Conference on Modelling Fluid Flow (CMFF'03)
The 12th International Conference on Fluid Flow Technologies
Budapest, Hungary, September 3 - 6, 2003

SIMULATION OF THE FORMATION AND SPREAD OF PHOTOCHEMICAL AIR POLLUTANTS IN HUNGARY

István **LAGZI**[1], Tibor **NAGY**[1*], Tamás **TURÁNYI**[1], László **HASZPRA**[2], Alison S. **TOMLIN**[3]

[1]Department of Physical Chemistry, Eötvös University (ELTE), Budapest, Hungary

[2]Institute for Atmospheric Physics, Hungarian Meteorological Service, Budapest, Hungary

[3]Department of Fuel and Energy, The University of Leeds, UK

*Corresponding author: H-1518 Budapest, P.O. Box 32, Hungary
Tel.: (+36 1) 209 0555 x1509, Fax: (+36 1) 209 0602, Email: nt2001@freemail.hu

ABSTRACT

An adaptive grid model has been developed to describe the formation of photochemical air pollutants in the Central European region. The modelled region covers an area of 1500 *km* × 1500 *km* with Hungary in the centre. Grid resolution in critical places can be as fine as 6 *km*. Vertical stratification of the troposphere, up to 3000 meters, is described by using four layers. The meteorological data used were obtained from the weather forecast model ALADIN of the Hungarian Meteorological Service. Simulation results are presented for a smog episode of 3rd and 4th August, 1998 and these show good agreement with the measured surface ozone concentrations.

Key Words: adaptive grid model, reactive flow, photochemical air pollution

NOMENCLATURE

c_i	$[cm^{-3}]$	concentration
$\mathbf{c}(t_n)$	$[cm^{-3}]$	numerical approximation to $\mathbf{c}$ at t_n
$\dot{\mathbf{c}}(t_n)$	$[cm^{-3}\ s^{-1}]$	time derivative of $\mathbf{c}(t_n)$
E_i	$[cm^{-3}\ s^{-1}]$	emission source
g	$[m\ s^{-2}]$	gravitational constant
h	$[s]$	time step
k_d	$[s^{-1}]$	dry deposition rate coefficient
K_x, K_y	$[m^2\ s^{-1}]$	turbulent diffusivity coefficient
L	$[m]$	Monin-Obukhov length
N	[-]	cloud coverage
r_a	$[m^{-1}\ s]$	aerodynamic resistance
r_b	$[m^{-1}\ s]$	quasi-laminar transfer resistance
r_c	$[m^{-1}\ s]$	surface resistance
R_i	$[cm^{-3}\ s^{-1}]$	chemical source term
q	$[W\ m^{-2}]$	sensible heat flux
t	$[s]$	time
T	$[K]$	surface layer temperature
T_*	$[K]$	dynamic temperature
u_*	$[m\ s^{-1}]$	friction velocity
v_g	$[m\ s^{-1}]$	dry deposition velocity
v_x, v_y	$[m\ s^{-1}]$	horizontal wind velocity
x, y	$[m]$	spatial coordinates
z_o	$[m]$	roughness length
κ	[-]	von Karman constant
Θ	$[rad]$	solar zenith angle

Subscripts

i	chemical species number
k	reaction number
n	time point n
x	direction x
y	direction y

1. INTRODUCTION

During the summer air-pollution episodes, very high ground-level ozone concentrations can be observed in Central Europe, including Hungary. The ozone burden is well beyond the tolerable level for both the population and the vegetation. An important tool in the management of ozone problems is a computational model, which calculates the concentration of air pollutants from

emission and meteorological data, via the solution of the appropriate PDEs based on first principle physical and chemical laws. Such a model can be used to test the effect of emission control strategies both in a short time scale (during smog episodes) and in a long term (legislation of the permission of new major emission sources). In cooperation among The University of Leeds, the Eötvös University (Budapest), and the Hungarian Meteorological Service a numerical model was developed. This paper describes the model and reports simulation results on a smog episode in early August, 1998.

