Bivariate Applications and Extensions of the Quadrature Method of Moments (QMOM)

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Summary of the Approach

Recent applications of the QMOM to the simulation of bivariate/multivariate particle populations [1,2,3] are presented, together with a preliminary description of several powerful extensions currently under development through 'hybridization' of the QMOM with principal components analysis (PCA), the spectral method called orthogonal collocation (OC), and empirical orthogonal function (EOF) methods. Applications of the PCA-QMOM will focus on: (i) bivariate coagulation and sintering of irregular-shaped particle aggregates in laminar flames, and (ii) general mixing of multicomponent/ multivariate particle populations under atmospheric conditions. Hybridization seeks to combine the best features of PCA, OC, and EOF with the QMOM. PCA provides an efficient and systematic way to assign quadrature points in higher dimensions. OC and EOF are fixed grid methods with generally higher resolution than the QMOM, which uses a smaller number of quadrature points optimally determined by the lower order moments of the particle distribution function. Thus, the QMOM has the intrinsic advantage of being an adaptive grid method. Combining these theoretical/numerical approaches should yield higher-resolution adaptive grids for the efficient simulation of multivariate nanoparticle populations evolving under complex environmental conditions including the nonlinear process of Brownian coagulation, with applications to combustion engineering, industrial crystallization, and atmospheric science.

Orthogonal collocation and moment methods are complementary approaches to particle population balance that might seem at first glance to be unlikely candidates for combining into a more powerful hybrid approach. The key concepts, illustrated as follows for the univariate case, are readily extendable to higher dimensions. Moment methods track *integrals* over the particle distribution function (PDF) and not the distribution itself. For a univariate distribution these moments take the form:

$$\mu_k = \int x^k f(x)dx \approx \sum_{i=1}^{n} x_i^{\ k} w_i \qquad (1)$$

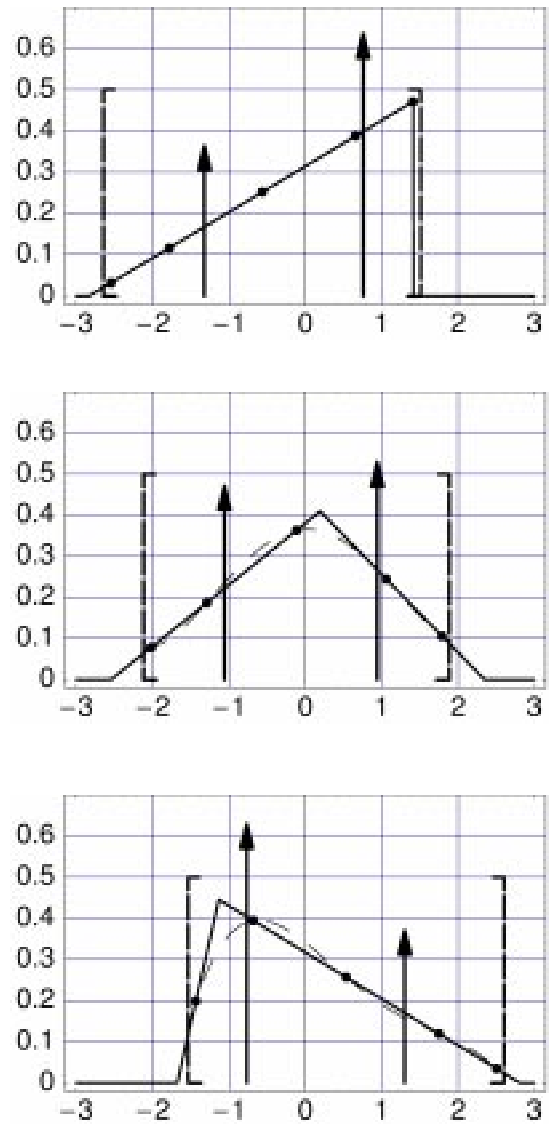

Figure 1: Location of the ROI (dashed vertical lines) based on Eq. 2 with Δ =2 for a triangular distribution function (solid line) with $\mu_0 = 1,\ \mu_1 = 0,$ and $\sigma = 1$, for three different values of the skewness: S = -0.565685 (top), S = -0.115685 (center) and S = 0.534315 (bottom). The arrows are located at the abscissae given by the QMOM and have lengths given by the corresponding weights. The five dots highlighted in the figure correspond to the Chebyshev nodes used for the OCM approximation (dashed line) almost superimposed to the exact PDF.

for the k^{th} moment. The approximate equality gives the *n*-point Gaussian quadrature approximation. The OCM, on the other hand, is a distribution function method. It attempts to represent and track the full pdf represented by polynomial interpolation over a discrete finite grid of sampling points at which the polynomial is constrained to match the PDF. Thus the OCM generates an approximation to the full pdf, whereas the QMOM provides very accurate tracking of moments but no representation of the PDF itself.

