886 \times 10^{-12} \exp(-1370/T)$	[R4]
RP +RP $\rightarrow$ RP	$k_5 = 6.7673 \times 10^{-12}$	[R5]
RP + $NO_2 \rightarrow$ SGN	$k_6 = 1.00 \times 10^{-13}$	[R6]
RP + $NO_2 \rightarrow$ SNGN	$k_7 = 1.00 \times 10^{-13}$	[R7]

Note: T is the temperature and Θ is the solar zenith angle.

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 to a system of ordinary differential equations of one independent variable, time. The model uses the flux limited, cell-centred finite volume scheme of Berzins and Ware (1994, 1995). The system of ordinary differential equations is integrated by the code SPRINT2D (Berzins and Furzeland, 1992; Berzins and Ware, 1996; Berzins et al., 1989). Operator splitting is carried out at the level of the non-linear equations by approximating the Jacobian matrix. Further details of the method are presented in Tomlin et al. (1997).

The initial unstructured meshes 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 identifies the regions of large error by comparison with a user-defined tolerance for the concentration of one or several species. 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 and 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 that is considered over all npde PDEs using user supplied absolute and relative tolerances:

$$\mathrm{serr}_j = \sum_{i=1}^{\mathrm{npde}} \frac{e_{i,j}(t)}{\mathrm{atol}_i / A_j + \mathrm{rtol}_i c_{i,j}}$$

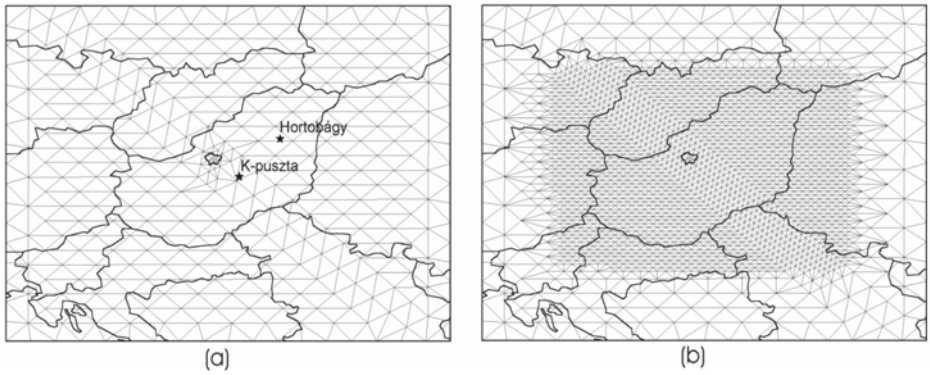
where atol and rtol are the absolute and relative error tolerances, respectively, $e_{i,j}(t)$ is the local error estimate of species i over element j, $c_{i,j}$ is the concentration of species i over triangle j and A_j is the area of jth triangle. This formulation for the scaled error provides a flexible way to weight the refinement towards any PDE error (Berzins, 1994; Berzins et al., 1992; Ware and Berzins, 1995). In the calculations presented, a combination of errors in species NO and NO_2 were used as a refinement indicator, because these are primary species and also because their concentrations are very closely related to ozone production. Estimation of the local spatial error of ozone concentration is not an efficient choice, because it would be too late to make refinement decisions on the basis of the detection of a large error in the concentration of a secondary pollutant. On the other

hand, concentrations of the VOCs are locally dominated by emissions, and since the available emission's inventory for VOCs has a coarse resolution (50 km × 50 km), the use of VOC concentration as an error indicator is not appropriate. Tomlin et al. (2000) previously demonstrated the success of using the local spatial error of the concentrations of nitrogen oxides for appropriate mesh refinement for a reactive plume from a NO_x ($NO + NO_2$) source. Each triangle that is flagged for refinement is split into four similar triangles. Refined triangles may later be coalesced into the parent triangle when coarsening the mesh.

