Extension of the graphical technique for estimation of particle size distribution parameters for the consistent intercomparison of diverse sets of multiwavelength lidar derived optical coefficients

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In applying the graphical technique to the estimation of the particle size distribution (PSD) parameters, determination of proper bounds surrounding the solution space for a particular confidence level is essential to the consistent intercomparison of diverse multiwavelength lidar optical data sets. The graphical technique utilizes ratios of backscatter and/or extinction coefficients, and it is shown that if the correlation between ratios is not taken into account in calculating the error bounds, the solution space will be overestimated, resulting in relatively larger discrepancies for a larger number of optical coefficients. A method for correcting the bounds, to account for the correlation is developed for various numbers of wavelengths. These improved bounds are then applied, for the case of a monomodal lognormal PSD, with an assumed refractive index, to assess the role additional Raman extinction channels play in improving retrieval capability of a typical three-channel backscatter lidar (1064, 532, and 355 nm) under varying noise levels. Applying the same formalism to underlying bimodal distributions of coarse and fine particles can result in false monomodal solutions. However, when both Raman optical extinction channels are available, no solution is obtained. This can potentially serve as a quick and simple method, prior to a more complex regularization analysis, to differentiate between cases in which the fine mode is dominant versus the cases in which the contribution from the coarse mode is significant. © 2005 Optical Society of America

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1. Introduction

The use of multiwavelength optical data to determine the microphysical parameters of aerosols has been applied to a variety of multispectral sensing techniques such as sun–sky radiometry. In these cases, retrieval of microphysical parameters involves the inversion of the ill-posed Fredholm integral equations, which are numerically unstable, and additional constraints need to be imposed to regularize the solutions. If there are sufficient spectral data, positivity and curvature constraints may be imposed for meaningful microphysical retrieval. On the other hand, if the number of optical measurements is small, regularization is imposed by parameterizing the PSD, assuming an a priori shape distribution function, and the retrieval problem is reduced to an estimation of the PSD parameters that are fundamentally related to its lowest moments. A somewhat similar approach showed that multispectral extinction measurements and their derived Ångstrom spectrum could be applied to estimate effective particle parameters, such as the total volume and effective radius, without any underlying shape assumption. However, these estimates become more uncertain and multivalued if the refractive index is unknown. This approach has also been extended to the extraction of the fine mode Ångstrom parameter (and hence size parameters) of a bimodal distribution, albeit with high uncertainty. These uncertainties are further magnified because it is the spectral derivative of the Ångstrom coefficient that dominates the error analysis making the Ångstrom analysis approach useful only for a sufficiently large number of optical data measurements with sufficiently small optical depth uncertainties.
While shape-independent linear optimization approaches may be appropriate for hyperspectral passive sensors where a large number of optical data sets are available, and for highly sophisticated lidar systems (such as the 6-wavelength, 11-channel aerosol lidar at the ITR), more typical lidar systems are limited by a smaller number of measurements and large uncertainty errors. Recent work using a variety of regularization methods seems to show that stable regularized PSD solutions may be obtainable using more conventional and accessible lidars. In Ref. 6, it was shown that by using a hybrid regularization approach and higher-order splines a $3\beta + 1\alpha$ sensing configuration that is obtainable by using the harmonics of the Nd:YAG laser plus a nitrogen Raman channel for an independent extinction measurement, which may be extrapolated to 532 nm ($\beta = 355, 532, 1064; \alpha = 532$), can accurately retrieve the PSD effective parameters (effective radius, surface, volume, and total number) but requires an a priori refractive index. Reference 7 shows that by adding another Raman channel for extinction coefficient at 355 nm, significant constraints on the refractive index can be obtained. In Ref. 8, a statistical regularization approach where no single solution is used but an average of regularized solutions (based on the principle of minimum discrepancy) is employed in a somewhat empirical fashion seems to significantly reduce retrieval uncertainties. This method was further extended to bimodal distributions with unknown complex refractive indices, but in such cases retrieval errors in the PSD moments significantly increase (~50% for 10% measurement noise levels). It must be pointed out that all of the regularization methods must use criteria for determining a smoothing parameter. This determination reduces to finding a local minimum, which in some cases does not exist, or in other cases there are multiple local minima, (especially in the case of the generalized cross validation method used in Ref. 7) and much effort must go into the elimination of spurious retrievals. These issues are even more of a problem with a smaller number of optical measurements. Therefore it is certainly useful to explore more direct methods that can be applied to smaller optical data sets. In addition, even in situations where regularization methods may be suitable, time constraints as well as difficulties in eliminating spurious inversions will significantly benefit from simpler approaches that may be regarded as first estimates on the PSD parameters.

