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A Pseudo-Non-Time-Splitting Method in Air Quality Modeling

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This paper presents a pseudo-non-time-splitting (PN) method that combines the different processes in air quality modeling, such as advection, diffusion, and emission, with chemical reaction computations. The purpose is to reduce the errors generated by the time-splitting method by taking the advantage of the fact that the integration time steps in the chemical reaction computation are much smaller than those used in other processes. An example of a cosine hill rotation with chemical reactions is used to demonstrate this new method. We show that the PN method produces results with similar accuracy to those from the conventional time-splitting method but consumes about 20% less execution CPU time.

1. INTRODUCTION

In air quality modeling, the equation describing the advection, diffusion, and chemical reactions of the chemical species is

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{u} c_i) = \nabla \cdot (\mathbf{K} \nabla c_i) + f_i + e_i, \tag{1}$$

where c_i is the concentration of the ith pollutant species, **u** is the wind velocity field, **K** is the diffusivity tensor, f_i is the chemical reaction rate, and e_i is the emission source.

Several factors cause difficulty in solving this system of partial differential equations. First, the modeling region is often three-dimensional and is very large. Second, the number of pollutant species also is not small. Finally and most importantly, the chemical reaction part f_i is normally very stiff; that is, the characteristic reaction times for different species differ greatly. In the case in which only chemical reactions take place, an accurate computation would require a backward differentiation scheme and very small integration step sizes. On the other hand, the advection, diffusion, and emission parts can be computed accurately with much larger time steps.

In order to exploit the fact that almost all the processes except the chemical reaction process may be integrated with much larger integration time steps, in practice, the approach is to use the time-splitting method, or the method of fractional steps. This method was first introduced by Yanenko [1] and described in Marchuk [2, 3] and Yanenko *et al.* [4]. In this approach, (1) is replaced by the following equations:

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{u} c_i) = 0, \tag{2}$$

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{K} \nabla c_i) = 0, \tag{3}$$

$$\frac{\partial c_i}{\partial t} = e_i, \tag{4}$$

$$\frac{dc_i}{dt} = f_i(c_1, \ldots, c_n), \quad i = 1, \ldots, n. \tag{5}$$

These equations are solved sequentially and each equation takes the results from the previous one as its initial conditions. A time step Δt, often 6 to 15 min in real modeling situations, is used to carry the concentrations c_i one step forward in Eqs. (2), (3), and (4). However, in the chemical reaction equation (5), this single time step Δt has to be cut into many small sub-integration time steps, called chemical integration time steps δt_j, such that $\Sigma_{j=1}^{M} \delta t_j = \Delta t$. The number M depends on the particular chemical solvers used, the error tolerance required, and the initial conditions of $c_i, i = 1, \ldots, n$. For some less accurate but fast solvers, such as the quasi-steady-state assumption (QSSA) solver [5], M typically is between 20 and 50. For some accurate solvers like the Livermore solver for ordinary differential equations [6] (LSODE), M is often greater than 30.

The time-splitting method separates the original simultaneous complex processes into sequential steps. The errors it introduced, in general, interact with the errors from the computation of other process in a complex fashion. In this note, a pseudo-non-time-splitting method is proposed. We then apply this method to a cosine hill rotation with chemical reactions example to demonstrate its accuracy and the computational time. In Section 2, we describe the methodology, and in Section 3, we present the results and analyze

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the differences between our results and those from the time-splitting method. The conclusions are drawn in the last section.

2. METHODOLOGY

In this section, we describe the method combining the advection and the chemical processes. The combination of other processes (such as diffusion, emission, and cloud formation) with the chemical process may be treated in a similar manner.

In the time-splitting method, the advection equation

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{u}c) = 0 \tag{6}$$

is computed and the results are used as the initial condition in the chemical reaction computation. In a finite difference scheme in both space and time, the concentration of species is represented as

$$c^n_{ijk} = c(n\Delta t, i\Delta x, j\Delta \mathrm{y}, k\Delta z). \tag{7}$$

After the computation of (6), the new species concentration at time $(n + 1)\Delta t$ can be expressed as

$$c^{n+1}_{ijk} = c^{\mathrm{n}}_{ijk} - \Delta t \nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}, \tag{8}$$

where

$$\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk} = G(\mathbf{u}, c, t = \Delta t, \boldsymbol{\nabla} \cdot (\mathbf{u}c))^n_{ijk}. \tag{9}$$

The specific form of $\nabla \widehat{\cdot (\mathbf{u}c)}$ depends on the advection solver used.

