BNL-79149-2007-JA

Numerical advection of correlated tracers: Preserving particle size/composition moment sequences during transport of aerosol mixtures

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Abstract. Nonlinear transport algorithms designed to reduce numerical diffusion fail to preserve correlations between moments, isotope abundances, etc. when these scalar densities are transported in models as separate tracers. In case of the particle size/composition coordinates of an aerosol, such loss can give rise to unphysical moment sets. New statistical approaches to aerosol dynamics, which involve tracking moments directly, offer highly efficient alternatives to sectional and modal methods for representing aerosols in climate models, but it is essential that moment set integrity be preserved throughout a simulation. In this paper we review the problem and weaknesses of previous attempts at solution, including vector transport – a scheme in which the moments, as internal aerosol coordinates, are transported together with a single lead tracer such as number or mass. A non-negative least squares (NNLS) solution that finally eliminates the problem without requiring modification of the transport algorithm itself is presented. Following each transport step, new moment sets are resolved into sums of previously validated sets with non-negative coefficients using NNLS. Transport errors are removed and the now guaranteed-to-be-valid moment sets are ready for passage to the aerosol dynamics module. In addition to moment set validation, the new scheme reduces numerical diffusion during transport and provides greater accuracy for the source apportionment of aerosol mixtures. The method is not limited to moment transport - similar improvements in accuracy are expected using NNLS in conjunction with modal and sectional methods.

1. Introduction

The most common approaches to atmospheric aerosol modeling use “modal” or “sectional” methods. Modal methods divide the aerosol into a small number of modes of prescribed shape having uniform composition within each mode. The method is efficient but generally not very accurate. Sectional methods divide the aerosol into a number of size classes. High accuracy requires good size resolution, and for this many class variables have to be carried in the model. Both approaches usually involve tracking of only number (N) and mass (M). For example, the modal method typically uses N, M, and a prescribed variance to parameterize a lognormal mode. In the sectional method, the particle population in each size class is characterized by its values of N and M (Fig. 1a).

The method of moments (MOM) offers a statistically based alternative that is more efficient than sectional methods and more accurate than modal methods. Efficiency derives from the fact that key moments of the particle population, which can include N, M, lower-order radial moments, and even the mixed moments of a multivariate representation (Fig. 1b), can be tracked directly without having to know the distribution itself. Aerosol physical and optical properties are obtained directly from the moments using quadrature (Q) methods. The resulting QMOM has found wide application in the engineering community where for many applications transport models can afford to operate using fine grids and linear differencing schemes. Nonlinear transport algorithms, designed originally to model shock fronts, have become the mainstay of atmospheric models. These reduce numerical diffusion over coarse grids, but can destroy consistency between moments and other correlated properties when these are individually transported like chemical tracers. Indeed, the problem has until now been the most serious impediment to widespread use of the QMOM in atmospheric models. Various ways around the moment transport problem have been suggested [1], but none is entirely satisfactory. In this paper we present a new approach – one that finally eliminates the consistency problem with the added bonus of providing a much more accurate scheme for source apportionment and transport of aerosol mixtures.

Transport nonlinearity is never fatal to an aerosol module requiring only transport of number and mass, although M/N ratios are subject to large errors when calculating means. With higher-order moment sequences the problem can (albeit infrequently) give rise to un-physical moment sets (e.g. the variance, equal to $\mu_2 - (\mu_1)^2$ for a normalized distribution, see Fig. 1 caption for definition of moments, will on occasion turn out negative)! A positive-definite and conservative transport scheme only guarantees that individual tracers remain positive and conserved – registry between the different moments of an aerosol, with their generally different spatial gradients, is not guaranteed when these are transported separately like chemical species.

(a)
$$\begin{pmatrix} N_1 \\ M_1 \\ N_2 \\ M_2 \\ N_3 \\ \vdots \\ N_l \\ M_l \end{pmatrix} \quad \begin{array}{l} \left.\begin{array}{l} \\ \\ \end{array}\right\} \\ \left.\begin{array}{l} \\ \\ \end{array}\right\} \\ \\ \\ \\ \left.\begin{array}{l} \\ \\ \end{array}\right\} \end{array}$$

modes or sections
N = number
M = mass

(b)
$$\begin{pmatrix} N_1 \\ \mu_1 \\ \mu_2 \\ \vdots \\ \mu_{i,j,\cdots} \\ N_2 \\ \mu_1 \\ \mu_2 \\ \vdots \\ \mu_{i,j,\cdots} \end{pmatrix} \begin{array}{l} \left.\begin{array}{l} \\ \\ \\ \\ \\ \end{array}\right\} \\ \left.\begin{array}{l} \\ \\ \\ \\ \\ \end{array}\right\} \end{array}$$

moments including for multiple populations (super-modes)