As described above, in situations where the number of optical data channels is insufficient to apply linear regularization effectively, shape constraints need to be used. In particular, the distribution $n(r)$ is supposed to be determined by a small set of parameters $p$ that can be connected to the lowest moments. Such decompositions are very useful in stabilizing and simplifying the retrieval, and it has been shown that the aerosol optical properties can be well described in terms of these lowest-order moments. This observation has led to useful approaches that directly connect microphysical moments to optical multiwavelength measurements such as those from SAGE III and simulated backscatter lidar data. Of course, this result is most useful in the case of stratospheric aerosols with less-variable refractive indices.

For a given parameterized PSD, within the Mie scattering approximation valid for spherical particles, the optical backscatter ($\beta$) and extinction ($\alpha$) coefficients can be written in the form

$$\begin{align*}
\{\beta(\lambda_i; p)\} &= \int_0^\infty \left\{K_{\beta}(r, \lambda_i)\right\} n(r) dr, \\
\{\alpha(\lambda_i; p)\} &= \int_0^\infty \left\{K_{\alpha}(r, \lambda_i)\right\} n(r) dr,
\end{align*}
$$

where $K_{\beta,\alpha}$ are proportional to the Mie backscatter–extinction efficiencies.

A typical form for the parameterized particle size distribution function is the lognormal distributions given by

$$n_{\text{lognormal}}(r, p) = \frac{N_d}{\sqrt{2\pi} \ln(\sigma)r} \exp\left(\frac{-\ln(r/\bar{r})^2}{2(\ln(\sigma)^2}\right). \quad (2)$$

The parameter set consists of the particle density $N_d$, the median radius $\bar{r}$, and the geometric standard deviation (GSD) $\sigma$. However, it is clear that simplifications result, if instead of the optical coefficients, optical coefficient ratios are used, thereby allowing a reduced parameterization that depends only on $p = [\bar{r}, \sigma]^{16-17}$.

Solving Eq. (1) then becomes a matter of finding the distribution parameters within the mode parameter space that best match the optical data set in a least squares sense. However, such analytic techniques suffer from difficulties due to possible local minima, and convergence. Furthermore, error assessment of the results requires time-consuming bootstrap Monte Carlo simulations. To avoid these issues, a graphical approach was proposed that can be used to estimate the PSD parameter values as well as the uncertainty in the retrieval. Rather than applying numerical algorithms, this approach utilizes the search of pregenerated look up tables (LUTs) that map optical coefficient ratios to a discretized PSD parameter space. The solution space is graphically visualized as the intersection of contours in the parameter space. The framework for this paper uses this graphical methodology that is based on a lognormal monomodal PSD and an assumed index of refraction. In Ref. 16, it was tacitly assumed that the error bounds would directly follow from the optical coefficients, extinction and backscatter coefficients but the ratio of these (independent) quantities. Such optical coefficient ratios are not Gaussian distributed or independent and this can have a significant effect on the total cumulative distribution function and therefore, the uncertainty bounds for a given confidence level needs to be redefined. This extension of earlier work is necessary to ensure that comparisons made...
between different optical coefficient sets are based on the same confidence level. Only then can the graphical technique be meaningfully applied to investigate improvements in the retrieval of PSD parameters with increased numbers of optical coefficients. Unfortunately direct comparisons with Refs. 16−17 cannot be performed since no working definition of the confidence bounds were given. In this work, we examine the potential benefits from the addition of nitrogen Raman extinction channels to a lidar system based on a typical three-wavelength Nd:YAG laser. The results obtained are consistent with recent studies using regularization techniques reviewed earlier but are obtained in a more direct manner.6

In addition, the graphical method was previously applied only to retrieve the mean radius and GSD parameters with little attention to obtaining estimates of the total number density.16,17 We outline a procedure for retrieval of both the number density and its uncertainty following the determination of PSD parameters. Finally, while the graphical technique is based on a single-mode PSD assumption, we also apply it to underlying bimodal distributions to see if quick insight can be gained into the nature of the distribution.

In Section 2, the basics of the graphical inversion are reviewed and methodology for determining confidence bounds is outlined. Further, a procedure to retrieve the number density and its uncertainty is introduced. In section 3, we apply the method to study the retrieval capabilities of a typical three-wavelength backscatter lidar using a Nd:YAG laser transmitter (355, 532, and 1064 nm) for an a priori known index of refraction and compare the results with those obtained from adding additional extinction coefficients from Raman channels. We also explore the possibility of using the graphical method to distinguish monomodal from bimodal distributions. In section 4, we summarize our results.