Since the concentrations are advanced at the time step Δt, which is much larger than the chemical integration time steps δt_j, $\Sigma^M_{j=1}\ \delta t_j = \Delta t$, efficiency is achieved in this computation.

On the other hand, the errors introduced from the time-splitting method are obvious. Consider computing the whole equation governing both the advection and chemical reactions

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{u}c) = f \tag{10}$$

from time $t = 0$ to $t = \Delta t$. The stiffness of the chemical reaction operator f restricts that we use chemical integration time steps δt_j, $\Sigma^M_{j=1}\ \delta t_j = \Delta t$. After time step δt_j is advanced, the concentration c changes due to both the advection and the chemical reaction. This change can be expressed as

$$c^{(j+1)} = c^{(j)} + \delta c^{(j)}_{\mathrm{ad}} + \delta c^{(j)}_{\mathrm{ch}}, \tag{11}$$

where $\delta c^{(j)}_{\mathrm{ad}}$ and $\delta c^{(j)}_{\mathrm{ch}}$ represent the increase of the concentration due to the advection and the chemical reaction, respectively. In the next step computation δt_{j+1}, $c^{(j+1)}$ contains both the $\delta c^{(j)}_{\mathrm{ad}}$ and $\delta c^{(j)}_{\mathrm{ch}}$ part and moves with the wind velocity and changes through the chemical reaction. In other words, in this computation, $\delta c^{(j)}_{\mathrm{ad}}$ will immediately take part in the chemical reaction and will make contributions to $c^{(j+2)}$, but not wait to do so until all the chemical reactions are finished at $t = \Delta t$. Therefore, physically speaking, the changes due to the advection and any other physical process should immediately be included in the chemical reaction.

We may make these different processes happen together in the following way. That is, we may still compute the advection process separately according to (6), but do not update the concentrations. Instead, we may store terms $\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}$ obtained from this computation and then retrieve them in the chemical reaction computations. In this way, the original equation

$$\frac{\partial c}{\partial t} + \nabla \cdot (\mathbf{u}c) = f \tag{12}$$

has been replaced by

$$\left(\frac{\partial c}{\partial t}\right)_{ijk} + \nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk} = f_{ijk}, \quad n\Delta t \le t \le (n + 1)\Delta t, \tag{13}$$

and like the time-splitting method, the computation is also carried out in each of the grid cells. We call this strategy the pseudo-non-time-splitting method. This method will bring changes due to the advection and, similarly, any other processes, immediately into the chemical reaction. Meanwhile, the advantages of the time-splitting method are preserved since $\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}$ terms are still computed in large time steps.

Moreover, for a lot of the advection schemes, the order of the accuracy is higher than one. In these cases, the advection term $\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}$ is a function of time. For a lot of the advection solvers, such as those generated from the linear multistep methods, $\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}$ has a Taylor expansion form

$$\begin{aligned} \nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}(t) = {} & (\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk})^{(0)} + (\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk})^{(1)} t \\ & + (\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk})^{(2)} t^2 \cdots. \end{aligned} \tag{14}$$

In the time-splitting method, $t = \Delta t$. However, in (14), since the chemical integration time steps are much smaller than Δt, $\nabla \widehat{\cdot (\mathbf{u}c)}^n_{ijk}$ should be evaluated at these chemical integration time steps where the solution is computed. We

TABLE I

Reaction	Rate
$HC + OH \rightarrow 4RO_2 + 2HCHO$	$k_1 = 6.0 \times 10^{-12}$
$HCHO + h\nu \rightarrow 2HO_2 + CO$	$j_2 = 7.8 \times 10^{-5} e^{-0.87/\cos\theta}$
$RO_2 + NO \rightarrow NO_2 + HCHO + HO_2$	$k_3 = 8.0 \times 10^{-12}$
$NO + HO_2 \rightarrow NO_2 + OH$	$k_4 = 8.3 \times 10^{-12}$
$NO_2 + h\nu \rightarrow NO_2 + O_3$	$j_5 = 1.0 \times 10^{-2} e^{-0.39/\cos\theta}$
$NO + O_3 \rightarrow NO_2 + O_2$	$k_6 = 1.6 \times 10^{-14}$
$O_3 + h\nu \rightarrow O_2 + O^1D$	$j_7 = 1.9 \times 10^{-4} e^{-1.9/\cos\theta}$
$O^1D + H_2O \rightarrow 2OH$	$k_8 = 2.3 \times 10^{-11}$
$NO_2 + OH \rightarrow HNO_3$	$k_9 = 1.0 \times 10^{-11}$
$CO + OH \rightarrow CO_2 + HO_2$	$k_{10} = 2.9 \times 10^{-13}$

should use $\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk}(t_m)$ instead of $\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk}(\Delta t)$, where $t_m = \Sigma_{j=1}^{m} \delta t_j$.

