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RETRIEVAL OF AEROSOL SIZE DISTRIBUTION MOMENTS WITH MULTI-WAVELENGTH LIDAR

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INTRODUCTION

Stratospheric aerosol characterization is needed e.g. to quantify heterogeneous chemical reactions[1], to correct methods based on differential absorption[2] (DIAL, SAOZ, TOMS), and to quantify radiative transfer. Among others, quantities of interest are[3]: composition, shape, number density (i.e. size distribution), surface density, and spectral scattering characteristics. The most common methods for determining the particle size distribution (PSD) out of the lidar signal consist of fitting to the data either predefined functions[4], or the PSD represented as a histogram[5]. The former method is affected by "a priori" assumptions on the shape of the PSD, while the latter is sentitive to data errors.

The moments M_i of a PSD $n(r)$, r being the particle radius, are defined as:

$$M_i = \int_0^\infty r^i n(r) dr \qquad (1)$$

M_o is the integrated number density, $\mu=M_1/M_0$ the mean radius, $\sigma^2=M_2/M_0-\mu^2$ the standard deviation, πM_2 the surface density, $\pi M_2/M_0$ the mean surface, etc. Compared with searching for the dependance of the PSD on particle radius, integrated quantities such as its moments have the following advantages: no "a priori" assumption on the PSD is required, such as its shape or mean radius; they are often the significant quantities (see above); they allow for computation of scattering characteristics at other than the lidar wavelenghts (which is needed for radiative transfer calculations). The drawback is that from the PSD moments its functional form cannot be determined, and as a consequence e.g. multi-mode PSD can hardly be discriminated from single-mode PSD.

Twomey and Howell[6] first presented theoretically a method for calculating PSD moments from scattering characteristics. Modification of this method was applied by Livingston and Russel[7] to extinction measurements by SAGE II sun photometer. The present method proposes the following extensions of Livingston method: no need of preliminary fit of predefined functions; computation of M_o, μ and σ (and not related quantities such as effective radius or effective variance, defined as ratios of M_2 and M_3); eigenvalue analysis[6] of the propagation of errors.

The basic assumptions in this work are: spherical particles, single scattering, known wavelength-dependent particle refractive index. These apply well e.g. in the case of stratospheric aerosol probing (see below).

DESCRIPTION OF RETRIEVAL METHOD

It is assumed that n extinction and n backscattering coefficients have been computed from the lidar signal[9] (which requires e.g. assumption on extinction-to-backscatter ratio), n being the number of lidar wavelengths. Out of these, an optimal set of m coefficients is determined using eigenvalue technique[6,10] (the latter allows prediction of information content of a set of measurements with given level of errors). After expansion[6,7] of each function r^i, $0 \le i \le m-1$, on the Mie extinction and/or backscattering cross sections $C(r,\lambda_t)$[11], integration on radius from 0 to ∞ yields the size distribution moments M_i:

$$M_i = \sum_{j=1}^{m} a_{ij} \int_0^\infty C(\lambda_j, r)\, n(r)\, dr + E_i^{(M,t)} = \sum_{j=1}^{m} a_{ij}\, \alpha(\lambda_j) + E_i^{(M,t)} \quad ,0 \le i \le m-1 \qquad (2)$$

where $\alpha(\lambda_j)$ is the backscattering or extinction coefficient corresponding to wavelength λ_j, and $E_i^{(M,t)}$ is the error caused by truncation of the serie after m terms. Details about computation of the expansion coefficients a_{ij} are found in ref. 7. It is seen from (1) that in order to minimize the truncation error $E_i^{(M,t)}$, approximation of r^i by the $C(r,\lambda_l)$ is to be performed in the vicinity of the peak values of $n(r)$. This makes the system of equations (2) non-linear through dependence of the a_{ij} on $n(r)$, and hence on the M_i. Ideally, the a_{ij} should depend on all the M_i sought. However, in order to simplify the resolution, dependence only on mean radius μ and standard deviation σ has been considered. Consequently, a non-linear system of two equations is to be solved for μ and σ:

$$(\mu^2+\sigma^2)\sum_{j=1}^{m} a_{0j}\,\alpha(\lambda_j) = \sum_{j=1}^{m} a_{2j}\,\alpha(\lambda_j)$$
$$(\mu^2+\sigma^2)\sum_{j=1}^{m} a_{1j}\,\alpha(\lambda_j) = \mu\sum_{j=1}^{m} a_{2j}\,\alpha(\lambda_j) \qquad (3)$$

The remaining moments (M_o and M_i, i=3,...,m-1) are calculated from (2).