2. Graphical Retrieval of PSD Parameters

Unlike numerical search algorithms that may have difficulties in converging, the graphical method does not need to find local minima. Therefore the technique can be used to retrieve PSD parameters as well as provide an estimate of retrieval errors without the need to perform numerical minimization. In this section, we describe the general graphical approach to determining parameter uncertainty estimates.

To examine how a set of multiwavelength optical coefficients can be used to determine underlying distribution parameters and some of the difficulties involved, we illustrate the graphical approach as presented by Post16 and applied to three-wavelength lidar.

An optical coefficient set comprising extinction ratios can be given as

\[
\begin{align*}
R_1 &= \frac{\alpha_{352}}{\alpha_{355}}, & R_2 &= \frac{\alpha_{1064}}{\alpha_{355}}, & R_3 &= \frac{\alpha_{1064}}{\alpha_{352}},
\end{align*}
\]

Similar ratio sets can be considered for the backscatter β or lidar ratio (S = α/β) parameters. The main steps in the graphical approach are as follows:

1. For a given point in the parameter space \( p(\tau_0, \sigma_0) \), where the zero subscript denotes the parameters used in generating the optical coefficients, a forward calculation can generate the corresponding calculated optical coefficient ratios \( R_k(p) \). The parameter space is discretized into cells \( p(\tau_i, \sigma_i) = p_m \), and the corresponding values of the optical coefficient (ratios) are evaluated and stored in three matrices, \( R^{i,j}_k \), \( k = 1−3 \) (for a three-wavelength lidar), which will then serve as look-up-tables (LUTs).

2. Given ratios of initially assumed optical coefficients (for the purpose of this paper, or calculated coefficients from lidar data) and the associated ratio uncertainty bounds (\( \Delta R_k \)), the preconstructed database (LUTs) is searched to determine the cells \( (i, j) \) that satisfy

\[
R_k - \Delta R_k \leq R^{i,j}_k \leq R_k + \Delta R_k, \quad k = 1−3.
\]

This can be thought of as finding the intersection of the six curves in the parameter space \( p \).

In this way, a visual representation of the solution space can be constructed and different optical measurement schemes compared. This procedure can be generalized to \( n \) distinct optical coefficients by forming the set of resulting \( n(n−1)/2 \) ratios \( R^{i,j}_k = \chi_j/\chi_i \), \( j = i, i = 1−n \).

In this work, the \( n(n−1)/2 \) precalculated ratio matrices (LUTs) \( R^{i,j}_k \) were generated on the following grids: \( \tau \) was divided into small-size (from 0.002 to 0.2 μm in 0.002 steps) and large-size (from 0.2 to 0.9 μm in 0.01 steps) ranges; \( \sigma \) ranged from 1.01 to 2.4 in steps of 0.01. Unless otherwise noted, we limit ourselves to a fixed complex refractive index of \( m = 1.5 + 0.0j \) (representative of nonabsorbing tropospheric aerosols).

To illustrate the general idea, we plot in Fig. 1 the contours for 2% uncertainty for different types of optical coefficient ratios (extinction, backscatter, and \( S = \alpha/\beta \)) generated from a lognormal PSD with \( \tilde{\tau}_0 = 0.65 \) μm, \( \sigma_0 = 1.4 \). In this figure, we can see the significant improvement in PSD retrieval using backscatter coefficients over extinction coefficients, which is well known in scattering theory and pointed out in Ref. 16. Finally, while it may seem that the third element in the ratio set is redundant and supplies no independent information, when the complete error space is assessed, it can make a significant difference.

A. Estimates of Confidence Bounds

In order to calculate the correct confidence bounds, we need to first develop the statistics for the ratio of random variables (RVs) and then combine the variances of the resulting set of ratios. Performing tedious but straightforward calculations based on the PDF of bivariate functions18 results in a probability density function for the fractional optical coefficient ratio deviation \( \delta = (R - \tilde{R})/\tilde{R} \) that is given by
Fig. 1. Example of graphical determination of PSD parameter solution space from three optical coefficients generated from an underlying lognormal distribution with $r_0 \approx 0.65 \mu m$, $\sigma_0 = 1.4$ and with the addition of 2% uncertainty for (a) extinction coefficient ratios, (b) backscatter coefficient ratios and (c) S ratios.