2. DESCRIPTION OF THE MODEL

The model describes the spread and the reactions of reactive air pollutants on a 2D unstructured triangular grid. The model represents the horizontal domain using Cartesian coordinate system through the stereographic polar projection. The model domain covers Central Europe including Hungary with a domain size of 1500 km × 1500 km. Vertical resolution of the pollutants is approximated by the application of four layers representing the surface, mixing, reservoir layers and the free troposphere. Meteorological data are processed for each layer within each triangular cell. The local wind speed and direction, temperature, cloud coverage and relative humidity were considered as a function of space and time. These data were obtained from the meso-scale meteorological model ALADIN [1], which provides data with a time resolution of 6 hours and a spatial resolution of 0.10 × 0.15 degrees. The model domain for ALADIN covers the Central European region from latitude 43.1° to 52.0° and longitude 10.35° to 25.1°. The data from ALADIN were interpolated using mass conservative methods to obtain the data relevant to a given space and time point on the model grid. The eddy diffusivity coefficients for the x and y directions were set to 50 m^2s^{-1} for all species.

One major sink for several species is the surface deposition. The dry deposition of species depends on its concentration and a deposition velocity (v_d), which is calculated [6] according to the following equation:

$$v_g = \frac{1}{r_a + r_b + r_c} \tag{1}$$

Parameter r_b is equal to $2.6/\kappa u_*$. The aerodynamic resistance r_a was calculated according to equation:

$$r_a = \frac{0.74}{\kappa u_* \left(\log \frac{z}{z_o} + \chi(z_o) - \chi(z) \right)} \tag{2}$$

where

$$\chi(z) = \begin{cases} -6.4\frac{z}{L} & \text{if } L > 0 \\ 2\ln\left(\sqrt{1-9\frac{z}{L}}+1\right) & \text{if } L < 0 \end{cases} \tag{3}$$

The roughness length z_o is assumed to be $0.25m$, representing medium size vegetation as typically found in an agricultural region. The Monin-Obukhov length L was calculated according to the following method:

(i) At night (R_g = 0, where R_g is the global radiation)

$$T_* = \min(T_{*1}, T_{*2}) \tag{4}$$

$$T_{*1} = 0.09(1 - 0.5N^2) \tag{5}$$

$$T_{*2} = \frac{TC_{dn}V}{4\beta_M sg} \tag{6}$$

β_M = 4.7 and

$$C_{dn} = \frac{\kappa}{\ln(z) - \ln(z_o)} \tag{7}$$

$$V = \sqrt{u^2 + v^2} \tag{8}$$

$$u_* = \begin{cases} \frac{C_{dn}V}{2}\left(1 + \left(1 - corr^2\right)\right)^{1/2} & \text{if } corr \le 1 \\ \sqrt{\hat{L}\frac{T_*}{T}g\kappa} & \text{if } corr > 1 \end{cases} \tag{9}$$

where $\hat{L}$ = 5m and

$$corr = \frac{2u_o}{C_{dn}^{1/2}V} \tag{10}$$

$$u_o = \left(\frac{\beta_M sgT_*}{T}\right)^{1/2} \tag{11}$$

(ii) During the daytime (R_g > 0)

$$u_* = \begin{cases} \dfrac{\kappa u}{\ln(s) - \ln(z_o)} & \text{if } V \geq 0.5 \ \ ms^{-1} \\ 0.05 \ \ ms^{-1} & \text{if } V < 0.5 \ \ ms^{-1} \end{cases} \quad (12)$$

$$T_* = -\frac{q}{\rho c_p u_*} \quad (13)$$

where ρ=1.2754 kg m^{-3} is the density of air and c_p=1.005 J kg^{-1} K^{-1} is the specific heat of air. The sensible heat flux q was calculated from the global radiation.

The Monin-Obukhov length L is calculated from the following equation:

$$L = \frac{Tu_*^2}{g\kappa T_*} \quad (14)$$

Fumigation describes the process, which occurs as the height of the mixing layer changes. Species exchange takes place between the mixing layer and the reservoir layer (if it exists) and the upper layer if not. This process is modelled by the equation:

$$\frac{d\left(H_m c_m\right)}{dt} = \frac{dH_m}{dt} c_{r,u} \quad (15)$$

where $c_{r,u}$ is the concentration in the reservoir or upper layer.

