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Development and analysis of a non-splitting solution for three-dimensional air quality models

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Abstract

This paper examines the errors introduced by operator splitting techniques in air quality models. Results are presented for different time steps used in the splitting schemes as well as for different ordering in which the operators are computed. Furthermore, a non-splitting technique is developed to analyze the performance of operator splitting techniques in air quality models. Convergence rates of operator splitting schemes are determined. Research indicates splitting techniques provides at most linear convergence. For fast-reacting species like N_2O_5, the convergence is not achieved when using splitting methods and time steps as small as 10 s. Symmetric and non-symmetric operator splitting does not provide significant difference in accuracy. Furthermore, operator splitting ordering with stiff operators computed last does not produce better results than with non-stiff operators computed last. The non-splitting method developed achieves convergence by reducing time steps, adapting time steps to insure convergence, and eliminating operator splitting.

Keywords: Operator splitting; Convergence; Operator ordering; Stiff integrator; Air quality models; Numerical techniques

1. Introduction

Operator splitting methods (Yanenko, 1971) are mathematical techniques used to solve partial differential equations. Splitting methods are commonly employed in three-dimensional air quality models to reduce the computational effort required to solve the governing equations. One of the first air quality models to use operator splitting is described by McRae et al. (1982). Operator splitting methods reduce chemically reactive three-dimensional transport equations into a series of one-dimensional transport equations and decouple chemical kinetics from transport dynamics. Some current models that split transport and chemistry are GATOR (Jacobson, 1997), Models3/CMAQ (Binkowshki and Shankar, 1995), and UAM-AERO (Lurmann et al., 1997).

Three-dimensional air quality models solve the atmospheric dispersion equations:

$$\frac{\partial c_i}{\partial t} + \nabla \cdot (\mathbf{u} c_i) = \nabla \cdot (\mathbf{K} \nabla c_i) + R_i(c_1, c_2, \dots c_n) \tag{1}$$

subjected to initial and boundary conditions

$$c_i(x_{\partial\Omega}, y_{\partial\Omega}, t) = c_{i\partial\Omega}, \tag{2}$$

$$\frac{\partial c_i(z=0,t)}{\partial z} = S_i^+ + v_g c_i, \tag{3}$$

$$\frac{\partial c_i(z=H,t)}{\partial z} = 0, \tag{4}$$

$$c_i(x,y,z,0) = c_{i0}, \tag{5}$$

where c_i, x, y, z, t, $\mathbf{u}$, $\mathbf{K}$, R_i, S_i^+, v_g, H, and $\partial\Omega$ are the concentration of species i, space variables, time variable, wind fields, diffusion fields, chemical kinetics, source, settling velocity, inversion height and domain

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boundary, respectively. McRae et al. (1982) used the methods of Strang (1968) to split equation (1) into the following symmetric operators: advective transport A, diffusion transport D, chemistry and vertical transport C, and source terms S.

$$\frac{\partial c_i}{\partial t} = A_{x,y} + D_{x,y} + C_z + S. \tag{6}$$

The numerical solution is constructed to preserve second-order accuracy by symmetric decomposition (Strang, 1968).

$$\begin{aligned} c_i(t + \Delta t) = {} & L_A(\Delta t/2)L_D(\Delta t/2)L_{C-Dz}(\Delta t) \\ & \times L_S(\Delta t)L_D(\Delta t/2)L_A(\Delta t/2)c_i(t), \end{aligned} \tag{7}$$

where L_x represents the integration of the operator x. Solving Eq. (1) as a sequence of sub-steps provides clear advantages: decoupling, higher degree of software modularity, and performance. For example, splitting a three-dimensional transport equation in space reduces the problem into a set of three one-dimensional transport equations. The total cost of solving the new system is approximately $3N$. On the other hand, the total cost of solving the non-split three-dimensional transport is approximately N^3. Furthermore, splitting allows the use of algorithms tailored to each operator. For example, a stiff integrator can be used to solve the chemistry operator and a flux scheme can be employed for the transport operator.