In the application of the pseudo-non-time-splitting method, the advection term $\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk}(t)$ has to be obtained from the particular advection solver used. For a crude application, this term can simply be set as a variable in space, but constant in time from the formula

$$\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk} = \frac{c^{n+1}_{ijk} - c^n_{ijk}}{\Delta t}, \tag{15}$$

where c^n_{ijk} and c^{n+1}_{ijk} is the concentration before and after the advection equation is computed. However, whenever possible, the advection term should be worked out as a variable in both space and time so that it can be evaluated at the chemical integration time steps.

3. TEST EXAMPLE

We need to use an example and compute the results from both the pseudo-non-time-splitting method and the time-splitting method, and then compare them with some known accurate results, respectively. The test example we use is the well-used simplified chemical mechanism with 10 chemical species. This example was previously used by Hov *et al.* [7], Odman and Russell [8], and Chock and Winkler [9]. The advection of the specise is in a two-dimensional domain with 32 × 32 grid cells and the wind velocity is the rotational flow velocity around the center of the domain. The chemical reactions are given in Table I, where the zenith angle θ is held constant at 71.5° throughout the computations. The initial conditions for the six species, namely, OH, HO_2, RO_2, O_3, O^1D, and CO, are that they are initially constant in the whole domain. The other four species, HC, HCHO, NO, and NO_2, will have a cosine hill shape distribution with the center at grid cell (24,16). In the rest area, they have constant values that are the same as the values at the foot of their cosine hills. This initial condition information and the H_2O value used are contained in Table II. The x and y rotational velocity component is $u = -\omega(y - y_c)$ and $v = \omega(x - x_c)$, respectively, where $x_c = 16$, $y_c = 16$ is the center of the domain. The angular velocity ω is set up such that one full rotation needs 24 h. The time increment, Δt, is chosen to be 300 s.

The advection solver we use is the accurate-space-derivative (ASD) method [10]. In ASD, the advancement of the species is carried through a truncated Taylor series

$$c^{n+1} = c^n + \frac{\partial c^n}{\partial t}\Delta t + \frac{\partial^2 c^n}{\partial t^2}\frac{\Delta t^2}{2!} + \cdots. \tag{16}$$

where the partial derivatives are obtained from the advection equation by using Fourier transformations. This expression is ready to be used in the pseudo-non-time-splitting method with only a small change. In a third-order scheme, the advection term $\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk}(t)$ we need is just

$$\begin{gathered}\nabla \cdot \widehat{(\mathbf{u}c)}^n_{ijk}(t) = \frac{\partial c^n_{ijk}}{\partial t} + \frac{\partial^2 c^n_{ijk}}{\partial t^2} + \frac{\partial^3 c^n_{ijk}}{\partial t^3}\frac{t^2}{2!}, \\ t = t_m = \sum_{j=1}^{m} \delta t_j, \quad 1 \le m \le M, \quad \sum_{j=1}^{M} \delta t_j = \Delta t.\end{gathered} \tag{17}$$

Besides the easy adjustment of the advection term from the ASD to the pseudo-non-time-splitting method, we also chose ASD as the advection solver because this method is the most accurate advection solver, as reported by Chock and Winkler [9]. We use LSODE as the chemistry solver.

4. RESULTS

4.1. *Accuracy*

After 24 h one full rotation is completed and the cosine hills come back to their original positions. Therefore, the

TABLE II

Initial	Conditions	Molecules/cm^3
Species	Background	Peak
HC	2.50×10^9	1.00×10^{11}
HCHO	1.25×10^{10}	5.00×10^{11}
NO	2.50×10^9	1.00×10^{11}
NO_2	2.50×10^9	1.00×10^{11}
OH	1.00×10^5	
HO_2	1.00×10^6	
RO_2	1.00×10^6	
O^3	5.00×10^{11}	
O_1D	1.00×10^{-3}	
CO	1.00×10^{12}	
H_2O	2.50×10^{15}	Fixed constant

