

Berkvens, P.J.F. and Botchev, M.A. and Verwer, J.G. (2000) On the Efficient Treatment of Vertical Mixing and Chemistry in Air Pollution Modeling. In: Proceedings of the 16th IMACS world congress, August 21–25, 2000, Lausanne, Switzerland. IMACS. ISBN 3–9522075–1–9.

On the Efficient Treatment of Vertical Mixing and Chemistry in Air Pollution Modelling

P.J.F. Berkvens*

M.A. Botchev*

J.G. Verwer*

March 1, 2000

Abstract

Reaction-Advection-Diffusion problems occurring in Air Pollution Models have very stiff reaction terms and moderately stiff vertical mixing (diffusion and cloud transport) terms which both should be integrated implicitly in time. Standard implicit time stepping is by far too expensive for this type of problems whereas widely used splitting techniques may lead to unacceptably large errors for fast species. Recently proposed alternatives are the so-called source splitting and the ROS2 method combined with approximate matrix factorization (AMF).

In this paper we compare these two methods with the standard Strang splitting method in a framework of an operational global dispersion model. The results of the tests favour the ROS2-AMF scheme.

Key words: Air pollution modelling, operator splitting, stiff ODEs, Rosenbrock methods.

AMS subject classifications: Primary: 65M06, 65M20. Secondary: 62Y20.

1 Introduction and Motivation

Typical reaction-advection-diffusion problems occurring in air pollution modelling have the form

$$(1) \quad \begin{aligned} \frac{\partial c}{\partial t} + \nabla \cdot (\underline{u}c) &= \mathcal{V}(c) + \mathcal{R}(c), \\ c &= c(\underline{x}, t), \quad c \in \mathbb{R}^M, \quad \underline{x} \in \Omega \subset \mathbb{R}^3, \end{aligned}$$

where M is the number of chemical components (tracers) in the model, the term $\mathcal{V}(c)$ describes vertical mixing by turbulent diffusion and subgrid transport (so-called cumulus convection):

$$(2) \quad \mathcal{V}(c) = \nabla \cdot (\underline{K}\nabla c) + \mathcal{V}_{\text{conv}}(c),$$

and $\mathcal{R}(c)$ is the reaction (chemistry) term.

For grid resolutions and number of tracers M ($M \sim 50$) used nowadays, spatial finite-volume discretization in (1) leads to a huge system of nonlinear stiff ODE's to integrate in time. Assume we have discretized (1) spatially on the grid $\{x_{ijk}\}$ with index k ($1 \leq k \leq K$) corresponding to the vertical coordinate. In the three-dimensional grid, let us choose a particular vertical column (i, j) that consists of the grid cells $\{x_{ijk}\}_{k=1}^K$. We denote by \mathbf{c}_{ij} the vector containing all concentration values of the column (i, j) for all the tracers $m = 1, \dots, M$:

$$(3) \quad \mathbf{c}_{ij} = (c_{ij1}^1, \dots, c_{ij1}^M, c_{ij2}^1, \dots, c_{ij2}^M, \dots, c_{ijK}^1, \dots, c_{ijK}^M),$$

*CWI, P.O.Box 94079, 1090 GB Amsterdam, the Netherlands, <http://www.cwi.nl>, [berkvens, botchev, janv]@cwi.nl

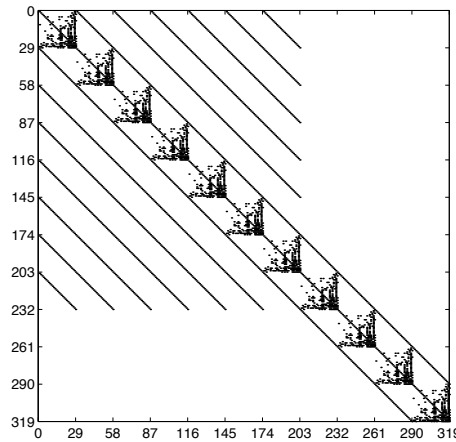


Figure 1: Sparsity structure of the coupled chemistry–vertical mixing Jacobian $\mathbf{A} = \mathbf{R}' + \mathbf{V}$ for $M = 29$ and $K = 11$. The diagonal blocks correspond to the chemistry Jacobians per grid point R'_1, \dots, R'_K .

