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## TECHNICAL NOTE

## COMPUTATION OF GLOBAL PHOTOCHEMISTRY WITH SMVGEAR II

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**Abstract**—A computer model was developed to simulate global gas-phase photochemistry. The model solves chemical equations with SMVGEAR II, a sparse-matrix, vectorized Gear-type code. To obtain SMVGEAR II, the original SMVGEAR code was modified to allow computation of different sets of chemical reactions for urban, free-tropospheric, and stratospheric regions during the same model run. SMVGEAR was also modified to allow grid cells in each region of the atmosphere to be reordered according to stiffness of the chemical equations, each time interval. Reordering speeds solutions by a factor of more than two compared to not reordering. Two 30 day simulations of chemistry coupled (with feedback) to radiation field calculations over a global grid, were performed. In one simulation, results were obtained for 49,680 grid cells (15-layers general circulation model grid), and in the other, results were obtained for 96,048 cells (29-layer grid). In both simulations, 200 “urban” (assumed below 900 mb) chemical equations were solved, 169 “free tropospheric” (225–900 mb) equations were solved, and 115 “stratospheric” (above 225 mb) equations were solved. The times required for the two 30 day simulations on a single processor of a Cray 90 were 6.2 and 11.6 h, respectively, at an average speed of approximately 362 megaflops.

**Key word index:** Gear code, photochemistry, global modeling, air pollution, ordinary differential equations.

## 1. INTRODUCTION

Models that simulate atmospheric photochemistry require the use of a stiff-ordinary differential equation solver. Because the number of model grid cells are often large and integration periods are often long, the numerical solver must be computationally fast. Also, the residual error from the solver must be small. Because most accurate solvers are relatively slow, modelers have often reduced computer time by either reducing the size of the modeling domain, reducing the number of species and reactions solved, or simplifying the chemical solver.

For example several excellent tropospheric or stratospheric chemical modeling studies have been carried out in two, instead of three, dimensions, because computer time was limited (e.g. Crutzen *et al.*, 1978; Logan *et al.*, 1981; Derwent, 1982; Isaksen and Hov, 1987; Austin, 1991; Hough, 1991; Kanakidou *et al.*, 1991; Garcia and Solomon, 1983, 1994; Ko *et al.*, 1993; Tie *et al.*, 1994; Strand and Hov, 1994). In addition, pioneering three-dimensional studies have often been carried out with either parametrized chemical solvers (e.g. Jacob *et al.*, 1989) or family solvers (e.g. Kaye and Rood, 1989; Rose and Brasseur, 1989; Kao *et al.*, 1990; Lefevre *et al.*, 1994; Elliott *et al.*, 1995).

In order to solve many chemical reactions in a large global grid with a high order of accuracy, either faster computers or faster integrators are required. Recently, Gear's predictor-corrector scheme (Gear, 1971) was modified with sparse matrix and vectorization techniques to obtain a fast integrator (Jacobson 1994; Jacobson and Turco, 1994). The resulting code, SMVGEAR (sparse matrix, vectorized Gear code), achieves over 360 megaflops on one processor of a Cray 90 computer, solves hundreds of chemical rate equations in large model domains, and maintains the accuracy of Gear's original code.

For the work described herein, SMVGEAR was modified to solve chemical rate equations over a global grid domain, where the stiffness of the equations varies significantly from place to place and time to time. In addition, SMVGEAR was modified to serially solve different sets of chemical equations for different regions of the atmosphere. For example, stratospheric, free tropospheric, and urban chemistry can be solved during the same model simulation. Specifying different chemical mechanisms for different regions of the atmosphere reduces computational requirements significantly. The modified version of SMVGEAR is called SMVGEAR II.

To simulate global photochemistry, SMVGEAR II was coupled to a radiative transfer model. Optical properties of all photodissociating gases in the model were used to calculate the radiation fields; consequently, feedback between chemistry and radiation occurred. Subsequently, computer timings were performed. In the following sections, SMVGEAR II is discussed and analyzed and computer timings are shown.

## 2. MODIFICATIONS TO SMVGEAR

## 2.1. Description of SMVGEAR

SMVGEAR is derived from Gear's, predictor-corrector method (Gear, 1971), which was based on the backward differentiation formula (BDF). The BDF can be written as (e.g. Byrne *et al.*, 1977; Hindmarsh, 1983)

$$y_n = h\beta_0 \frac{dy_n}{dt} + \sum_{j=1}^q \alpha_j y_{n-j} \quad (1)$$

where  $y_n$  is an array of  $N$  real variables at time  $t_n$  (and  $n$  denotes the time step number),  $dy_n/dt = f(t_n, y_n)$  is an array of first derivatives for each value of  $y_n$  at time  $t_n$ ,  $h = t_n - t_{n-1}$  is the current time step,  $q$  is the current order of the method ( $1 \leq q \leq 5$ ), and  $\alpha_j$  and  $\beta_0$  are scalar multipliers ( $\beta_0 > 0$ ) that depend on the current order. The boundary

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conditions,  $y_0 = y(t_0)$ , define the initial values for the problem.

A solution to equation (1) can be found by solving the modified Newton iterative equation, written as

$$P_n [y_{n(m+1)} - y_{n(m)}] = -y_{n(m)} + h\beta_0 f(t_n, y_{n(m)}) + \sum_{j=1}^q \alpha_j y_{n-j} \quad (2)$$

where

$$P_n \approx I - h\beta_0 J_n \quad (3)$$

is a predictor matrix,  $m$  is the iteration number,  $I$  is the identity matrix, and

$$J_n = J(t_n, y_n) = \left[ \frac{\partial f_n^i}{\partial y_n^j} \right]_{i,j=1}^N \quad (4)$$

is the Jacobian matrix of partial derivatives.