3 Results and discussion

The simulated period was the beginning of August 1998. During almost the whole period low cloud coverage, low wind speeds and the high temperatures resulted in high photo-oxidant levels in most of Europe. In Hungary, high ozone concentrations were measured at the K-puszta (48°58′N, 19°33′E) and Hortobágy (47°29′N, 20°56′E) monitoring stations of the Hungarian Meteorological Service. Three simulations, corresponding to three different spatial grid structures, were carried out. The first grid was a coarse fixed one that covered a part of Central Europe as seen in Figure 2(a). The resolution of this coarse grid (level 0) was characterised by an edge length of 70 km. The grid structure of the second type was a fixed fine nested grid (level 2) over Hungary, which had an edge size of 17.5 km (see Figure 2(b)). In the adaptive grid simulations, the refinement was restricted to the area of the nested grid in Figure 2(b) and limited to 2 levels. Therefore, the minimum grid size was identical to that of the fine grid in the nested grid calculations. The initial concentrations of the major species were 0.4 ppb for NO_2, 2.0 ppb for NO, 89.3 ppb for O_3 and 4.1 ppb for VOC. The initial concentrations were equal in each layer across the whole simulated domain.

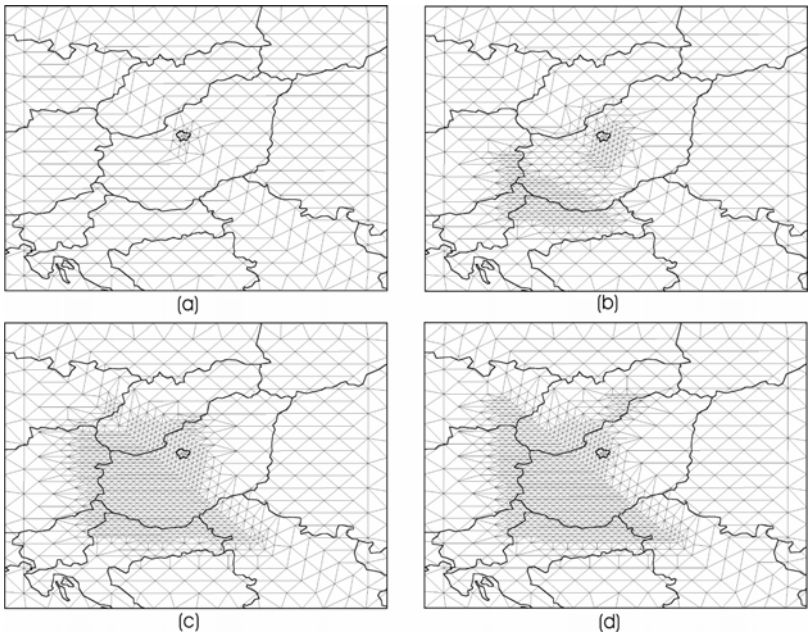
Figure 2 The structure of the coarse (level 0; with a nested grid around Budapest) and fine (level 2) grid. The symbols show the monitoring stations of the Hungarian Meteorological Service. The average mesh lengths are 70 and 17.5 km for the two cases, respectively



The simulation period from noon on 31 July to midnight on 3 August 1998 was chosen. Figure 3 illustrates the evolution of the adaptive grid in time. The adaptive grid was initially refined around Budapest, which is the main emission source of the primary pollutants in Hungary. High spatial gradients in NO_x concentrations are therefore likely

to have formed close to the Budapest region leading to an increase in spatial errors and therefore mesh refinement. This is in part due to the high-resolution inventory used in the simulations and raises interesting issues with regard to the influence of mesh resolution within an Eulerian framework. Clearly, where a coarse emission inventory is used, a certain level of averaging has already taken place, perhaps resulting in a lower sensitivity to solution grid resolution below a certain level, although meteorological events may still lead to steep spatial concentration gradients under certain conditions. The finer the resolution of the emissions inventory, the larger the spatial concentration gradients will be at the edges of the down wind plumes formed. The representation of large point sources represents a particular challenge to Eulerian models. As emissions inventories become finer therefore, the influence of grid resolution on solution accuracy may become more apparent, requiring the use of techniques such as transient grid adaption.

