CE ON AIR POLLUTION
ON 95

RMEN

ology, UK

os
oniki, Greece

ology, UK

ISORY COMMITTEE

. Lalas
.S. Longhurst
Lyons
Millan Millan
Moller
Tirabassi
Zannetti

Southampton, UK

ng,
loniki, Greece

e 44 which appears on the front

Air Pollution III - Volume 1

Air Pollution Theory and Simulation

EDITORS:

H. Power
Wessex Institute of Technology, UK

N. Moussiopoulos
Aristotle University of Thessaloniki, Greece

C.A. Brebbia
Wessex Institute of Technology, UK

Computational Mechanics Publications
Southampton Boston

1995

Efficient high resolution methods for air pollution models

A. Tomlin, J.M. Ware, J. Smith, M. Berzins, M.J. Pilling
Department of Fuel and Energy, School of Computer Studies and School of Chemistry, University of Leeds, Leeds LS2 9JT, UK

This paper investigates the use of time dependent adaptive gridding techniques for the solution of atmospheric reaction/flow problems. Preliminary 2D studies related to the effects of power station emissions on regional ozone levels have been carried out. The results show the importance of using adaptive grids in order to represent the interaction of the plume with background air over large distances. The adaptive mesh automatically reveals features of cross wind concentration profiles which would not be shown using the standard mesh sizes adopted in regional atmospheric calculations. As the level of adaptivity increases, and the mesh becomes locally refined in regions of steep spatial gradients, the total and peak ozone concentrations change quite significantly. The results demonstrate the advantage of using adaptive meshes in solving multi-scale atmospheric problems with spatially inhomogeneous source patterns.

1 Introduction

Computational models describing the chemical transformations and transport of species in the troposphere play an essential role in understanding the complex processes which lead to the formation of pollutants such as greenhouse gases, acid rain and photochemical oxidants. An accurate and detailed description of the distribution of pollutant concentrations is needed over large spatial regions in order to compare with field measurement calculations. Such comparisons, at high resolution, will inevitably promote a better understanding of the processes which lead to high concentrations in certain areas. One of the key issues which has to be addressed is the long range interaction between different emission sources, such as that between plumes generated from point sources and distributed urban and regional emissions in order to formulate abatement strategies.

Achieving high resolution in air pollution models is a difficult challenge because of the large number of species present in the atmosphere. For high resolution 3-dimensional calculations, detailed chemical schemes can become prohibitively large. In order to address the computational problems posed by complex atmospheric problems previous models have adopted two strategies. The first has been to keep the detailed chemistry necessary for understanding the many reactions of pollutants such as NO_x, SO_2 and volatile organic compounds (voc), and to use either 1-D trajectory models to simulate the reaction/transport problem [1], or coarse Eulerian grids. Such models have been essential in developing an understanding of how chemical species interact to form secondary pollutants such as ozone. They have not however, provided the spatial resolution which is needed to understand the complex

interactions between multiple sources, and require an oversimplification of the mixing processes that can occur. Other models use a simplified chemical scheme but with a high resolution grid, and are not able to assess the role of individual species on pollutant distribution. In order to achieve high spatial resolution and a detailed chemical description at the individual compound level, new modelling strategies are required.

Some steps have been taken in regional air quality models to include the use of variable sized grids, thus achieving better resolution in certain regions. Grid refinement in these telescopic models has been used in a predescribed way [2, 3, 4, 5]. This allows for refinement in, for example, high emission areas, but cannot take account of the spatial gradients resulting from the dispersion of pollutants through the atmosphere. Away from concentrated sources such models use large grids of up to 50 kilometres and therefore could still lead to inaccurate downwind profiles as the plumes are transported and averaged into the larger grids. This is a particular problem for species such as ozone, where the chemical time-scale of pollutant formation, is such that the main pollution episodes occur at very long distances downwind of both point and extended sources of photochemical precursors. Regions of steep spatial gradients of reactants, intermediates and products will move with time according to the wind-field and the spatial distribution of emissions. Given the non-linear nature of the chemical kinetics, reliable solution can only be obtained if the mesh can refine accordingly and there is therefore a need for the application of time dependent adaptive algorithms.

