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ABSTRACT

The coupled, non-linear system of continuity equations describing an air pollution model with non-linear chemistry is solved numerically using finite differences, finite elements and pseudo-spectral methods. A smoothing procedure is proposed to avoid negative concentrations. Several tests are performed: single puff transported parallel and not parallel to the co-ordinate axis, two puffs along parallel lines, a rotating puff and a rotating plume. The accuracy of the results of advection+chemistry+smoothing calculations is evaluated through the comparison with the results of box model calculations.

The concentration at the peak of the puff is compared and in the case with advection only, chemistry only and advection+chemistry after 24 hours integration. The relative errors made in the case where advection+smoothing+chemistry is applied does not exceed 5%. These errors are of the same magnitude as the errors at the peak of the puff for the case where advection only is performed.

For runs with discretization by second order finite differences, it is well known that the advection algorithms are neither able to preserve the shape of the puff nor to preserve the maximum concentrations in the puff. Our runs only confirmed this conclusion.

For runs with the Smolarkiewicz algorithm, the results are slightly better than with the algorithm based on second order finite differences. However, the improvement of the accuracy is negligible compared with the increase of the computing time spent.

The runs with the finite elements (CHAPEAU) advection algorithm shows that the accuracy of this advection algorithm is worse than that of the pseudospectral advection, but it is faster than the latter algorithm with regard to computing time.

The second order finite differences algorithm is about 5 times faster than the pseudospectral algorithm when the advection time only is taken into account. In the same situation the Smolarkiewicz algorithm is only a little better than the pseudospectral algorithm, while the finite elements (CHAPEAU) algorithm is about 2.5 times faster. The differences are less when advection+smoothing+chemistry is applied.

CONTENTS

	Page
ABSTRACT	1
1 INTRODUCTION	4
2 TREATMENT OF NEGATIVE CONCENTRATIONS	5
2.1 Matching advection with chemistry	5
2.2 Numerical treatment of the advection	7
2.3 Numerical treatment of the chemical part	9
2.4 Test Examples	10
3 NUMERICAL RESULTS	14
3.1 Results obtained with the pseudospectral method	14
3.2 Runs with other advection algorithms	23
3.3 Comparison of the computing times spent for the different algorithms	27
3.4 Further work	29
4 REFERENCES	29

AIR POLLUTION MODELS WITH NON-LINEAR CHEMICAL REACTIONS

1 INTRODUCTION

The continuity equation is the mathematical basis for the modelling of pollutant transport, transformation and deposition: The equation is used on the following semiempirical form

$$\frac{\partial c_i}{\partial t} + \sum_j \frac{\partial u_j c_i}{\partial x_j} = \sum_j \frac{\partial}{\partial x_j} (K_j \frac{\partial c_i}{\partial x_j}) + R_i + S_i, \quad i = 1, 2, \dots, q \quad (1.1)$$

q being the number of species studied (9 in this model), c_i , R_i and S_i being the concentration, the chemical reaction term and the source term for the i 'th pollutant, respectively, u_j and K_j being the wind velocity and the diffusion coefficient along the j 'th co-ordinate axis. In this case the model is two-dimensional ($j=1,2$). The reaction term R_i also includes ground removal.

The continuity equations are coupled, forming in general a non-linear system through the chemical reaction terms. This system can be solved using finite differences, finite elements or pseudo-spectral or other numerical methods for the space discretization.

In the PHOXA-project, where an Eulerian model for studying photochemical reactions and transport of air pollutants in Northern and Central Europe is developed, the advection terms in (1.1) are treated numerically using the SHASTA method (Boris and Book, 1973). As an alternative the more accurate Zalesak algorithm (Zalesak, 1979), which is more expensive with regard to computing time, is used.

In order to facilitate the computational process, splitting and fractional time-steps are often used. For example, in the 3-dimensional Eulerian model for studying the regional transport of photochemical oxidants and their precursors developed by Carmichael et al. (1986), (1.1) is split into one dimensional sub-models that are semi-discretized by finite elements and then Crank-Nicholson fractional time steps are successively performed. A linearization procedure is applied to the chemical part and the linearized chemical part is solved analytically.

One of the schemes developed by Smolarkiewicz (Smolarkiewicz, 1983) has been adopted in the treatment of the advection part of the acid deposition model developed at NCAR (Chang et al., 1987). One of the schemes proposed by Smolarkiewicz (1985) is also used in the present (see Section 2).

The numerical solution of the continuity equations should not contain negative components when non-linear chemical terms are involved in the model. At the same time the algorithm used in the treatment of the advection terms should not produce excessive numerical diffusion. It is difficult to satisfy these requirements simultaneously. The straightforward attempt to avoid negative concentrations by a low order finite difference scheme leads to a situation where the numerical diffusion is significant. This will be demonstrated in the following sections.

The treatment of the continuity equation in presence of photochemical reactions by different space discretization algorithms; finite differences, finite elements, a pseudospectral algorithm and one of Smolarkiewicz' algorithms are discussed here.

2 TREATMENT OF NEGATIVE CONCENTRATIONS

The matching of the advection part and the chemical part of the continuity equation is a difficult numerical problem when the chemistry is non-linear.

A solution to the problem and numerical tests of its accuracy and efficiency are discussed in this section.

2.1 MATCHING ADVECTION WITH CHEMISTRY

The discretization of the advection part of the continuity equation usually leads to some negative concentrations except in the case where an artificial diffusion is introduced either directly or implicitly. A good numerical method suitable for the advection part and which does

not introduce artificial diffusion, will produce negative concentrations that are small and remain small during the whole integration process. If the numerical method produces negative concentrations that are small and remain small and if the chemical reactions are linear, the negative concentrations can be tolerated. This is not the case when non-linear chemical reactions are introduced. Then the negative concentrations will normally cause instability and, therefore, must be avoided. This can be achieved by the use of different kinds of smoothing or by introducing artificial diffusion in some way. The first procedure is the most attractive one, because small negative concentrations appear only locally. Negative concentrations occur as a rule only in regions where the concentrations are small, or even equal to zero, and are due either to truncation errors, to rounding errors or both. It is therefore sufficient to perform smoothing only in regions where negative concentrations appear. On the other hand, if artificial diffusion is introduced, then very often this will also affect the regions with high concentrations, which may be undesirable. We have chosen the first approach and developed a simple procedure by which smoothing is carried out only in the regions with small concentrations, while the high concentrations remain untouched.

The following smoothing is performed:

Step 1 Calculate the quantity GMAX equal to the maximum concentration of the s 'th compound obtained after the advection step. Set $SFACT = 0.05 * GMAX$.

Step 2 Denote by Cmean the set of all concentrations that are smaller than SFACT. Calculate the sum of the elements in set Cmean and denote it by GMEAN. Let the number of elements in set Cmean be IMEAN. Set the quantity GLAT equal to the maximum of $GMEAN/IMEAN$ and $0.01 * SFACT$.

Step 3 Set all concentrations of the compound under consideration that are smaller than the quantity GLAT equal to GLAT.

This smoothing is by no means perfect. It is not mass conservative, but it is simple. However, the advection method to which the smoothing algorithm is attached should be a good one, in the sense that when the

smoothing is not carried out, and when only chemical reactions are involved, the advection method produces small negative concentrations that remain small during the whole computation. The pseudospectral discretization of the space derivatives of first order is an example of such a method. A short description is given in the next paragraph of some numerical methods that are often used in the numerical treatment of the advection part of models for long-range transport of air pollutants.

2.2 NUMERICAL TREATMENT OF THE ADVECTION

The smoothing described in the previous paragraph has been tested in connection with four numerical methods for handling the advection. The basic properties of these algorithms are outlined below.

The pseudospectral algorithm. Approximation of the value of the first order space derivative at the grid points is obtained by trigonometric interpolation. Fast Fourier Transforms are used to accelerate the computation. The algorithm was proposed by Kreiss and Oliger (1972), see also Orszag (1971) or Fornberg (1975), and has been used for air pollution studies by Prahm and Christensen (1977), Zlatev et al. (1983a,b,c) and Zlatev (1985). An advantage of the pseudospectral algorithm is the preservation of high concentrations during advection. There are two disadvantages; the non-physical requirement of periodicity of the concentrations on the boundaries of the space domain under consideration, and the computing time requirement. The algorithm is much more time consuming than most of the other algorithms.

Second-order central finite differences. This is a classical algorithm. Central differences are used in the discretization of the first order space derivatives at the grid points. To get an approximate value of the derivative under consideration at a given grid-point, the values of the concentration in two neighbour grid-points are applied. On the boundaries often one side first-order differences are used. The algorithm is cheapest with regard to computing time among the four algorithms considered here. Concentration peaks are strongly smoothed out when central differences are used, and the artificial smoothing

worsens with the integration time. More details about this algorithm can be found, for example, in Orszag (1971).