and by \bar{c} the vector containing all the unknowns in the 3D grid, so that $\bar{c} = \{c_{ij}\}_{ij}$. Then the discrete equations for the column (i, j) can be written in the form

$$(4) \quad \dot{c}_{ij} + (A[\bar{c}])_{ij} = \mathbf{V}_{ij}c_{ij} + \mathbf{R}_{ij}(c_{ij}),$$

where $A[\bar{c}]$ is the discretized advection operator, nonlinear because of the high-order flux limited approximation used [7, 12]. \mathbf{V}_{ij} is a vertical transport $(KM) \times (KM)$ matrix for all of the tracers. Matrix \mathbf{V}_{ij} can be represented as a Kronecker product of a $K \times K$ matrix V_{ij} describing vertical transport of each tracer and $M \times M$ identity matrix I_M :

$$(5) \quad \mathbf{V}_{ij} = V_{ij} \otimes I_M.$$

Finally, $\mathbf{R}_{ij}(c_{ij})$ in (4) is the chemistry operator that acts per grid cell:

$$\mathbf{R}_{ij}(c_{ij}) = (R_{ij1}(c_{ij1}^1, \dots, c_{ij1}^M), \dots, R_{ijK}(c_{ijK}^1, \dots, c_{ijK}^M)).$$

Standard stiff ODE solvers are by far too expensive for solving (4) in terms of the CPU time. A widely used feasible alternative is operator splitting; this means that the advance in time for the whole system is a sequence of time substeps to advance the various processes (advection, vertical mixing and chemistry).

One of the simplest splitting scenarios used in many state-of-the-art codes is (a) advance advection with an explicit higher order upwind scheme [7, 12]; (b) advance vertical mixing with an implicit scheme; (c) advance chemistry with an implicit scheme. It is necessary to use implicit integration at steps (b) and (c) since both chemistry and vertical mixing are stiff processes. Chemistry has characteristic time scales that range from seconds to minutes to months and, thus, is extremely stiff.

A major problem with splitting is that it can give rise to a large time-integration error for fast reacting species. This splitting error may not only adversely affect the global time-integration error but also, due to the stiffness of the problem, introduces transients that hinder the integrators used at each of the split steps. Splitting of the stiff processes leads to the most severe errors and therefore coupling steps (b) and (c) would be most beneficial. In this paper we consider two techniques for efficient coupling of steps (b) and (c). Therefore, in what follows we omit in (4) the advection term $(A[\bar{c}])_{ij}$ and the subindices ij . Thus, we are left with the ODE system

$$(6) \quad \dot{c} = f(c) = \mathbf{V}c + \mathbf{R}(c),$$

that describes vertical mixing and reactions in a particular vertical column of the grid. For details on treating the advection and coupling it with the other processes we refer to [7, 17].

Straightforward application of a fully implicit scheme would be too inefficient for the following reason. At each grid point the chemistry Jacobian is a sparse matrix and this sparsity can be preserved to a large extent in the LU factorization. This makes uncoupled implicit treatment of the chemistry quite economical. However, when the chemistry is coupled with the vertical mixing the structure of the Jacobian $\mathbf{A} = \mathbf{R}' + \mathbf{V}$ is such that its LU factorization would lead to, generally speaking, complete fill-in. Loss of sparsity, even only within the diagonal blocks to the chemistry Jacobians (see Figure 1), leads to a dramatical increase of the computational work and hence is unacceptable.

Note that the presence of the term $\mathcal{V}_{\text{conv}}(c)$ in (2) makes matrix V ($= V_{ij}$ in (5)) full rather than tridiagonal; however, even in the tridiagonal case the block sparsity in the LU factorization of \mathbf{A} would be lost.

A number of authors [9, 16, 18, 10] have proposed schemes that avoid operator splitting and are much cheaper than the coupled fully implicit treatment of vertical mixing and chemistry. Among these new schemes, the second order Rosenbrock method ROS2 with approximate factorized Jacobian appeared to be quite promising. However, the experience observed was not convincing enough to state that the new schemes are generally superior. More realistic tests and theoretical understanding are still needed here.

In this paper we compare one standard operator splitting scheme with ROS2 with approximate matrix factorization and with a so-called source-splitting method. The experiments are made in a large scale global air pollution model. It should be stressed that in the tests described in [9, 16, 18], only diffusion was taken into account in the vertical mixing (cf. (2)). Incorporation of the term $\mathcal{V}_{\text{conv}}(c)$ in our tests leads to a non-local coupling among the vertical layers, so that the matrix V is no longer tridiagonal. This makes (partially) implicit solution more cumbersome.