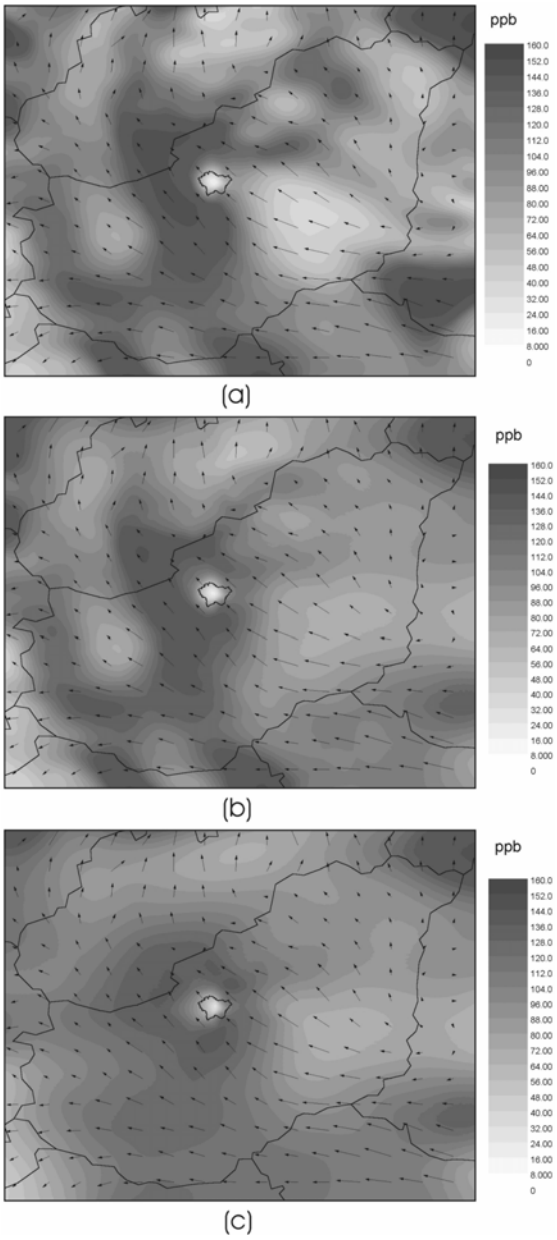
Figure 3 The time evolution of the adaptive grid: (a) t_o, (b) t_o + 24 hr, (c) t_o + 48 hr and (d) t_o + 72 hr



After one and a half simulation days, the whole western region of the domain became refined, because this region is more industrialised and emits higher levels of ozone precursors than the other regions of the simulated area. Figure 4 shows the calculated ozone concentrations after 3 days of simulation on the 3 August 1998 at 17.00, as a result of three simulations each using a different type of grid. During the simulated period, the southern winds transported the ozone precursors towards the north from Budapest. The simulated ozone concentrations using the fine (Figure 4(a)) and the adaptive grids (Figure 4(b)) are very similar. Both simulations show high ozone concentrations in a wide north and north-west region around Budapest, but in the city the ozone concentration is much lower due to the high local NO emission. In these simulations, the plume is bent, follows the direction of the wind and extends up to about 150 km from the city at 17.00. There are significant differences in the predicted peak ozone concentrations