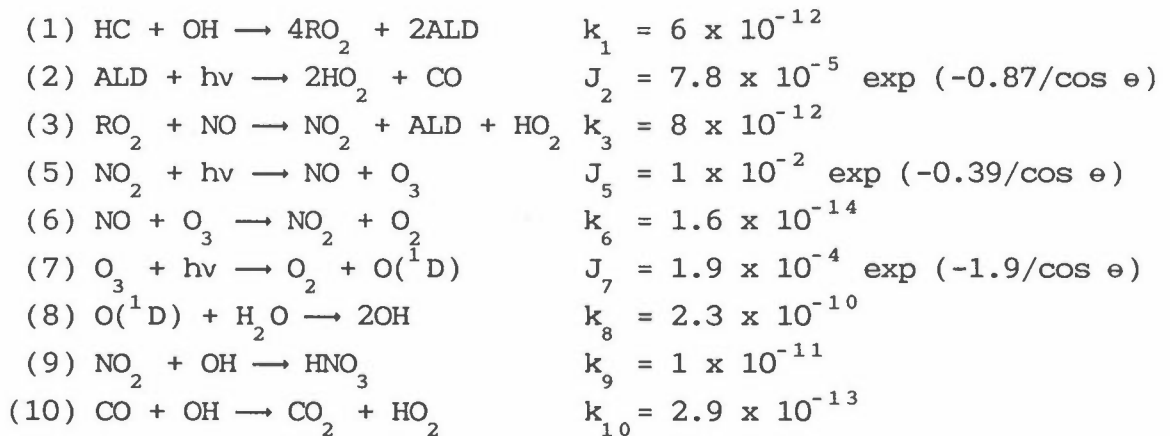
Finite elements. The particular algorithm used by Chock and Dunker (1983), see also Pepper and Baker (1979), or Pepper et al., (1979), is used in the discretization of the first-order derivatives (but the time-integration algorithm used in the publications referenced above is for the present study replaced by the time-integration algorithm described by Zlatev (1984) and Zlatev et al. (1984)). The discretization is based on piece-wise linear finite elements defined along the coordinate axes, which means that the two-dimensional advection operator is split into two one-dimensional operators. Boundary conditions are easy to establish. The method is more time-consuming than the use of central differences, but is cheaper than the pseudospectral algorithm. Its accuracy is much better than the accuracy of the central differences, but it is not as good as that of the pseudospectral algorithm.

The Smolarkiewicz algorithm. This algorithm is proposed by Smolarkiewicz (1985). The first-order space derivatives are discretized with central differences. The space domain is considered as two-dimensional (in the second algorithm splitting is applied in order to consider the derivatives on grid-lines parallel to the co-ordinate axes). The classical forward Euler time-integration algorithm is modified in order to achieve order two, by adding several correction terms containing approximations to the concentrations from one time level only, for the computation of the concentrations at the grid points of the next time level. Thus, this is a one level scheme, which is important in the solution of non-linear problems but is not important when the numerical treatment of the advection part is separated from the numerical integration of non-linear chemical terms. The scheme is much more time consuming than the ordinary central differences. It is ~~nearly as expensive as the pseudospectral algorithm.~~ Its accuracy is not much better than the accuracy of the central differences. Also here the grid-points close to the boundaries have to be treated in a special way.

2.3 NUMERICAL TREATMENT OF THE CHEMICAL PART

The chemistry in the model is defined to be as simple as possible so that the number of equations (components) could be kept at a minimum at the same time as processes known to be the most difficult ones to handle numerically, were included. The chemistry is not claimed to be realistic in the sense that the computed concentrations should agree with atmospheric measurements. The chemical scheme is defined to be realistic for numerical testing. The scheme is given in Table 1.

Table 1: Chemical scheme used in the model



θ is the solar zenith angle. k 's are in units $\text{cm}^3/(\text{molecule} \times \text{s})$, J 's in s^{-1} . HC is a lumped hydrocarbon molecule, ALD formaldehyde.

The same algorithm (discussed in Hesstvedt et al., 1978) is used in the treatment of the chemical part of the model in connection with all four advection algorithms. This is possible, because the chemical part is handled independently from the advection part. The continuity equation is split into several parts, and each part is integrated separately at each time-step. The main principles that are applied in the integration of the chemical part are discussed in the following. Consider an arbitrary time step and assume that the advection part has been performed by any of the four methods discussed in the previous paragraph. The chemistry part of the continuity equation can be written in the form

$$dc/dt = -Qc + P \quad (2.1)$$

where Q and P vary in space and with time and depend on the concentrations of the other species at the grid-point and the time under consideration. This means that the equation (2.1) is in general non-linear, although it looks linear. It is clear that in our case with ten species and 32×32 grid points, we have to solve $10 \times 32 \times 32 = 10240$ such equations at each time step. The solution is found by the use of three different algorithms. An attempt to choose automatically the optimal algorithm for each of the 10240 equations is carried out during the integration process by examining the stiffness of the equation system. This is done by checking the magnitude of Q . If Q is sufficiently small, then the equation (2.1) is solved by the classical forward Euler formula. If Q is not very small but not very large either, then (2.1) is assumed to be linear (its coefficients are considered as constants) and the exact solution of the linearized form of (2.1) is used as an approximation to its real solution. If Q is very large, then (2.1) is replaced by $-Qx + P = 0$ and an approximation to the solution is obtained by solving the latter equation. The criteria actually used are given in Hesstvedt et al. (1978). This algorithm is numerically very robust and provides good results. The disadvantage of the algorithm is that it uses too many IF statements in the FORTRAN code (at each chemical time step and for each of the 10240 equations the magnitude of Q must be checked to select the proper sub-algorithm). Therefore the algorithm performs poorly on a vector computer. We are considering some ways to improve the algorithm for use in vector machines.

2.4 TEST-EXAMPLES

Several experiments are performed to evaluate how well the smoothing procedure works and how accurate the numerical solution is. The tests are selected to give solutions by solving the equations analytically or by using physical arguments.

The following test-examples are used in the experiments:

- (1) Single puff advection parallel to one of the co-ordinate axes.
- (2) Single puff advection not parallel to any of the co-ordinate axes.
- (3) Advection of two puffs with parallel trajectories, not necessarily parallel to one of the co-ordinate axes.
- (4) A rotating puff (proposed simultaneously by Molenkamp, 1968, and Crowley, 1968).
- (5) A rotating plume with a trajectory identical to that of the rotating puff (see (4)).

If advection only is performed, then the solution of each of these test-examples is well-known. If both advection and chemistry are carried out, then a good approximation of the solution can be found by applying a "box-model" in which the initial concentrations are defined in an appropriate way. In a box-model the continuity equation is solved with chemical terms only. To illustrate how this works, let us consider test (3) with a constant wind velocity. Assume that the highest concentration at time t_1 is c_1 and is located at the grid-point (x_1, y_1) . At time t_2 the same concentration c_1 is moved to the grid-point (x_2, y_2) if advection only is applied. If chemistry only is applied (the box-model), then at time t_2 the concentration at grid-point (x_1, y_1) is changed (due to the chemical reactions) from c_1 to c_2 . Now it is clear that if both advection and chemistry are carried out, then the solution at time t_2 and at grid-point (x_2, y_2) should be c_2 . Moreover, it is also clear that if this is not so, then the only reason for the discrepancy is the smoothing algorithm. Thus, the tests can be applied to check the influence of the smoothing algorithm on the results of the global method.

These arguments are valid for the first four test-examples only. But the behaviour of the plume in the fifth test-example can be predicted by using physical arguments. Thus, also the fifth test-example gives

some information about the performance of the combination advection + smoothing + chemistry.

Some results obtained for the solution of the rotation test are presented in the next section. The rotation test is a representative test-example, which is commonly used in tests with advection algorithms. This is probably the first application of this test-example in the verification of a combined advection-chemistry scheme.

The initial distribution used in the rotation test-example for the concentration of nitrogen oxide is given in Fig. 1. It is seen that the center in the puff is located at the grid-point (8,16). Background concentrations (2.5% of the highest concentration; rounded to 2 in the figure) are assumed in the whole domain outside the area where the puff is located. The distance between two grid-points is 150 km along both co-ordinate axes. The wind-velocity field is defined so that a full rotation is performed in 24 hours. The size of the time step in the advection part was 150 s in most of the runs. This means that the rotation is completed in 576 advection steps. The chemical part was integrated with a step size of 30 s (five chemical steps are performed for each advection step).