System of equations (3) is solved graphically and considering errors on the $\alpha(\lambda_j)$: at each point of a grid (μ,σ), left (L) and right (R) side of each equation is computed, as well as errors ΔL and ΔR; these are calculated by standard error propagation techniques[12] (i.e. propagation of error by the coefficients a_{ij}) and estimation of the trunction error $E_i^{(M,t)}$. The point (μ,σ) is considered as part of the solution if $L\pm\Delta L$ meets $R\pm\Delta R$. Most probable solution is obtained by setting errors on the input $\alpha(\lambda_j)$ to 0, whereas its uncertainty is determined from extension of the cluster of solution points. It has to be noticed however that the concept of *most probable solution* is delicate to handle in this context; on the other hand, the cluster of solution points defines all *possible solutions*, and therefore has clearer interpretation.

The solution is constrained using expansion of the $C(r,\lambda_l)$ on the r^i, $0\leq i\leq m-1$, (instead of the reverse, equ. (1)):

$$\alpha(\lambda_i) = \sum_{j=0}^{m-1} b_{ij} M_j + E_i^{(\alpha,t)}, \quad 1\leq i\leq n \qquad (4)$$

in which propagation of errors (by expansion coefficients b_{ij}) and truncation error $E_i^{(\alpha,t)}$ are generally lower than in (2). The same way as for (3), solution is rejected if left and right side of (4), with respective errors, do not meet. With the help of this system of equations, all the extinction and backscattering coefficients retrieved from the lidar signal have been used (instead of only optimal set, needed for (3)). The obtained solution is eventually constrained by general mathematical properties concerning distribution moments[13].

In principle, equation (4) can also be used to calculate the extinction coefficient (with error) at other than the lidar wavelengths, which is useful for radiative transfer calculations, as well as for correction of the aerosol effect on differential absorption instruments.

It has to be stressed that no arbitrary regularization constraint (in term of ill-posedness of the inverse scattering problem[14]) has been used. This way, *all solutions (in term of size distribution moments) of the inverse scattering problem have been pointed out*, by the way defining the information content of the measurements with their error. However, the delicate point of this retrieval method resides in the estimation of propagation and (especially) truncation errors in (3) and (4), as well as errors on the extinction and backscattering coefficients themselves.

RETRIEVALS FOR SIMULATED MEASUREMENTS

The retrieval method has been first tested on simulated measurements calculated from two different PSD. The first is a Junge-type distribution taken out of standard set of stratospheric aerosol PSD[15], while the second is a two-mode lognormal PSD retrieved from Pinatubo stratospheric sounding with in situ particle counter[16]. Wavelength-set (0.355, 0.532, 0.750, 0.850 μm) of the lidar based at Sodankylä (67ºN, 26ºE) during EASOE were considered[4].

Eigenvalue analysis indicates that the set of 4 backscattering coefficients is best suited for resolution of system of equations (3). The set of 4 extinction coefficients is characterized by lower propagation and truncation errors, but resulted in dispatched clusters of solution points, indicating that more than one solution type is compatible with the measurements. Combination of more than 4 ext./back. coefficients resulted in much higher propagation errors. Such an evaluation of the information content can be performed without assuming any particuliar PSD.

Fig. 1 shows that "most probable" inferred values are within 25% for a relatively narrow PSD as the Junge-type (σ/μ=39%), whereas it can reach discrepencies as high as 50% in the case of the broader lognormal distribution (σ/μ=86%). In fact, generally speaking, the broader the distribution, the broader also will be the set of possible solutions. Uncertainties on the inferred values (not shown on the figure) can be quite large (typically 50% of the true value, and sometimes exceeding 100%). This is explained partly by the way errors propagate, as well as the truncation error (both connected to the particuliar wavelength set), but also by the fact that no assumption was made on the PSD, which consequently extends the solution set compared to situations in which the PSD is constrained.

