Aerosol dynamics using the quadrature method of moments: Comparing several quadrature schemes with particle-resolved simulation

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Abstract. The method of moments (MOM) is a statistically based alternative to sectional and modal methods for aerosol simulation. The MOM is highly efficient as the aerosol distribution is represented by its lower-order moments and only these, not the full distribution itself, are tracked during simulation. Quadrature is introduced to close the moment equations under very general growth laws and to compute aerosol physical and optical properties directly from moments. In this paper the quadrature method of moments (QMOM) is used in a bivariate test tracking of aerosol mixing state. Two aerosol populations, one enriched in soot and the other in sulfate, are allowed to interact through coagulation to form a generally-mixed third particle population. Quadratures of varying complexity (including two candidate schemes for use in climate models) are described and compared with benchmark results obtained by using particle-resolved simulation. Low-order quadratures are found to be highly accurate, and Gauss and Gauss-Radau quadratures appear to give nested lower and upper bounds, respectively, to aerosol mixing rate. These results suggest that the QMOM makes it feasible to represent the generallymixed states of aerosols and track their evolution in climate models.

1. Introduction

Gaussian quadrature provides a systematic method for approximate evaluation of integrals of the form

$$I = \int_0^\infty \sigma(m) f(m) dm \approx \sum_{i=1}^{N} \sigma(m_i) w_i \,. \tag{1}$$

Here $\sigma(m)$ is a known kernel function, in this case of particle mass. The weight function $f(m)$ is the aerosol distribution function, and I is some integral property of the distribution. The approximate equality gives the quadrature approximation with abscissas and weights, m_i and w_i, respectively, determined to give correct values for selected moments of the distribution:

$$\mu_k = \int_0^\infty m^k f(m) dm . \tag{2}$$

A key feature of the quadrature method of moments (QMOM) is that one does not need to follow the distribution itself to make the approximation on the right-hand side of equation (1) – only its selected moments [1]. This makes for a highly efficient method. For example, given the 2N integral moments for k = 0 through 2N-1, the N quadrature points (N abscissas and N weights in correspondence with these moments) are determined. Efficient algorithms are available do the inversion, and these are well conditioned for small N (here we consider N values of 1, 2, and 3). Furthermore, the approximate equality of equation (1) becomes exact for polynomial kernels of degree less than or equal to 2N-1, as is most easily seen by substitution into equation (1) and expanding the integral in the moments defined by equation (2).

1.1 Aerosol mixing state

The remarkable efficiency of the QMOM makes this method ideal for use in atmospheric models. The QMOM has been used to represent aerosols in chemical transport models (CTMs) on the subhemispheric and regional scale, wherein several types of aerosols were tracked (e.g., sulfate, dust) using six moments/three quadrature points per type. Later the QMOM was applied to multicomponent, internally mixed populations, with remarkable levels of accuracy demonstrated in comparisons with high-resolution sectional models [2]. Even with chemical resolution capability, the QMOM was limited to internal mixtures and distribution functions defined along a single radius or mass coordinate. Capturing the general mixing states of an evolving multicomponent particle population requires, instead, multidimensional representation. It is here that statistical approaches, most prominently Monte Carlo-based particle resolved simulation and the QMOM, come into their own to offer the greatest advantage over sectional and modal methods. Monte Carlo is best for benchmark simulations, especially for complex multivariate processes in high dimension, whereas the QMOM provides economy of representation and great computational speed.

The QMOM has been applied to bivariate populations of combustion particles undergoing simultaneous coagulation and sintering. Two coordinates, surface area and volume, represent particles of mixed size and nonspherical shape [3]. Calculations were benchmarked bu using a CPU-intensive 2D model of $150 \times 150 = 22500$ sections and by Monte Carlo simulation [4]. The theory of QMOM extension to higher dimensions has been described and illustrated for the general mixing of multicomponent aerosols [5]. Model validation in higher dimension is extremely problematic and often limited to analytic test cases. Particle-resolved (PR) simulation, developed by two of us (Riemer and West), finally provides a platform for benchmarking the QMOM under realistic conditions where other approaches, for example, high-resolution sectional calculations, are impractical to carry out.