Some air pollutants (called the primary pollutants) are directly emitted to the atmosphere. Such pollutants are the hydrocarbons and other volatile organic compounds, and the nitrogen-oxides. Other pollutants, like the ozone (O_3), are formed by chemical reactions in the troposphere. A photochemical air pollution model has to describe the chemical kinetics of the transformations of the air pollutants. Our model uses the GRS (Generic Reaction Set) gas-phase chemical kinetic scheme that was proposed by Azzi and Johnson [7]. This mechanism represents 7 species interacting in 7 reactions:

$$\text{ROC} + h\nu \rightarrow \text{RP} + \text{ROC} \quad \text{(R1)}$$

$$\text{RP} + \text{NO} \rightarrow \text{NO}_2 \quad \text{(R2)}$$

$$\text{NO}_2 + h\nu \rightarrow \text{NO} \quad \text{(R3)}$$

$$\text{NO} + \text{O}_3 \rightarrow \text{NO}_2 \quad \text{(R4)}$$

$$\text{RP} + \text{RP} \rightarrow \text{RP} \quad \text{(R5)}$$

$$\text{RP} + \text{NO}_2 \rightarrow \text{SGN} \quad \text{(R6)}$$

$$\text{RP} + \text{NO}_2 \rightarrow \text{SNGN} \quad \text{(R7)}$$

where ROC denotes the reactive organic compounds, RP is the radical pool, SGN denotes the stable gaseous nitrogen products and SNGN stands for the stable non-gaseous nitrogen products. Temperature dependent rate constants are represented by standard Arrhenius expressions. The photolysis rates were parameterised by the following function, where a_k and b_k are appropriate constants.

$$J_k = \left(1 - 0.75N^{3.4}\right) a_k \exp\left(-b_k \sec\Theta\right) \quad (16)$$

The model uses the Emission Inventory of the Council of Budapest, the Hungarian National Emission Inventory and the inventory data of the European Monitoring and Evaluation Program (EMEP). The spatial distribution of the data are (1 km × 1 km), (20 km × 20 km), and (50 km × 50 km) for Budapest, Hungary and Europe, respectively. Special care was needed for the appropriate nesting of the three emission inventories. These data include the annual emission of NOx, CO, and non-methane hydrocarbons (NMHCs). The actual emission rates of species were calculated from the annual emission data using assumed typical daily emission time profiles.

3. THE NUMERICAL METHODS

The transport, and the physical and chemical interactions of species in the atmosphere can be described in two space dimensions by equation

$$\frac{\partial c_i}{\partial t} = -\frac{\partial (v_x c_i)}{\partial x} - \frac{\partial (v_y c_i)}{\partial y} + \frac{\partial}{\partial x}\left(K_x \frac{\partial c_i}{\partial x}\right) + \frac{\partial}{\partial y}\left(K_y \frac{\partial c_i}{\partial y}\right) + R_i + E_i - k_{di} c_i \quad (17)$$

The first two terms on the right hand side represent the transport (advection) of species by a wind field. The second two terms describe the diffusion due to turbulent eddies. The next two terms account for the chemical production and the emission of species. The last term denotes the dry deposition of pollutants.

The Method of Lines is very popular to solve advection–reaction problems. This approach reduces the set of PDEs in three independent variables (x, y, t) to a system of ordinary differential

equations (ODEs) in one independent variable, time. The two main steps of the Method of Lines are the spatial discretisation and the time integration. The spatial discretisation of the PDEs derived from the atmospheric transport-diffusion equation Eq. (17) is usually performed by using either the finite volume or the finite element approaches. Our model uses the flux limited, cell centred finite volume scheme of Berzins and Ware [3-6] on an unstructured triangular mesh. 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. The model domain is represented by an unstructured mesh of triangular elements surrounding each grid point, thus forming a small volume over which the solution is averaged. The system of ODEs can then be solved as an initial value problem using code SPRINT2D [3-6] with the Theta integration method. This method defines the numerical solution by equation

$$\mathbf{c}(t_{n+1}) = \mathbf{c}(t_n) + (1-\Theta) h \dot{\mathbf{c}}(t_n) + \Theta k \mathbf{F}_N \left(t_{n+1}, \mathbf{c}(t_{n+1})\right) \quad (18)$$

where $t_{n+1} = t_n + h$, $\Theta = 0.55$, and $\mathbf{F}_N$ represents the transport and the source terms.