N = number

μ_k = k^{th} radial or mass moment

$\mu_{i,j}$ = mixed moments (multivariate distributions)

Figure 1. Vector arrays containing sequences of transported scalar densities that can be used to represent aerosols in climate models. Examples in (b) include $\mu_k = \int r^k f(r)dr$, which are the radial moments of size distribution $f(r)$ [2]. Multiply indexed quantities such as $\mu_{i,j} = \int\int m_1{}^i m_2{}^j f(m_1, m_2, \cdots, m_n) dm_1 \cdots dm_n$, where m_i is a species mass coordinate, describe higher-order statistical properties of a generally mixed, n-component aerosol [3]. Even within a moments framework it is generally useful to partition the aerosol into population classes, having separate moments for each class. A major goal of our research is to utilize advanced statistical methods to determine optimal aerosol partitioning and optimal moment sequences assuming that the next generation climate models will be limited to carrying only about 30-40 aerosol coordinates.

The leading solution to the problem, which we implemented in several chemical transport models (CTMs), uses "vector transport" (VT) [1,2]. Here a sequence of moments is normalized to a selected lead tracer, usually number or mass, and only that one tracer is transported. The remaining vector components, and in general there will be a different vector for each grid cell, are transported with the same mixing coefficients as the lead tracer, thereby preserving moment ratios within the sequence. Most VT schemes use number vector transport (NVT) or volume vector transport (VVT), depending on the lead moment [1]. The main advantage of the VT schemes is that they preserve valid moment sequences. The idea here is that any linear combination of valid moment sequences with positive coefficients is itself a valid moment sequence; a theorem, not difficult to prove, that is also behind the new approach. Also the number of transported quantities is reduced, but the accuracy is not as good as found when more than one moment is transported (e.g. as with Wright's NVVT scheme [1]). The main disadvantage, other than loss of accuracy, is that the VT schemes require some change to the transport algorithm to explicitly call out the cell-to-cell mixing coefficients at each time step (c.f. the coefficients c_j in Eq. 3 below). The aerosol module should be readily interchangeable with any advection scheme, and the modifications required by VT have proven tedious and time consuming. A least-squares solution to the problem is now suggested.

2. A least-squares method for preservation of moment sequence integrity during transport

2.1. Description of the method and its application to source apportionment of aerosol mixtures:
Least squares (LS) provides optimal resolution of a length *m* moment sequence, $\mathbf{b}^{\mathbf{I}}$, which includes any error from transport, into a valid basis thereby yielding a valid moment set. Consider, for example, three valid aerosol source vectors $\{\mathbf{v}_1, \mathbf{v}_2, \mathbf{v}_3\}$ labeled R, G, and B respectively. Form from these the columns of an $m \times 3$ matrix $\mathbf{A} = [\mathbf{v}_1, \mathbf{v}_2, \mathbf{v}_3]$. LS solves the following linear system:

$$\mathbf{Ac} = \mathbf{b}^{\mathbf{I}} + \mathbf{e}, \qquad (1)$$

obtaining the coefficient vector $\mathbf{c} = (c_1, c_2, c_3)^{\mathbf{T}}$ that minimizes the Euclidean norm of the error residual, $\mathbf{e}$. From this we obtain both a valid moment vector $\mathbf{b} \approx \mathbf{b}^{\mathbf{I}}$, closest in the least squares sense to $\mathbf{b}^{\mathbf{I}}$, and its source resolution:

$$\mathbf{b} = \mathbf{c}_1\mathbf{v}_1 + \mathbf{c}_2\mathbf{v}_2 + \mathbf{c}_3\mathbf{v}_3. \qquad (2)$$

Using non-negative least squares (NNLS) [4] insures non-negative coefficients $\mathbf{c}_i$. Validity of $\mathbf{b}$ follows immediately from the theorem stated in Sec. 1. Figure 2 demonstrates the method for source apportionment. Consider again the three aerosol types {R, G, and B}, each characterized by a column vector of 4 radial moments, $(\mu_0, \mu_1, \mu_2, \mu_3)^{\mathbf{T}}$. We use moments from the lognormal distributions tested previously in the VT schemes by Wright (Fig. 2 of [1]). Let the initial distribution contain pure R in grid cell 3, pure G in cell 4, and a uniform backgound of pure B elsewhere. The mixture is transported in Fig. 2 below either by NVT or as individual tracers resulting in vectors $\mathbf{b}^{\mathbf{I}}$ (one for each cell), which are then resolved into R, G, and B components after 70 time steps using NNLS. Figure 2a shows the NVT scheme to be quite diffusive even though the number distribution itself shows relatively little dispersion. This appears to be due to the fact that N_R, N_G, N_B, and each of the remaining moments, have spatial gradients that differ from those of the lead tracer, *N*. Figure 2b shows much improved results when all four moments are separately transported and thus contribute more or less equally to determining $\mathbf{b}^{\mathbf{I}}$.

