Numerical Algorithms for Transport-Chemistry Problems

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1. Introduction

Ever increasing emissions of pollutants in the atmosphere, ground water and surface water, as a consequence of the growing population and economic activities, are affecting our environment. The negative effects become more and more noticeable: smog in urban regions (see also figure 1), ground water contamination, the growth of algae in surface water, and even atmospheric climate change has to be feared. Since physical experiments are in general too costly, or even impossible, mathematical simulations become increasingly important to study the long term effects of these emissions and to predict the efficiency of policy decisions to reduce the effects.

Once emitted, the pollutants will be transported by wind or water and dispersed by molecular diffusion and turbulence, while at the same time complex chemical reactions take place. The mathematical description of these processes is given by a large system of time dependent 3-dimensional partial differential equations of the advection-diffusion-reaction type involving the pollutants and all other substances that play a role in the reactive chain. For the numerical solution of these systems the availability of efficient numerical algorithms and the access to large computers is crucial. In fact, computer capacity is, and will remain, a critical factor. Though computer power continues to expand, the computational requirements for high resolution transport models with full chemistry are still out of range for many applications.

Figure 1. Smog above Rotterdam Harbour (Photo: Geosens).

Numerical analysis can assist the development of new models by providing more efficient numerical methods, tailored to the application at hand, and the full exploitation of the evolving computer architectures, such as advanced multi vector processors and, in the near future, massively parallel processing systems.

Below the numerical aspects will be outlined and discussed. Thereafter
266 a short description will be given of three projects presently carried out at CWI concerning transport of chemically reactive substances. These projects are part of CWI's research programme Mathematics & the Environment.

2. NUMERICAL RESEARCH

The basic mathematical equations describing transport and chemistry consist of a system of s partial differential equations for the unknown concentrations $c_k(x,t)$, $k = 1,\ldots,s$, which depend on time t and space $x = (x_1, x_2, x_3)$ in a 3-dimensional domain Ω. The equations, derived from mass balances, are given by

$$\frac{\partial}{\partial t}c_k(x,t) + \sum_{i=1}^{3}\frac{\partial}{\partial x_i}\left(u_i(x,t)c_k(x,t)\right) =$$

$$\sum_{i=1}^{3} \frac{\partial}{\partial x_i}\left(d_i(x,t)\frac{\partial}{\partial x_i}c_k(x,t)\right) + f_k(x,t,c_1(x,t),\ldots,c_s(x,t)),$$

with suitable initial and boundary conditions. The quantities $u_i(x,t)$ represent the velocities of the transport medium, such as water or air. These are either given in a data archive or computed alongside with a Navier-Stokes or hydrodynamical shallow water program. The diffusion coefficients $d_i(x,t)$ are constructed by the modellers and include also parametrizations of turbulence. The final term $f_k(x,t,c(x,t))$, which gives a coupling between the various substances, describes the nonlinear chemistry and also emissions (sources) and depositions (sincs). In actual models these equations are augmented with other suitable sub-grid parametrizations and coordinate transformations.

The number of substances s can be large, for example up to 100 in current atmospheric models. Also the spatial domain Ω can be very large, which implies that many grid points are needed for a proper spatial resolution, say hundred thousand to millions. Moreover, often one is interested in long term effects, so that the equations have to be integrated over long time intervals.

The huge size of the problems makes them difficult to implement, even on modern supercomputers. In order to obtain numerical solutions, without using excessive computer time or memory, fast and efficient numerical algorithms are crucial. The numerical results should be reliable and the numerical errors should be well below the errors introduced by physical approximations in the model. These demands cause a number of outstanding numerical challenges, some of which are discussed below.

2.1. Efficient chemistry schemes

In many applications the chemistry is stiff, which means that the reactions take place on very different time scales. To obtain accurate solutions it would be unnecessary to represent the very fast reactions, but for numerical stability this is required when using explicit methods. For this reason, such equations are commonly handled with implicit methods, where at each time step a system of nonlinear algebraic equations arises. Usually these implicit relations are solved with a Newton type iteration, which requires calculation of Jacobians and the solution of large linear systems, with much computer storage needed.

