A nonnegativity preserved efficient algorithm for atmospheric chemical kinetic equations

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ABSTRACT

Air pollution models plays a critical role in atmospheric environment research. Chemical kinetic equations is an important component of air pollution models. The chemical equations is numerically sticky because of its stiffness, nonlinearity, coupling and nonnegativity of the exact solutions. Over the past decades, numerous papers about chemical equation solvers have been published. However, these solvers cannot preserve the nonnegativity of the exact solutions. Therefore, in the calculation, the negative numerical concentration values are usually set to zero artificially, which may cause simulation errors. To obtain real nonnegative numerical concentration values, very small step-size has to be adopted. Then enormous amount of CPU time is consumed to solve the chemical equations. In this paper, we revisit this topic and derive a new algorithm. Our algorithm Modified-Backward-Euler (MBE) Method can unconditionally preserve the nonnegativity of the exact solutions. MBE is a simple, robust and efficient solver. It is much faster and more precise than the traditional solvers such as LSODE and QSSA. The numerical results and parameter suggestions are shown at the end of the paper. MBE is based on the P-L structure of the chemical equations and a deeper view into the nature of Euler Methods. It cannot only be used to solve chemical equations, but can also be applied to conquer ordinary differential equations (ODEs) with similar P-L structure.

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1. Introduction

Air pollution models is critical in atmospheric environment research. Chemical kinetic equations is an important component of air pollution models. In air quality simulations, the concentrations of various pollutants are generally described by a set of Reaction-Diffusion-Advection partial differential equations (PDEs). By using operator splitting techniques, the effects of different processes such as chemical reactions, diffusion, advection are firstly calculated separately and then combined in some way [9]. Air quality simulations concern computational nodes in a three-dimensional (3D) space and for each node there is a set of chemical kinetic ODEs.

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The numerical difficulty of these chemical ODEs arises from the stiffness, nonlinearity, coupling and nonnegativity of the exact solutions. A balance between calculation speed and accuracy is the essential problem. Generally, Step-size restriction and a great number of spatial nodes lead to the fact that enormous amount of CPU time is consumed to solve the chemical ODEs. Therefore, efficient numerical methods for the chemical equations are significant.

Various numerical methods for these ODEs have been proposed over the past decades, including QSSA, MQSSA, HYBRID, MHYBRID, Gong & Cho, LSODE, SMVGEAR, etc [1,3,5,6,8–14,16]. These methods are generally classified into two groups: fast-slow species dependent algorithms (e.g., QSSA, MQSSA, HYBRID, MHYBRID, Gong & Cho) and species independent algorithms (e.g., LSODE, SMVGEAR). In the former case, the species are divided into different groups in each time step and different numerical schemes are applied to them separately. In the latter case, all the species share the same numerical scheme. Among the existing algorithms, two solvers are frequently used: QSSA and LSODE. QSSA is based on Forward Euler Method and numerical approximation, which is considered as a simple, fast but less accurate solver. Whereas LSODE is based on Gear Method, which is complicated, comparatively slow but more precise.

Although there are all sorts of solvers, these solvers cannot preserve the nonnegativity of the exact solutions. Therefore, in the calculation, the negative numerical concentration values are usually set to zero artificially, which may cause simulation errors. The cause is that zero and a very small positive value have entirely different mathematical properties: any value times zero is zero; a relatively large positive value times a small positive value may equal a value which cannot be neglected. To obtain real nonnegative numerical concentration values, very small step-size has to be adopted (e.g., with QSSA, the step-size should be smaller than 1 s; with LSODE, the step-size should be even smaller than what QSSA can accept). Then enormous amount of CPU time is consumed to solve the chemical equations.

A solver which can conquer stiffness and at the same time unconditionally preserve the nonnegativity is intensely desired. Our Modified-Backward-Euler (MBE) Method, which is simple, robust and efficient, can fully meet such requirement. MBE is much faster and more precise than the traditional solvers such as QSSA and LSODE.

The objective of this paper is to present a nonnegativity preserved efficient algorithm for the chemical ODEs and to put the algorithm into practice. We analyze the P-L structure of the chemical ODEs and the nature of Euler Methods. Based on the analysis, we present our MBE Method. In our simulations, carbon bond mechanism CBM-Z [15] is adopted, which is a set of chemical ODEs including 67 species. The numerical results and parameter suggestions are shown at the end of the paper.

2. Model analysis

We suppose that there are m species, then the chemical kinetics at each spatial node can be described by the following form

$$\frac{dC_i}{dt} = P_i(C_j, t) - L_i(C_j, t)C_i, \tag{1}$$

$$C_i(0) = C_i^0, \tag{2}$$

where $i, j = 1, 2, \ldots, m$; $P_i \geq 0, L_i \geq 0$; C_i represents the concentration of species i; P_i and L_iC_i are the chemical production and loss rates of the species, respectively; C_i^0 is the initial value.

Now we discuss the initial-value problem (3) and (4), in which the subscript i is dropped for convenience.

$$\frac{dC}{dt} = P - LC, \tag{3}$$

$$C(0) = C^0, \tag{4}$$

where $P \geq 0, L \geq 0$.

Firstly, we suppose that P and L in (3) are constants, then system (3) and (4) has the exact solution

$$C(t) = (C^0 - \frac{P}{L})e^{-Lt} + \frac{P}{L}, \qquad L > 0; \tag{5}$$

or

$$C(t) = C^0 + Pt, \qquad L = 0. \tag{6}$$

From (5) we have

$$\lim_{t \to +\infty} C(t) = \frac{P}{L} \tag{7}$$

and

$$C^0 > \frac{P}{L} \Rightarrow C(t) > \frac{P}{L}, t > 0, \tag{8}$$

$$C^0 < \frac{P}{L} \Rightarrow C(t) < \frac{P}{L}, t > 0, \tag{9}$$

$$C^0 = \frac{P}{L} \Rightarrow C(t) = \frac{P}{L}, t > 0. \tag{10}$$