2.2. Preserving moment sequences after each advection step:
In the previous example the basis vectors (and $\mathbf{A}$) were constant. Modifying the same procedure, Eqs. 1 and 2 are used to obtain valid updated moment sequences at each time step. Begin by assuming that a valid moment set, $\mathbf{v}_j(t)$, already exists in cell j at time t. The procedure generates a valid set at the

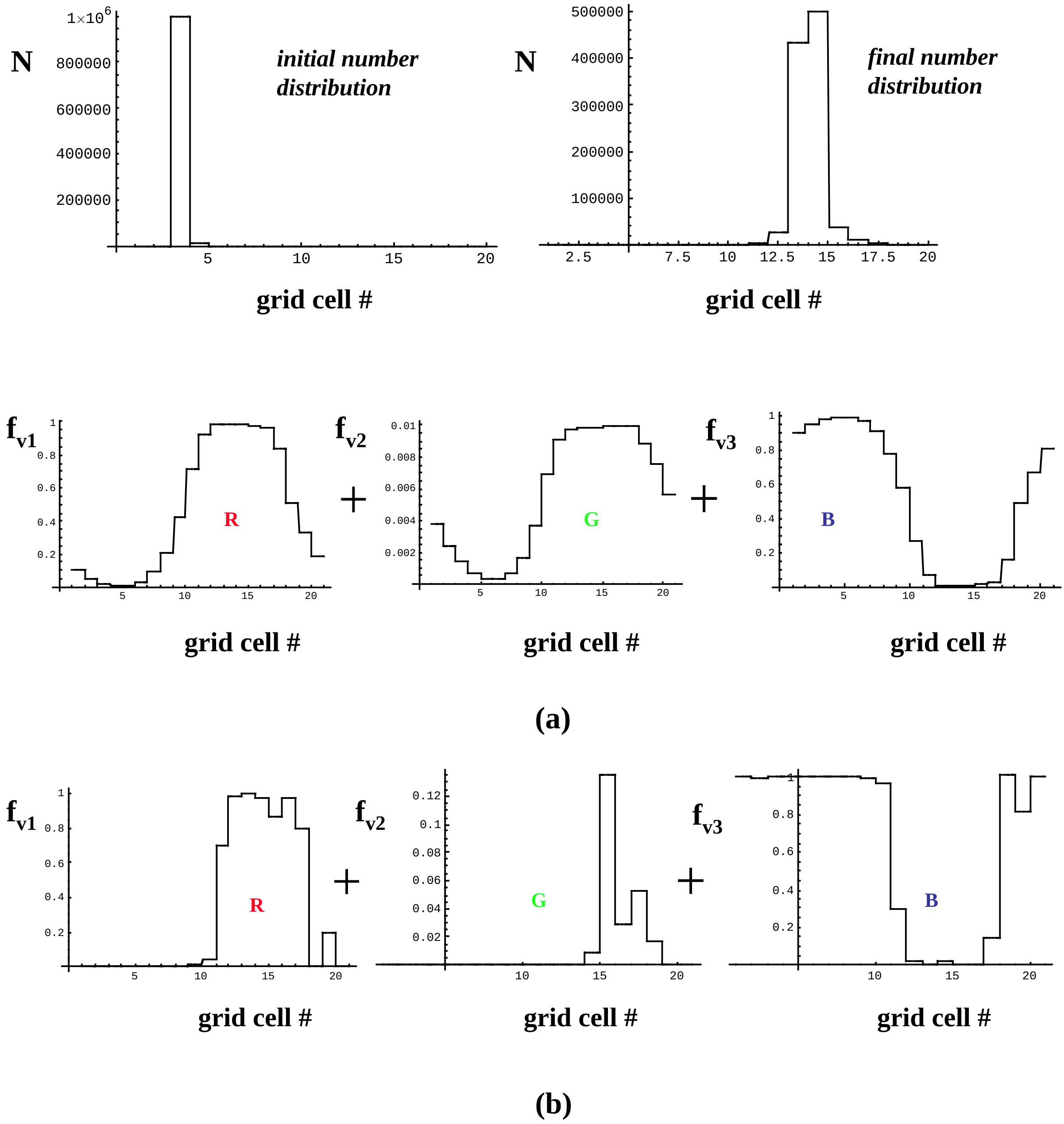

Figure 2. Decomposition of transported moments from an aerosol mixture into three component fractions here labeled R, G, and B. Top. Initial distribution (left) and final distribution of total particle number ($N \equiv \mu_0$) after 70 advection steps with a spatially uniform Courant number of 0.15 and a periodic boundary after 20 grid cells (right). Only cell-averaged concentrations are shown. See text for initial color distribution. The exact final distribution, not shown, is simply the initial distribution translated $10.5 = 70 \times 0.15$ units to the right. Transport was carried out using the quadratic upstream method with flux limitation [5]. (a) NNLS decomposition of final distribution after number vector transport (NVT) with moments 1-3 linked to particle number. (b) NNLS decomposition of the final distribution after separate advection of all 4 moments (Eqs. 1 and 2).

next time step $t+\Delta$, where Δ is the time increment. The method was tested using the quadratic upstream method with flux limitation [5]. Neighboring grid cells are illustrated schematically in Fig. 3. For this case $\mathbf{A} = [\mathbf{v}_{j-1}(t), \mathbf{v}_j(t)]$ consist of just 2 column vectors, each a valid moment set, and similarly for all grid cells $\{j\}$ at time t. First each moment is transported as an independent tracer and

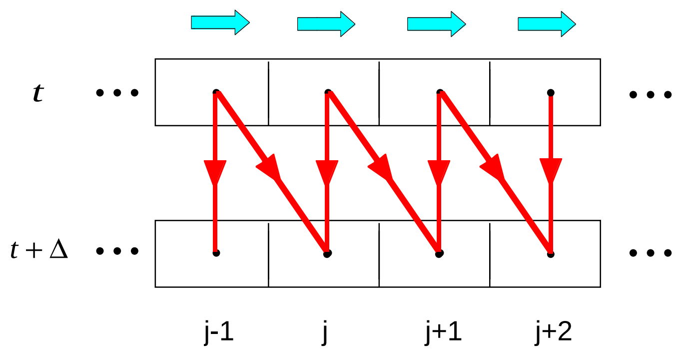