Operator splitting has a subtle disadvantage: one must use small time steps to capture coupling dynamics. Failure to comply with the time-step restrictions might result in substantial errors.

The errors produced by operator splitting motivate the two objectives of this paper:

- To develop a non-splitting approach for the solution of a three-dimensional airshed model, the CIT Airshed Model (Harley et al., 1993). Yanenko (1971) constructed theoretical frameworks for simpler linear transient systems. However, the rigorous theory for nonlinear systems is only developed for simplified cases. Lanser and Verwer (1998) showed that splitting errors arise for non-commutative operators in air quality modeling. Thus, a non-splitting solution is necessary to benchmark splitting techniques.
- To compare and analyze the discrepancies between splitting and non-splitting approaches. Numerical experiments are performed to quantify the errors introduced by operator splitting. These experiments with operator splitting show that convergence of some aerosol precursors and particulate matter concentrations is difficult to achieve as the time steps decrease. Furthermore, for those species that converge, the optimal time steps vary among species.

2. Numerical operator splitting analysis

2.1. Strang splitting

Lanser and Verwer (1998) proved that when operators commute, Strang (symmetric) splitting techniques do not produce splitting errors. Advection and diffusion operators commute if the velocity and diffusion field are independent of space. Operators in air quality models do not commute. Sportisse (1998) showed that the order of operations in splitting schemes is important to minimize errors from non-commutative operators. In particular, Sportisse recommended the integration of stiff operators at the end of the splitting step. Chemical transformations represent the stiff operator and the horizontal transport represents the non-stiff operator. This section analyzes numerical splitting techniques to quantify differences in the order of operations.

Most airshed models use symmetric splitting methods. The operation order of the CIT Airshed model is computed by TCT which denotes horizontal transport, chemistry-vertical transport and horizontal transport, respectively. Namely, $T = L_A L_D$ and $C = L_{C-Dz} L_S$. The order of operations studied in this research are: TCT, CTC, TC, and CT to determine accuracy of different ordering schemes. The time steps between operators vary from 5 to 30 min. Fig. 1 shows the time steps as a function of time for a typical day. Small time steps are taken when wind speeds and photolysis rates are large. Convergence rate of different order of operations are computed using time steps of 5 and 25 times smaller than the time step used in the base case.

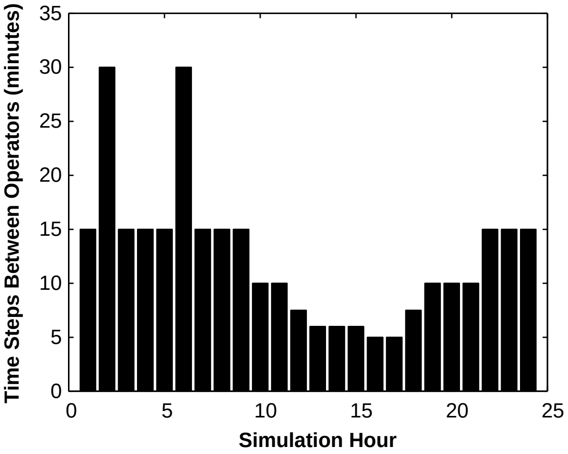

Fig. 1. Time steps between operators in the California Institute of Technology (CIT) Airshed Model. During high wind and chemically intensive hours of late afternoon, the CIT Airshed Model computes with smaller time steps. Convergence of operator splitting methods are performed by reducing these time steps by factors of 5, 25, 125, 625 and 1250.