A way to overcome the problem of implicitness has been developed at CWI [4]. Instead of using a Newton iteration, one employs a Gauss-Seidel iteration. By using the special form of the chemical equations this leads to numerical schemes that are essentially explicit but still have the favourable stability properties of implicit schemes. Recent comparisons have shown that this approach can lead to codes which are for practical accuracy de-

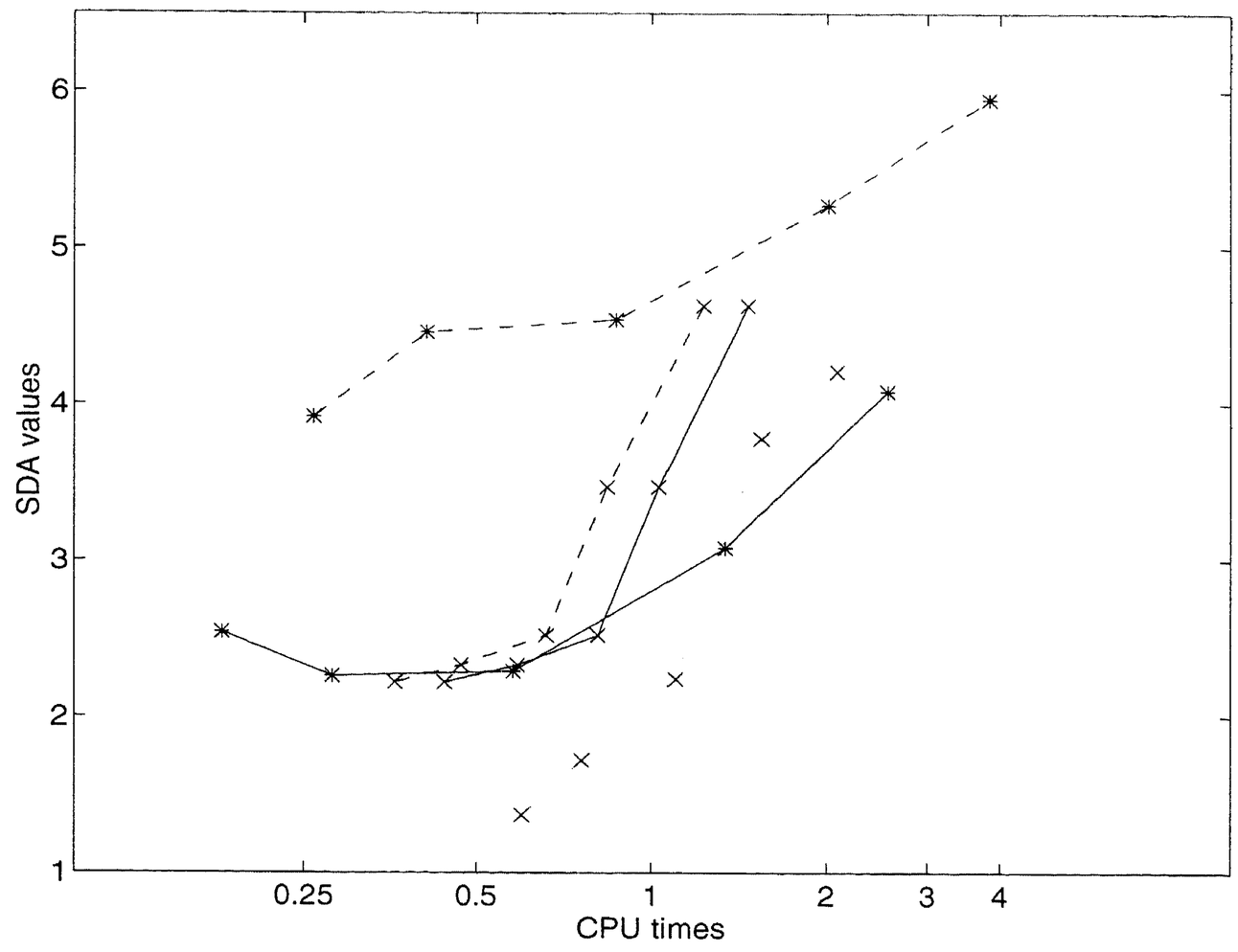

Figure 2. Results for TWOSTEP1 (*, solid), TWOSTEP2 (*, dashed), VODE1 (x, dotted), VODE2 (x, solid), VODE3 (x, dashed).

mands at least competitive with modern codes for general stiff problems, such as VODE, and for problems where the number of chemical substances is not too large significant gains in computing time (CPU) have been achieved.

