10.7 RETRIEVAL OF PARTICLE SIZE DISTRIBUTION FROM OPTICAL MEASUREMENT: INFLUENCE OF NONSPHERICITY

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1. INTRODUCTION

Optical remote sensing has been increasingly used to measure particle size distributions, including multispectral extinction measurements (Arnott et al. 1997), multispectral backscattering measurements (Ben-David and Herman 1985) and multi-angular scattering measurements (Heintzerberg 1978). Despite the subtle differences, most remote sensing methods require solving a Fredholm integral equation of the first type (Twomey 1977). Take the inversion of multi-spectral extinction measurements as an example. The optical depth (b) of a particulate cloud can be expressed as

$$\int_{D_{min}}^{D_{max}} K(y,D)x(D)dD = b(y)\,, \tag{1a}$$

$$K(y,D) = LC_e(y,D)\,, \tag{1b}$$

where y represents the wavenumber of the electromagnetic wave, D is the particle diameter, D_{min} and D_{max} are the smallest and largest diameter respectively, x(D) is the desired size distribution, $C_e(D,y)$ the extinction cross section of a particle with diameter D at wavenumber , and L is the physical path. It is well known that Eq. (1) is illposed in that a large family of x(D) produce similar b(y) (Twomey 1977; Hansen 1998).

Atmospheric particles such as aerosol and ice crystals are often nonspherical. There is abundant evidence that the Mie theory is inadequate to approximate scattering properties of nonspherical particles (Mishchenko et al. 1996). It is therefore anticipated that the Mie theory-based instruments may produce spurious size distributions in the presence of nonspherical particles.

The anomalous diffraction theory (ADT) is also widely used to approximate light scattering by nonspherical particles (van de Hulst 1957). Some researchers have argued that ADT may be better than the Mie theory to approximate light scattering by nonspherical particles (Baran et al. 1998). Recently, new ADT expressions for finite circular cylinders were presented (Liu et al. 1998).

In this paper a new retrieval algorithm is presented. Then it is compared with an iterative algorithm and the method of truncated singular value decomposition (TSVD). The influences of particle nonsphericity on size distribution retrieval are investigated by use of both the Mie theory and ADT.

2. NEW PROCEDURE FOR RETRIEVAL

2.1. Retrieval Algorithm

In practice, one needs to discretize Eq.(1) in order to solve it numerically. Without loss of the generality, the discrete version of Eq. (1) is described by the matrix equation:

$$\mathbf{Ax} = \mathbf{b}\,, \tag{2}$$

where A is a M x N base matrix with the component $A_{ij} = \int_{D_j-\Delta D/2}^{D_j+\Delta D/2} K(y_i,D)dD$, **x** is a N x 1 vector whose components x_j represent the particle concentration at D = D_j, **b** is a M x 1 vector whose components b_i are the measured signal at y = y_i, M is the number of wavenumbers and N the number of size bins.

A cursory examination may lead one to consider that the retrieval problem is trivial because standard methods are available to solve Eq.(2) (Lawson and Hanson 1995). This is true when **A** is well-conditioned. Unfortunately, this is not true for our problem, because of the inherent illposedness carried over from the original integral equation. In order to obtain a useful solution, it is necessary to incorporate additional constraints about the desired solution. The most commonly used constraints on the size distribution are nonnegativity and smoothness.

A natural way to combine the smoothness constraint with Eq. (2) was introduced independently by Phillips (1962) and Tikhonov (1963), and some modifications of Phillips' work was suggested by Twomey (1963). Briefly, with a Lagrange multiplier λ, the constrained least squares problem becomes

$$\min_x \left(\|\mathbf{Ax} - \mathbf{b}\|^2 + \lambda \|\mathbf{Lx}\|^2 \right), \tag{3}$$

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where **L** is a P x N smoothing matrix with P ≤ N that is usually a discrete representation of a differential operator, and $\|\mathbf{w}\| = \left(\sum_i w_i^2\right)^{1/2}$ is the Euclidean norm of the vector **w.** In this paper, we use the name of "PTT" to represent this method. Although PTT has found wide applications, its direct use may lead to nonphysical negative values of number concentration (Steele and Turco 1997).

On the other hand, to obtain a nonnegative solution, Lawson and Hanson (1995) developed an algorithm to solve a nonnegative least squares problem (NNLS) :

$$\min_{x}\left(\|\mathbf{Ax}-\mathbf{b}\|^2\right), \text{ subject to } \mathbf{x} \geq \mathbf{0}. \qquad (4)$$

This method has received some applications in atmospheric sciences. For example, Yau and Rogers (1984) applied it to inferring the size distribution of precipitation areas from raingauge measurements. Kim and Boatman (1990) used this algorithm to correct size distributions directly measured with an Forward Scattering Spectrometer Probe (FSSP). Although the NNLS guarantees a nonnegative solution, the solution may have unrealistic fluctuations and spikes when the inverse problem is seriously illposed, such as retrieving size distributions from multispectral optical depth.