Figure 3. Removing advection errors using non-negative least squares (NNLS). Advection is in the direction of the green arrows. Begin by assuming that the moment sets at time *t* are valid. Cell *j* at time $t+\Delta$ then contains valid contributions from itself, i.e., material remaining from time *t*, a valid upwind contribution from cell *j*-1, but also any error, **e**, incurred during the transport step. In the NNLS correction process, **e** is minimized and removed. Equation 3 then insures valid updated moment sequences at the next time step, $t+\Delta$, for passage to the aerosol module.

results combined to form the uncorrected intermediates $\{\mathbf{v}^{\mathbf{I}}{}_j(t+\Delta)\}$ (vectors $\mathbf{b}^{\mathbf{I}}$ of Sec. 2.1). Finally, these are resolved as in Eq. 1 using NNLS and the (already minimized) error removed to obtain the final (now valid) result for cell *j*:

$$\mathbf{v}_j(t+\Delta) = c_{j-1}\mathbf{v}_{j-1}(t) + c_j\mathbf{v}_j(t) \qquad (3)$$

In the VT scheme the c-coefficients are culled from the transport model, but only for the lead tracer. In the present example all four tracers were advected, each having (because their spatial gradients are different) a different set of unknown coefficients. So which set should we use if we had them? NNLS provides the natural answer – none of them! Instead, NNLS yields a different set of coefficients, with each tracer having a “voice” in the result. The new scheme, which operates independently of the advection routine, insures that only valid moment sets are passed to the aerosol module. Efforts are currently underway to implement NNLS with the BNL aerosol module MATRIX – a highly flexible QMOM platform for use in both regional scale and global climate models.

Acknowledgements

The author thanks Dr. Dong Huang of BNL for pointing out the NNLS algorithm described in Ref. 4. This work was supported by the BNL/SUNY-SB SciDAC Science Application Partnership (SAP) “Statistical Approaches to Aerosol Dynamics for Climate Simulation” under Contract NO. DE-AC02-98CHI-886 with the U.S. Department of Energy.

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