



Mass conservative, positive definite integrator for atmospheric chemical dynamics

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ABSTRACT

Air quality models compute the transformation of species in the atmosphere undergoing chemical and physical changes. The numerical algorithms used to predict these transformations should obey mass conservation and positive definiteness properties. Among all physical phenomena, the chemical kinetics solver provides the greatest challenge to attain these two properties. In general, most chemical kinetics solvers are mass conservative but not positive definite. In this article, a new numerical algorithm for the computation of chemical kinetics is presented. The integrator is called Split Single Reaction Integrator (SSRI). It is both mass conservative and positive definite. It solves each chemical reaction exactly and uses operator splitting techniques (symmetric split) to combine them into the entire system.

The method can be used within a host integrator to fix the negative concentrations while preserving the mass, or it can be used as a standalone integrator that guarantees positive definiteness and mass conservation. Numerical results show that the new integrator, used as a standalone integrator, is second order accurate and stable under large fixed time steps when other conventional integrators are unstable.

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1. Introduction

The use of operator splitting in the atmospheric advection–diffusion equation separates the solution of pollution dynamics into four key operators: the advection–diffusion operator, the chemical dynamics, the emissions and deposition, and the aerosol transport and formation (Carmichael et al., 1986; Lansner and Verwer, 1999).

It is important that numerical solutions to all processes describing pollutant formation should be mass conservative and positive definite. The advection–diffusion operator (Nguyen and Dabdub, 2001; Bott, 1989), the emissions, and the aerosols operators (Nguyen and Dabdub, 2002; Landry et al., in press) are solved with algorithms that ensure higher-order, accurate, positive definite and mass conservative properties through flux formulations. The exception is the operator related to the numerical solution to stiff chemical dynamics, for which these properties are more difficult to obtain, as illustrated in Sandu et al. (1997a,b). Most traditional solutions to chemical dynamics are mass conservative, as in Verwer et al. (1998), but the addition of the strict positive

definiteness property is difficult to achieve. There has been some research presented in the literature; for example, Sandu (2001) developed a method using projection to convert negative concentrations into non-negative concentrations via an optimization routine under a mass conservative constraint.

This paper presents a novel alternative algorithm to attain mass conservative and positive definite integration of chemical dynamics. The algorithm developed here solves exactly each chemical reaction, and uses an operator splitting approach (Yanenko, 1971) to combine them in order to solve the entire system of reactions. We prove that the exact solution to each chemical reaction is positive definite and mass conservative, and consequently the solution to the entire system also shares those properties. This new method is called *Split Single Reaction Integrator* (SSRI). It can be used independently of other integrators or in conjunction with an host integrator, in which SSRI is invoked only when the host integrator provides negative concentrations. SSRI is second order accurate (Strang, 1968)—which is the limit of the order of accuracy of all air quality models that employ the operator splitting theory. Numerical results show that the method is stable for integrations with large fixed time steps.

The structure of this article is as follows. Section 2 develops the conservation statements. In Section 3, SSRI is presented and the properties of mass conservation and positiveness are detailed. In

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Section 4, test cases and numerical results are presented to show the efficiency and stability of SSRI.

2. Chemical dynamics and conservation statements

In order to gauge the mass conservation capabilities of a chemical kinetics solver, a set of conservation measures must be established. Establishing such conservation relations defines the quantities to conserve and provides useful constraints for the solvers.

To illustrate the framework of conservation relations, consider a simple set of atmospheric reactions as in Seinfeld and Pandis (1998):



Let the time-dependent concentration vector be $c(t) = (\text{NO}, \text{NO}_2, \text{O}, \text{O}_3, \text{O}_2)^T$ and the vector of reaction rates be $R(t) = (k_1 \text{NO}_2, k_2 \text{OO}_2, k_3 \text{O}_3 \text{NO})^T$, where t denotes the time. The system of ordinary differential equations governing the evolution of the chemical species in this set of reactions can be represented as

$$\frac{dc(t)}{dt} = \omega R(t), \quad \text{where} \quad \omega := \begin{pmatrix} 1 & 0 & -1 \\ -1 & 0 & 1 \\ 1 & -1 & 0 \\ 0 & 1 & -1 \\ 0 & -1 & 1 \end{pmatrix}. \quad (4)$$