The shortcomings of PTT and NNLS can be circumvented by coupling both together in two steps. First, the minimization problem of Eq. (3) is equivalent to solving the corresponding normal equation:

$$\left(\mathbf{A}^t\mathbf{A} + \lambda\mathbf{L}^t\mathbf{L}\right)\mathbf{x} = \mathbf{A}^t\mathbf{b}, \qquad (5)$$

where the superscript t indicates the matrix transpose. Second, applying NNLS to Eq. (5) yields a smoothed nonnegative solution. This new algorithm is named the smoothing-constrained NNLS and is denoted by SCNNLS.

2.2. The L-Curve Method

The nonnegativity constraint is physically realistic because particle number concentration can not be negative. However, the smoothness constraint needs to be used with caution. Size distributions of various smoothness may occur in the atmosphere, depending on the physical processes and scales involved (Liu and Hallett 1998). Therefore, as in PTT, finding an optimal Lagrange multiplier is as important as the algorithm itself. A too large multiplier leaves out information available in the measurement, while a too small multiplier produces a solution significantly contaminated by errors. Furthermore, the optimal Lagrange multiplier depends on errors in measurements, properties of the base matrix **A**, and the smoothness of the desired size distribution. In reality, all these things are hardly known in advance. Therefore, an objective approach that does not require such pre-information is desirable for choosing the Lagrange multiplier. The so-called L-curve method has proved to serve this purpose well (Hansen 1998; Schimpf and Schreier 1997) and has been proved for PTT (Hansen 1998). However, applications to inverse problems constrained by both smoothness and nonnegativity have not been reported. We have made numerical experiments of applying the "L-curve" idea to SCNNLS. Our results indicate that although the "L-corner" behavior may not be as sharp as that in PTT, depending on the specific problem, a λ that gives an optimal tradeoff between the smoothing degree and the residue can still be located. An example of the L-curve for SCNNLS is shown in Fig. 1.

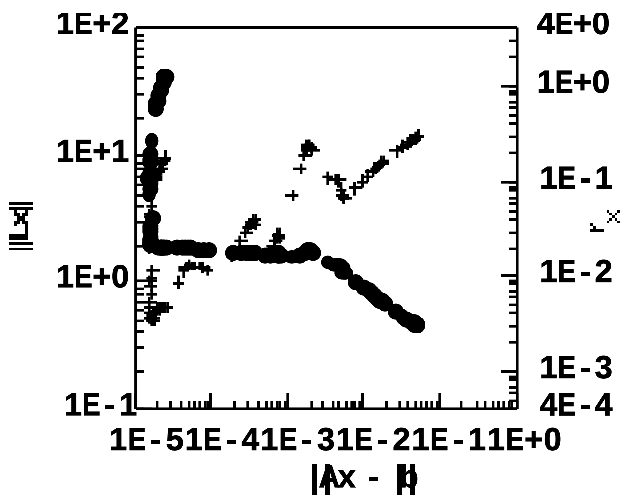

Fig.1. An example of the L-curve for SCNNLS. Dots are ||**Lx**||, a measure of size distribution smoothness. Smaller values represent more smoothing. Crosses are relative size distribution error, $r_x = \|\mathbf{x} - \mathbf{x}^*\| / \|\mathbf{x}^*\|$, where **x*** is the true size distribution. ||**Ax**-**b**|| is the residual error in original matrix equation. The L-curve characterizes the relationship between the smoothness of the solution and the residue error as a function of λ. Note that the λ corresponding to the corner of the L-curve gives a good size distribution retrieval (low r_X) .

3. EVALUATION OF SCNNLS

To evaluate SCNNLS, numerical studies are made for spherical particles. We use Mie theory and the refractive indices for ice (Warren 1984) to generate the base matrix **A** and a synthetic multispectral optical depth for a given size distribution. Then a corresponding size distribution is retrieved by inverting the synthetic optical depth. To demonstrate the advantages of the new procedure, size distribution retrievals are also made for the same data set by use of the iterative algorithm presented in Arnott et al. (1997) and TSVD. Figure 2 is an example of the retrieval. It can be seen from these figures that the size distributions retrieved by all the three methods are sufficiently good to the human eyes in terms of the size distribution as well as the retrieved optical depths. However, the iterative algorithm is much slower than the other two. The TVSD is as fast as SCNNLS. However, a careful examination reveals that it produces unrealistic negative concentrations, particularly at both ends of particle sizes. These results demonstrate that SCNNLS outperforms the other two algorithms.