RETRIEVALS FOR 4-WAVELENGTH LIDAR DATA

Retrieval method was then applied to data of the Sodankylä lidar on 13th Feb. 1993. This date was particularly chosen because measurements with in situ particle counter by T. Deshler[16] were performed the same day, though situated some distance away (Kiruna, Sweden). Lidar depolarization measurements[4] indicate that the aerosol was composed mostly of spherical particles above 17 km, while low optical depth[4] (0.1-0.3) suggests that multiple scattering is negligible. Retrieval of the extinction and backscattering coefficients with the so-called Klett algorithm is described in ref. 4 or 17. Error of 10% was considered on both types of coefficients at all the wavelengths. The retrieval routine assumed sulfuric acid in water; calculation of wavelength-dependent refractive indices, out of water vapor and temperature profiles obtained by sonde measurements, is desribed in ref. 17.

Fig.2 shows results from the method of moments, the algorithm of Stein et al.[4] which consists of fitting a lognormal distribution on the backscatter profile, and the in situ measurements of Deshler (out of two-mode lognormal fit to the counter data). It is noticed that for all the represented parameters, excepting the standard deviation, there is good agreement between method of moments and Stein's algorithm. This fact is not obvious "a priori", because the latter algorithm assumes a monomodal distribution. Such an agreement is not met with Deshler measurements, especially at lower altitudes. For the case of number density, the discrepency can be attributed to the fact that different air masses were probed[17]. Also, sulfuric acid assumption may no longer be valid in the lower stratosphere. As for the standard deviation, amplification of error from lognormal parameters (i.e. for Stein and Deshler results) should be taken into account.

Error bars on results of the method of moments are rather large (20 to 50%, sometimes more than 100% for the number density). Notice that these represent also all possible moments compatible with the measurements and their error. This is coherent with the fact that almost all uncertainties meet Deshler or Stein results.

CONCLUDING REMARKS

As shown when applied to synthetic measurements of model or measured particle size distributions (PSD) with up to 20% error, the method of moments retrieves the correct values with precision of ~25% to 50%, depending on the broadness of the distribution (e.g. monomodal or bimodal). However, in most of the cases it successfully predicts the uncertainty in inferred solution. The latter seems confirmed when applied to lidar measurements, in that the uncertainty in solutions agree with in situ measurements and another inversion method of the lidar profile.

As a conclusion, the present method seems most suited for cases when uncertainty about the functional form of the PSD makes the usual predefined function fitting algorithms suspicious. In that case it provides, above all, bounds for the PSD moments. The latter can e.g. further be used as constraints for other inversion techniques.

Future development comprises extensive comparison with classical methods (which usually require "a priori" assumptions), in order to determine the situations for which a method with no assumption on the PSD is needed. Such en effort is in particuliar scheduled in the frame of MOANA project (Measurement and modelling of Ozone and Aerosols in the Northern Atmosphere), as part of SESAME campaign.

REFERENCES

1. D. R. Hanson, A. R. Ravishankara,, J. Geophys. Res., **96**, 17307 (1991).
2. W. Steinbrecht, A. I. Carswell, 16th International Laser Radar Conference, NASA Conference Publication 3158 (1992).
3. R. M. Measures, *Laser Remote Sensing*, Wiley, 1984, chap. 9.1.6.
4. B. Stein, M. Del Guasta, J. Kolenda, M. Morandi, P. Rairoux, L. Stefanutti, J. P. Wolf, L. Wöste, Geophys. Res. Let., EASOE special issue, in press.
5. H. Müller, H. Quenzel, Appl. Optics, **24**, 648 (1985).
6. S. Twomey, H. B. Howell, Appl. optics, **6**, 2125 (1967).

7. J. M. Livingston, P. B. Russell, Jou. Geophys. Res., **94**, 8425 (1989).
9. J. A. Reagan, M. P. Mc Cormick, J. D. Spinhirne, Proceedings of the IEEE, **77**, 433 (1989).
10. G. Viera, M. A. Box, Appl. Optics, **26**, 1312 (1987).
11. C. F. Bohren, D. R.Huffman, *Absorption and Scattering of Light by Small Particles*, Wiley, 1983, chapter 3.4.
12. P. R. Bevington, *Data Reduction and Error Analysis for the Physical Sciences*, McGraw-Hill, New-York (1969).
13. J. A. Shoat, J. D. Tamarkin, *The Problem of Moments*, American Mathematical Society, Providence, Rhode Island 02904 (1943).
14. S. Twomey, *Introduction to the Mathematics of Inversion in Remote Sensing and Indirect Measurements*, Elsevier (1977).
15. P. B. Russell, T. J. Swissler, M. P. McCormick, W. P. Chu, J. M. Livingston, T. J. Pepin, Jou. Atmo. Sci., **38**, 1279 (1981).
16. T. Deshler, Geophys. Res. Let., EASOE special issue, in press
17. B. Stein, PhD dissertation, Free University of Berlin, Germany (1994).