Particle-resolved simulation of three populations interacting through coagulation is described in the following section and used to benchmark QMOM accuracy for Gauss and Gauss-Radau quadratures of varying complexity (numbers of tracked moments). We find Gauss quadrature tends to overestimate (and Gauss-Radau quadrature underestimate) coagulation rate, nevertheless yielding nested bounds and very rapid convergence to the PR-simulation result.

2. Calculations

2.1 Interacting particle populations: Description of the model

Aerosol aging is an important atmospheric process that tends to make particles more hydrophilic and thus better able to serve as sites for cloud droplet condensation, thereby affecting cloud properties and resulting in shorter particle lifetimes. Coating of hydrophobic soot particles with sulfate is one such aging process. Here we consider an initial test condition consisting of two distinct particle populations: one enriched in soot (mass composition 90% soot and 10% sulfate) the other in sulfate (90% sulfate and 10% soot). These subsequently interact through coagulation to produce a third, generally mixed, population evolving ultimately to an internally mixed final state. The situation is depicted schematically in figure 1 (I and II are the initial sulfate and soot enriched populations, respectively, and III is the mixed population at some intermediate stage of development). The coagulation kernel is set proportional to the sum of the volumes of the coagulating particles – for simplicity we assume particle densities of unity and use mass and volume interchangeably. The sum kernel allows for a well-known analytic solution, but only for the total population – not for the individual subpopulations. The initial populations are taken to have identical size distributions, differing only in number and composition. The total initial population is set at 100,000 particles. As coagulation proceeds and particle number is reduced, the population is doubled several times through "cloning" to maintain good statistics throughout the PR-simulation. Figure 2 shows the evolution of particle number (corrected for cloning) and comparison with the QMOM result, which is exact for the total population. Agreement supports the excellent statistics achievable with the PR-simulation method.

2.2 Moments and quadratures
Because populations I and II are each of uniform composition, they can be treated by using the univariate moments and quadrature point assignments of equations (1) and (2). Population III, on the other hand, is generally mixed and is treated by tracking bivariate moments of the form

$$\mu_{pq} = \int_0^\infty \int_0^\infty m_1^p m_2^q f(m_1, m_2) dm_1 dm_2 . \tag{3}$$

Particle coordinates m_1 and m_2 are the masses of sulfate and soot, respectively. Quadrature points are easily assigned [5] to give correct values for the lower-order moments listed in figure 1. The blue and green points give two such assignments for population III, each capable of reproducing the particle number, distribution mean, and covariance matrix elements for that population. Figure 2 compares results using 1 point/mode quadrature (red points), tracking number and mass composition for each mode, and a more extensive calculation (blue quadrature points) that tracks 14 moments including the covariance matrix. 1-point quadrature tracking of number and mass, for each of a number of populations, is now in use in the GISS climate model [6] and plans are underway to incorporate the more extensive scheme, tracking four moments for each univariate mode and the covariance matrix for generally mixed populations (the 14 moment case of the test calculation).

Figure 3 shows the effect of using more moments and odd numbers of moments; previous applications of the QMOM all used Gauss quadrature and even numbers (per mode) of moments. Gordon [7] showed in the sixties how to obtain nested upper and lower bounds to the Laplace transform of a positive distribution function using only that functions moments. His approach is reformulated in [8] in the language of Gauss and Gauss-Radau (GR) quadratures. Briefly, the GR scheme places an abscissa at zero (the lower boundary of the distribution function domain) thereby using one less moment. The method is tested here for sequences of 1–6 univariate moments separating the total distribution now into two populations: population I and others (II and III), left panel; population II and others (I and III), right panel. Two separate calculations were performed. It is not clear that the true population decay can be written as the Laplace transforms of any fixed distribution function. Hence, the behavior illustrated in figure 3 may be fortuitous and needs to be explored for other kernels to establish its generality. Nevertheless the calculations clearly show rapid convergence of the quadrature approximation, equation (1), even with low numbers of moments.