If advection only is performed, the accuracy is checked by comparing with the field in Fig. 1 for nitrogen oxide and the corresponding fields (not given here) for the other species. The box-model (chemistry only) was run for 24 hours with a step size of 30 s and the results were compared with the results of the combined advection-chemistry scheme. Runs with different advection algorithms were performed both in the mode where advection only was performed and in the mode where advection + smoothing + chemistry was applied. The numerical results from these runs are given in the next section.

all concentrations are divided by the initial concentration and multiplied by 100
 the largest concentration is: 1.0000×10^{11}

the initial distribution is given in this plot

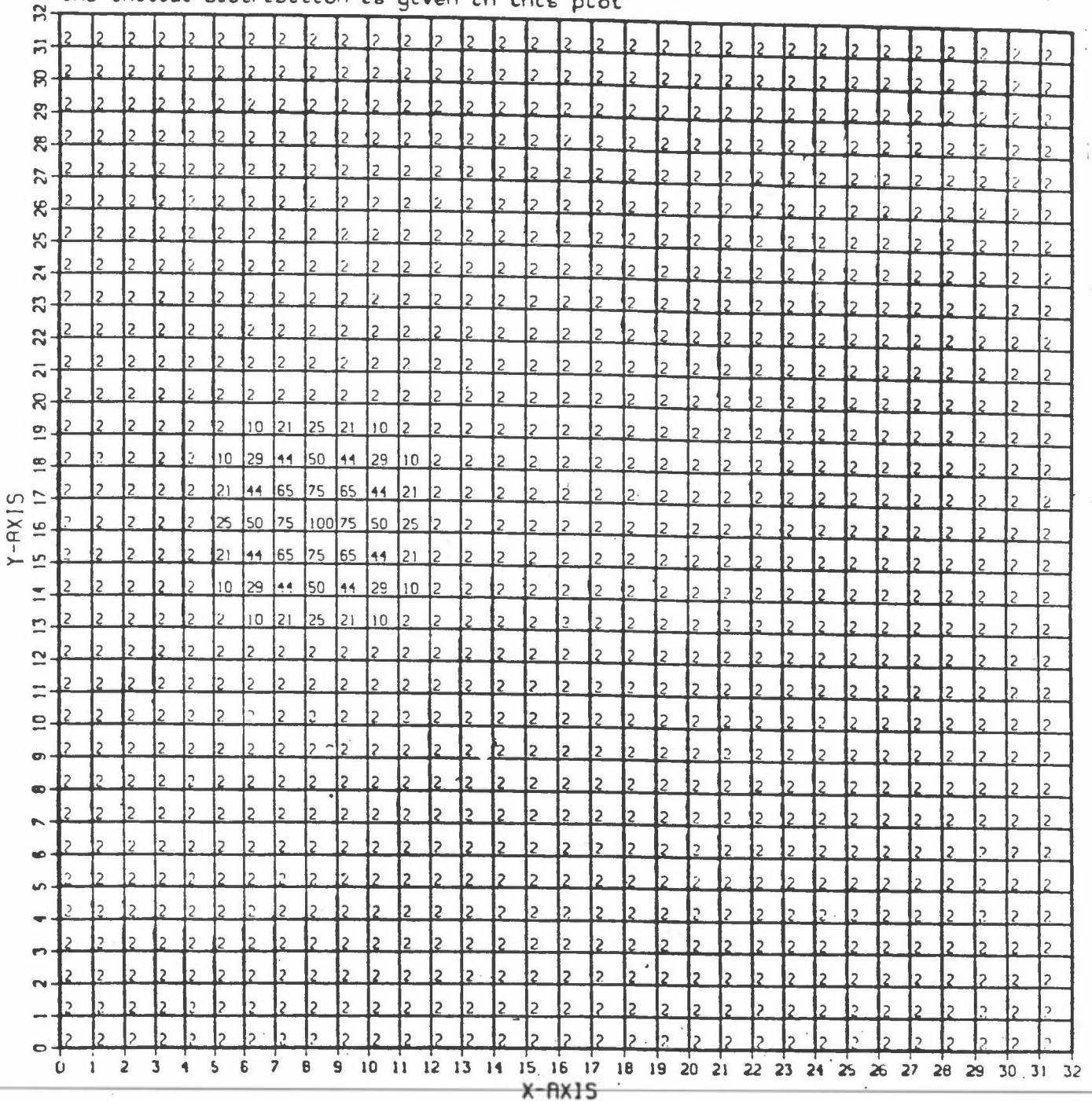


Figure 1: The initial field for nitrogen oxide in the rotation test and scaled by 10^{11} (molecules/cm³).

3 NUMERICAL RESULTS

3.1 RESULTS OBTAINED WITH THE PSEUDOSPECTRAL METHOD

Most of the experiments carried out were performed applying the pseudospectral algorithm to the advection part. A good performance should be expected (with regard to the accuracy achieved) when the pseudospectral algorithm is used with a good smoothing device. Therefore, the pseudospectral algorithm is a good test for the smoothing device introduced in the previous section. The results obtained in the rotation test will mainly be discussed here, but it should be emphasized that the conclusions made are based on results obtained in all five tests listed in the previous section.

The results of the rotation test are presented in three-dimensional plots. The initial distribution for nitrogen oxide is given in Fig. 2. The result obtained when one rotation only is performed by using advection only is shown for nitrogen oxide in Fig. 3. One should study very carefully Fig. 2 and Fig. 3 in order to discover the discrepancies between them. The results obtained for the box-model (run separately for each grid-point) are shown in Fig. 4. It is seen that the chemical reactions (i) change completely the shape of the pattern when nitrogen oxide is considered and (ii) change the magnitude of the concentration at the peak of the puff with several orders. Therefore, it should be expected that the numerical algorithms perform poorly in this case. It is seen, Fig. 5, that the results obtained by advection + chemistry are not as good as the results obtained with chemistry only, Fig. 4. The smoothing procedure is satisfactory, however. The differences arise because of (i) and (ii)

Also for nitrogen dioxide there is a change in shape when rotation + chemistry is performed, and the peak concentration drops by several orders of magnitude. The changes are smaller than for nitrogen oxide, however, and the comparison between chemistry only for 24 h (Fig. 6) and rotation + chemistry (Fig. 7) is much better than for NO.

It should be mentioned that the initial distribution for nitrogen dioxide is quite similar to that of nitrogen oxide shown in Fig. 2. If

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100 :

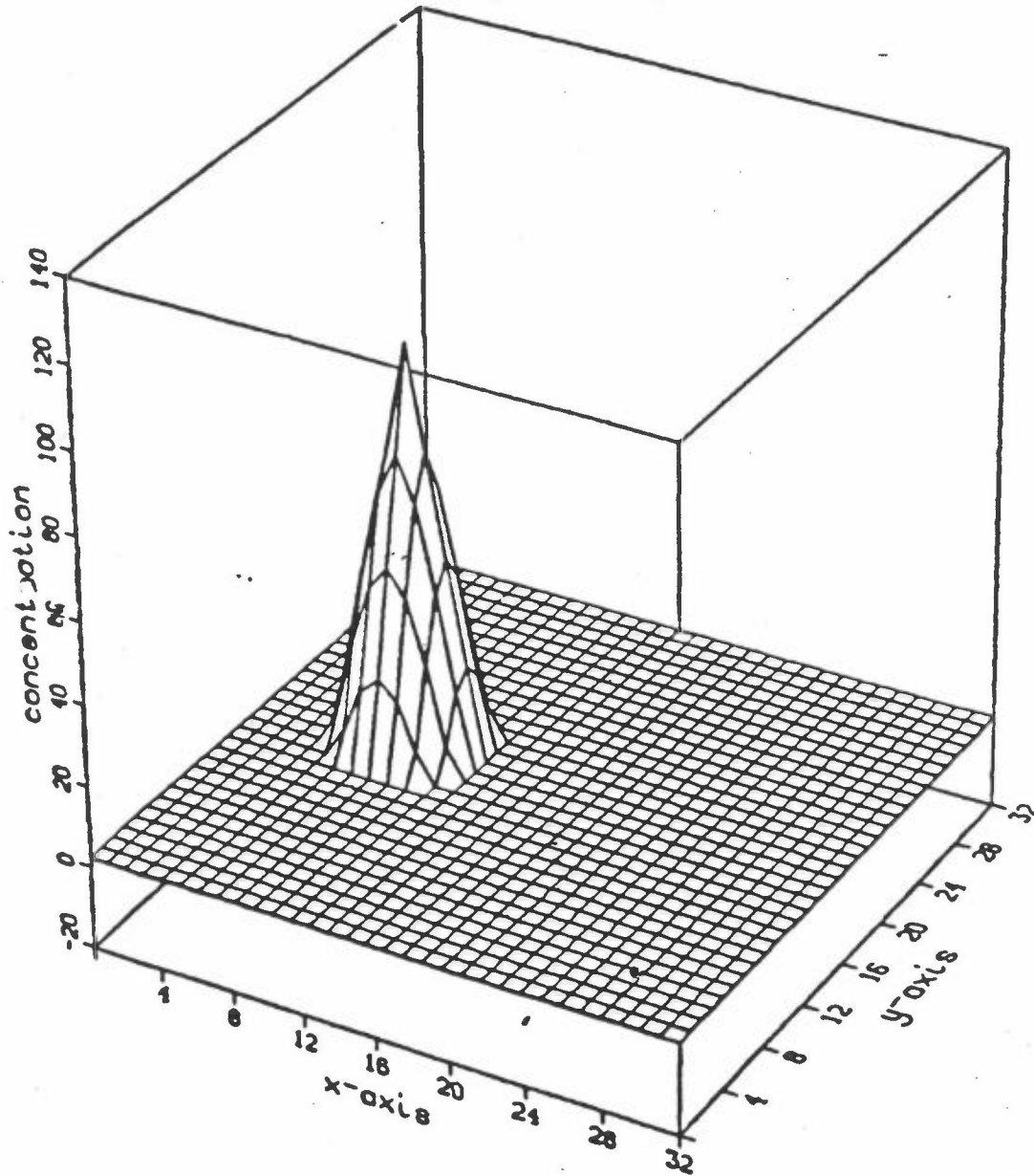


Figure 2: The initial distribution for nitrogen oxide, scaled by 10^{11} (in molecules/cm³).