An example is presented in figure 2 where the number of significant digits (SDA $= -\log_{10}(\text{error})$) is plotted against the CPU times for several versions of the codes TWOSTEP and VODE applied to a chemical ozone system with 15 substances; TWOSTEP is based on the BDF2 method with Gauss-Seidel iteration, whereas VODE uses a range of BDF methods with Newton iteration. The versions TWOSTEP1 and VODE1 are used as black-box solvers. In TWOSTEP2 some substances are grouped together which are known a-priori to have strong interactions. In VODE2 an analytic expression for the Jacobians is provided (calculated with the formula manipulation program MAPLE) and VODE3 uses also a manual reordering of the equations to exploit sparsity patterns for the linear algebra. In this comparison TWOSTEP1 needs less CPU time to reach the 1% error level than its black-box counterpart VODE1. The code TWOSTEP2 gives here a much better accuracy for given CPU time than the other VODE schemes.

2.2. Operator splittings

The different parts of the equations can be solved most efficiently by different numerical methods, but the different modules then have to be combined after each time step. This will lead to additional numerical errors, the so-called splitting errors. If the equations are strongly advection dominated, as they usually are in practice, these splitting errors can be strongly reduced, with little computational work, by solving the chemistry equations along the transport paths (characteristics).

It is still an open question which parts of the equations should be treated separately. In general, it is expected that numerical errors can be reduced by the simultaneous solution of processes with comparable time scales. In a recent research report (supported by the CRAY Research Grant Program) it has been shown that chemistry and vertical diffusion can be solved simultaneously in an efficient way by using a Gauss-Seidel approach, similar as for the chemistry only, combined with tri-diagonal solvers for the vertical diffusion.

2.3. Advection transport schemes

The accurate solution of advective transport is still a difficult numerical problem, due to the following requirements:

- The equations are mass conservative and a numerical method should mimic this behaviour.
- Concentrations are nonnegative, of course, but to maintain this property in the numerical solution the standard discretizations in space of order 2 or higher cannot be used.
- Strong gradients may be present in the concentration profiles. These gradients should not lead to oscillations (wiggles) but they should also not be smeared out by the numerical method.

A good compromise seems to be a combination of accurate spatial discretizations and flux limiters, together with explicit Runge-Kutta time integration, see [1]. However, if the advection is coupled with diffusion coefficients that are not small or if the grid spacing is fine in certain regions, then numerical stability will necessitate very small time steps and this will cause a degradation of efficiency.

Several ways to avoid such small time steps have been examined at CWI. With moderate diffusion coefficients one can use a Hopscotch type splitting to solve advection and diffusion simultaneously [3]. With small grid spacings in certain regions a dimension splitting approach can be advantageous, where the multi-dimensional advection problem is replaced by a series of 1-dimensional problems, which can be solved easily without any time step restriction for stability [2].

2.4. Local grid refinements
On the spatial domain there will be regions with smooth solutions and little chemical activity as well as regions with strong gradients and large chemical activity, caused by strong local emissions for instance. In the latter regions a high resolution is needed to avoid large numerical errors, whereas in the other regions less resolution would lead to better efficiency. This can be accomplished with local grid refinement where either the user specifies the high-resolution regions or where such regions are computed adaptively using suitable monitor functions. The use of local refinements for regional air-pollution models is being examined at present in the project EUSMOG.

2.5. HPCN: high performance computing and networking
The very large number of equations needed for an accurate description of transport and chemistry in many applications requires full use of advanced supercomputers, such as the CRAY C90, with possibilities of vector and parallel processing. At present, models are frequently simplified by restricting the number of chemical substances and spatial resolution in view of available computer capacity and speed. With the fast growth of the available meteorological and physical data, for example through remote sensing, it is expected that in the near future massively parallel computer systems will become increasingly important for the realization of new models.

The full use of such computer systems will force numerical analists to rethink the setup of algorithms and implementations. Although the design of massively parallel architectures is still in motion, numerical research in this direction is necessary to anticipate the future developments. At CWI a pilot project has started which examines the possibilities of the CRAY T3D system for use in regional atmospheric models.

3. PROJECTS AT CWI

At present three projects are carried out at CWI concerning transport of pollutants with various application backgrounds.

3.1. EUSMOG
The project EUSMOG is carried out in close cooperation with scientists of the Air Laboratory of RIVM – the Dutch National Institute of Public Health and Environmental Protection. RIVM also gives financial support for the project.