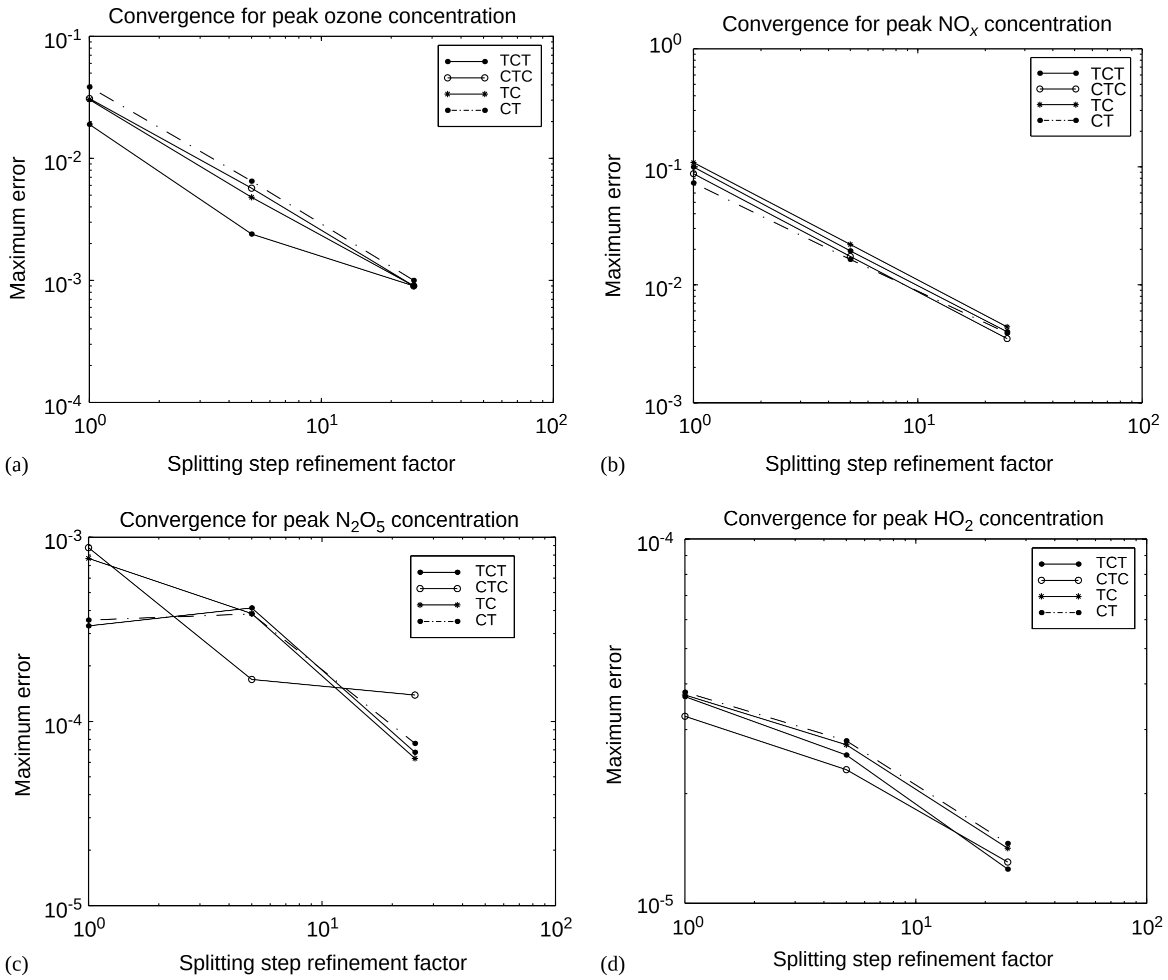

Plate 1. (a) Maximum error in the peak ozone concentration as the time steps between operator splitting decreases. *T* and *C* represent horizontal transport and chemistry-vertical diffusion, respectively. Strang splitting, *CTC* does not perform better than a non-symmetric split, *TC*. Convergence for all cases is at most linear. (b) Maximum error in the peak NO$_x$ concentration as the time steps between operator splitting decreases. *T* and *C* represent horizontal transport and chemistry-vertical diffusion, respectively. Placing stiff operators at the end of a time split, *TC*, does not produce more accurate results than solving a non-stiff operator at the end of a time split, *CT*. Convergence for all cases is only linear. (c) Maximum error in the peak N$_2$O$_5$ concentration as the time steps between operator splitting decreases. *T* and *C* represent horizontal transport and chemistry-vertical diffusion, respectively. Convergence of fast reacting species like N$_2$O$_5$ is not achieved even after reducing the splitting time steps by a factor 625. This species is important to aerosol formation in the South Coast Air Basin of California. (d) Maximum error in the peak HO$_2$ concentration as the time steps between operator splitting decreases. *T* and *C* represent horizontal transport and chemistry-vertical diffusion, respectively. Convergence of radicals like HO$_2$ is slow requiring excess of 25 times reduction in operator splitting time steps for satisfactory convergence.