$$P(\delta; \Delta_n, \Delta_d) = \frac{1}{2\pi\Delta_n\Delta_d} \exp\left(-D/2\right) \left\{ \frac{2}{A} \exp\left[-\frac{A}{2}\right] \right. \\
\times (B - 1)^2 \left. + (B - 1) \left[ \frac{2\pi}{A} \exp\left[-\frac{A}{2}\right] \right] \right\},$$

$$A = \frac{1}{\Delta_d^2} + \left(1 + \delta^2\right) \Delta_n^2, \hspace{1cm} B = A^{-1} \delta + \delta^2 \Delta_n^2,$$

$$D = \frac{\delta^2}{\Delta_n^2} - AB^2,$$

where $\Delta_d = \text{std}[\chi_d / \bar{\chi}_d]$ is the fractional standard deviation of the optical coefficient in the denominator and $\Delta_n = \text{std}[\chi_n / \bar{\chi}_n]$ that in the numerator of the optical coefficient ratio $R$. To verify the PDF expression in Eq. (4), we plot in Fig. 2 the above expression together with a numerically generated frequency plot of the ratio statistics for the case of $\Delta_d = \Delta_n = 0.1$. The PDF shows significant asymmetry about zero, illustrating that Gaussian formulations of confidence bounds are incorrect. If the ratios are uncorrelated, once the single-ratio PDF $P$ is given, the total PDF for $N$ ratios scales as $P_N = (P)^N$. Similarly, if we define the ID cumulative distribution function as

$$C(\delta) = \int_{-\delta}^{\delta} P(\delta') d\delta',$$

and the $N$-dimensional cumulative distribution function as

$$C_N(\delta) = \int_{-\delta}^{\delta} \ldots \int_{-\delta}^{\delta} P_N(\delta_1 \ldots \delta_N) d\delta_1 \ldots d\delta_N,$$

then $C_N^{\text{uncorr}}(\delta) = C(\delta)^N$, where $C_N(\delta)$ is the probability that all ratios lie within $R(1 - \delta) \leq R(1 + \delta)$. When each ratio set within the $N$ dimensional domain defined by Eq. 5(b) will result in a parameter retrieval inside the contour curves defined in Eq. (3) (i.e., when the contours bound a closed area in solution space), then the percentage of ratios inside the $N$-dimensional domain will be equal to the percentage of parameter retrievals inside the bounding contour curves. This allows us to define the uncorrelated confidence bound as the numerical (graphical) solution of

$$C_N^{\text{uncorr}}(\delta) = C_{\text{level}},$$

where $C_{\text{level}}$ is the desired confidence level.

However, because correlations in the optical coefficient ratios are present, $C_N^{\text{corr}}(\delta) \neq C(\delta)^N$. To illustrate the effect of correlations between the optical coefficient ratios, the cumulative distribution func-

![Fig. 2. Verification of PDF distribution function formula [Eq. (4)] with a numerically generated histogram of the ratio statistics for the case of $\Delta_d = \Delta_n = 0.1$.](image-url)
A correlated set of optical coefficient ratios obtained from numerical simulations is illustrated in Fig. 3. In this figure, the cumulative PDF \( C_N(\delta) \) of the set of correlated optical coefficient ratios valid for 3, 4 and 5 optical channels is compared with the cumulative PDF generated from the same number of uncorrelated optical coefficient ratios \( C_N^{\text{uncorr}}(\delta) \). Both curves were calculated by using uncorrelated random number generators that directly generate appropriate uncorrelated optical coefficient ensembles with 10% uncertainty. From these, the set of correlated or uncorrelated optical coefficient ratio ensembles were formed and the cumulative distribution function constructed. It should be pointed out that numerically calculating the cumulative PDF is far less intensive than trying to calculate the actual PDF distribution.

We note that a significant increase is observed in the difference between the correlated and the uncorrelated cases as the number of measurements increases. This implies, as intuitively expected, that a larger set of correlated data has the information content of a smaller set of uncorrelated data. By assuming that all the ratios are uncorrelated, the confidence bounds are overestimated depending on the size of the optical coefficient set (3, 4, or 5).

To verify the general approach, we performed conventional Levenberg–Marquart inversions on an ensemble of three-channel backscatter coefficients generated from \textit{a priori} PSD parameters and with equal uncorrelated fractional variances of 10%. The inversions were then compared in Fig. 4 with the graphical method solution space by using the confidence level \( C_{\text{level}} = 0.65 \), but with uncorrelated bounds. The scatter points represent the inversion results, and the solid curves are the confidence boundaries.