The goal of this project is to develop fast numerical algorithms for the prediction of smog episodes. The model uses a 4-layer parameterization in the vertical direction and a chemical ozone model with 15 chemical substances. The numerical research in this project is mainly devoted to the development of new chemical schemes and the use of local grid refinements

to represent local emission sources and to follow strong gradients in space and time.

Figures 3 and 4 are produced with CWIROS, the code which is being developed at CWI for summer and winter smog. Figure 3 gives a 5-day prediction of sulfur-dioxide SO_2 concentrations over Europe using emission data and meteorological input parameters from RIVM. Figure 4 gives the computational grid at ground level used at the end of the prediction. The grid refinement procedure is fully automatic and the grid adjusts itself to the solutions. Since the grid is strongly refined at small regions only, this procedure is much more efficient than using overall refinements.

3.2. TRUST

In the TRUST project we study the simulation of transport and bio-chemical interaction of salinity, pollutants, suspended material (such as sediment or mud), etc. in shallow seas. These seas play an important role as a link between land and ocean and usually have a highly productive ecosystem. As an example, we may consider the North Sea, which is partly surrounded by highly industrialized countries; as a consequence, it is subject to a large number of effects, such as natural sewage in urban sewer outlets, pollution by rivers, recreation, heavy transport, and, in this example, also the exploration of minerals (gas and oil), and fishery. Moreover, the interaction with the ocean and the atmosphere has a significant influence on the ecosystem. Although we are still far away from the final goal, i.e., a thorough understanding, and control, of the marine ecosystem dynamics in shallow seas, the TRUST project tries to contribute in pushing the problem one step further into the direction of its final solution. The research at CWI concentrates on the development of fast, parallel algorithms for the time integration of the advection-diffusion-reaction equations [3]. In contrast to air-pollution models, the chemical reactions in water are usually quite slow, implying that the reaction terms do not need special concern, and can be treated by standard explicit methods. For the advection-diffusion part, we developed a so-called Hopscotch method, which seems to be very promising, both with respect to numerical properties as well as with respect to vectorization/parallelization capabilities. An other point of interest in this context is that various regions require different spatial grid resolution. For example, to capture the local phenomena in coastal regions and estuaries, a much finer grid is needed. This is clearly illustrated in figure 5, showing the distribution of suspended material in the North Sea. Since the information shown in this figure is very costly and time-consuming to obtain, it is clear that efficient numerical simulations are of utmost importance. In the TRUST project, there is a close co-operation with the research groups at the TUD (Delft University of Technology), RIKZ (National Institute for Coastal and Marine Management), and Delft Hydraulics. Financial support is obtained from the CRAY