These operators are denoted *TCT*5 and *TCT*25, respectively. The convergent concentrations are computed using a sequence of smaller time steps computed by *TCT*125, *TCT*625 and *TCT*1250.

The gaseous components examined are O$_3$, N$_2$O$_5$, NO$_x$ and HO$_2$. These species are not computed using pseudo-steady-state approximations nor are they part of a lumped assumption. Ozone is important to overall air quality. N$_2$O$_5$ and HO$_2$ represent challenging small time-scale species. NO$_x$ represents emitted sources important to the formation of ozone and aerosols.

Plate 1 shows the convergence rate of various operator orderings, *TCT*, *CTC*, *CT*, and *TC*. The convergence rate is computed by the maximum error norm of scheme *X* versus scheme *TCT*1250 given by

$$\text{error} = \text{Max}_t\left\{\left|\text{Max}_{x,y,z}\, X - \text{Max}_{x,y,z}\, TCT1250\right|\right\}. \quad (8)$$

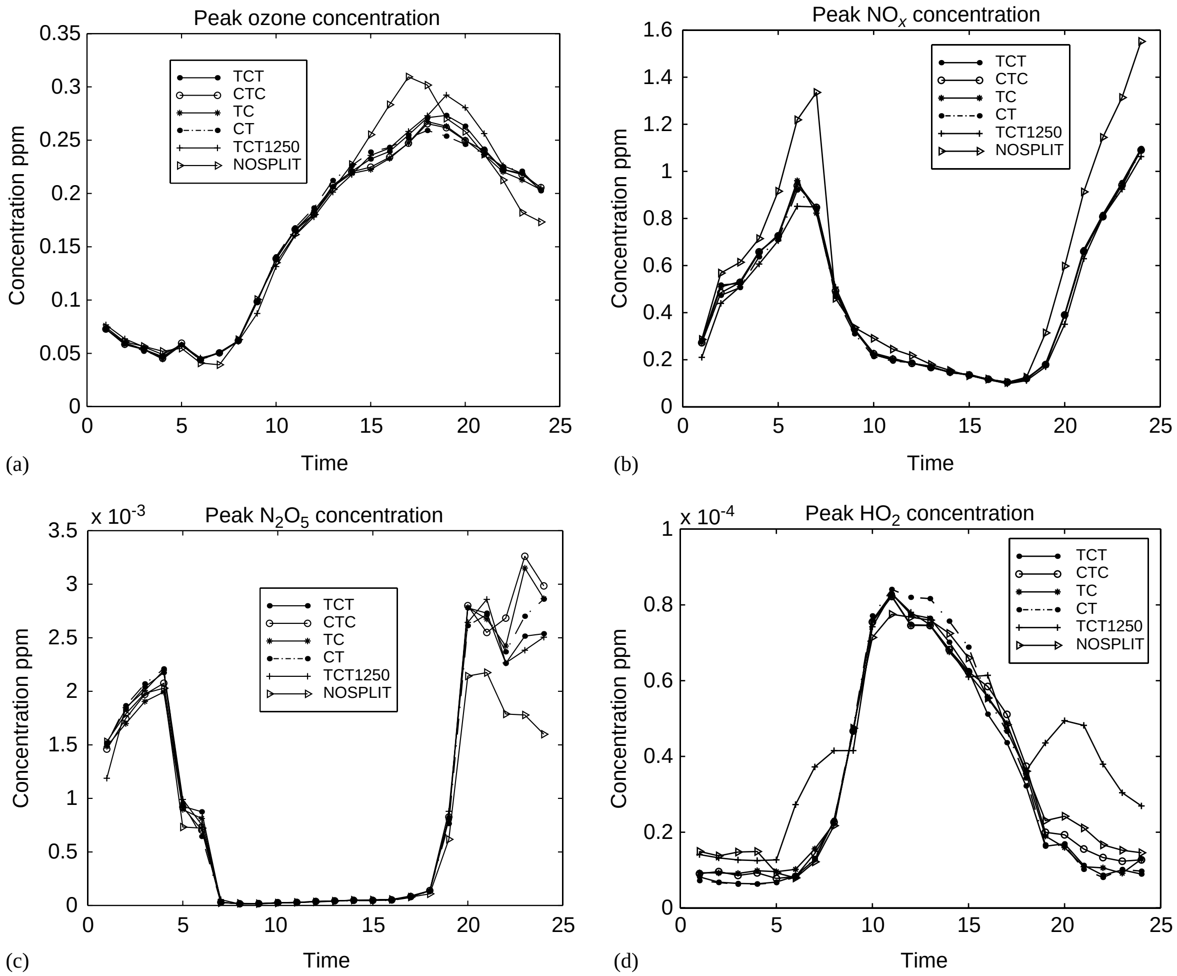

Plate 2. (a) Peak ozone concentrations for the simulation hours indicate differences between operator order, splitting techniques, and non-splitting techniques. *TCT*1250 represents the splitting techniques when reducing the base case time step by a factor of 1250. Maximum ozone concentrations occur at different time and obtain different magnitude between splitting and non-splitting approaches. Peak night time ozone decreases substantially faster in the non-splitting approach. (b) Peak NO$_x$ concentrations for simulation hours indicate differences between splitting techniques and non-splitting techniques. Due to high concentrations from emissions during morning hour traffic, splitting techniques are not able to capture rapidly changing dynamics. (c) Peak N_2O_5 concentrations for the simulation hours indicate differences between splitting techniques and non-splitting techniques. Splitting techniques do not converge and report higher N_2O_5 concentrations than the non-splitting approach for nighttime hours. The reduction in N_2O_5 is attributed to the rapid decline of the maximum ozone concentrations at night in (a). (d) Peak HO_2 concentrations for the simulation hours indicate significant differences between highly resolved splitting technique *TCT*1250 and other splitting techniques. This is expected due to the fast-reacting mechanisms dictating the behavior of HO_2.