The results of the intercomparison are shown in Table 1, which provides the percentage of numerical inversion solutions within the graphical solution.
space. As is clear from the table, the graphical estimates were fairly accurate for calculating the confidence region as seen by the agreement between the fraction of inversions within the confidence region and \( C_{\text{level}} = 0.65 \). However, a small but significant overestimation of the confidence region results. This is easily explained by observing that the true cumulative distribution \( C_N^{\text{corr}}(\delta_{\text{uncorr}}) \) evaluated with the uncorrelated bounds results in \( C_N^{\text{corr}}(\delta_{\text{uncorr}}) = 0.71 \), which is in good agreement with all the table entries except case \( d \). For this case, the solution space included the boundary of the look-up-tables. Therefore some of the numerical inversions were attracted to the boundary, that is inside the confidence region (see Fig. 4(d)). This leads to an additional artificial increase of the percentage of inversions within the confidence zone.

While we have assumed equal optical coefficient ratio uncertainties, and therefore equal confidence bounds in our discussion, the determination of confidence bounds for unequal uncertainties is not difficult. The more general unequal uncertainty case can be reduced to that of the equal uncertainty case by simply rescaling the fractional optical coefficient ratio deviations \( \delta_{i} = s_{i}\delta_{i} \), where \( s_{i} \) is the factor describing the modification in uncertainty for the \( i \)th optical ratio that leads directly to the modified confidence bounds \( \Delta R_{i} = s_{i}\Delta R \).

### B. Number Density Estimation

When optical coefficient ratios are used, information on the number density is not directly accessible. However, once a confidence region is determined, the number density distribution may be obtained numerically by using the following procedure:

1. For each parameter cell \( p(\bar{r}_{i}, \sigma_{i}) \) within the confidence zone, we calculate the associated number density \( N_{k}^{\text{corr}}/N_{0} \) for the \( k \)th optical coefficient \( \chi_{k} \) as the ratio \( N_{0}\chi_{k}/\chi_{0}^{\text{corr}} \), where \( N_{0} \) is the reference number density (= 1 for this work) used to generate the optical coefficient look-up tables \( \chi_{k}^{\text{corr}} \). Therefore the relative error \( N_{k}^{\text{corr}}/N_{0} \) ascribed to each cell in parameter space is \( \chi_{k}/\chi_{0}^{\text{corr}} \).

2. From the relative number density error the average number density uncertainty can be obtained for each cell as \( N_{\text{avg}}^{\text{corr}}/N_{0} = 1/k\sum_{k} N_{k}^{\text{corr}}/N_{0} \) (averaging over all optical coefficient ratios \( k \)). This averaging of the number density inversions is expected to help reduce large deviations in the retrieval. Of course, if the variances of the optical coefficients are different, an appropriate scaling (as described at the end of subsection 2.A) of each channel should be performed.

3. Construct a histogram from the set of \( N_{\text{avg}}^{\text{corr}}/N_{0} \) values calculated from all the parameter cells that were in the solution space.

This method was applied to optical coefficients generated from an example PSD (\( \bar{r}_{0} = 0.5, \sigma_{0} = 1.4 \)) with an uncertainty level of 10%, and confidence level set to 65%. The confidence bounds are shown in Fig. 5(a), and the resultant fractional number density histogram is given in Fig. 5(b). In particular, besides obtaining the general statistics, we see how averaging of the number density over all optical channels compares with that obtained by taking each channel separately and combining all number density retrievals into one histogram (union). From the comparison, we see that the averaging has a positive effect on the large deviation tail, taking probability away from the large deviation events and transferring it to smaller deviating solutions. Of course, once the numerical PDF is obtained, all meaningful statistical moments can be calculated for a given confidence level.

### 3. Applications of Graphical Inversion Assessment

#### A. Parameter Retrieval Improvement with Extinction Coefficients

A quick measure of the overall potential for a set of optical coefficients to retrieve the PSD mode parameters is the size of the retrieval area \( A \) defined by the confidence bound contours in parameter space. Besides limiting the aerosol mode mean \( \bar{r} \) and variance \( \sigma \), reducing the retrieval area obviously reduces the uncertainty in the number density retrieval as well. Therefore it is natural to study how \( A \) varies over the parameter space. Since errors are usually reported as fractional errors, it is useful to define a normalized retrieval area \( \Delta A_{N} = A/(\bar{r}_{0}\sigma_{0}) \), where \( (\bar{r}_{0}, \sigma_{0}) \) are the underlying PSD parameters used to calculate the optical coefficients before the introduction of uncertainty. For completeness, we also normalize deviations of the mode mean and mode variance as \( \Delta \bar{r}_{N} = \Delta \bar{r}/\bar{r}_{0}, \Delta \sigma_{N} = \Delta \sigma/\sigma_{0} \), where \( \Delta \bar{r} \) and \( \Delta \sigma \) are the difference between the maximum and minimum retrieval of the mode radius and the variance within area \( A \), and we explore their variation over the entire lognormal parameter space for the important reference case of the elastic backscatter signal from the three harmonics of the Nd:YAG transmitter at 1064, 532, and 355 nm (i.e., three backscatter but no extinction) with an assumption of 10% uncertainty in the optical coefficients. This error level is commonly used as the benchmark by the community but is still quite difficult to achieve.