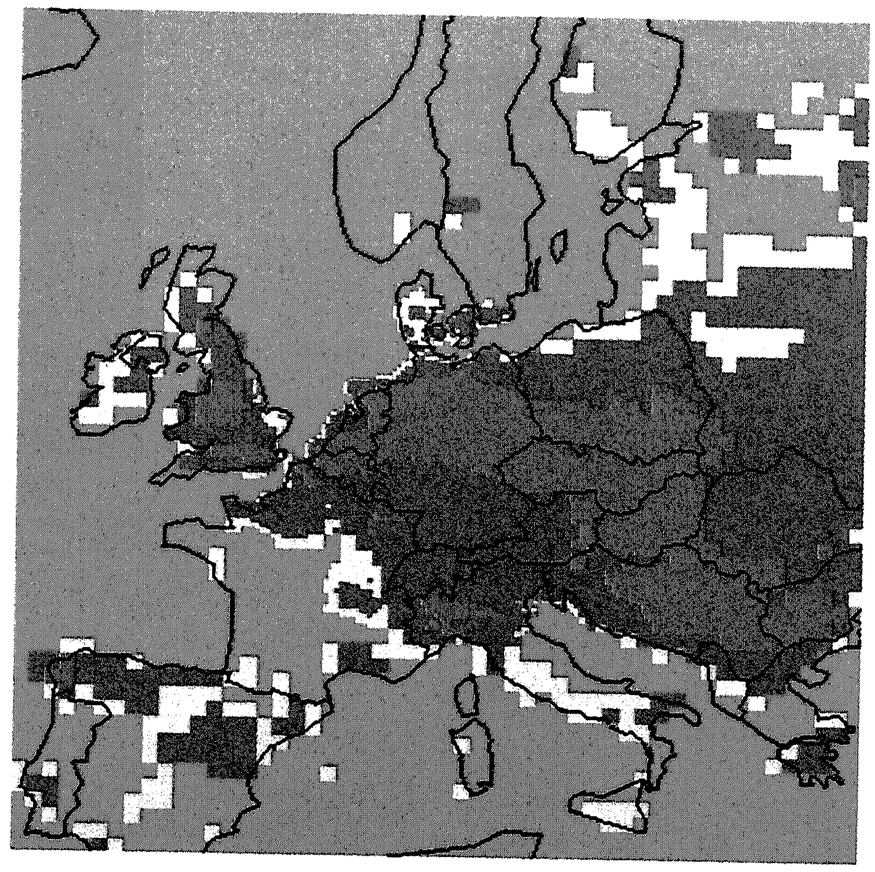

Figure 3. SO_2 concentrations over Europe, 5-day prediction.

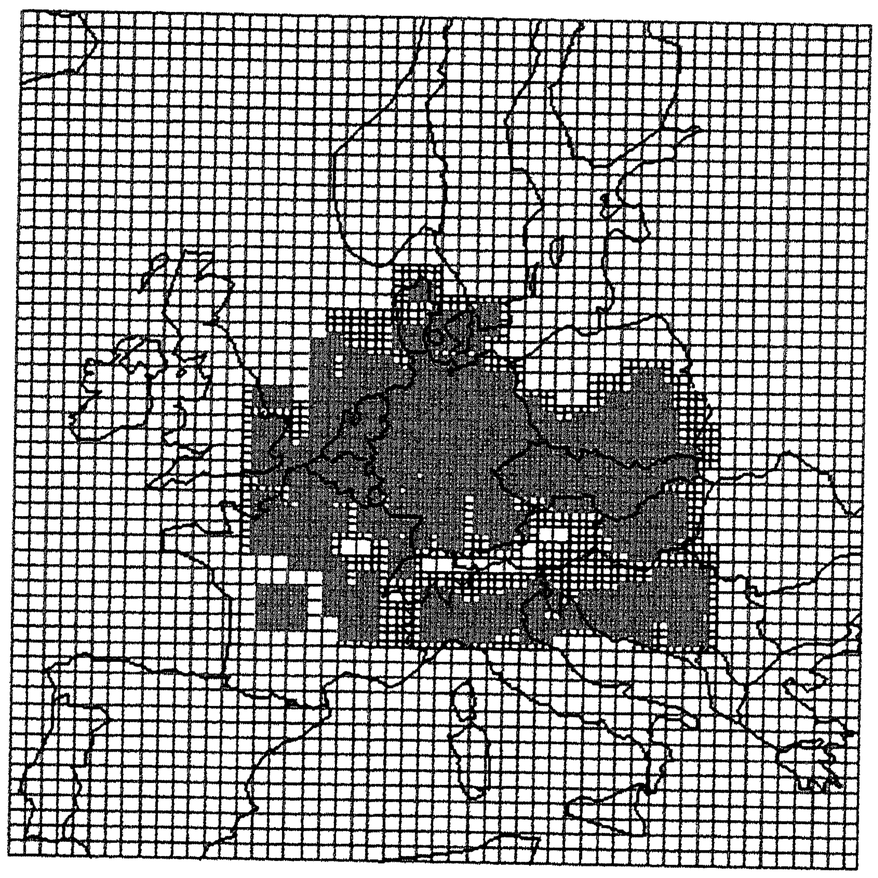

Figure 4. Horizontal grid at day 5 in calculation of Figure 2.