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 1.0000×10^{11}

one rotation when advection only is performed

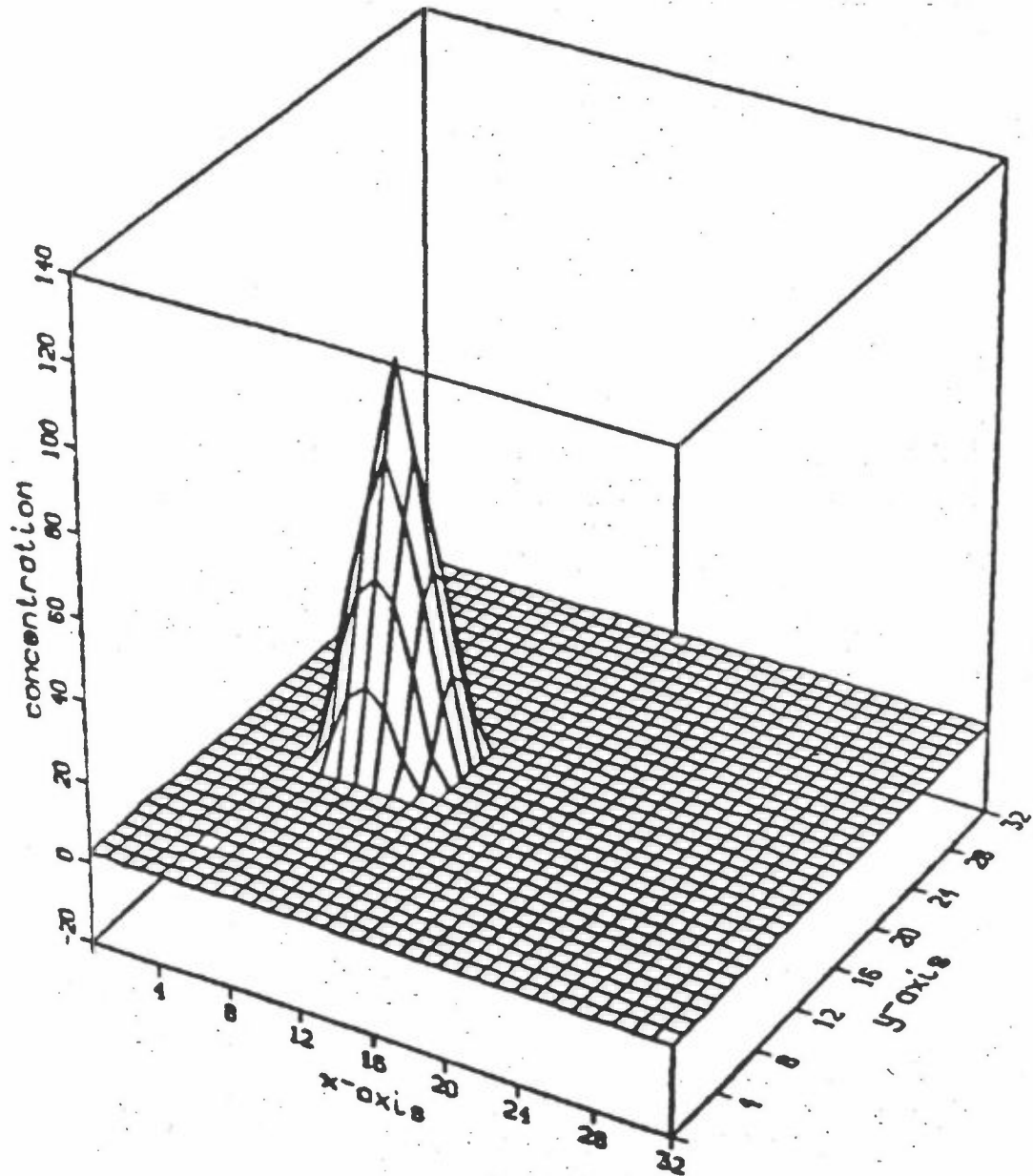


Figure 3: The solution for nitrogen oxide when advection only is carried out. The advection algorithm used is the pseudo-spectral algorithm (concentrations scaled by 10^{11} , in molecules/cm³).

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 1.0564×10^6

one rotation when chemistry only is performed

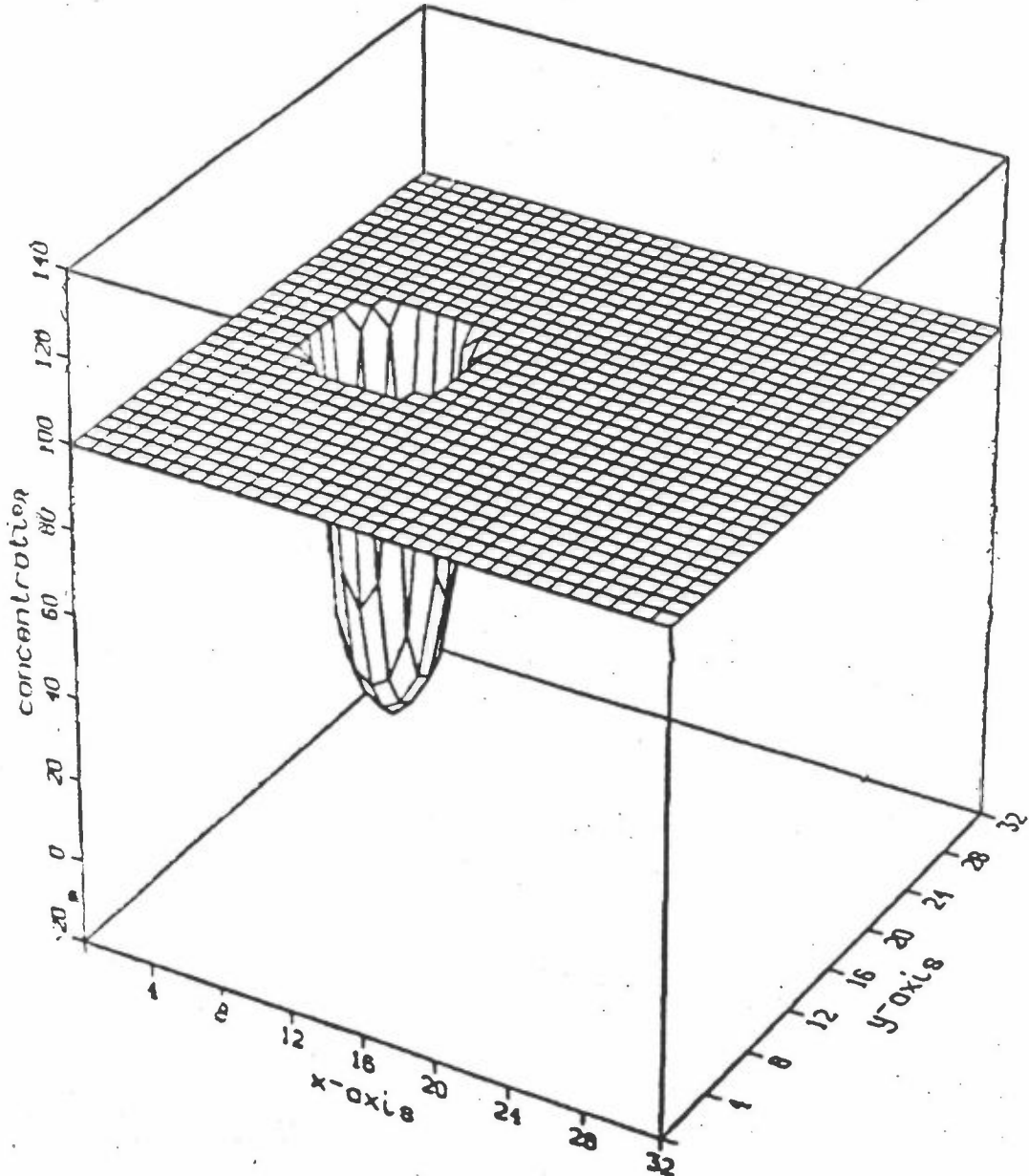


Figure 4: The solution for nitrogen oxide when chemistry only is carried out. Concentrations in molecules/cm³.