The Raman lidar technique allows for additional optical data in the form of extinction coefficients at the nitrogen Raman-shifted wavelengths generated from laser transmitter output at 355 and 532 nm signals (at 387 and 607 nm respectively), which can be extrapolated to the elastic lidar wavelengths with the appropriate power law wavelength-dependent extinction models. However, the usefulness of the additional extinction data is not clear and seems to depend on whether the complex refractive index is known. It has been shown in a number of studies that a single extinction coefficient is necessary for stabilization when regularization is used, and that a single extinction is needed for PSD with a known complex index. To evaluate the information content in a given set of optical coefficients, using the confidence level \( C_{\text{level}} = 0.65 \), for each value of the parameters \((\bar{r}_{0}, \sigma_{0})\)
we calculate the set \( \Delta r_N, \Delta \sigma_N, \text{ and } \Delta A_N \) as the relevant performance metrics.

To illustrate the importance of proper definition of confidence bonds, the calculations are initially carried out by assuming that the optical coefficient ratios are uncorrelated. Figure 6 shows the performance metric for the case of three-wavelength backscatter lidar. It is immediately apparent that the parameter space can be divided into a region of good parameter retrieval and a region of poor parameter retrieval that occurs for distributions with simultaneously large mode radius and mode width. This is consistent with the general observation that large particle modes cannot be inverted unless the exact shape is known, and as illustrated, even then, the uncertainties grow very large.

Unfortunately, a practical problem that needs to be addressed in the graphical method is that finite LUT calculation windows bias the retrieval as the confidence region interacts with the edges of the computation window. Therefore only the portion of parameter space that is sufficiently far from the calculation window boundary has confidence regions totally contained in the window unclipped, while for parameter values near the boundary, the confidence region is clipped. However, in our case, significant clipping occurs mostly for PSDs, which are already determined to be difficult to retrieve (large particle modes), so it does not greatly affect the qualitative division of the parameter space.

While the three-backscatter optical coefficient set is capable of retrieving PSDs for a known index of refraction for the case of lognormal distributions for certain well-defined regions of parameter space as illustrated by Fig. 6(c), a natural extension is used to assess the usefulness of extra Raman extinction coefficients. It has already been shown with regularization methods\(^8,9\) that two extinction parameters can stabilize a solution in the presence of unknown refractive index, while when considering PSD retrieval with known refractive index, it was shown\(^7\) that a single extinction coefficient is sufficient for monomodal distributions.

To study the improvements in the retrieval with an increased number of optical coefficients and the effect of correlation in the optical coefficient ratios, we plot in Fig. 7 the enhancement factors as defined by

\[
E_{4,3} = \log_{10} \left( \frac{\left( \frac{\Delta A_N}{\Delta A_N'_{4,3}} \right)}{\left( \frac{\Delta A_N}{\Delta A_N'_{3,3}} \right)} \right) \text{ for } 3\beta \text{ versus } 3\beta + 1\alpha,
\]

\[
E_{5,4} = \log_{10} \left( \frac{\left( \frac{\Delta A_N}{\Delta A_N'_{5,4}} \right)}{\left( \frac{\Delta A_N}{\Delta A_N'_{4,3}} \right)} \right) \text{ for } 3\beta + 1\alpha \text{ versus } 3\beta + 2\alpha,
\]

first using uncorrelated bound estimates. From these definitions, positive enhancement factors result in improved retrieval uncertainty. In particular, we make the following observations that are valid where the refractive index is assumed known \textit{a priori}.

1. There is a definite region where \( E_{4,3} > 0 \), indicating that an additional extinction coefficient helps reduce parameter retrieval uncertainty [Fig. 7(a)].

2. However, since \( E_{5,4} < 0 \) for nearly all parameter values, we come to an anomalous result that suggests that more coefficients may lead to an increased uncertainty in parameter retrieval [Fig. 7(b)].