The present work describes how such algorithms, which automatically refine the mesh in regions of high spatial error, can improve on the telescopic approach. The aim is to show that the use of adaptivity could lead to a better understanding of the complex multiscale phenomena that arise in regional scale models. We have not attempted to develop a comprehensive regional model, but rather to apply a set of numerical modelling tools to a particular test case in order to demonstrate their advantages over traditional techniques. We study here the interaction of a power plant plume with background emissions. We will illustrate, by comparison with a base mesh of the order commonly found in regional scale models, how new features of the cross plume concentration profile are revealed by the adaptive solution.

2 Model equations

The general form of the equations used to describe atmospheric flow problems is the atmospheric diffusion equation:

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

where c_s is the concentration of the s'th compound, u and w are wind velocities, K_x and K_y are diffusivity coefficients and k_{1s} and k_{2s} are dry and wet deposition velocities respectively. E_s describes the distribution of emission sources for 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 where each is coupled through the nonlinear chemical reaction terms.

3 Numerical Methods

We now briefly describe the technique used for the discretisation and solution of the above p.d.e.s in two space dimensions. A more detailed discussion of the numerical methods can be found in references[6, 7, 8]

3.1 Discretisation on unstructured triangular meshes

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. This method, 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, typically time. The system of o.d.e's can then be solved as an initial value problem and the accuracy of the method depends on the discretisation scheme chosen. In the present application a finite volume method has been used to discretise in space, where the solution is represented as as series of piecewise constant elements. A cell centered scheme is used, where the p.d.e., is integrated over the triangular element and the divergence theorem is applied to convert the area integral over the element to a line integral over the edges of the triangle. The midpoint quadrature rule is then used to reduce each line integral to a flux evaluation at the midpoint of each edge.

A popular class of discretisation schemes are those based on using (approximate) Riemann solvers normal to the edges of the mesh to calculate the flow between mesh elements. How the solution values inside the Riemann solver are chosen imparts different properties to the scheme. A simple first-order scheme would just use the solution value associated with the mesh element in the Riemann solver. Berzins and Ware [8, 9] have developed a more accurate second order scheme that still maintains the physical range of solution around steep gradients and discontinuities. This is preferable to the first order scheme which is too numerically diffusive and smoothes out features in the solution. This sophisticated approach is only applied to the convective terms. A more traditional centrally differenced approach is used for the diffusive terms. The above spatial discretization scheme results in a system of o.d.e.s in time which are integrated using a modified form of the stiff o.d.e. solver DASSL method[10] with a Newton-Krylov iterative method[11] to solve the large systems of nonlinear linear equations.

3.2 Mesh Generation and Adaptivity.

The initial meshes are created from a geometry description using the Geompack [12] mesh generator. These meshes are then refined and coarsened using data structures that enable efficient mesh adaptation. Refinement is achieved by subdividing each triangle into four triangles for each level of adaptivity as shown in Figure 1. Triangles on the edge of the adapted region must be

bisected to avoid the appearance of hanging nodes. The criteria for adaption is based on a local spatial error estimate for the solution which 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, [8]. An integer refinement level indicator is calculated from the scaled error to indicate the number of times the triangle should be refined or derefined.

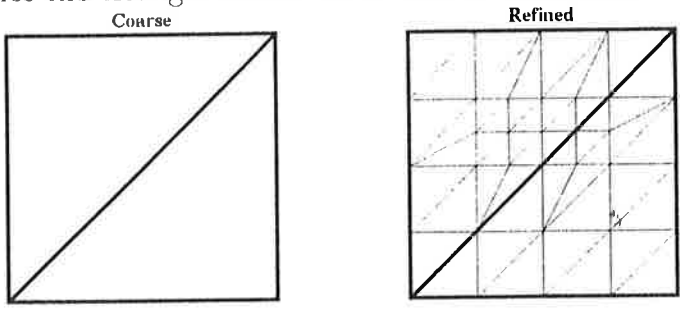

Figure 1: Method of refinement for unstructured triangular mesh

4 Application - plume dispersion

The test case describes the dispersion of a plume originating from a concentrated pollution source such as a power station chimney[13]. The purpose of choosing such an application is to look at a multi-scale problem and to compare the fully adaptive scheme described above with the sort of mesh generally used in regional scale models. We have not therefore used a comprehensive atmospheric model but have simply chosen to represent the main features which would commonly be found in such a model including slow and fast nonlinear chemistry, concentrated source terms, advection and diffusion.