4. SIMULATION RESULTS

The model was tested on the simulation of a photochemical air pollution 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 grid included large triangles outside Hungary (typical size was 100 *km*) and a rectangle containing a nested fine grid that covered Hungary. The typical grid size in the nested grid was 6 km. This initial grid is depicted in **Figure 1**.

The simulations started on 3rd August, 1998 at 0.00. The initial composition was 0.04 ppb for NO_2, 1 ppb for NO, 40 ppb O_3, 4 ppb for NMHCs. This is a typical summer midnight composition of surface air. Accurate estimation of the initial concentrations is not critical because after about one simulation day the calculation results converge to similar values.

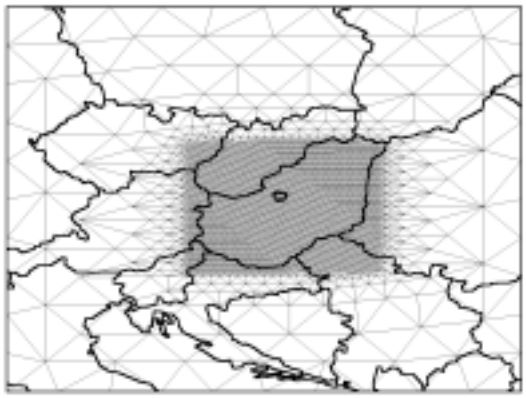

Figure 1. The initial nested grid.

Figure 2 shows the surface temperature in the modelled region on 4th August, 1998 at 12.00. It is well visible that in almost the whole Carpathian Basin the temperature is around 35 °*C*. The thermal atmospheric chemical reactions are fast at such a high temperature and it promotes the production of secondary air pollutants (like ozone) by chemical reactions.

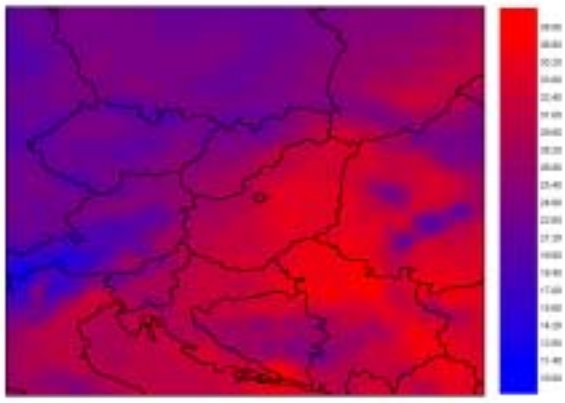

Figure 2. Surface temperature (°*C*) in the modelled region on 4th August, 1998 at 12.00.

Figure 3 shows the cloud coverage at the same time, which was below 2 % in almost the whole region. Similar cloud coverage patterns were obtained during the two simulated days, which means that the intensity of sunshine was high during these days, and therefore not only the thermal, but also the photochemical reactions were fast, allowing significant production of secondary pollutants.

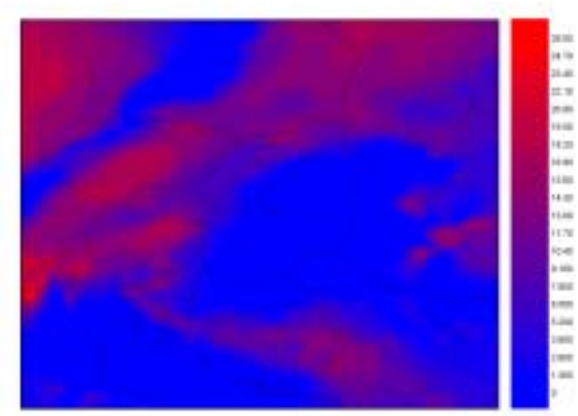

Figure 3. Cloud coverage on 4th August, 1998 at 12.00.