In Section 2 we describe the methods used, in Section 3 the results of the numerical experiments are presented and our conclusions follow in Section 4.

2 Methods

All three methods used in the tests are derived from a particular Rosenbrock method (which we will call ROS2) from the stiff ODE field [3, 5]. Let $\gamma = 1 + \frac{1}{2}\sqrt{2}$. The Rosenbrock method reads

$$(7) \quad \begin{aligned} \mathbf{c}_{n+1} &= \mathbf{c}_n + \frac{3}{2}\mathbf{k}_1 + \frac{1}{2}\mathbf{k}_2, \\ (\mathbf{I} - \gamma\tau\mathbf{A})\mathbf{k}_1 &= \tau\mathbf{f}(\mathbf{c}_n), \\ (\mathbf{I} - \gamma\tau\mathbf{A})\mathbf{k}_2 &= \tau\mathbf{f}(\mathbf{c}_n + \mathbf{k}_1) - 2\mathbf{k}_1. \end{aligned}$$

This two-stage method is second order consistent for any matrix \mathbf{A} . This matrix is supposed to be an approximation to the Jacobian matrix $\mathbf{f}'(\mathbf{c}_n)$. When using the exact Jacobian, the method has a stability function which is L -stable and positive on the negative real axis. In [17, 2] it is argued that this low order, linearly implicit method is suitable for solving stiff atmospheric chemical kinetics problems (box models), provided the costs for the linear system solutions and the Jacobian updates are minimized by using optimal sparsity routines. Such routines are provided by the chemical kinetics preprocessor KPP [13]. For the reasons already mentioned, as it stands, method (7) should not be applied to the coupled vertical-transport-chemistry problem as the costs for the linear systems solutions with matrix $\mathbf{I} - \gamma\tau\mathbf{A}$ are too high.

Strang splitting

Let $\Phi_V(t; \tau)$ and $\Phi_R(t; \tau)$ denote the integrator (7) applied to $\dot{\mathbf{c}} = \mathbf{V}\mathbf{c}$ and $\dot{\mathbf{c}} = \mathbf{R}(\mathbf{c})$, respectively. A Strang-type operator splitting method for system (6) using a step size τ and ending with a chemistry step [14], then can be formulated as

$$(8) \quad \mathbf{c}_{n+1} = \Phi_R(t_{n+1/2}; \frac{1}{2}\tau) \Phi_V(t_{n+1/2}; \frac{1}{2}\tau) \Phi_V(t_n; \frac{1}{2}\tau) \Phi_R(t_n; \frac{1}{2}\tau) \mathbf{c}_n.$$

This method is 2nd-order consistent. The leading local error term is the sum of the leading local errors made by the Rosenbrock method and the leading local splitting error. In what follows, we refer to method (8) as STRANG.

Stability properties of the STRANG method are largely determined by those of each of its substeps. Since at each substep the L -stable ROS2 method is applied, the STRANG method is L -stable.

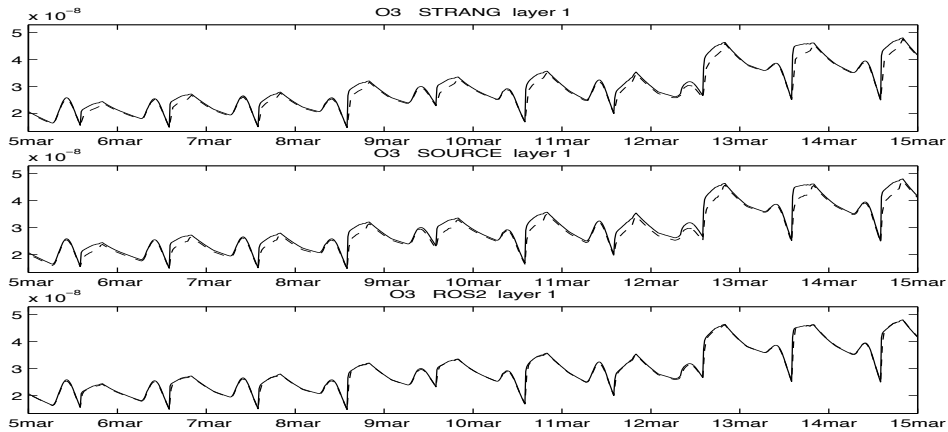
Source splitting method

In the source splitting method [9, 8], one of the split processes is approximated at each time step as a constant source to be applied with the other split process. More precisely, we replace (6) with