4.1 Chemical Mechanism

For initial code development the simple scheme shown in Table 1 containing 10 species was used. Despite its simplicity it represents the main features of a tropospheric mechanism, namely the competition of the fast equilibrating inorganic reactions, and VOC chemistry occurring on a much slower timescale. The VOC reactions were represented by reactions of a single species, formaldeyde. The photolysis rates were parametrised as a function of the solar zenith angle θ in agreement with those used by Derwent et al. [1]:

$$J_i = a_i \exp(-b_i \sec\theta),$$

where i is the reaction number. Future developments of the code will include simplified chemical mechanisms which have been obtained from large comprehensive schemes via systematic reduction methods [14].

Source terms are included for NOx and formaldehyde. For the sake of simplicity the source term for formaldehyde is the same over all spatial points although it is possible to include unstructured emissions data by interpolation onto the triangular grid. The source term for formaldehyde is chosen so that the background concentration remains roughly constant.

Table 1 - Simplified chemical mechanism

	O_2		
NO_2	$\rightarrow$	$O_3 + NO$	$a_1 = 1.45 \times 10^{-2}$, $b_1 = 0.4$
$NO + O_3$	$\rightarrow$	$NO_2 + O_2$	$k_2 = 6.53\times 10^{-11}$
O_3	$\rightarrow$	$O^1D + O_2$	$a_3 = 2.0 \times 10^{-4}$, $b_3 = 1.4$
$O^1D + H_2O$	$\rightarrow$	$2OH$	$k_4 = 2.2 \times 10^{-10}$
$OH + NO_2$	$\rightarrow$	HNO_3	$k_5 = 1.5\times 10^{-11}$
$OH + CO$	$\rightarrow$	$CO_2 + H$	$k_6 = 2.2\times 10^{-13}$
$H + O_2 + M$	$\rightarrow$	$HO_2 + M$	$k_7 = 5.1 \times 10^{-32}\ (T/300)^{-0.9}$
$HO_2 + NO$	$\rightarrow$	$NO_2 + OH$	$k_8 = 8.3\times 10^{-12}$
$O^1D + M$	$\rightarrow$	O_3	$k_9 = 2.0\times 10^{-11}\ e^{(100/T)}$
$HCHO + OH$	$\rightarrow$	$HO_2 + CO + H_2O$	$k_{10} = 1.0\times 10^{-11}$
$HCHO$	$\rightarrow$	$2HO_2 + CO$	$a_{11} = 3.32\times 10^{-5}$, $b_{11} = 0.56$
$HCHO$	$\rightarrow$	$H_2 + CO$	$a_{12} = 5.54 \times 10^{-5}$, $b_{12} = 0.79$

The power station is taken to be the only source of NOx and the concentration in the chimney corresponds to an emission rate of NOx of 400 kg hr^{-1}. We have considered only 10% of the NOx to be emitted as NO_2, the rest being NO. Representations of horizontal transport were kept simple so that the effect of grid resolution could easily be seen We have assumed a uniform and constant wind speed of 5ms^{-1} in the x-direction. The eddy diffusion parameter has been set at $3\times10^4 m^2s^{-1}$ in the y-direction only, for all species. Deposition has not been included.

The initial mesh was generated with only 100 elements. This resulted in a largest element with a side length of about 50km, i.e. comparable to the size of mesh generally used in regional scale atmospheric models. Close to the chimney the base mesh was much smaller, of the order of 1-5kms. This initial approach of choosing a smaller mesh close to the source is similar to the telescopic approach. For each calculation the refinement indicator was based on the spatial error in NO. In general large spatial errors occur where steep spatial gradients exist so that a large degree of refinement is expected close to the chimney. For sources which behave almost like point sources steep spatial gradients are likely to persist down to very high levels of refinement. This would have the consequence that the number of elements on which the p.d.e.s had to be discretised would become prohibitively large. Therefore the maximum level of refinment was limited to 3. The actual size of the mesh elements used at each computational step depends on the size of elements in the initial mesh and on the maximum level of refinment allowed.