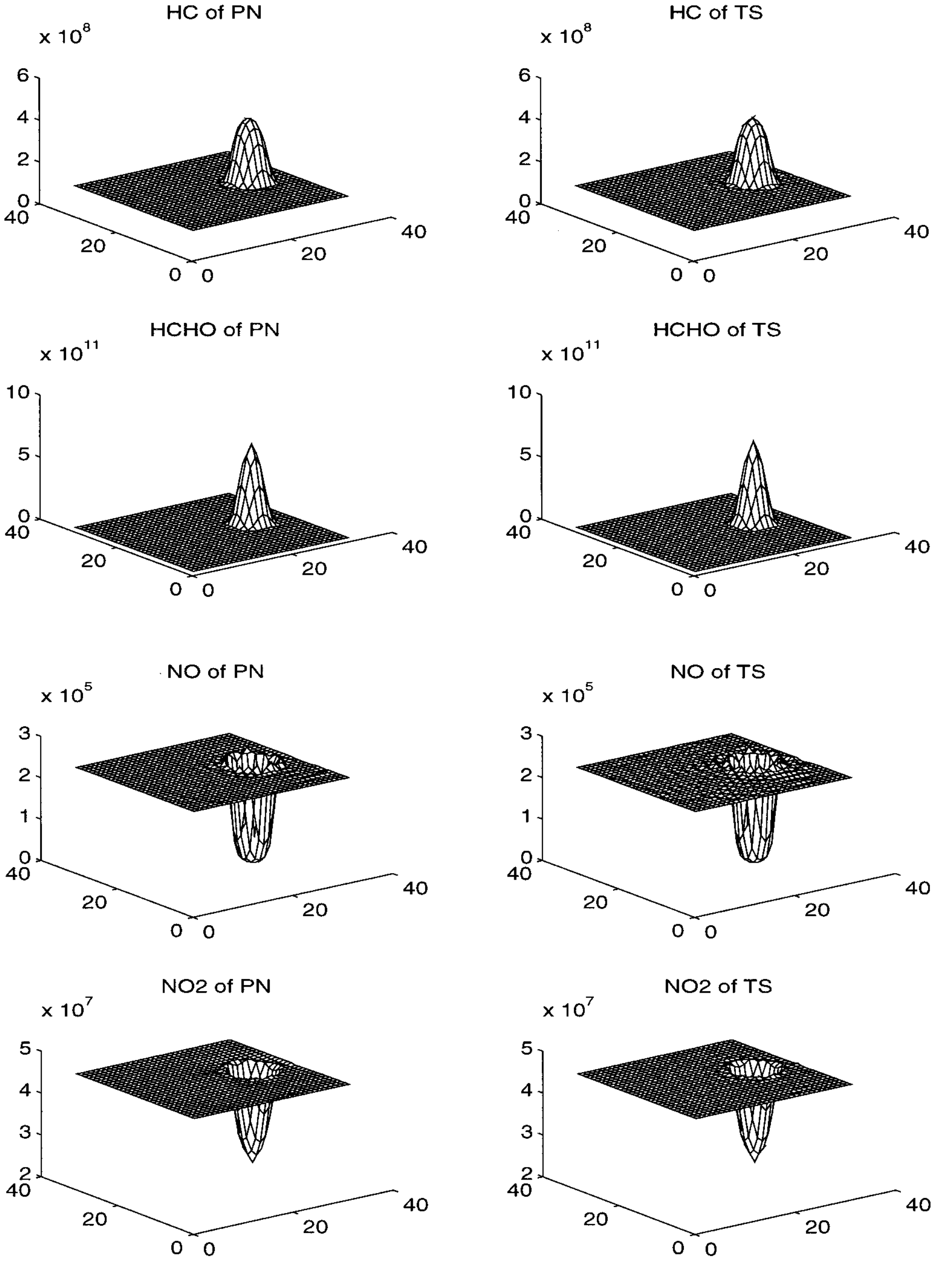

FIG. 1. Comparison of the concentration distributions after one full rotation in 24 h. PN: pseudo-non-time-splitting method; TS: time-splitting method.

species concentrations after one full rotation of advection and chemical reactions are compared against the "exact" results. The "exact" results we define here are the results computed from only the chemical reactions in 24 h. In this "exact" results computation, the absolute tolerance *atol* in LSODE is set to zero and the relative tolerance *rtol* is set to $1 \cdot e^{-9}$. In the computation of the pseudo-non-time-splitting method and the time-splitting method, *atol* in LSODE is set to zero and *rtol* is set to $1 \cdot e^{-4}$.