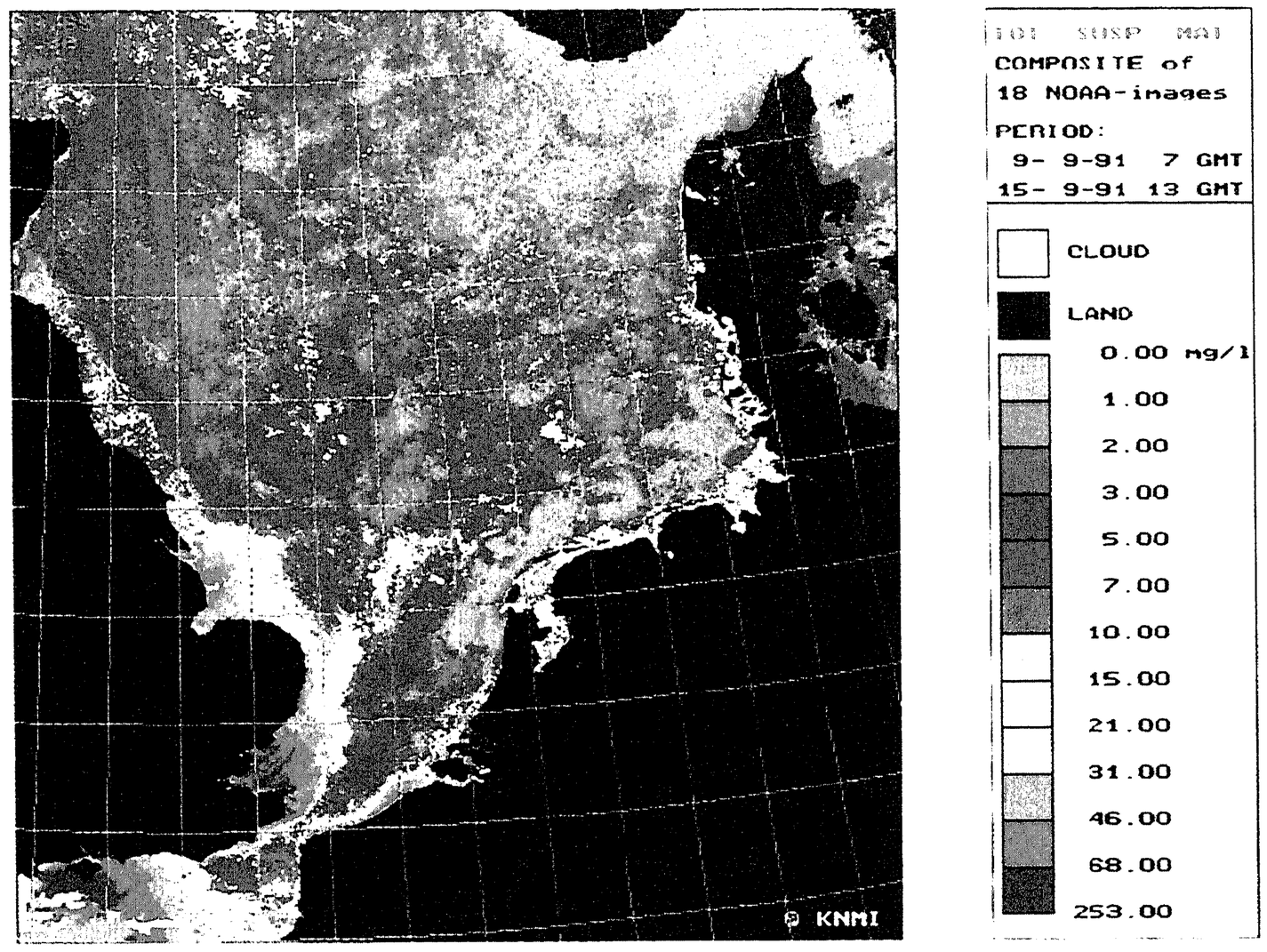

Figure 5. Remote sensing observations of the North sea area show that suspended fine-grained particles are predominantly concentrated in relatively narrow zones along the coastal boundaries.

Research Grant Program and the NOWESP project, which is part of the MAST II program of the EU.

3.3. CIRK

CIRK is a joint research programme of CWI, IMAU (Institute for Marine and Atmospheric Research) , RIVM (National Institute of Public Health and Environmental Protection) and KNMI (Royal Netherlands Meteorological Institute). The goal of the programme is to develop a model for global transport and chemistry of trace constituents in the troposphere, which should improve present models by using more advanced numerical methods and more refined parameterizations of physical and chemical processes. The development of such global models, which cover the whole troposphere and part of the stratosphere, is becoming increasingly important for atmospheric

scientists to study the long-term effects of regional pollution sources.

The research at CWI is financially supported by RIVM, and is directed to numerical problems associated with advection on spherical geometries, fast chemical solvers, computational aspects of operator splitting, and vectorization and parallelization aspects. This work is combined with the research at IMAU, RIVM and KNMI concerning, among other things, troposphere-stratosphere exchanges and sub-grid parameterizations for vertical transport and cloud systems.

Below some of the relevant references are given. More references for the projects EUSMOG and CIRK can be found at the WWW site
' http://www.cwi.nl/cwi/projects/nw1.4.html '.

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