5 Results

Each run has been carried out over a period of 48 hours so that the diurnal variations can be observed. We present here only a selection of the results which illustrate the main features relating to the adaptivity. Figure 2a shows contour levels for O_3 for a level 2 solution on the afternoon of the second day when peak concentrations occur. In this picture the nodes of the polygons represent triangle centres in the unstructured mesh. The initial domain was 500 x 300km and the figure shows that maximum refinement occurs close to

the chimney stack and along the edges of the plume. This reveals heightened ozone concentrations along the plume edges, a feature which was not revealed by the base mesh. In Figure 2b we present a cross plume profile of the NO_2

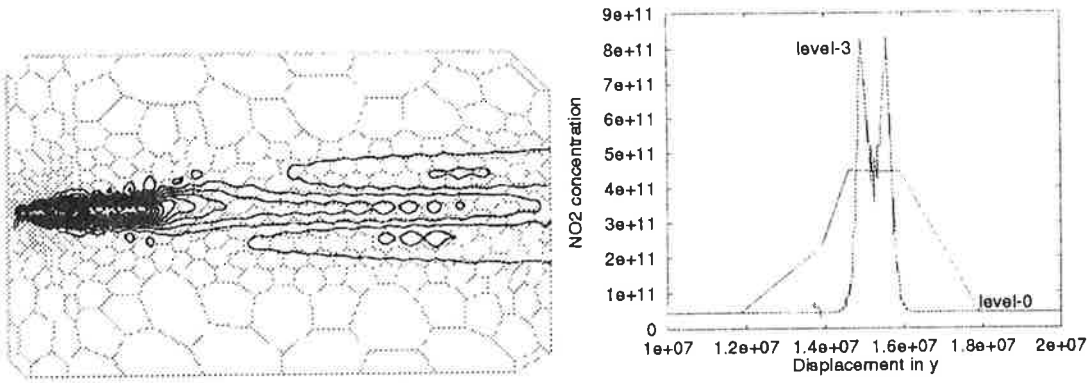

Figure 2: Contour levels for ozone and the effect of adaptivity on NO2 levels.

concentrations at a distance of 5km downwind of the chimney stack. The figure clearly shows the features at the edge of the plume which are revealed by the adaptive solution. From the base mesh, where the distance between elements along the y-axis is 20km, it appears that the concentration of NO_2 rises to a peak in the centre of the plume. If the mesh is refined to higher levels then we start to see the true structure of the plume emerging. From the level 3 solution we can see that the peak concentrations are actually found along the edges of the plume and that the concentration of NO_2 drops at the plume centre. The area under these curves can easily be calculated and it is found that there is a 30% difference between the level 0 and the level 3 solutions. This shows that not only the peak concentrations, but the total integrated concentrations are very different for the different levels of adaptivity. It is clear therefore that using a coarse grid in regions of steep spatial gradients can lead to an over estimate of total pollutant concentrations.

6 Discussion

We have shown above that there are key features which cannot be represented by the coarse meshes generally used in regional scale models. Steep concentration gradients close to the stack, such as the variations in NO_2 concentration, might be captured by telescopic models which provide a higher resolution in concentrated emission areas. However, features such as the peak ozone concentrations, which occur at the edges of the plume for a long distance downwind of the source, may not be captured by such techniques. For example, there is a strong possibility that peak ozone levels will occur in low emission regions such as rural areas. Telescopic models would not refine the mesh in such regions and a fully adaptive algorithm provides a good way to resolve such features. We have also shown that as well as inaccurately describing the position of peak concentrations, coarse meshes can also lead to inaccurate estimates of average or integrated concentration levels. This is important if we are to use models to predict total budgets for individual

species. One of the key reasons why such errors appear is the nonlinearity in the chemical reaction rates. Unless we are refining to very high levels the nonlinear chemistry will be mesh dependent. Since it is impossible in atmospheric models to reach a uniform level of refinement where convergence can be achieved, then this will almost certainly be true in some regions of the model, particularly regions where steep gradients occur.

For example, consider the simple nonlinear reaction, $A + B \rightarrow C$. If we compare the value of the rate of this reaction k[A][B] for a single element and an element which has been refined into two sub-elements, we can see that the two rates are not generally equal. The concentrations of A and B in the large element are assumed to be the average of that in the two smaller elements:
$A_{avg} = 1/2\ (A_1 + A_2)$ and $B_{avg} = 1/2\ (B_1 + B_2)$.
The reaction terms for the large element can now be evaluated as:
rate = $k\ A_{avg}B_{avg} = k/4(A_1 + A_2)(B_1 + B_2)$
Considering the two smaller elements, the average reaction term is given by:
rate = $k/2(A_1B_1+A_2B_2)$.
The above two expressions will only be close in value if the concentrations in the two cells are almost equal. In regions of steep spatial gradients the solution values in neighbouring cells will be quite different, so that the reaction terms will be different for the coarse and fine meshes. These spatially varying reaction rates may then feed back to further increase the differences between coarse and fine mesh average concentrations. The interaction of mesh refinement and nonlinear chemistry may thus have a significant effect on solution profiles.