All cases exhibit a lower maximum ozone concentrations than that computed by $TCT1250_{\text{ozone}}$ as shown in Plate 1. The worst results are those computed by *CT* with 13% error. The best results are those computed by *TCT* with 6.5% error. Plate 2 shows that NO$_x$ errors produced by the various operator ordering are similar to each other. The largest variations are seen for stiff components, N_2O_5 and HO_2. Eigenvalues of stiff components require smaller time scales to resolve. The time scales that are represented in Fig. 1 cannot capture the transient responses of these fast-reactive species. Maximum errors occur during nighttime when N_2O_5 dominates the formation of aerosol for the South Coast Air Basin of California.

Sportisse et al. (2000) and Blom and Verwer (1999) recommended that stiff operators be computed last. Data presented in Plate 1 show that the best ordering is not evident. While *CTC* ordering is closer to the converged concentrations than *TCT* for NO$_x$, it is farther away for ozone. Furthermore, symmetric Strang splitting might not produce better results than non-symmetric splitting. The performance of *TC* surpasses that of *CTC* by up to 16% for ozone. This behavior is also observed by Sportisse et al. (2000) using a box model. Results indicate convergence of N_2O_5 is not achieved. The time scale of N_2O_5 requires further reduction in time steps beyond the factor 625. The large reduction in time step required is due to fast reactive radicals like NO_3. The convergence of N_2O_5 plays a pivotal role in aerosol dynamics. Namely, N_2O_5 is responsible for the formation of HNO_3 at night

$$N_2O_5 + H_2O \rightarrow 2HNO_3. \tag{9}$$

The resulting nitric acid combines with ammonia to produce ammonium nitrate aerosols. Numerical runs with aerosol operators (not shown) indicate that reducing splitting time steps by a factor of 5 and 10 do not produce convergent results for nitrate and ammonium particulates. For those species that do converge, the convergence rate is at most linear and not quadratic. The slope of the logarithm plots indicates at most unity slope for all ordering schemes. HO_2 shows less than linear convergence. Although symmetric Strang split should produce a quadratic convergence in time, the inclusion of stiff chemistry reduces the order by one. This reduction effect for linear systems is also shown by Sportisse (1998).