SMVGEAR uses Gear's solution method; however, sparse matrix and vectorization techniques were applied to improve computational speed significantly. About half of the speedup of SMVGEAR was due to sparse-matrix techniques and half was due to vectorization. The primary sparse-matrix technique was to reorder the matrix of partial derivatives, then to eliminate all subsequent multiplies by zero during matrix decomposition and back substitution. The primary vectorization technique was to divide the grid domain into blocks of approximately 500 grid cells each, then to solve equations in each block together. In such cases, almost every inner loop in the code was vectorized around the grid-cell loop, which had length equal to the number of grid cells in a grid block.

The average size of a grid block in the model was chosen as a compromise between competing factors. For example, when a grid block is large, speed from vectorization increases. However, the number of total operations also increases because more grid cells are tied together whenever a single cell causes many iterations for the block as a whole. A compromise grid block size of 500 was chosen because it is large enough to result in 90% maximum vectorization speed on Cray computers (NAS, 1992) but small enough not to cause too many excess calculations. Nevertheless, excess calculations do occur. Thus, to reduce excess calculations, a method was developed here to reorder grid cells so that cells with similar stiffness are placed in the same grid blocks.

## 2.2. Predicting stiffness and reordering grid cells

In SMVGEAR, grid-cells were grouped arbitrarily into blocks of approximately 500 cells each. For example, a grid with dimensions  $40 \times 50 \times 10$  (20,000 grid cells) gave rise to 40 grid blocks. Cells for each block were gathered sequentially, from the southwesternmost corner of the surface layer towards the east, then to the next latitudinal row, and then to the next vertical layer.

While this method of cell-gathering was convenient, it resulted in grid blocks containing groups of adjacent grid cells with different stiffnesses. Examples of where stiffness varies among adjacent cells are regions where sunrise and sunset occur, regions where photodissociation rates differ vertically, and regions where strong concentration gradients occur (such as near emissions).

To reduce excess computations caused by grouping grid cells with varying stiffness together, a technique was developed to regroup grid cells each chemistry time interval according to stiffness. A chemistry time interval is defined as a period during which photochemistry (or photochemistry plus emissions) is solved alone in the model. Before and after a chemistry time interval, transport and other processes are allowed to affect concentrations; however, during a chemistry interval, chemistry is integrated alone, with variable integration time steps. In typical model applications, chem-

istry time intervals vary between 300 and 1800 s while integration time steps vary between  $10^{-8}$  and 1800 s.

In SMVGEAR II, grid cell stiffness is determined in two ways. First, for each grid cell, except for those in which sunrise occurs during the time interval, stiffness is estimated with

$$S_{k,n} = \frac{1}{N} \sum_{i=1}^N \left( \frac{dy_{i,k,n}/dt}{y_{i,k,n} + E_{Ai,k,n}} \right)^2 \quad (5)$$

where  $y_{i,k,n}$  and  $E_{Ai,k,n}$  are the concentration and absolute error tolerance, respectively, of species  $i$  at time  $n$  in grid cell  $k$ , and  $N$  is the order of the matrix of partial derivatives. After the stiffness is estimated for each grid cell with equation (5), the cells are reordered from smallest to largest values of  $S$  with the *heapsort* sorting routine (Press *et al.*, 1992), which is an  $N \log_2 N$  process. Subsequently, the reordered cells are placed sequentially into grid blocks. Thus, if each grid block contains 500 cells, then the first block will contain the 500 cells with the lowest predicted stiffness values and the last block will contain the 500 cells with the highest predicted stiffness values.

To test the effects of reordering, a 4 h simulation over a global grid domain was performed and statistics were gathered. Figure 1 shows the correlation between the minimum stiffness predicted for each grid block over all time intervals vs the resulting number of matrix back-substitution calls. The number of back-substitution calls correlates positively to stiffness. The figure shows that, in general, the larger the minimum predicted stiffness for the block, the greater the number of back-substitution calls (thus the greater the actual stiffness of equations in the block). Consequently, grouping grid cells that have similar initial stiffnesses reduces excess calculations. To test this hypothesis further, comparisons of results with and without reordering were made. Table 1 compares statistics from global simulations. The table shows that the number of back-substitution calls decreased by a factor of over two when cells were reordered compared to when they were not reordered.

When sunrise occurs, equation (5) is not a good predictor of stiffness since the equation predicts stiffness at the beginning of the interval, before sunrise occurs, and stiffness changes rapidly after sunrise. Instead, sunrise cells are ordered here by time of sunrise. Further, since all sunrise cells face similar stiffness difficulties as each other, they are sequestered into their own blocks. Sunset cells are also given their own blocks. However, they are ordered by stiffness, as predicted from equation (5), since stiffness of sunset cells only decreases between the beginning and end of a time interval. Table 1 shows that the computer time and number of back-substitution calls decreased when cells were sequestered and reordered during sunrise and sunset as opposed to when they were not sequestered or reordered at all.

## 2.3. Solving stratospheric, tropospheric, and urban chemistry serially

The second new feature of SMVGEAR II is a modification that allows it to solve different sets of chemistry for different regions of the atmosphere. For example, many chemical reactions that are important in the stratosphere are not so important in the troposphere. Similarly, many reactions that are important in urban regions are not so important in the free troposphere or stratosphere.

The chemical mechanism used for urban air consist of 200 inorganic and organic reactions (176 kinetic and 24 photolysis reactions). The tropospheric mechanism consists of 169 reactions (147 kinetic and 22 photolysis), which include all the urban reactions except for aromatic reactions. Finally, the stratospheric mechanism consists of 115 reactions (81 kinetic and 34 photolysis), which include inorganic chemistry, light organic chemistry, and chlorine and bromine chemistry. Most reactions in all three mechanisms were