In the case of the QMOM, the pdf provides a nonstandard weight function for which the abscissas and weights are obtained and updated as a function of time from the moments. Equations for updating the moments are constructed in terms of the abscissas and weights thereby resulting in closure for the system of evolving moments (or evolving abscissas and weights) [4]. The QMOM has the advantage of being fast and remarkably accurate [5,6], and the disadvantage of not furnishing the pdf – although for many applications only integrals over the pdf are required and here the method can be quite accurate [7]. OC traditionally works on a fixed grid of N points. The pdf is defined and updated using these points, and interpolated by the polynomial of degree N-1 constrained to pass through the N points [8]. If N is sufficiently large (e.g. 5-15) the method provides an accurate representation of the size distribution (see Fig. 1), provided the points are co-located with the pdf. The OCM is not constrained to reproduce moments and does not have the adaptive QMOM property of evolving abscissas, which are always optimally placed to reproduce moments, with changes in shape and location of the pdf. This means that if there is large change in the pdf, e.g. due to a large growth in particle size, the fixed grid must be initially established broad enough to cover any anticipated change. Thus a great many more OC points may be required than would otherwise be necessary to fit the pdf– if one had and adaptive grid and knew the region of interest (ROI) as a function of time. This is the main idea behind the hybrid approach. Finally we have the advantage that neither the QMOM or the OCM appear to suffer from the well-known problems of 'numerical diffusion' inherent in sectional methods, and neither invokes assumed size distributions as do the 'modal methods' and early moment closure methods used prior to development of the QMOM.

We next describe the new approach that combines the best features of the QMOM and OC methods into a hybrid method that eliminates many of the weaknesses of the individual methods [9]. In application, the hybrid QMOM–OCM runs these two complementary methods in parallel using the lowest-order moments tracked in the QMOM to determine the ROI as a function of time for the OCM. Thus the QMOM is totally independent of the OCM, whereas the latter uses results from the QMOM to update the region of interest. Figure 1 illustrates the hybrid method for a triangular pdf of evolving skewness, using 5 collocation points and evolution of 4 moments or, equivalently, 2 quadrature abscissas and weights. The boundaries *a* and *b* of the ROI are determined using:

$$a = \bar{x} - \Delta\frac{\sigma}{2}\left(\sqrt{4+S^2} - S\right), \quad b = \bar{x} + \Delta\frac{\sigma}{2}\left(\sqrt{4+S^2} + S\right) \qquad (2)$$

where $\bar{x} = \mu_1 / \mu_0$ denotes the mean of the distribution,

$$\sigma = \sqrt{\frac{\mu_2}{\mu_0} - \left(\frac{\mu_1}{\mu_0}\right)^2}$$

is the standard deviation, and

$$S = \frac{1}{\sigma^3}\left(\frac{\mu_3}{\mu_0} - 3\frac{\mu_1}{\mu_0}\frac{\mu_2}{\mu_0} + 2\left(\frac{\mu_1}{\mu_0}\right)^3\right)$$

is the skewness. Δ, is a fixed parameter that plays that role of the radius of the 'confidence interval', and need not be symmetrical if *S* is not equal to zero. Based on the Chebyshev inequality this parameter should be restricted to values greater than unity and a convenient approximation would be given by setting Δ in the 2-4 range (the higher value for broad distributions such as the lognormal).

The QMOM calculation my be improved by tracking more moments. We will present results from additional tracking of the next higher moments μ_4 and μ_5, using in the first case Gauss-Radau quadrature to obtain a new quadrature abscissa located at the ROI left boundary (a), and Gauss-Lobatto quadrature incorporating both higher moments to obtain new abscissas at both boundaries (a and b). Thus six moments (0 through 5) suffice to give (uniquely) two quadrature points within the ROI and one at each boundary. A significant advantage of including Gauss-Radau and Gauss-Lobatto quadratures is the fact that one can now compute upper and lower bounds for CERTAIN INTEGRALS over ANY DISTRIBUTION within the ROI [10].

Bivariate Extension

Both the QMOM and the OCM can be extended to higher order problems. The first successful extension of moment methods to particle distribution functions characterized by more than a single mass (or radius) coordinate was achieved using the QMOM [11]. Calculations were made for a well-known bivariate (volume/area) model of particles undergoing simultaneous coagulation and sintering, developed by Koch and Friedlander, and compared with benchmark calculations using a discrete model with 150 size classes along each of the two coordinates (22500 gid points in all). A bivariate QMOM simulation can be carried out in seconds on using a ‘personal computer’ whereas the benchmark calculation required about 10 calendar days on a Sun Spark Enterprise workstation. Thus adding in the parallel QMOM simulation requires only minor computational burden that, by identifying the ROI, results in a much smaller N-values being required by the OCM and much less computational burden overall.

The great computational efficiency of the QMOM makes this method an ideal candidate for extention to bivariate and multivariate simulations. One powerful approch that has been recently developed is an extension of the QMOM using principal components analysis (PCA) to assign the quadrature points [12, 13]. The bivariate moments are of the mixed form:

$$\mu_{kl} = \int_0^\infty \int_0^\infty x_1^k x_2^l f(x_1, x_2) dx_1 dx_2 \qquad (3)$$

where x_1 and x_2 are particle volume and surface area, respectively, and $f(x_1, x_2)$ is the bivariate number distribution function. The simplest bivariate calculation tracks six mixed moments representing particle number concentration, μ_{00}, the centroid of the distribution $\{\mu_{10}, \mu_{01}\}$, and the elements of the 2x2 covariance matrix $\{\mu_{20}, \mu_{02}, \mu_{11}\}$ in coordinates centered about the means. In the hybrid PCA-QMOM/OCM (QMOM-OCM for short) the ROI is optimally defined in the principal coordinate frame. Here the ROI is centered at the coordinate centroid and has an ellipsoidal geometry with axes range several times the PCA variances obtained as the eigenvalues of the covariance matrix. The corresponding PCA eigenvectors determine the principal axes, or orientation of the ellipsoid. The hybrid method yields both moments (from the QMOM) and an accurate and computationally efficient representation of the pdf (from the OCM). Illustrative calculations will be carried out both for a univariate case (as in Fig. 1) and for a bivariate simulation using the irregular-shaped particle, volume-area coordinates of the coagulation-sintering model.

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