2.2. Non-splitting approach

Operator splitting performs well for most gaseous species. However, due to the non-converging fast-reacting species, a non-split approach is implemented. Any non-split approach is computationally intensive due to the coupling of hundred of thousands to millions of ordinary differential equations. Implicit methods are not reasonable in such large cases. The non-split approach is thus extended from an explicit chemical integrator used in the CIT Airshed Model (Harley et al., 1993). The integrator is a second-order iterative, asymptotic integrator (Seinfeld and Pandis, 1998). The asymptotic integrator solves the following dynamical system,

$$\frac{dc_i}{dt} = P_i - L_i c_i \equiv P_i - \frac{c_i}{\tau_i}, \tag{10}$$

$$\frac{dc_i(z=0,t)}{dz} = S_i^+ + v_g c_i, \tag{11}$$

$$\frac{dc_i(z=H,t)}{dz} = 0, \tag{12}$$

where P_i and L_i are the production and loss terms. The asymptotic integrator first computes the predictor by assuming the loss and production terms are slow changing over a period of Δt. $P(t+\Delta t) \approx P(t)$ and $\tau(t+\Delta t) \approx \tau(t)$ which results in the prediction

$$c^*(t+\Delta t) = \frac{c(t)(2\tau(t)-\Delta t) + 2\Delta t\tau(t)P(t)}{2\tau(t)+\Delta t}. \tag{13}$$

With the prediction, $P^*(t+\Delta t) = P(c^*)$ and $\tau^*(t+\Delta t) = \tau(c^*)$ are computed and used for the corrector

$$c(t+\Delta t) = \frac{c(t)(\tau(t+\Delta t)+\tau(t)-\Delta t) + 0.5\Delta t(P(t+\Delta t)+P(t))(\tau(t+\Delta t)+\tau(t))}{\tau(t+\Delta t)+\tau(t)+\Delta t}. \tag{14}$$

An iterative method insures that chemical species do not change drastically under operations represented in the integration. If the predictor and corrector are not sufficiently close, the time step is reduced. The fast reacting chemical kinetics dictate the time steps. This is unlike operator splitting where splitting steps are determined by advection (McRae et al., 1982; Blom and Verwer, 1999). Including all processes and species into an integrator permits accurate and efficient determination of optimal time steps. Again, this is unlike operator splitting, where a uniform reduction is applied.

Most urban airshed models couple chemistry and vertical diffusion to reduce splitting errors (Lanser and Verwer, 1998). In asymptotic chemical integrators, operators are decomposed into production and loss terms. Emissions are incorporated in the production term and integrated in time. Emission rates are assumed constant for each time step. Transport operators can also be decomposed as such. For example, the contribution of diffusion using a second-order finite difference approximation is distributed to the production and loss terms

$$P_i = k_{zz}c_i(z+dz) + k_{zz}c_i(z-dz), \tag{15}$$

$$L_i = 2.0k_{zz}, \tag{16}$$

where k_{zz} and dz are the vertical diffusivity and cell height, respectively. By noting the sign of the coefficients in a discretization scheme, multidimensional transport is implemented into the asymptotic integrator. However, the allocation of the transport terms into production and loss components may not produce mass conservative properties. An alternative is to assign the transport contribution to the production term. Flux methods like that of Bott (1989) and Nguyen and Dabdub (2001) can then be used to compute $dc_i/dt = (F^{k+1} - F^k)/dx$, where F_i^k denotes the flux at boundary k of species i. Further simplification in computation is made by a local support interpolating a uniform two-dimensional flux. For example, consider interpolating a flux, $f = uc, vc$, to second-order accuracy at a point with its immediate neighbors, $\{f(x,y), f(x+\mathrm{d}x,y), f(x-\mathrm{d}x,y), f(x,y+\mathrm{d}y), f(x,y-\mathrm{d}y)\}$