This observation is counterintuitive since adding extra optical coefficient ratios should reduce the retrieval uncertainty. This comes about because, as noted earlier, the above calculation was done assuming uncorrelated optical coefficient ratios. In this approximation, the magnitude of the uncertainty...
bound applied to each ratio is larger than it should be if statistical dependence is taken into account. To adjust for this affect, we can simply apply a correction factor
\[
\frac{f}{H_{2084}} \frac{C}{H_{2085}} \frac{R}{H_{2084}} \frac{corr}{H_{2084}} \frac{C}{H_{2085}} \frac{R}{H_{2086}} \frac{uncorr}{H_{2084}} \frac{C}{H_{2085}} \frac{R}{H_{2086}}
\]
to convert the uncorrelated confidence bounds for a given confidence level to the correct correlated bounds utilizing the results in Fig. 3. Examination of Fig. 3 shows that very little adjustment is needed for the case of three optical coefficient ratios (i.e., \( f \sim 1 \)). However, a significant adjustment of confidence bounds is needed when considering more than three optical coefficients. This correction has been verified by calculating the percentage of numerical inversions contained in the correlated confidence bounds for a variety of confidence levels and comparing with the solid curves in Fig. 3.

Applying this correction to the calculation of enhancement factors \( E_{4,3} \) and \( E_{5,4} \) to account for correlations leads to a much more reasonable quantitative comparison as illustrated in Fig. 8. Note that nearly all the anomalous negative enhancement regions have been eliminated. The final observations are that:

1. There is a definite region where \( E_{4,3} > 0 \), where an additional extinction coefficient helps reduce parameter retrieval uncertainty significantly [Fig. 8(a)].

2. Since \( E_{5,4} = 0 \) for most parameter values [Fig. 8(b)], we see that including a second extinction parameter does not significantly help in reducing the parameter retrieval for the most part except when considering a fine urban mode (e.g., \( r \approx 0.15 \mu m, 1.4 \leq \sigma \leq 2.0 \)), where there is a moderate enhancement going from one to two extinction parameters as seen in the expanded view of Fig. 9.

While the calculations above were carried out with a 10% uncertainty in the optical coefficients, the efficacy of the graphical technique for various uncertainty levels can be easily examined. This is illustrated in Fig. 10, where the values of the normalized maximum and minimum PSD parameters within the solution space (\( \sigma_N^{\text{max,min}} = \frac{\bar{r}}{r_0} \), \( \sigma^{\text{max,min}} = \frac{\sigma_{\text{max,min}}}{\sigma_0} \)) are plotted as a function of the noise uncertainty level. All results were obtained for
Figure 10(a), representing larger particle modes \( r_0 = 0.8, \sigma_0 = 1.8 \), shows clearly that the retrieval error rapidly increases for relatively small uncertainty levels, and no improvement occurs with the addition of extinction coefficients. For more moderate particle sizes \( r_0 = 0.5, \sigma_0 = 1.6 \), three backscatter coefficients alone will result in large errors for moderate uncertainty levels in the optical coefficients, while significant improvement is observed with the addition of a single extinction channel. In this case, the second extinction channel adds little improvement. Finally, for smaller particles that are better representative of the urban accumulation mode \( r_0 = 0.3, \sigma_0 = 1.4 \) the addition of either one or two extinction coefficients significantly improves the retrieval.

B. Bimodal Distributions

While the single lognormal mode is very amenable to graphical analysis, the most realistic aerosol particle size distribution models are bimodal size distributions that combine fine particulate (accumulation mode) aerosols with a coarse lognormal particle mode. This model is consistent with the global cluster analysis from a very extensive set of AERONET (NASA Aerosol Robotic Network) measurement records across the globe.\(^{19}\) In that study, the analysis of the AERONET data set yielded six distinct clusters described by the column optical (refractive index) and physical (particle size distribution) properties and were identified as desert dust, biomass burning, rural (background), industrial pollution, marine, and dirty pollution. Each cluster has a unique set of mode parameters, total volume, mean radius, GSD, and complex refractive index \( \nu_{f,c}, (r_{f,c}, \sigma_{f,c}, (m_r)_{f,c}, \nu_{c}) \) for both fine (f) and coarse (c) parameters,

\[
\frac{dV}{d \log(r)} = \frac{V_f}{2\pi \ln(\sigma_f)} \exp\left[ -\frac{\ln(r/r_f)}{2 \ln(\sigma_f)^2} \right] + \frac{V_c}{2\pi \ln(\sigma_c)} \exp\left[ -\frac{\ln(r/r_c)}{2 \ln(\sigma_c)^2} \right].
\]

One issue that would be of interest is the possibility
of separating monomodal distributions from true bi-
modal distributions. As mentioned in the introduction,
this problem has been studied by using extinction co-
efficients alone,\(^4\) where it was shown that if the coarse
mode is not too abundant and that the fine mode Ang-
strom coefficient is sufficiently large (to extract from
the coarse flat Angstrom coefficient), such a separation
is possible.