A typical simulation result is provided in **Figure 4.** This figure shows the wind field on 4th August, 1998 at 17.00. It is well visible that in Southeast Hungary the northwest wind meets an air flow, coming from the east. The result is a large eddy and an almost stagnating air flow over south-east Hungary. Figure 4. also shows the calculated ozone concentrations. Three ozone plumes are observable, which are the results of the air pollution of Prague (see the upper left corner), that of Vienna (see middle of the left side) and that of Budapest. The latter was enhanced by the meteorological situation, because the air that was coming from Budapest, polluted by nitrogen-oxides and hydrocarbons, was trapped in the stagnating air zone and therefore large amount of ozone could be produced by the chemical reactions in this region. **Figure 5** shows the predicted and the measured ozone concentrations at the K-puszta monitoring station from 0.00 of 1st August to midnight 4th August, 1998. Initially, the simulation results show transient effects, but the agreement between the measured and the simulated ozone concentrations is good in the last two days.

4. SUMMARY

An adaptive grid model has been developed to describe the formation of photochemical air pollutants for the Central European region. The modelled region covers a 1500 km × 1500 km area with Hungary in the centre. Vertical stratification of the troposphere, up to 3000 meter, is described by using four layers. The local wind speed and direction, relative humidity, temperature and cloud coverage were obtained from the weather forecast model ALADIN of the Hungarian Meteorological Service. Our air quality model uses the Emission Inventory of Budapest, the National Emission Inventory for Hungary and the EMEP inventory data for outside the country.

Photochemical air pollution is a highly non-linear phenomenon and therefore accurate calculations of concentrations are needed with good spatial resolution. Also, very short execution times are needed if the model is used in real-time applications, like a decision supporting tool during air pollution disasters. These requirements were met by using triangular unstructured grids in each layer. The model automatically places a finer spatial resolution grid in regions were higher numerical error is predicted. Grid resolution in critical places can be as fine as 6 km.

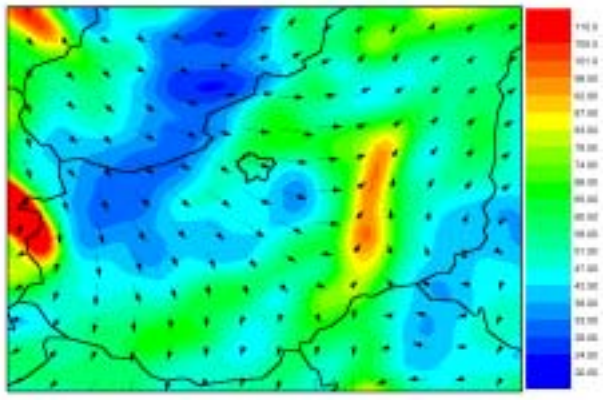

Figure 4. Surface layer ozone concentration (ppb) and wind field on 4th August, 1998 at 17.00 (on the 2nd day of the simulation)

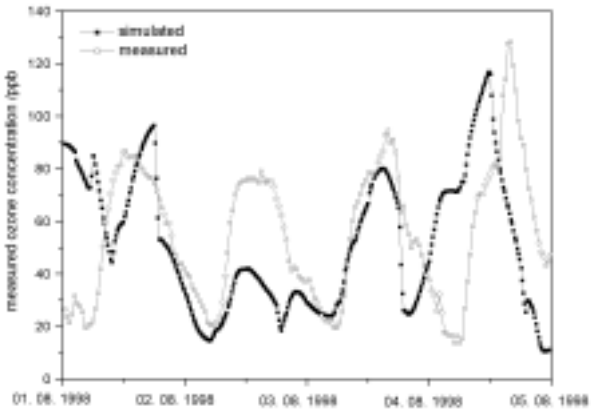

Figure 5. Comparison of simulated and measured ozone concentrations at the K-puszta monitoring station in the early days of August, 1998.

The model was tested on the 4th August, 1998 smog episode in Hungary. Simulated and measured ozone concentrations were compared at the K-

puszta monitoring station in Central Hungary from 0.00 of 1st August to midnight 4th August, 1998 and satisfactory agreement was found. Plotting the spatial distribution of the air pollutants in Central Europe shows that the primary air pollution of large cities, like Budapest, Vienna, or Prague, cause large concentration of ozone far away from the cities, in rural areas.

ACKNOWLEDGMENTS

The authors at ELTE acknowledge the support of OTKA Grant T043770.

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