The normalized absolute difference (NSD) introduced by Chock and Winkler [9] and the ratio of the peak values of various species at the center of the cosine hills are the main two criteria we used in the comparison. The NSD is the sum of the absolute values of concentration differences between the "exact" and the computed results over all the grid cells, divided by the sum of the "exact" concentrations over the cosine hill domain. The ratio of the peak values is the ratio of the computed peak values of different species over the corresponding values of the "exact" results at the center of the cosine hills. Besides the NSD and the ratio

TABLE III

	CPU time		Number of F and J Evaluations			
rtol	$1.e^{-4}$	$1.e^{-7}$	$1.e^{-4}$		$1.e^{-7}$	
PN	2677	5800	3010967(F)	963643(J)	9896162(F)	2196817(J)
TS	3280	7770	4149593(F)	1357955(J)	14668785(F)	2784341(J)
Saving	19%	25%	27%	29%	32%	21%

of peak values, the overall distributions of several species will also be plotted.

Figure 1 shows distributions of HC, HCHO, NO, and NO2 after one full rotation in 24 h from both the PN (pseudo-non-time-splitting) method and the TS (time-splitting) method. The distributions of other species are not shown here since the difference is invisible. It is easy to see that the distributions from the PN method and the TS method are very similar. Generally speaking, the results from the PN method have less ripples than those from the TS method. Figure 2a shows the ratio of peak values and Fig. 2b shows the NSD values for all the species. From Fig. 2a it is seen that the ratios of peak values from the PN method for all the species are slightly lower than the corresponding ratios from the TS method except that of NO. Figure 2b shows that the NSD values for the two methods are very close.

4.2. *The CPU Time*

At the beginning of the investigation, it was expected that the CPU time consumed by the PN method would be longer than that of the TS method. In the PN method there is some additional work. The advection terms have to be stored after the advection computation is done and, later on, be retrieved in the chemical rate equations. However, the CPU time actually consumed in the PN method is less than that of the conventional time-splitting method. Table III contains the CPU times spent and the numbers of the evaluation of the chemical reaction rate function F and the jacobian matrix J on the comparison runs in Figs. 1, 2, together with the similar information in the computation at $rtol = 1 \cdot e^{-7}$ case.

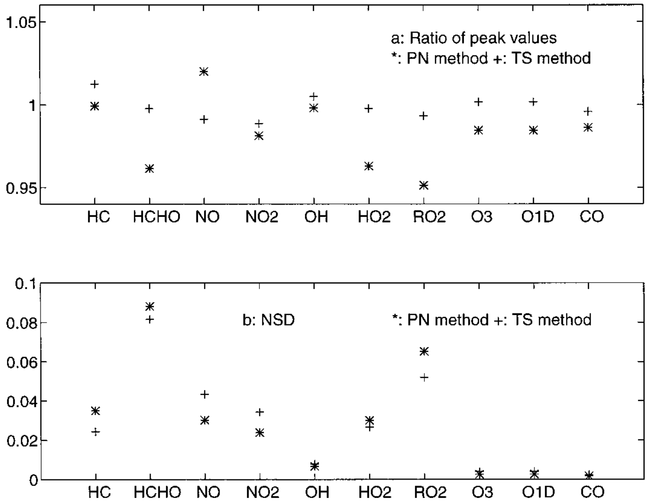

FIG. 2. (a) The normalized absolute differences (NSD) of concentrations after one full rotation in 24 h. (b) The ratios of the peak value at (24,16): *, PN; +, TS.

The CPU time saving from the PN method over the TS method is beyond our expectation. The reason for this saving is not very clear at this time. A tentative explanation is that since advection and chemical processes are combined together, the errors from the advection equation are reduced in the whole process since the chemistry integration time step δt_j is much smaller than Δt. For example, if the error from the advection computation is $err \sim at$, where a is a constant, the total error after Δt will be $err \sim a\Delta t$ in the TS method. On the other hand, in the PN method, we have $\Delta t = \Sigma_{j=1}^{M} \delta t_j$, where δt_j is much smaller than Δt. Therefore, after Δt time, the error accumulated will look like

$$err \sim \int_0^{\Delta t} atdt = a\frac{\Delta t^2}{2}.$$

Therefore, with smaller errors from the advection equation, the species concentrations are more accurate and thus converge much faster in the chemistry solver than in the TS method.

5. CONCLUSIONS

A PN method is presented and applied to the cosine hill rotation computation with the advection solver of ASD and chemistry solver of LSODE. It is shown that this new method can produce results of similar accuracy with the conventional time-splitting method, but consumes about 20% less CPU time. Also, the noises of the concentration distributions of the species are reduced.

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