Here ω is the stoichiometric matrix of the system of chemical reactions (1)–(3). In order to determine all appropriate conservation equations, all linear combinations of species that are conserved have to be found. For these species, the time derivative $dc(t)/dt$ vanishes. Thus we consider a matrix M that contains all linear combinations of derivatives that are zero.

$$M \frac{dc(t)}{dt} = 0, \quad (5)$$

and substituting (4) into (5) yields

$$M \frac{dc(t)}{dt} = M \omega R(t) = 0. \quad (6)$$

Since the relation (6) is true for all times, it follows that

$$M \omega = 0 \quad \text{and} \quad \omega^T M^T = 0.$$

Namely, M^T belongs to the null-space of ω^T , denoted by $\mathcal{N}(\omega^T)$, and therefore

$$M \in \mathcal{N}(\omega^T)^T.$$

The matrix M^T is a matrix containing a basis of the null-space $\mathcal{N}(\omega^T)$. In particular, the matrix M for the sample reactions (1)–(3) is given by:

$$M = \begin{pmatrix} 1 & 0 & -1 & 0 & 1 \\ -1 & 0 & 1 & 1 & 0 \\ 1 & 1 & 0 & 0 & 0 \end{pmatrix},$$

which implies that the mass conservation relations for this simple system (1)–(3) are

$$\text{NO} - \text{O} + \text{O}_2 = \text{NO}^0 - \text{O}^0 + \text{O}_2^0$$

$$\text{O} + \text{O}_3 - \text{NO} = \text{O}^0 + \text{O}_3^0 - \text{NO}^0$$

$$\text{NO} + \text{NO}_2 = \text{NO}^0 + \text{NO}_2^0,$$

where the superscript 0 denotes the initial concentrations at time $t = 0$. The relations given by M are used to quantify the mass conservation properties of the SSRI integrator. Note that there are more conservation relationships than just the obvious conservation of nitrogen and oxygen species.

The use of conservation relations can reduce the number of differential equations, by replacing them by algebraic relations. In addition, the use of conservation relations can transform non-mass conservative integrators into conservative integrators.

3. Numerical algorithm

3.1. An operator splitting approach

Among the existing chemical integrators, there are algorithms that are positive definite but not mass conservative, e.g. CHEMEQ2 (Mott et al., 2000). On the other hand, there are also many algorithms that are mass conservative but not positive definite (Verwer et al., 1998). It seems that having both properties is elusive for higher-order integrators in the literature.

The problem of losing the positive definiteness and mass conservative characters come either from the numerical approximation of the ordinary differential equations or from the interpolation and smoothing of the solution. These two artifacts arise since nonlinear systems of coupled ordinary differential equations are complex and exact solutions are virtually impossible to derive.

The proposed method relies on explicit exact solutions of a decoupled set of ordinary differential equations. The coupled set of chemical reactions is decoupled into a *sequence of single reactions*. Each reaction is represented by a system of ordinary differential equations that is considered independently. Moreover, the exact solution to each single chemical reaction can be derived and allows to construct a solution that is mass conservative and positive definite. The solution to the entire mechanism is computed via an operator splitting scheme using the exact solution of each chemical reaction. Since the solution associated with each chemical reaction is mass conservative and positive definite, as we prove later, the solution to the entire system shares the same properties.

To illustrate the decomposition according to the chemical reactions, consider three arbitrary chemical reactions:



Strang (1968) operator splitting techniques can be used to split fast chemical reactions from slow chemical reactions.

Let us explain briefly how the splitting of reactions can be achieved. Assume that the reaction (9) is slower than the reaction (8), which is slower than the reaction (7). The chemical dynamics can be split as follows. Let Δt be a given time step. First, we solve the fast reaction (7) for a time step of $\Delta t/2$ and the solution is used as the initial condition for solving the other fast reaction (8) for a time step of $\Delta t/2$. Then, the slow reaction (9) is solved for a time step Δt . Once the slow reaction (8) is solved, the split is completed by solving the fast reactions in reverse order, i.e. successively (8), then (7), for a time step of $\Delta t/2$. The splitting of the reactions into a fast-slow-fast sketch is a symmetric split.

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