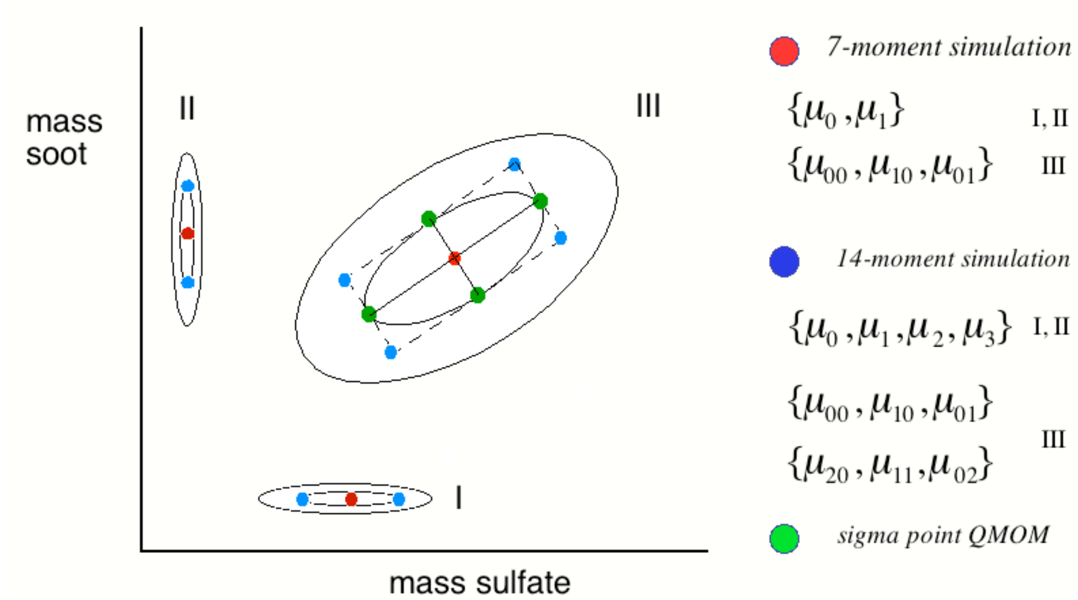

Figure 1. Schematic diagram showing the three particle populations (I, II, and III) and approximate quadrature point locations and population densities (1 and 2σ surfaces).