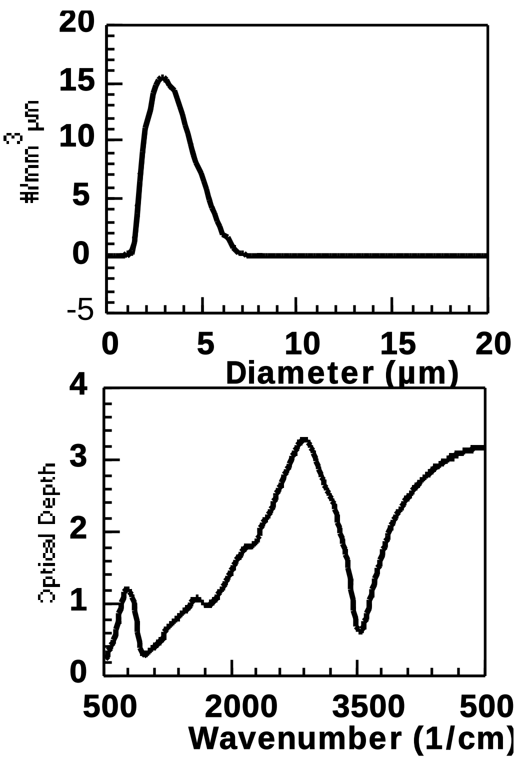

Fig.2. Retrieved size distributions (a) and corresponding optical depth (b) . True = thick solid, SCNNLS-retrieval = solid; Iterative-retrieval = dashed; TSVD-retrieval = dotted.

4. EFFECT OF MIE THEORY

The Mie theory has been extensively used in optically sizing instruments for measuring size distributions because it is difficult and expensive to compute scattering properties for nonspherical particles. Recently, the T-matrix method for nonspherical scattering has been significantly improved (Mishchenko et al. 1996). To study the influence of particle nonsphericity, we use the T-matrix method to generate the base matrix **A** and synthetic spectral optical depth for given size distributions of finite cylinders. The synthetic optical depth is then inverted by use of both the T-matrix-calculated **A** and that calculated based on Mie theory for spheres. It is noteworthy that even the T-matrix method is still so computationally intensive ("M times N" T-matrix calculations are needed to generate a M x N base matrix **A**) that the matrix **A** has been generated only for cylinders with the diameter-to-aspect ratio = 1.0 and M = 19 ; N = 129. The values of M =19 and N = 129 are not optimal; they are the results of the compromise between retrieval performance, size resolution and available computer resources. As shown in Fig. 3a, when the appropriate scattering theory (here T-matrix method) is used, the retrieved size distribution (thin solid curve) agree well with the true size distribution (thick solid curve). However, when Mie theory is used, the retrieved size distribution (dotted curve) significantly departs from the true size distribution. Spurious particles are generated at both size ends, and a spurious second mode of large particles occurs. Despite the significant differences in retrieved size distributions, only small deviations occur for retrieved optical depths.

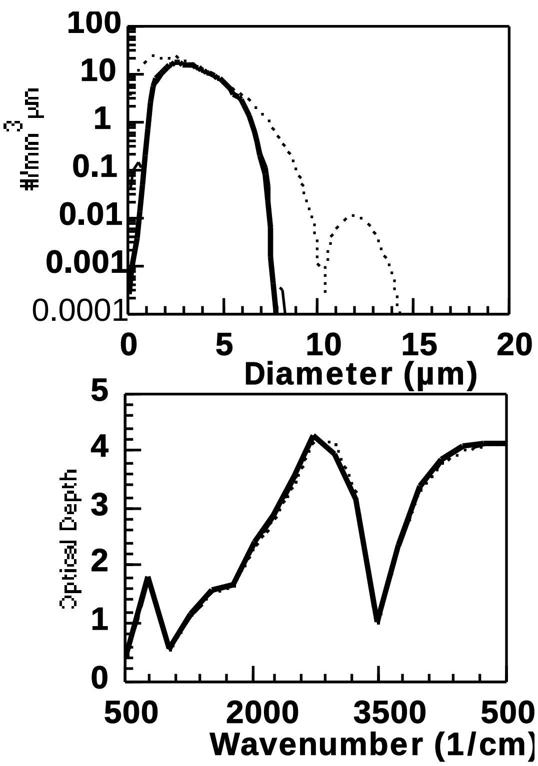

Fig.3. Effects of the Mie theory on size distribution retrieval. True = thick solid; T-matrix-retrieval = solid; Mie theory-retrieval = dotted. Note that T-matrix retrieved results almost overlap with the true values, and "diameter" for T-matrix and ADT refers to cylinder diameter.