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 1.1927×10^6

one rotation with both advection and chemistry

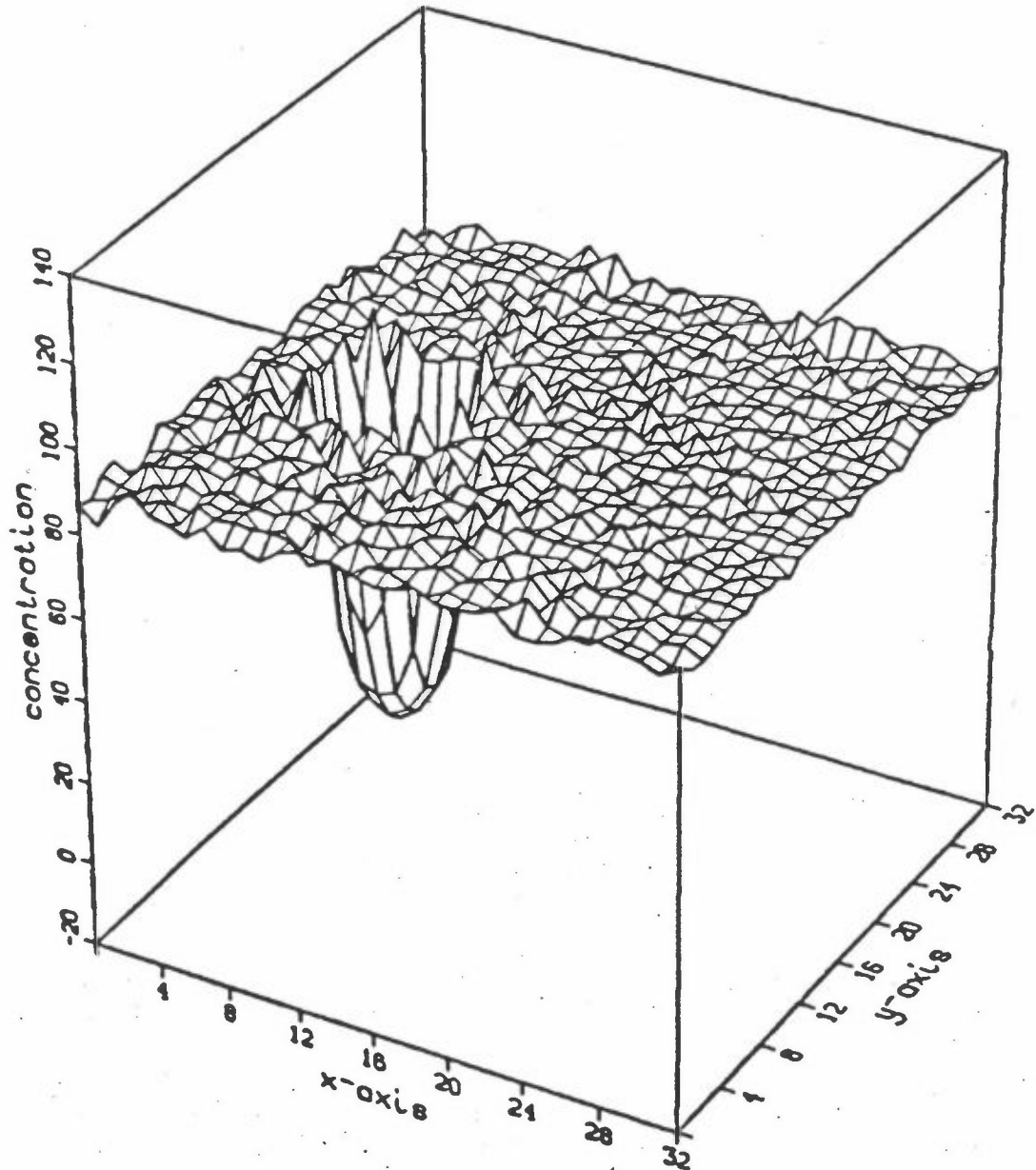


Figure 5: The solution for the nitrogen oxide when advection+chemistry is carried out. The advection algorithm used is the pseudo-spectral algorithm. Concentrations in molecules/cm³.

NITROGEN DI-OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 3.8836×10^8

one rotation when chemistry only is performed

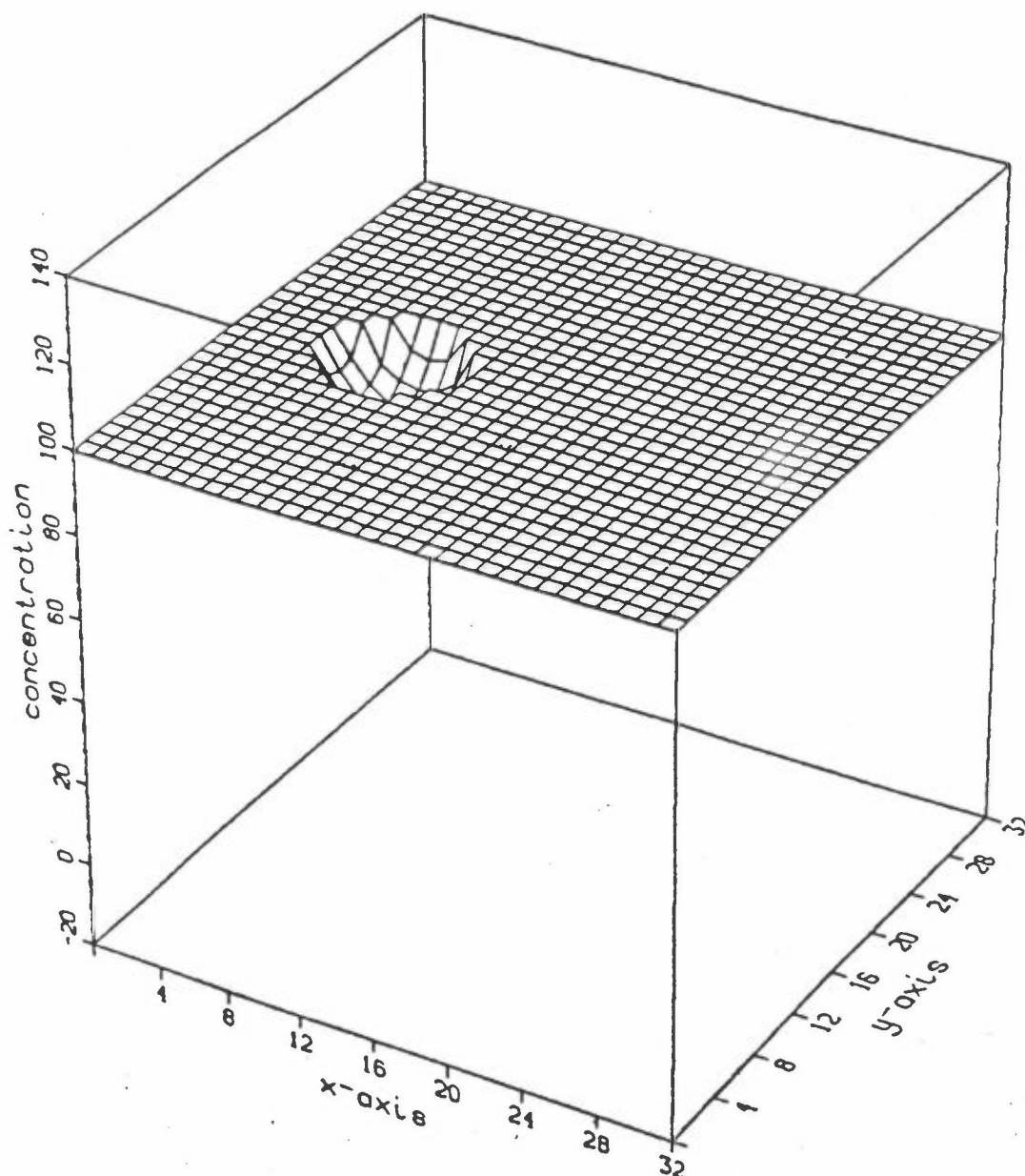


Figure 6: The solution for nitrogen dioxide when chemistry only is carried out. Concentrations in molecules/cm³.