$$(9) \quad \dot{\mathbf{c}} = \hat{\mathbf{f}}(\mathbf{c}) = \mathbf{R}(\mathbf{c}) + \mathbf{s}, \quad t \in [t_n, t_n + \tau],$$

where \mathbf{s} is the source term approximating vertical transport. The scheme can be written as

$$\begin{aligned} \tilde{\mathbf{c}}_{n+1} &= \Phi_V(t_n; \tau) \mathbf{c}_n, \\ \mathbf{s} &= \frac{\tilde{\mathbf{c}}_{n+1} - \mathbf{c}_n}{\tau}, \\ \mathbf{c}_{n+1} &= \Phi_{\hat{\mathbf{f}}}(t_n, \tau) \mathbf{c}_n. \end{aligned}$$

Figure 2: The O₃ (ozone) solution.

Formally, splitting has been removed because the step $\Phi_{\hat{F}}$ is in fact done for the unsplit system (9) that approximates the full system (6). Independently of the order of the methods used per substep in Φ , this S-SPLIT method is only first order consistent. Observe that $\mathbf{s} = \mathbf{V}\mathbf{c} + \mathcal{O}(\tau)$ for any $\mathbf{c} \in [c_n, \tilde{c}_{n+1}]$. Just as in standard splitting, in S-SPLIT there is essentially the same freedom left to use one's favorite combination of algorithms. Higher order generalizations of source splitting are considered in [10].

We note in passing that it is profitable to have the substep corresponding to the stiffest process, chemistry, at the end of the splitting sequence [14]. This is the case for methods STRANG and S-SPLIT.

Simple analysis reveals nice stability properties of the S-SPLIT scheme: it is L -stable with respect to the chemistry term and A -stable with respect to the vertical mixing term. However, the stability function is not monotone along the negative real axis as it is for ROS2.

Approximate factorization

The ROS2-AMF method is the ROS2 scheme (7) applied to the whole system (6) with an inexact matrix $\mathbf{I} - \gamma\tau\mathbf{A}$. For $\mathbf{I} - \gamma\tau\mathbf{A}$ we take Approximate Matrix Factorization (AMF) in the form:

$$(10) \quad \mathbf{I} - \gamma\tau\mathbf{A} = (\mathbf{I} - \gamma\tau\mathbf{V})(\mathbf{I} - \gamma\tau\mathbf{R}')$$

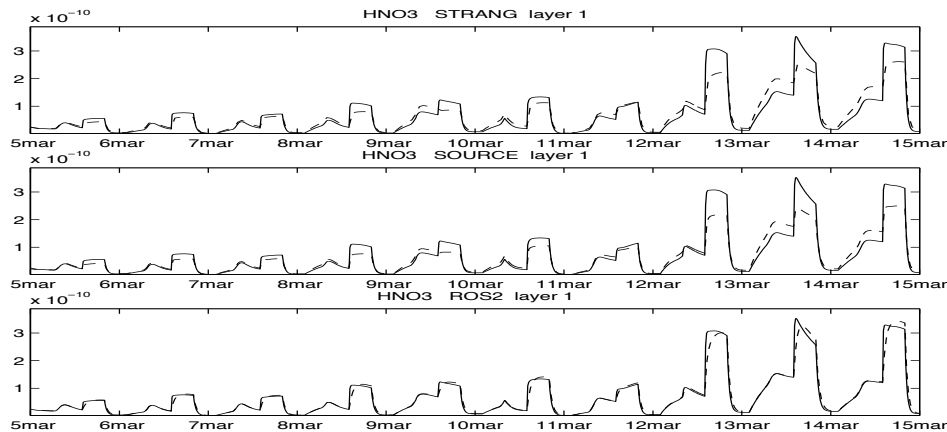
This ROS2-AMF method was introduced in [18] and recently tested in [2]. The approximate matrix factorization is known at least since the papers [4, 1] and is reminiscent of the splitting technique already used in the 1950s in more conventional alternating direction methods [11]. A recent survey can be found in [15]. In [6] and references therein, interesting theoretical stability results are given revealing some limitations of approximate matrix factorization.