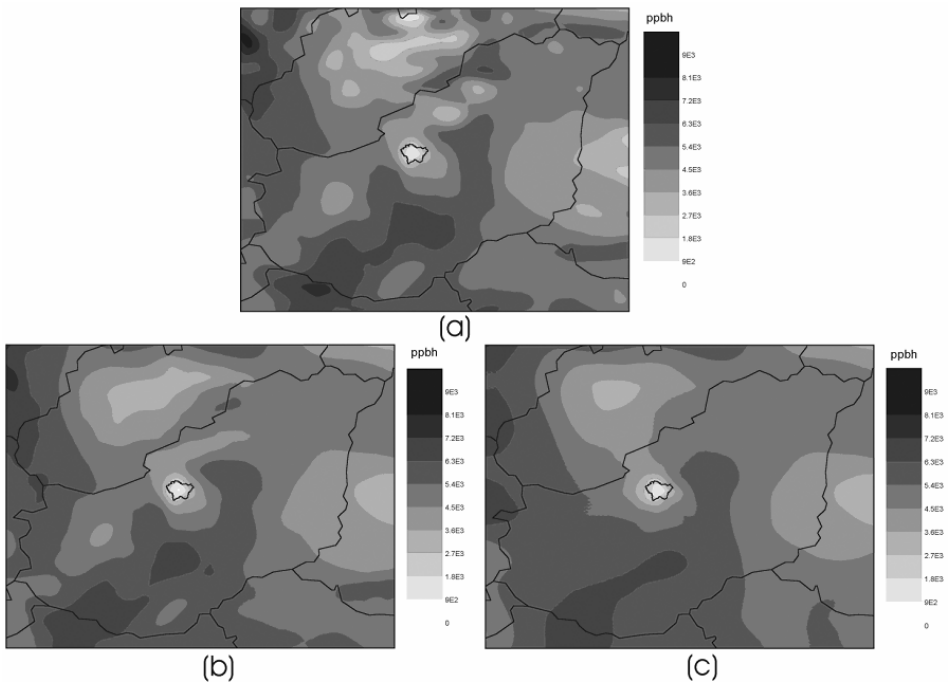
between the coarse grid (Figure 4(c)) and the fine (and adaptive) grid simulations. In general, the simulations using higher resolution grids predict higher peak ozone concentrations than the low resolution ones. The simulations using the coarse grid predict an 'ozone ring' around the city and smooth the concentration peaks due to numerical diffusion. The detailed structure of the plume in the South West region of the country is lost in the low-resolution simulations.

Figure 4 Calculated ozone concentrations on the 3 August 1998 at 17.00 with wind field originated from the ALADIN weather prediction model: (a) application of fixed fine nested grid, (b) adaptive grid and (c) fixed coarse grid