NITROGEN DI-OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 3.9245×10^8

one rotation with both advection and chemistry

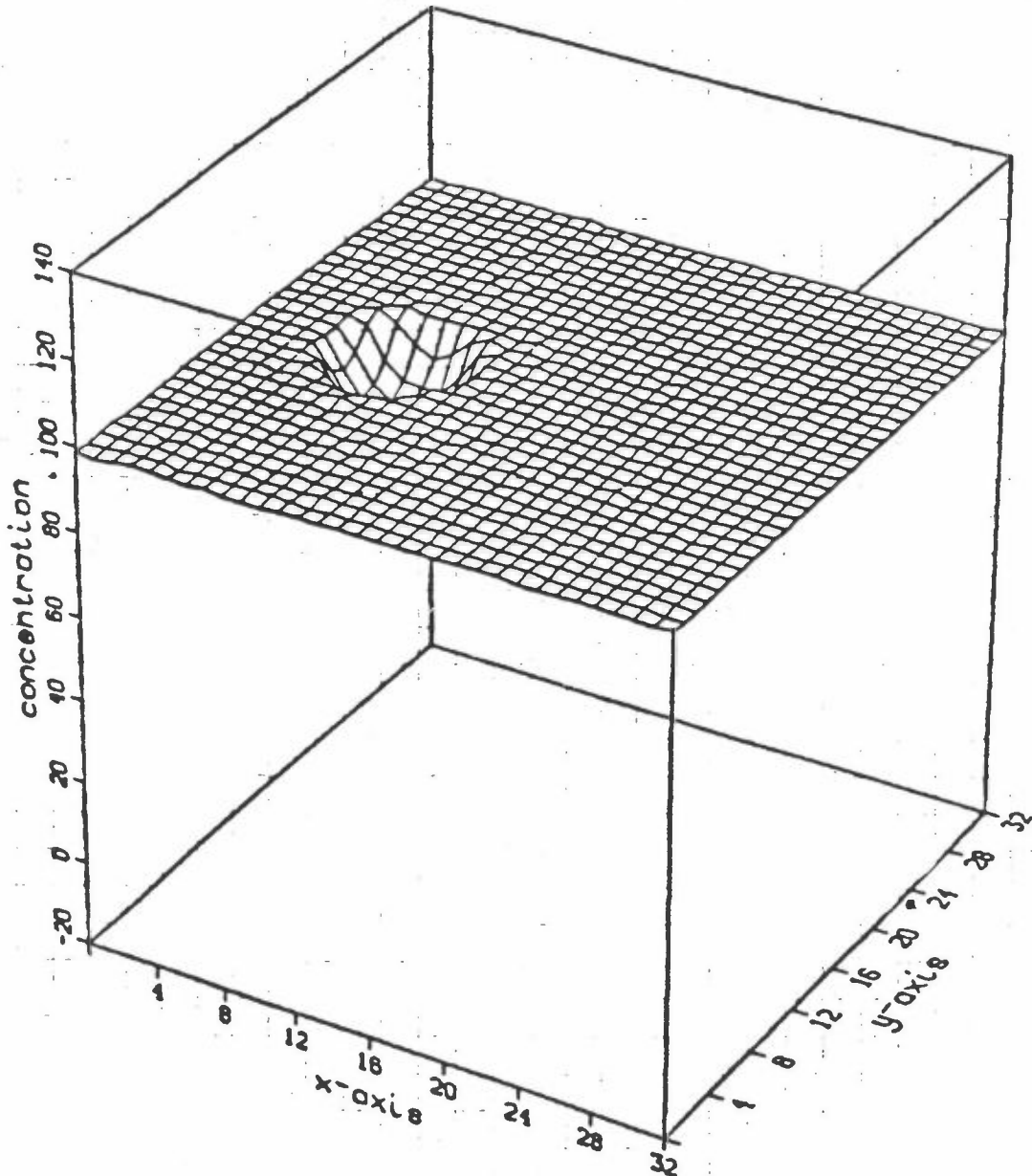


Figure 7: The solution for nitrogen dioxide when advection+chemistry is carried out. The advection algorithm used is the pseudo-spectral algorithm. Concentrations in molecules/cm³.

the chemical reactions do not change the shape of the distribution or the magnitude of the highest concentration significantly, then the results for advection + chemistry are nearly as good as those obtained by advection only; see Fig. 8. The conclusion is that in general the smoothing procedure does allow us to get good results also for species with significant non-linear chemistry terms.

It is interesting to compare the concentration at the peak of the puff initially and in the case where advection only, chemistry only and advection+chemistry are performed for 24 h. This comparison is made in Table 2. It is seen that the relative errors made in the case where advection+smoothing+chemistry is applied do not exceed 5%. These errors are of the same magnitude as the errors at the peak of the puff for the case where advection only is performed.

Table 2: The values of the concentrations at the peak of the puff. The modifications (in percent) due to the advection and advection+smoothing are given in brackets. The advection algorithm used is the pseudospectral algorithm. All fields after the first four contain the same concentrations at all grid-points at the beginning, therefore advection only is performed without error.

Pollutant	Initial value	Advection only	Chemistry only	Advection + chemistry
NO	1.0×10^{11}	0.95×10^{11} (95%)	1.16×10^5	1.19×10^5 (103%)
NO ₂	1.0×10^{11}	0.95×10^{11} (95%)	3.11×10^8	3.25×10^8 (105%)
HC ₂	1.0×10^{11}	0.95×10^{11} (95%)	1.97×10^9	2.01×10^9 (102%)
HCHO	5.0×10^{11}	0.475×10^{10} (95%)	2.47×10^{11}	2.42×10^{11} (98%)
O ₃	5.0×10^{11}	5.0×10^{11} (100%)	1.60×10^{12}	1.60×10^{12} (100%)
HNO ₃	1.0×10^{10}	1.0×10^{10} (100%)	2.10×10^{11}	2.06×10^{11} (98%)
HO ₂	1.0×10^6	1.0×10^6 (100%)	8.77×10^{11}	8.61×10^{11} (98%)
RO ₂	1.0×10^6	1.0×10^6 (100%)	4.02×10^{10}	3.90×10^{10} (96%)
OH ₂	1.0×10^5	1.0×10^5 (100%)	9.85×10^5	9.96×10^5 (100%)
O(¹ D)	1.0×10^{-3}	1.0×10^{-3} (100%)	4.42×10^{-4}	4.43×10^{-4} (100%)

ALDEHYDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 2.4188×10^{11}

one rotation with both advection and chemistry

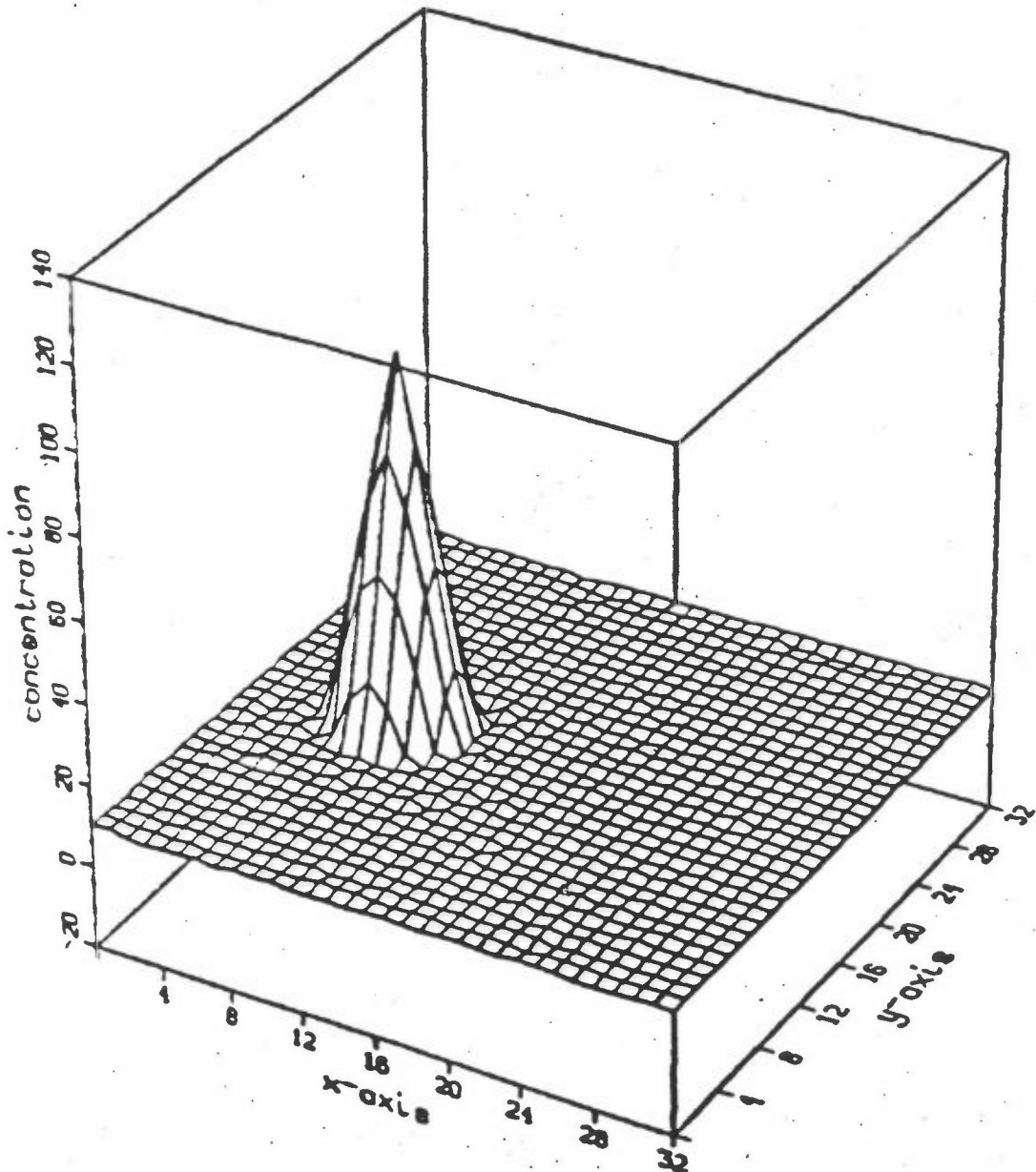


Figure 8: The solution for aldehyde when advection+chemistry is carried out. Concentrations in molecules/cm³.

3.2 RUNS WITH OTHER ADVECTION ALGORITHMS

The tests listed at the end of Section 2 have also been run with the other advection schemes. The results of these runs are briefly discussed in this paragraph.

(a) Runs with discretization by second order finite differences. It is well known that these advection algorithms are neither able to preserve the shape of the puff nor to preserve the maximum concentrations in the puff. Our runs only confirmed this conclusion. Moreover, the puff is nearly completely distorted when both advection and chemistry are performed (compared with the corresponding pattern for the case where only chemistry is applied).

(b) Runs with the Smolarkiewicz algorithm. The results become slightly better when the Smolarkiewicz algorithm is applied instead of the algorithm based on second order finite differences. However, the improvement of the accuracy is negligible compared with the increase of the computing time spent. Our conclusion is that the Smolarkiewicz (1985) algorithm should not be recommended for air pollution problems of the type considered in this paper.

(c) Runs with the finite elements (CHAPEAU) advection algorithm. This algorithm deserves more attention. It is less accurate than the pseudospectral algorithm, but this is by no means prohibitive. The results for nitrogen oxide in the cases where advection only and advection+smoothing+chemistry are applied are given in Fig. 9 and Fig. 10, respectively (these results are to be compared with the results obtained by the pseudospectral algorithm; see Fig. 3 and Fig. 5). The values of concentrations in the peak are given in Table 3 (to be compared with Table 2). While the accuracy of this advection algorithm is worse than that of the pseudospectral advection, it is faster than the latter algorithm with regard to computing time. The computing times will be compared in the next paragraph.

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 1.0000×10^{11}

advection only by finite differences CHAPEAU

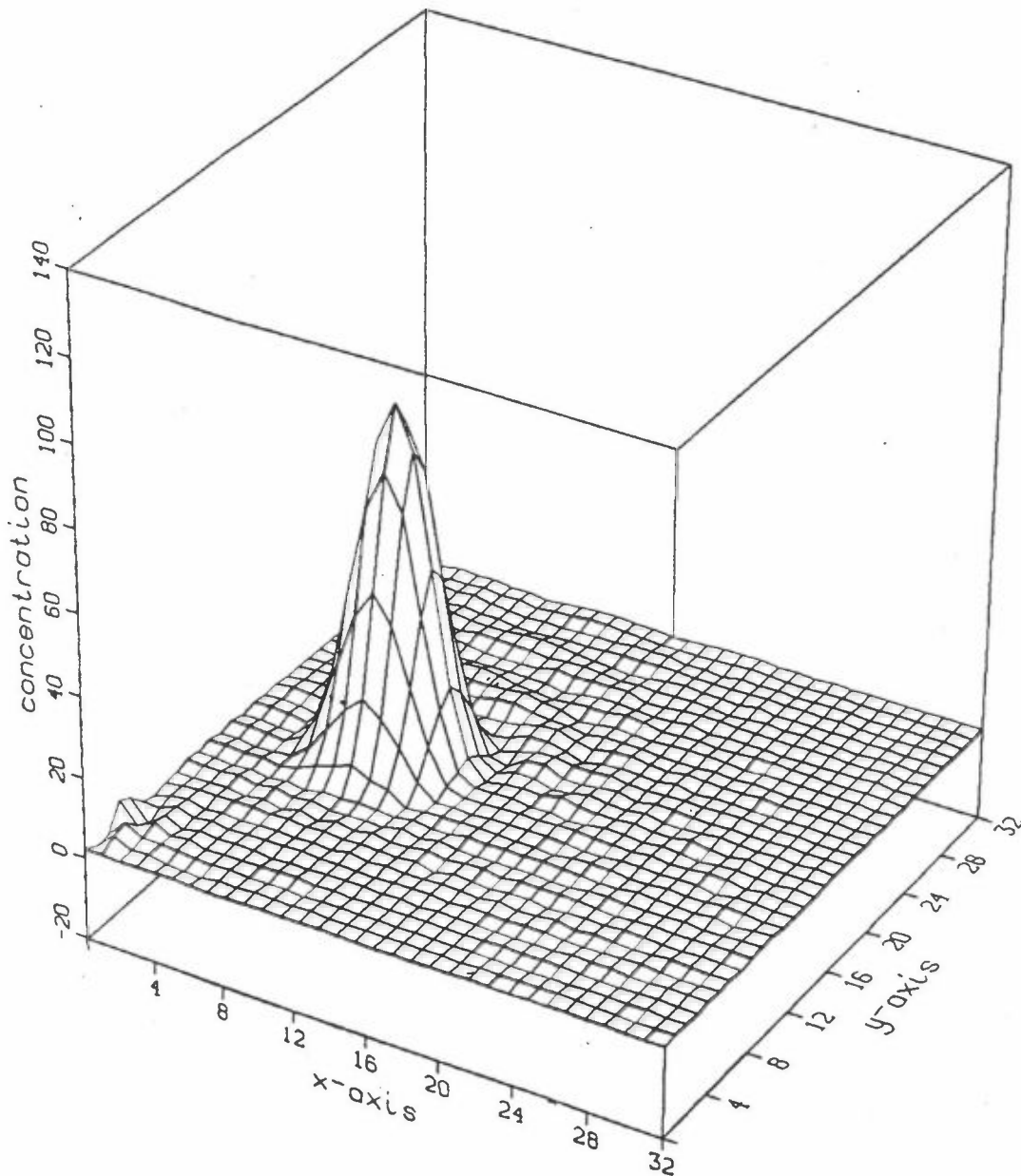


Figure 9: The solution for nitrogen oxide with advection only using the finite elements (CHAPEAU) algorithm. Concentrations in molecules/cm³.

NITROGEN OXIDE

all concentrations are divided by the largest concentration and multiplied by 100