The ROS2-AMF method is split-free because the 'splitting' appears only at the level of the linear solves with matrix $\mathbf{I} - \gamma\tau\mathbf{A}$. However, the method has retained the ease of implementation of the standard splitting, i.e. the algorithmic ingredients, among which the linear solves with matrices $\mathbf{I} - \gamma\tau\mathbf{V}$ and $\mathbf{I} - \gamma\tau\mathbf{R}'$ are the most essential, remain basically the same as in conventional splitting. Although the ROS2 method (7) is a W-method [5] which means it remains order two consistent with an arbitrary matrix \mathbf{A} , stability of the scheme with \mathbf{A} not being exact Jacobian is corrupted. Approximation (10), however, leads to only moderate loss of stability: in fact, the scheme maintains its A -stability, only the L -stability is lost [18].

3 Numerical Experiments

For our experiments we have taken one grid column of the large scale global air pollution model TM3¹. We have chosen a time interval of 10 days in March, 1997 and have used the real meteorological data for this time period (among the data are air pressure and temperature, turbulent diffusion and cloud parameters, deposition rates and surface emissions, etc.) The vertical column consists of $K = 19$ grid cells. The chemical reaction set used is CBM-IV having $M = 29$ tracers.

¹See URLs <http://www.cwi.nl/cwi/projects/TM3/> and <http://www.phys.uu.nl/~peters/TM3/TM3S.html>

Figure 3: The HNO_3 (nitric acid) solution.

Computational costs Methods S-SPLIT and ROS2-AMF require the same work per time step, and two steps of either of the methods are equal in cost to one step of STRANG. In our experiments we use the same constant step size $\tau = 30$ min for S-SPLIT and ROS2-AMF and twice as large a step size for STRANG (so that each of the substeps in (8) are performed with the stepsize 30 min). This makes these three methods equal in cost per physical time unit.

The work per step of S-SPLIT or ROS2-AMF consists essentially of one evaluation of the chemistry Jacobian \mathbf{R}' , one sparse LU factorization of $\mathbf{I} - \gamma\tau\mathbf{R}'$, two linear system solves with each of the matrices $\mathbf{I} - \gamma\tau\mathbf{R}'$ and $\mathbf{I} - \gamma\tau\mathbf{V}$, two chemistry function evaluations and two matrix-vector multiplication with vertical transport matrix \mathbf{V} . Matrices \mathbf{V} and $(\mathbf{I} - \gamma\tau\mathbf{V})^{-1}$ are updated every 6 hours.

Results In Figures 2–4, we have plotted concentration² of tracers O_3 (ozone), HNO_3 (nitric acid) and OH (hydroxyl radical) in the surface grid cell as a function of time. Ozone O_3 is a long living tracer, whereas OH is a fast varying tracer, a so-called radical. The surface grid cell is most interesting and important in the model since most of the emissions and depositions take place here. For each of the three methods, the solution curve is plotted dashed against a solid line of a reference solution. This reference solution was obtained by the STRANG method with a tiny step size $\tau = 6$ sec. As we see on the plots, the error of ROS2-AMF is considerably smaller than those of the other two methods. This is also the case for the other tracers and grid cells.

4 Conclusions

In our tests within the operational global dispersion model TM3, the ROS2-AMF solver appeared to be notably more accurate for a wide range of tracers. Since the use of the ROS2-AMF method does not mean an increase in the computational costs as compared to conventional splitting methods, ROS2-AMF seems to be an attractive alternative to splitting in large scale operational dispersion models. The source-splitting method S-SPLIT appeared to be less accurate in most cases than the standard splitting method STRANG.

Acknowledgments The authors would like to thank Maarten Krol and Wouter Peters from the Institute for Marine and Atmospheric Research Utrecht (IMAU) for model advice.

References

- [1] R. M. Beam and R. F. Warming. An implicit finite-difference algorithm for hyperbolic systems in conservation-law form. *J. Comput. Phys.*, 22:87–110, 1976.
- [2] J. G. Blom and J. G. Verwer. A comparison of integration methods for atmospheric transport-chemistry problems. CWI Report MAS-R9910, Amsterdam, 1999. To appear in *J. Comp. Appl. Math.*
- [3] K. Dekker and J. G. Verwer. *Stability of Runge-Kutta methods for stiff non-linear differential equations*. North-Holland Elsevier Science Publishers, 1984.

²This is the ratio of the tracer mass in a grid cell to the air mass in the cell.