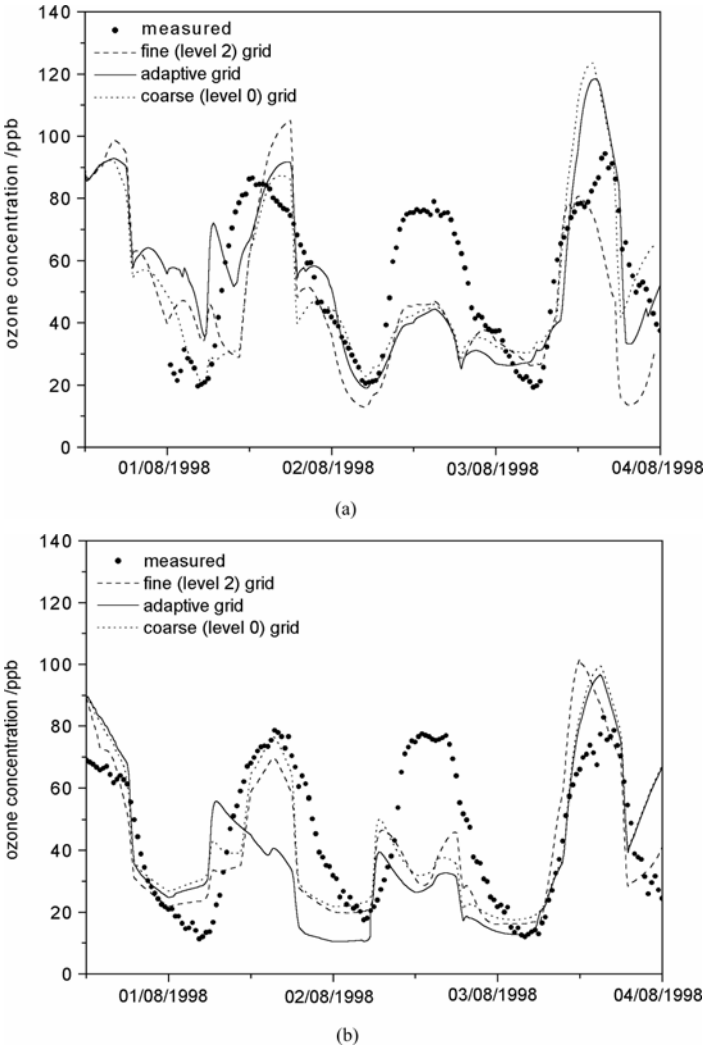
The comparisons of integrated ozone concentrations across the simulation period are displayed in Figure 5. We obtain very similar integrated ozone patterns using the fixed nested fine and adaptive grids. In both cases, a plume-shaped region of lowered ozone concentrations is present to the North East of Budapest with a peak of much higher integrated concentrations at the end of the plume close to the Slovakian border. The ability to model this depletion of ozone results from the use of a high-resolution emissions inventory for Budapest coupled with a grid resolution that is high enough to resolve the resulting plumes. This enables the resolution of nitrogen oxide concentrations in this region leading to the depletion of ozone. In the coarse grid simulation, the integrated ozone concentrations are smoothed in comparison and the plume-shaped structure is not present. Hence, although the fixed coarse grid model is the fastest to simulate, it misses key features related to integrated concentrations and would therefore be unsuitable for estimating the long-term impact of ozone.

Figure 5 Calculated integrated ozone concentrations for the simulation period: (a) application of fixed fine nested grid, (b) adaptive grid and (c) fixed coarse grid



The simulated and measured ozone concentrations at the K-puszta and Hortobágy monitoring stations are shown in Figure 6 as a function of time. The agreement between the simulated and the measured concentrations is relatively good for most of the episode. On the third simulation day (2 August 1998) however, each of the models underestimated the ozone concentration. The difference between the models is quite small indicating that the problem does not lie with the gridding strategy but more likely with errors in the input data such as meteorological conditions and or emissions. On the fourth day, the simulated results had good accordance with the measured ones.

Figure 6 Comparison of model predictions using several grid structures with the measured ozone concentrations at: (a) the K-puszta and (b) the Hortobágy monitoring stations.



Note: Measured data are not available for K-puszta on 31 July, 1998.

The ratio of CPU time requirements and number of grid cells for the coarse, adaptive and fine grid models is given in Table 2. The fine and the adaptive models provided similar results, but the former method required 1.5 times more computer time and a significantly higher number of grid cells. Therefore, the adaptive grid model provides an efficient method for the prediction of secondary air pollution formation at the regional scale. Such model could be successfully used for operational purposes due to lower CPU costs.