the largest concentration is: 1.6006×10^6

adv + chem by finite elements: CHAPEAU

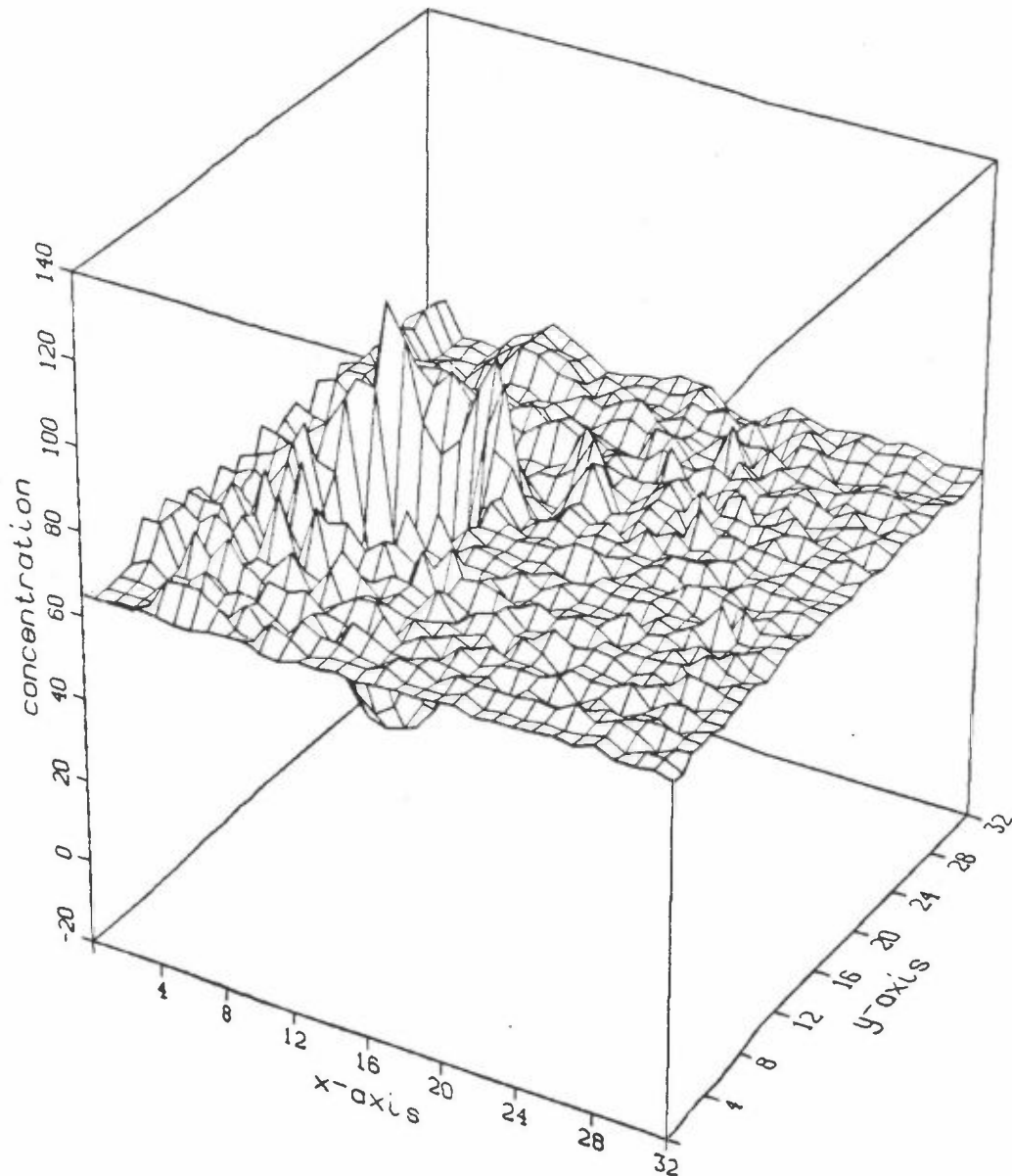


Figure 10: The solution for nitrogen oxide when advection and chemistry only is carried out. The advection algorithm used is the Chapeau finite elements algorithm. Concentrations in molecules/cm³.

Table 3: Concentrations at the peak of the puff. The modifications (in percent) due to the advection and advection+smoothing are given in brackets. For the advection the finite elements (CHAPEAU) algorithm is used. All fields after the first four contain the same concentrations at all grid-points at the beginning, therefore the advection is performed without error.

Pollutant	Initial value	Advection only	Chemistry only	Advection + chemistry
NO	1.0×10^{11}	0.88×10^{11} (88%)	1.16×10^5	1.44×10^5 (124%)
NO ₂	1.0×10^{11}	0.87×10^{11} (87%)	3.11×10^8	3.16×10^8 (102%)
HC ₂	1.0×10^{11}	0.87×10^{11} (87%)	1.97×10^9	1.99×10^9 (101%)
HCHO	5.0×10^{10}	0.87×10^{10} (87%)	2.47×10^{11}	2.15×10^{11} (87%)
O ₃	5.0×10^{11}	5.0×10^{11} (100%)	1.60×10^{12}	1.54×10^{12} (103%)
HNO ₃	1.0×10^{10}	1.0×10^{10} (100%)	2.10×10^{11}	1.84×10^{11} (88%)
HO ₂	1.0×10^6	1.0×10^6 (100%)	8.77×10^{11}	7.74×10^{11} (88%)
RO ₂	1.0×10^6	1.0×10^6 (100%)	4.08×10^{10}	3.35×10^{10} (82%)
OH	1.0×10^5	1.0×10^5 (100%)	9.95×10^5	1.01×10^6 (102%)
O(¹ D)	1.0×10^{-3}	1.0×10^{-3} (100%)	4.42×10^{-4}	4.27×10^{-4} (97%)

Table 4: Computing times (with a constant stepsize) obtained with four different space discretization algorithms. Number of the advection steps: 576. Number for the chemical steps: 5 per advection step. The stepsize in the advection part: 150 seconds. The stepsize in the chemical part: 30 seconds. The computer used in the runs: Univac 1110/82.

Space discretization algorithm	Only advection	Advection + chemical reactions	Only chemical reactions
Finite differences	333	2933	2600
Smolarkiewicz	1378		
Pseudospectral	1523	4169	2646
Finite elements	683	3308	2625

3.3 COMPARISON OF THE COMPUTING TIMES SPENT FOR THE DIFFERENT ALGORITHMS

Eulerian models are rather expensive to run with complex chemistry, and a large computer is required. The storage requirements for the different algorithms are not large. The computer memory is normally sufficient and the inclusion of additional arrays does not cause difficulties. On the other hand, the computing time is a crucial factor when long runs are to be performed.

The computing times for the four algorithms are given in Table 4. In all cases the time step size is 150 s for the advection part and 30 s for the chemical part. Thus, in all cases 576 advection time-steps are carried out, while 5 chemical steps are performed at each advection step.

It is seen from Table 4 that computing times for the chemical part are nearly the same (in fact these should be precisely the same since the same work is carried out during the chemical part for each advection algorithm; the differences are caused by the uncertainties connected with the subroutine that calculates the computing time). The second order finite differences algorithm is about 5 times faster than the pseudospectral algorithm when the advection time only is taken into account. In the same situation the Smolarkiewicz algorithm is only a little better than the pseudospectral algorithm, while the finite elements (CHAPEAU) algorithm is about 2.5 times faster. The differences are less when advection+smoothing+chemistry is applied.

The computing times shown in Table 4 indicate that only episodes can be treated numerically by the model at present, keeping in mind also that only a very simple chemical description is given. If more realistic runs are to be carried out (in order to calculate mean concentrations on monthly, seasonal and yearly bases), then these computing times must be reduced at least by a factor of 100. The question is: can we achieve such a great reduction? We are not prepared to give the answer to this question now. However, we shall discuss two ways in which such a reduction could be achieved as well as the results obtained when the computational performance of the code was attempted to be made more efficient.

The first way in which a reduction in computing time could be achieved is by improving the algorithms used and/or by running the algorithms with a longer time step. We have tried the latter approach. The chemical part has been run with a step size of 150 s (one chemical step per one advection step). In this way the computing time for the chemical part is reduced by a factor of 5. It is clear that in this situation the importance of the choice of an advection algorithm is increased considerably. The accuracy of the results becomes poorer, but remains acceptable. More experiments are needed to decide the maximum step size that can be allowed. The photochemical reactions cause considerable changes in the concentration field during sunrise and sunset (say from 6 to 10 am and from 6 to 10 pm). A small step size could be used in these periods, and a longer step size in the other periods.

The second way involves the use of a fast vector processor. However, the present code is not vectorized. The chemical integration procedure contains many IF statements. Moreover, the chemical integration is performed in the traditional sequential manner (successively one grid-point after another). Some transformations that are formally quite simple, are necessary. Many improvements are needed also in the advection part. Nevertheless, it is worthwhile to do these changes. The following examples illustrate that it is worthwhile to do modifications. The rotation test has also been run on AMDAHL VP100. The computing time for the case where the pseudospectral algorithm is used (together with smoothing and chemistry) was about 215s. This is more than 19 times faster than the time for the corresponding run in Table 4. For the finite elements (CHAPEAU) advection discretization the corresponding figure is 105 s (this is a reduction by a factor of about 30 compared with the computing time obtained by the same algorithm on UNIVAC 1100/82; see Table 4). Therefore the experiments indicate that the main aim stated in this paragraph, to reduce the computing time by a factor at least equal to 100, is not unrealistic.

3.4 FURTHER WORK

The calculations reported here were not dependent on the conditions applied at the grid boundary. To be able to do model calculations of a realistic situation, the boundary conditions will become important for the results calculated in the interior of the grid. It is well known that the establishment of boundary conditions is particularly difficult for the pseudo-spectral method, where it has been necessary to introduce strong damping of the concentration on the boundary in linear chemistry models. The introduction of non-linear chemistry will necessitate a refinement of the boundary conditions already in use in pseudo-spectral models. This work is now underway.

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TITLE
ABSTRACT (max. 300 characters, 7 lines) The coupled, non-linear system of continuity equations for an air pollution model is solved numerically using finite differences, finite elements and pseudospectral methods for the advection term. The accuracy of the solution is evaluated with emphasis on the rotating puff test. The pseudo-spectral and finite element methods give satisfactory results.

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