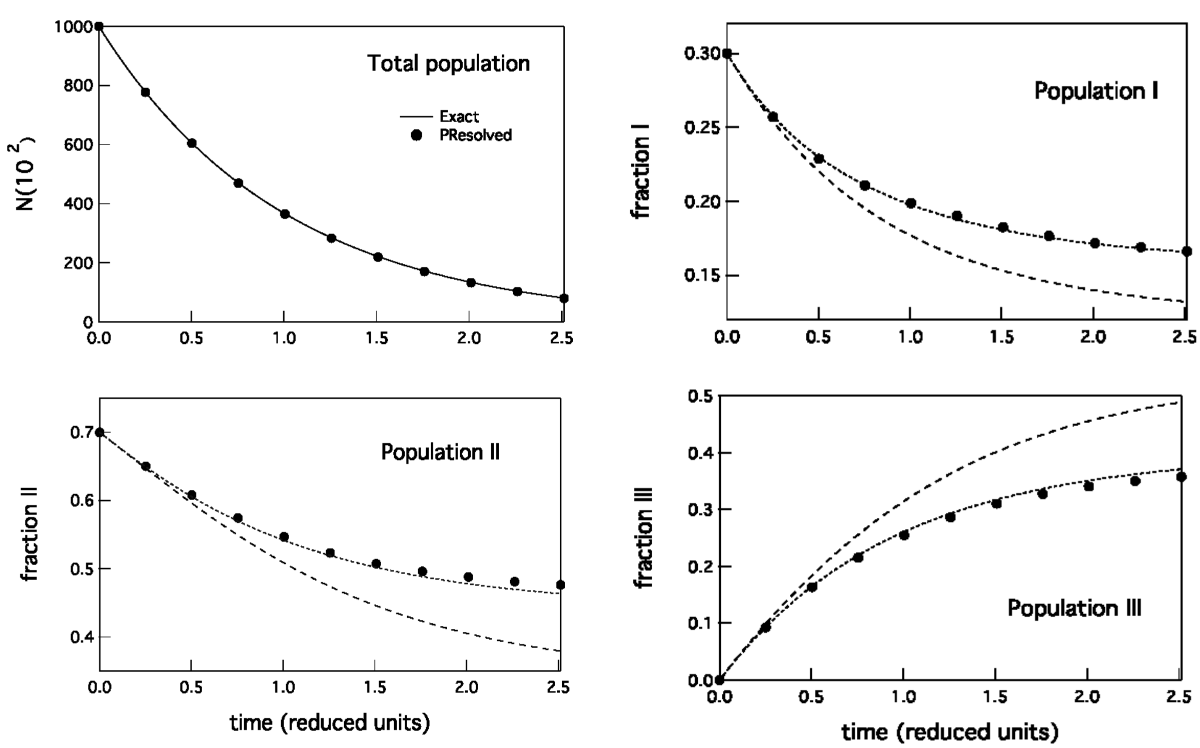

Figure 2. Evolution of total particle number (upper left panel) showing exact QMOM (curve) and particle-resolved simulation (markers) results. Remaining panels show particle number fraction for each population: long-dashed curves, QMOM with 7-moment tracking; short dashed curves, QMOM with 14-moment tracking; markers, particle resolved simulation.

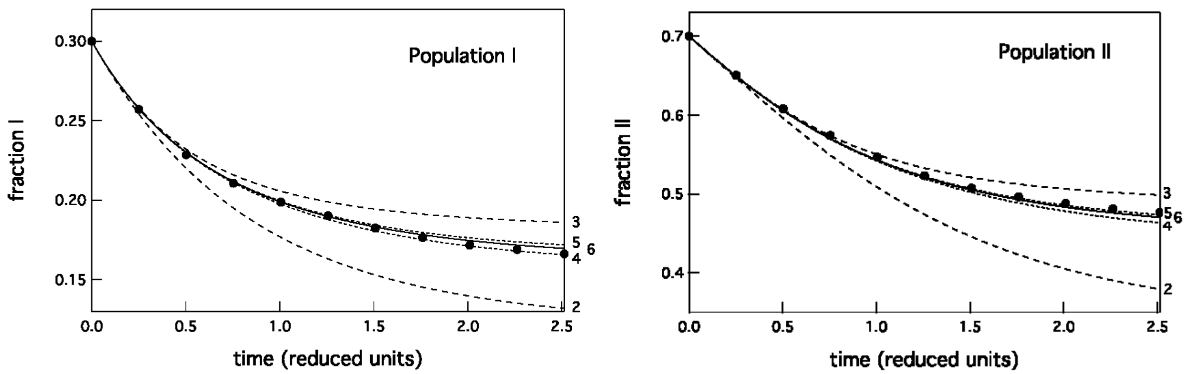

Figure 3. Testing higher-order Gauss and Gauss-Radau quadrature schemes. Curves are labeled by the number of univariate moments for each initial population used in the calculation. Gaussian quadratures use an even number of moments; Gauss-Radau quadratures an odd number. One point Gauss-Radau quadrature (not shown) yields the trivial upper bound – no decay. Note that the use of Gauss-Radau (and Gauss-Lobatto) quadratures in addition to providing bounds extends the capability of QMOM to allow for accurate integrations over boundary-limited domains [9].

A more detailed description of these results will follow in future publications.

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