$$f(x,y) = \sum_{0 \geqslant i+j \geqslant 2} a_{ijk} x^i y^j. \tag{17}$$

In fact the derivative of f in each direction is the central difference,

$$\frac{\partial f(x,y)}{\partial x}(x,y) \approx \frac{f(x+\mathrm{d}x,y) - f(x-\mathrm{d}x,y)}{2\mathrm{d}x}, \tag{18}$$

$$\frac{\partial f(x,y)}{\partial y}(x,y) \approx \frac{f(x,y+\mathrm{d}y) - f(x,y-\mathrm{d}y)}{2\mathrm{d}y}. \tag{19}$$

An approximation for the advective contribution can be computed by

$$\frac{\mathrm{d}c}{\mathrm{d}t} \approx \frac{uc(x+\mathrm{d}x,y) - uc(x-\mathrm{d}x,y)}{2\mathrm{d}x} + \frac{vc(x,y+\mathrm{d}y) - vc(x,y-\mathrm{d}y)}{2\mathrm{d}y}. \tag{20}$$

Note that in operator splitting methods, each direction is solved separately,

$$\frac{\mathrm{d}c}{\mathrm{d}t} \approx \frac{uc(x+\mathrm{d}x,y) - uc(x-\mathrm{d}x,y)}{2\mathrm{d}x}, \tag{21}$$

$$\frac{\mathrm{d}c}{\mathrm{d}t} \approx \frac{vc(x,y+\mathrm{d}y) - vc(x,y-\mathrm{d}y)}{2\mathrm{d}y}. \tag{22}$$

Thus, the coupled equation (20) is the sum of each uncoupled equation. This research uses a fourth-order Bott interpolation in x and y and first-order forward difference in z. First-order interpolation in z is permissible since vertical advection is not dominant. Flux limitations are imposed for advection contribution but not for chemical dynamics. The asymptotic solver and higher-order chemical integrator do not guarantee positive definiteness. Negative mass arising from the asymptotic integration are made zero. This procedure is the same in the non-splitting case. In chemical systems that do not include stiff components, the semi-implicit asymptotic solver is purely explicit, thus preserving the original behavior of the explicit advection solver. For cases that include stiff components, the asymptotic integrator is semi-implicit. However, this semi-implicit behavior does not adversely affect the advection solver because the time steps used are dictated by the chemistry not advection. That is, the advection solver converges easily compared to the chemistry dynamics.

Dirichlet boundary conditions are applied for the horizontal boundaries and Neumann boundary conditions are applied for the vertical boundaries. The initial conditions are interpolated from observed data for some species and assumed zero for other species. Specific values of the boundary conditions, initial conditions, and chemical mechanisms are discussed Harley et al. (1993).

3. Results

The results presented in this section is for the modeling episode of 27 August 1987 for the South Coast Air Basin of California. The episode is a part the campaigned by the Southern California Air Quality Study (SCAQS). The episode has reported high smog concentrations and high temperatures which facilitate the formation of many pollutants (Jacobson, 1993; Meng et al., 1998).

Plate 2 represents the peak concentrations for each hour. *NOSPLIT* is the non-splitting concentrations. The iterative convergence factor for the coupled (*NOSPLIT*) dynamics-chemical asymptotic integrator is the same as the uncoupled (*TCT*) chemical asymptotic integrator. The difference is that *NOSPLIT* includes all processes: advection, diffusion and chemistry. Peak ozone concentrations in Plate 2 indicate agreement within 5% for all schemes during the early hours. However, maximum ozone concentration occur at different times between the split and non-split approaches. Peak ozone concentrations decreases faster at nighttime hours for the non-split case. This discrepancies occur because of the non-convergence of fast reacting species like $\mathrm{OH}, \mathrm{HO_2}$ and $\mathrm{N_2O_5}$ in splitting schemes. These species directly impact ozone formation. The concentration profiles for $\mathrm{N_2O_5}$ clearly show that all splitting schemes do not converge at nighttime. Difference of up 44% are reported among splitting schemes. Non-splitting approaches produce lower concentrations of $\mathrm{N_2O_5}$ at night due to lower nighttime ozone concentrations. Ozone is needed to oxidize $\mathrm{NO_2}$ to $\mathrm{NO_3}$ for the formation of $\mathrm{N_2O_5}$ for nighttime chemistry.