7 Conclusions and future directions

We have presented in this work a simple example in order to illustrate the importance of adaptive methods for the solution of atmospheric models. Using a case study of a power station plume we have illustrated the need for adaptive methods in order to accurately represent the interaction of a concentrated pollution source with background levels. The adaptive solution reveals features such as peak levels of NO_2 and O_3 which could not be detected using a coarse mesh. The change in mesh refinement also resulted in a change in overall or integrated concentration levels. This indicates that because of strongly nonlinear terms in the chemical reaction rates, the source terms in the p.d.e. will be mesh dependent and to reduce the effects of the nonlinearity it is important to refine the mesh at least in regions of steep spatial gradients. This has been partially addressed by the telescopic methods presently used in air quality models. However, the present test case has shown that steep gradients can occur downwind from the source, for example the change in ozone concentrations along the edges of the plume. Adaptive algorithms provide and ideal method of achieving accuracy in such regions and can do so in an automatic way.

The work here is preliminary in terms of using adaptive methods in a full air pollution model although it demonstrates the importance of high resolution meshes. Questions of whether adaptivity is necessary in the vertical direction, and whether to use structured or unstructured meshes have still

to be answered and the authors intend to address these questions in their future work.

References

[1] Derwent R. G. and Jenkin M.E. (1990) *Hydrocarbon involvement in photochemical ozone formation in Europe* AERE-report-R13736

[2] Moussiopoulous N. (1994) The EUMAC zooming model *Eurotrac94 - ISS Garmisch-Partenkirchen*

[3] Jakobs H.J. (1994) *EUROTRAC94 - report*

[4] Sunderam S., Logan J.A. and Wofsy. S.C. A Regional Scale Model for Ozone in the United States With Subgrid Representation of Urban and Power Plant Plumes. *Journal of Geophysical Research*, 95:5731–5748, 1990.

[5] Sillman S., Logan J. A., Wofsy S. C. (1990) *J. Geophys. Res.* **95**, 1837

[6] Berzins M. , Dew P.M. , and Furzeland R.M. Developing software for time-dependent problems using the method of lines and differential algebraic integrators. *Appl. Numer. Math.*, 5:375–397, 1989.

[7] Berzins M., Lawson J., and Ware J.M. Spatial and Temporal Error Control in the Adaptive Solution of Systems of Conservation Laws. In R. Vichnevetsky, D. Knight, and G. Richter, editors, *Advances in Computer Methods for Partial Differential Equations VII*, pages 60–66. IMACS, 1992.

[8] Berzins M.,(1995) Temporal error control in the Method of Lines for convection dominated p.d.e.s. *SIAM J. Sci. Comp.* May 1995.

[9] Berzins M. and Ware J.M. (1995). Positive cell-centered finite volume discretization methods for hyperbolic equations on irregular meshes. *Appl. Numer. Math.*, 16:417–438, 1995.

[10] Brenan K.E. , Campbell S.L. and Petzold L.R. (1989). Numerical Solution of Initial Value Problems in Differential-Algebraic Equations. *North Holland, New York.*

[11] Seward W.L. and Jackson K.R., (1993). Adaptive linear equations solvers in codes for large stiff systems of o.d.e.s. *SIAM J. Sci. Comp. 14*, 3, 800–823.

[12] Joe B. and Simpson R.B., (1991). Triangular meshes for regions of complicated shape, *Int. J. Numer. Meth. Eng. 23*, 987–997.

[13] Emissions of radiative gases and ozone precursors from coal use. Progress Report No 1, Contract No JOU-CT92-0229, Joule 2 Program, C. E. C.

[14] Tomlin A.S. , Pilling M.J., Turányi T., Merkin J.H. and Brindley J. (1992). *Combustion and Flame*, 91:107.