5. EFFECT OF ADT

Results from Liu et al. (1998) reveal that ADT only works better than the Mie theory at special wavelengths where relative absorption is strong. Because wavelengths other than the special ones are also needed in retrieving size distributions from multispectral extinction measurements, using ADT also causes errors in the base matrix **A**, and such errors are expected to distort the retrieved size distribution as well. To demonstrate the effect of ADT, size distributions of finite cylinders are retrieved by use of the ADT developed in Liu et al. (1998). Figure 4 shows the results, suggesting that the ADT approximation cause retrieval errors similar to the Mie theory .

6. DISCUSSION

The above results demonstrate that severe distortions occur when the Mie theory or ADT is used to retrieve size distributions of nonspherical particles. In fact, applying either the Mie theory or ADT to nonspherical particles results in systematic errors in the base matrix **A.** It is such systematic errors in **A** that conspire with the illposedness to cause the distortions of retrieved size distributions. As shown in Fig.5, the retrieval process actually transforms the error in the matrix **A** into the error in retrieved size distribution, yet makes the agreement between true and retrieved optical depths reasonably better. These studies suggest caution in interpreting size distributions measured by Mie theory-based instruments when nonspherical particles exist.

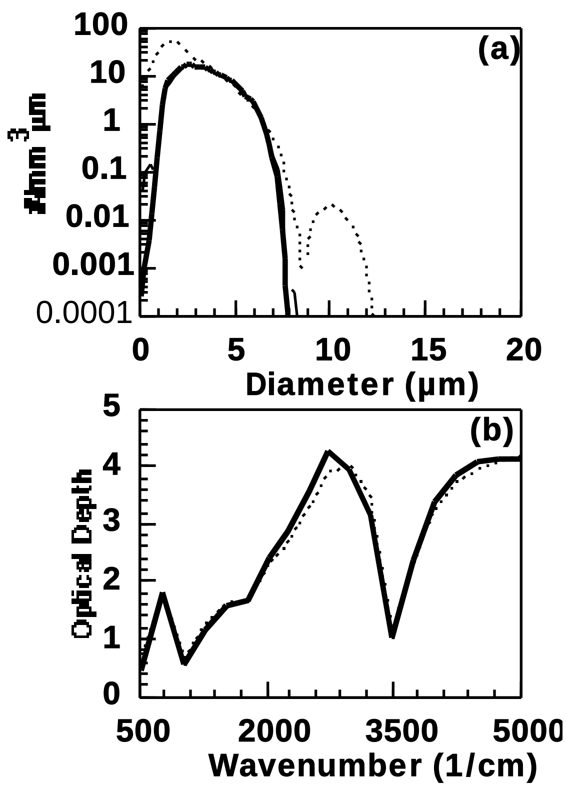

Fig.4. Effects of ADT on size distribution retrieval. True = thick solid; T-matrix-retrieval = solid; ADT-retrieval = dotted. Note that T-matrix-retrieved results almost overlap with the true values.

7. CONCLUDING REMARKS

A new procedure that incorporates speed, smoothness and nonnegativity to retrieve size distributions from multispectral extinction measurements was developed and evaluated by inverting synthetic multispectral optical depth. The advantages of the new procedure were demonstrated by comparing with an iterative algorithm and TSVD.

The influence of particle nonsphericity on size distribution retrieval was studied by using T-matrix method to calculate extinction cross sections of finite cylinders. The results show that severe distortions of retrieved size distributions occur when applying Mie theory to nonspherical particles. These results are similar to those obtained by Heintzenberg (1978) in inverting multi-angle scattering data and those by Gardiner and Hallett (1985) in analyzing the FSSP measurements in the presence of ice crystals. These results suggest that caution should be applied in analyzing and interpreting size distributions measured in the presence of nonspherical particles.

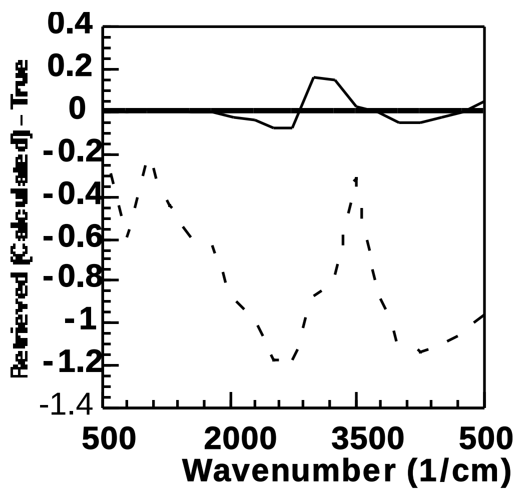

Fig.5. An example that retrieval processes transforms the error in **A** into error in retrieved size distributions. Thick solid = retrieved using T-matrix; Solid = retrieved using the Mie theory ; Dashed = calculated using the Mie theory.

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