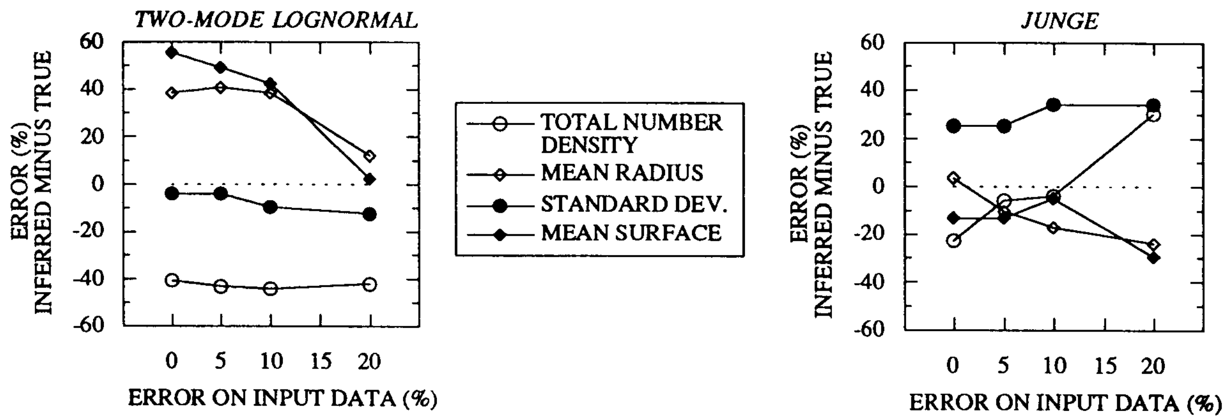

Fig.1. Error in retrieved moments for simulated measurements, calculated out of model size distribution[15] (Junge), or measured[16] (two-mode lognormal).

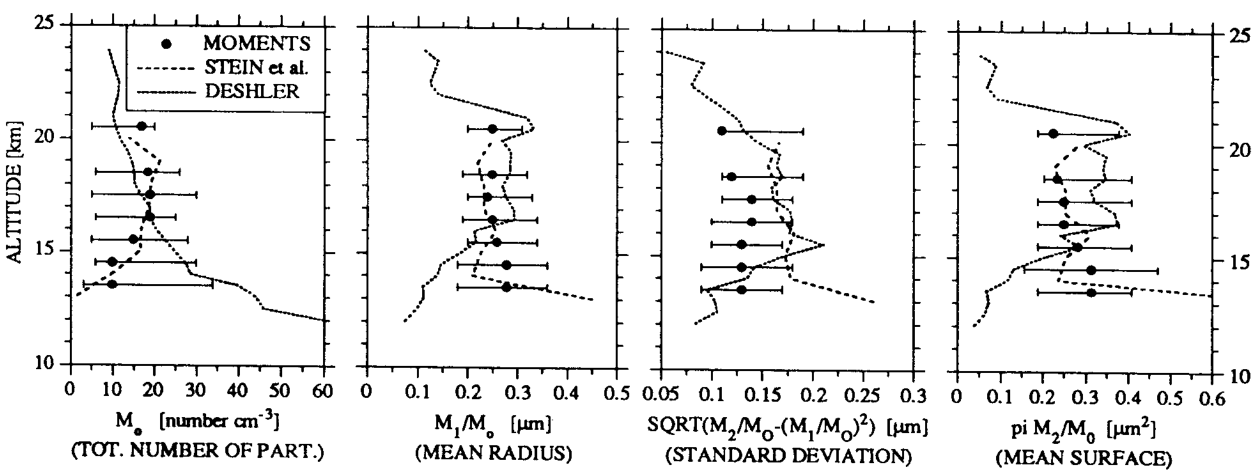

Fig. 2. Retrieval method applied to four-wavelength lidar data (13 Feb. 1992, Sodankylä, EASOE), as compared with in situ particle counter measurements (Deshler[16]), and another method for inferring size distributions from the lidar profile (Stein et al.[4]).