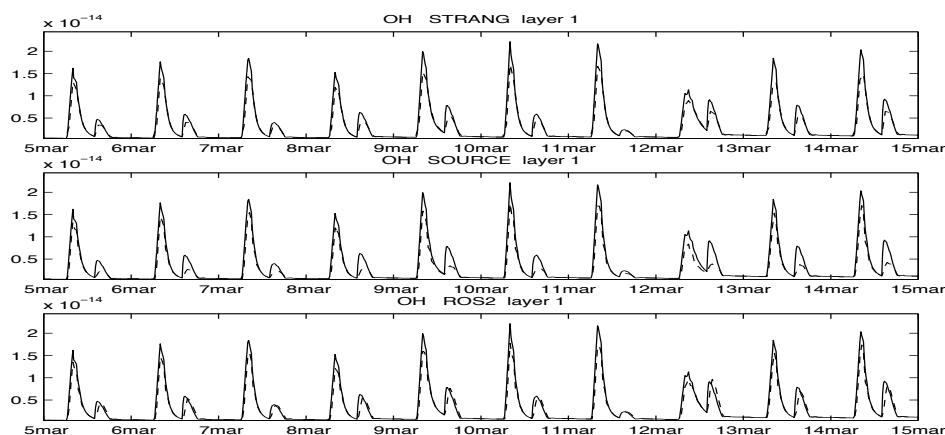


Figure 4: The OH (hydroxyl radical) solution.

- [4] E. G. D'yakonov. Difference systems of second order accuracy with a divided operator for parabolic equations without mixed derivatives. *USSR Comput. Math. Math. Phys.*, 4(5):206–216, 1964.
- [5] E. Hairer and G. Wanner. *Solving Ordinary Differential Equations II. Stiff and Differential–Algebraic Problems*. Springer Series in Computational Mathematics 14. Springer–Verlag, 1991.
- [6] W. Hundsdorfer. Accuracy and stability of splitting with stabilizing corrections. CWI Report MAS-R9935, Amsterdam, 1999.
- [7] W. Hundsdorfer, B. Koren, M. van Loon and J. G. Verwer. A positive finite-difference advection scheme. *J. Comput. Phys.*, 117:35–46, 1995.
- [8] C. Kessler. *Entwicklung eines effizienten Lösungsverfahrens zur modellmässigen Beschreibung der Ausbreitung und chemischen Umwandlung reaktiver Luftschadstoffe*. PhD thesis, Fakultät für Maschinenbau der Universität Karlsruhe, Aachen, 1995.
- [9] O. Knoth and R. Wolke. A comparison of fast chemical kinetic solvers in a simple vertical diffusion model. In S.-V. Gryning and M. Millán, editors, *Air Pollution Modelling and Its Application X*, pages 287–294, New York, 1994. Plenum Press.
- [10] O. Knoth and R. Wolke. Implicit-explicit Runge-Kutta methods for computing atmospheric reactive flows. *Appl. Numer. Math.*, 28:327–341, 1998.
- [11] D. W. Peaceman and H. H. Rachford, Jr. The numerical solution of parabolic and elliptic differential equations. *J. Soc. Indust. Appl. Math.*, 3:28–41, 1955.
- [12] A. C. Petersen, E. J. Spee, H. van Dop and W. Hundsdorfer. An evaluation and intercomparison of four new advection schemes for use in global chemistry models. *J. Geophysical Research*, 103(D15):19 253–19 269, 1998.
- [13] A. Sandu, F. Potra, G. Carmichael and V. Damian. Efficient implementation of fully implicit methods for atmospheric chemical kinetics. *J. Comput. Phys.*, 129:101–110, 1996.
- [14] B. Sportisse. An analysis of operator splitting techniques in the stiff case. Technical Report 98–127, ENPC-CERMICS, Champs sur Marne, France, 1998.
- [15] P. van der Houwen and B. P. Sommeijer. Approximate factorization for time-dependent partial differential equations. CWI Report MAS-R9915, Amsterdam, 1999.
- [16] J. G. Verwer and J. G. Blom. On the coupled solution of diffusion and chemistry in Air Pollution Models. *ZAMM (Special issue. Proceedings of ICIAM-95)*, 76 (suppl. 4):469–472, 1996.
- [17] J. G. Verwer, W. Hundsdorfer and J. G. Blom. Numerical time integration for air pollution models. CWI Report MAS-R9825, Amsterdam, 1998. To appear in “Surveys for Mathematics in Industry”.
- [18] J. G. Verwer, E. J. Spee, J. G. Blom and W. Hundsdorfer. A second order Rosenbrock method applied to photochemical dispersion problems. *SIAM J. Sci. Comput.*, 20:456–480, 1999.