In Fig. 11, we examine the situation where both modes
in the bimodal distribution are fine modes with mode 1
given as \(r = 0.8, \sigma_0 = 1.8, m_r = 1.5, m_i = -0.02,\)
and mode 2 as \(r = 0.7, \sigma_0 = 1.4, m_r = 1.5, m_i =
-0.02\) with a 10\% uncertainty level. The mixing ratio
was designed so that both modes would have equal
optical depths of 532 nm. This situation is compara-
tible with the bimodal distributions considered in
Ref 7. In that work, although the bimodal distri-
bution was successfully inverted, a larger optical data
set \((6\beta + 2\alpha)\) was needed. Applying the graphical
technique to optical coefficients \((3\beta + 2\alpha)\) derived
from the bimodal distribution resulted in a para-
ter retrieval space whose mean radius is between the
individual modes and whose GSD parameter is sig-
ificantly larger as would be expected by a smeared
out bimodal distribution. We conclude that it is not
possible to distinguish an underlying fine particle bi-
modal distribution by using the graphical technique
within the five optical coefficients \((3\beta + 2\alpha)\) scheme,
and more optical backscatter channels are needed.

However, the situation changes drastically if the
two modes are sufficiently separated (i.e., coarse and
fine mode). To see this, we chose four Aeronet PSD's
retrieved over New York City, which are illustrated
in Fig. 12. Figures 12(a) and 12(b) represent near-
single-mode distributions, while Figs. 12(c) and 12(d)
are highly bimodal examples. Instead of using the
actual retrieved refractive index that has wavelength
dependencies that cannot be implemented within the
context of the graphical technique, we processed all
retrivals with a complex refractive index of
\(m_r = 1.5, m_i = -0.02\). Figure 13 shows the results of the
graphical technique applied to optical coefficients de-
rived from the four PSDs with a 10\% error level. We
note that for the near-single-mode PSDs all three
lidar schemes \((3\beta + 2\alpha), (3\beta + 1\alpha),\) and \((3\beta)\) produce
reasonable results. However, with the strongly bi-
modal distributions, \((3\beta)\) produces a false monomodal
distribution in all cases, \((3\beta + 1\alpha)\) produces a false
monomodal distribution for the more moderate
course mode case, [Fig. 13(d)], and \((3\beta + 2\alpha)\) does not
produce any false monomodal distributions. This sug-

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suggests that the \((3\beta + 2\alpha)\) scheme can potentially flag bimodal distributions.

While the AERONET bimodal distributions were processed with the original \textit{a priori} refractive index of \(m_r = 1.5, m_i = -0.02\), it might be argued that false monomodal distributions may be retrieved if an incorrect refractive index is used. This has been investigated numerically by varying the refractive index of the retrieval LUT and observing that no choice in the complex refractive index will retrieve a false monomodal PSD for the \((3\beta + 2\alpha)\) set. This result is in accord with the observation that the perturbations in the optical coefficient spectra due to a coarse particle mode are wavelength insensitive, but changes in the refractive index would result in very large modifications in the optical coefficient spectra. Therefore refractive index changes cannot reproduce the modifications to the optical coefficient spectra due to an additional coarse mode.

4. Summary

The graphical technique employing lognormal particle size distribution parameters with an assumed refractive index is applied to the intercomparison of typical three-wavelength backscatter lidar (1064, 532, and 355 nm) and four and five wavelength systems augmented by additional nitrogen Raman channels that can yield estimates of extinction coefficients at 532 and 355 nm. We have demonstrated that to obtain correct quantitative comparisons, the correlations between optical coefficient ratios used in graphical technique must be included. This is accomplished by refining the confidence bounds to account for such correlations. We have also extended the graphical technique by introducing a procedure for the retrieval of particle number density and its uncertainty. The results of this intercomparison indicate that significant improvement in retrieval error results from the addition of a single extinction channel. The addition of a second extinction channel seems to mainly affect small particle (accumulation) modes. Finally, the application of the graphical technique to bimodal distributions can result in false monomodal solutions. If the coarse mode fraction is not large the technique can isolate and retrieve the fine mode parameter. It is demonstrated that when the coarse mode fraction is significant, the technique can potentially flag the bimodal distributions when both Raman optical extinction channels are available, by yielding no false single-mode retrieval. While we have not focused in this paper on simultaneous...
PSD parameter and complex refractive index retrieval, the same approach described here can be used to study these problems. In particular, since our confidence bound values are independent of the underlying parameterization of the aerosol PSD, the same numeric confidence bounds may be applied. Further work is underway to consider the correct solution space within the resultant 4D parameter space \( [\bar{r}, \alpha, m_r, m_i] \) by simultaneously satisfying all optical coefficient ratio bounds.

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