Discrepancy between splitting and non-splitting approaches for maximum NO_x concentrations is up to 32%. Differences occur during peak traffic hours in the morning when NO_x is emitted. It is believed that for large emissions, splitting techniques can produce errors since they do not capture coupling dynamics. Numerical experiments involving only advective processes and one-dimensional winds (horizontal), show that both splitting and non-splitting approaches produce almost identical results. However, three-dimensional winds are used, discrepancies occur in areas of large spikes or emissions. This is the case for NO_x dynamics.

Integration of *NOSPLIT* uses variable time steps. These time steps are determined internally by a convergence factors, ε. The time steps are reduced iteratively until the convergence criterion is met at the final time, t_f,

$$\left|\frac{c(t_\mathrm{f}, \Delta t_{n+1})}{c(t_\mathrm{f}, \Delta t_n)}\right| - 1 < \varepsilon. \tag{23}$$

The results for *NOSPLIT* in this research uses $\varepsilon = 0.001$. When ε is reduced by a factor of 5 from the base case, differences in the integrations are negligible.

All species, including N_2O_5 and HO_2, from the reduced ε simulation are indistinguishable from *NOSPLIT* in Plate 2. Computing order of convergence of *NOSPLIT* is complex since time steps vary. However, when ε is reduced by a factor of 5, the number of iterations in the coupled integration increased by 1.8. In fact, when ε is reduced by a factor of 25, the number of iterations is increased by approximately factor of 4. This might indicate second-order convergence in time, unlike any of the splitting schemes, even the symmetric Strang split. The computational time of the non-split approach is faster than *TCT*1250. Given that the base case requires 1 time unit to complete with the *TCT*, the non-split approach requires 38 time units to complete. Reducing the time step of the splitting approach by 1250 requires 372 time units and still does not converge. Unfortunately, non-splitting approaches do not facilitate the use of parallel computers since domain and operator decompositions are not possible.

4. Conclusions

Operator splitting in three-dimensional air quality models provides efficient and accurate solutions for most gaseous species. However, for some fast-reacting species, operator splitting methods require very small time steps for convergent solutions. This fact prompted two goals accomplished by this research. The first goal is to analyze the convergence rate of different operator splitting ordering schemes. The second goal is to develop a solution to the governing equations in air quality models without operator splitting.

Result presented here show that reducing time steps by a factor of 625 from the base case time steps does not produce convergence for N_2O_5 and HO_2. Results indicate errors of species like N_2O_5 using operator splitting can be as much as 44%. N_2O_5 behavior indicates a significant reduction in accuracy of aerosol dynamics is accrued with operator splitting. These fast-reacting species play a major role in formation of both ozone and particulate matter in the South Coast Air Basin of California. This research confirms that Strang splitting does not provide second-order convergence in air quality models. Order of convergence is at most one for splitting schemes studied here. Furthermore, the ordering of operator splitting schemes does not provide significant increase in accuracy. Research indicates that accuracy is better attained by reducing time steps between operators, adapting time steps for convergence, and eliminating operator splitting.

This research developed a convergent solution to the modeling equations by employing a reduced, adaptive time steps with a non-split formulation. Non-splitting approaches couple physical and chemical dynamics. Non-splitting approaches use time steps that are determined by error (convergence) bounds inside the integrator which consider advection, emissions, deposition, and chemical processes. This permits the use of optimal time steps during integration. Time steps in splitting approaches, however, are determined by advective scales and assume chemical scales can be resolved accordingly. Although the use of non-splitting techniques in fully developed models is computationally expensive, the solution obtained from non-splitting techniques provides a benchmark to gauge the performance of splitting techniques.

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