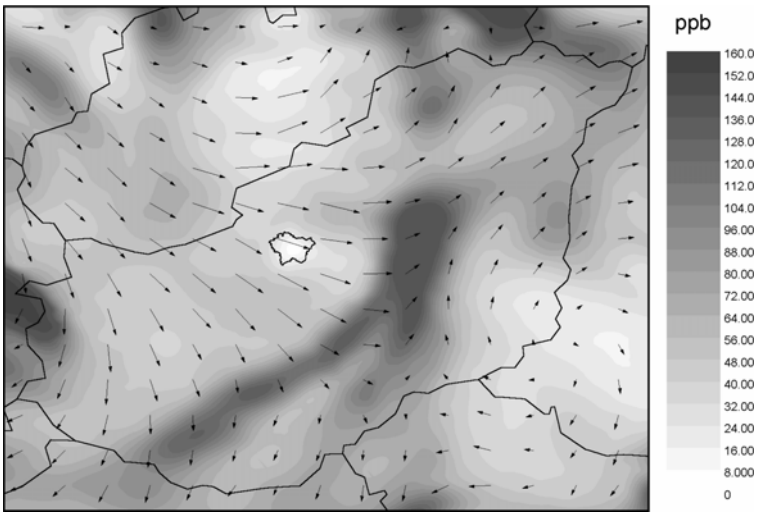
Figure 7 represents the ozone distribution on 4 August 1998 at 17.00. In this period the wind field was opposite to that on the previous day. The ozone plume structure differs in this case. The ozone peak is far from Budapest, but lower ozone concentrations can be observed near to Budapest in the downwind direction due to the relatively large

NO concentrations in the plume of Budapest. Elevated ozone concentrations can be observed in a south-easterly direction from Budapest and in the western part of the country (due to the Vienna plume). This ozone distribution resembles to the usually found, because the main wind direction in Hungary is north-westerly. This clearly demonstrates the long-range impact of the emission of large amounts of ozone precursors in the city on ozone concentrations downwind. The model presented may wrongly predict the distribution of pollutants near the major point sources and in the case of intensive vertical turbulent transport because of using a transport scheme with only a limited number of atmospheric layers. Additionally, the model cannot incorporate small-scale effects, which could produce transitionally incorrect predictions at point sources. Carpathian Basin ensures special meteorological conditions; therefore, the simplified vertical transport parameterisation can be predominantly used. Furthermore, the selected chemical mechanism is efficient to predict ozone concentration, but it can overpredict ozone concentrations in rural locations.

Table 2 Comparisons of CPU times and number of grid cells for each meshing strategy

Grid type	*Relative CPU time*	*Number of grid cells*
Coarse	1	353
Adaptive	8.8	353–1276
Fine nested	13.2	2298

Figure 7 Calculated ozone concentrations on the 4 August 1998 at 17.00 using fixed fine nested grid with wind field originated from the ALADIN weather prediction model



4 Conclusions

An adaptive grid model describing the formation and transformation of photochemical oxidants based on triangular unstructured grids has been developed and applied to the simulation of photochemical oxidant formation in Hungary. The model contains a

high-resolution emissions inventory for the Budapest region and during the simulation automatically places a finer resolution grid in regions characterised by high-concentration gradients and therefore by higher numerical error. Using the adaptive method, grid resolutions of the order of 10 km can be achieved in regional air pollution models without excessive computational effort. The overhead in using such a transient adaptive scheme stem from the need for interpolation of emissions and meteorological data as well as modelled concentrations onto the new grid structure following grid refinement or de-refinement. However, such overheads can be minimised if the grid refinement procedure is not performed for each simulation timepoint but is limited to a given time interval, which in the current application was 5 min. This procedure, coupled with the use of a refinement 'safety layer' was demonstrated in Tomlin et al. (2000) to provide an efficient method of tracking steep concentration gradients without leading to excessive overheads. Figure 3 demonstrates that in the current application, large parts of the nested domain did not require refinement using the chosen tolerances. If refinement overheads are limited the application therefore shows that transient adaption can provide an efficient alternative to traditional nested grid approaches.

The simulation of a photochemical episode that occurred during August 1998 demonstrates the influence of precursor emissions from Budapest on down-wind ozone concentrations up to 150 km from the city. The comparison of different grid strategies demonstrates that the use of a coarse grid has a tendency to smooth out key features in both local and integrated ozone concentrations due to numerical diffusion. This results in the underestimation of both ozone depletion in high NO_x regions and peak ozone concentrations. The adaptive model predicts similar features to the fixed fine grid model using less CPU time and grid cells. The results therefore indicate the potential for using adaptive models in an operational context for assessing the long-term impact of ozone within Europe.

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