

Aerosol size distribution retrieved from optical depth measurements in Tamanrasset and Blida

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(reçu le 30 Mars 2009 - accepté le 30 Mars 2012)

Abstract - *Since we are often faced with limited, insufficient or contaminated observations in remote sensing. We will employ a theoretical technique for the retrieval of the particle size distribution. In this study, the columnar aerosol size distributions have been inferred by numerically inverting particulate optical depth measurements obtained in Blida and Tamanrasset from AERONET database at wavelengths region between 0.340 - 1.640 μm . Our results are in good agreement with those found by other authors and the correlation between the aerosol optical thickness measurements and the reproduced AOTs by the inverted columnar aerosol size distribution is very good with a correlation coefficient more than 0.97.*

Résumé - *Car nous sommes souvent confrontés à des observations limitées, insuffisantes ou contaminées dans la télédétection. Nous allons employer une technique théorique pour l'extraction de la distribution granulométrique des particules. Dans cette étude, les distributions en taille des aérosols intégrés sur la colonne atmosphérique ont été obtenues par l'inversion numérique des mesures de l'épaisseur optique des particules établis à Blida et Tamanrasset à partir des données d'AERONET dans la région spectrale entre 0.340 - 1.640 μm . Nos résultats sont en bon accord avec ceux trouvés par d'autres auteurs et la corrélation entre les mesures d'épaisseur optique des aérosols et l'AOTs reproduites par la distribution en taille inversé est très bonne avec un coefficient de corrélation plus de 0.97.*

Keywords: Aerosol properties - AERONET - Size distribution - Inversion methods.

1. INTRODUCTION

Atmospheric aerosols are suspensions of small solid or liquid particles in the atmosphere, which plays an important role in atmospheric and environmental research since they take part in many physical and chemical processes in the atmosphere.

Because of the wide variety of sources, the properties of atmospheric aerosol particles, such as size, shape, chemical composition, and optical thickness, may be heterogeneous, and their temporal and spatial variation can be very large [1-5].

Knowledge of both aerosol optical properties (extinction, scattering cross section, phase function, and single-scattering albedo) and microphysical aerosol properties (particle size distribution function and the complex refractive index) is essential for the determination of the effect of atmospheric aerosols on the climate and for the control of the air quality [6].

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Meanwhile, aerosol particle size distribution and aerosol optical thickness are two important properties for characterizing the aerosol particles and the correction of the atmosphere [7].

While aerosol size distributions can be measured in-situ, in different size ranges, using a variety of instruments [8], they can also be derived from measured aerosol optical properties, such as the spectral aerosol optical depth [9], or backscattering [10], or a combination of angular scattering intensities and spectral optical depth [11, 12].

Various techniques have been developed for recovering the particle size distribution function $n(r)$, for example direct regularization methods [7, 13-17] and iterative methods [18-23].

This study deals only with the inversion of spectral aerosol optical thickness measurements, and its aim is to determine aerosol size distributions on Blida (36N, 2E) and Tamanrasset (22.78S, 5.53E) northern and southern Algeria respectively during 2006. To accomplish this, we use the inversion algorithm of King *et al.* [9].

2 THE INVERSION PROBLEM

Since the relationship between the size of atmospheric aerosol particles and the wavelength dependence of the extinction coefficient was first suggested by Ångström in 1929, the size distribution began to be retrieved by extinction measurements.

First, Ångström inferred that the parameters of a Junge size distribution could be obtained from the aerosol optical thickness (AOT) at multiple wavelengths, and he obtained the useful Ångström empirical formula of Junge size distribution $\tau_{ae} = \beta \times \lambda^{-\alpha}$, where τ_{ae} is the measured AOT, β is the turbidity coefficient, and α is the Ångström exponent reflecting the aerosol size distribution [24].

Assuming that aerosols are homogenous spheres, the relationship between the aerosol size distribution and AOT τ_{ae} can be written as Fredholm integral equations of the first kind,

$$\tau_{ae}(\lambda) = \int_0^{\infty} \pi r^2 \times Q_{ext}(r, \lambda, m) \times n_c(r) \times dr \quad (1)$$

where r is the particle radius, λ the wavelength of incident illumination, m the complex refractive index of the aerosol particles, $Q_{ext}(r, \lambda, m)$, the extinction efficiency factor from Mie theory, and $n_c(r)$ the columnar aerosol size distribution, that is, the number of particles per unit area per unit radius interval in a vertical column through the atmosphere.

Various methods that have been proposed for solving this equation include those in which an assumption about the analytical form of the size distribution to be retrieved is made [25, 26], as well as methods that make no assumption about the size distribution shape [9, 27].

3 METHOD OF SOLUTION

In this article, the constrained linear inversion is used to solving the equation (1); this method was introduced by Twomey [1] and developed by King *et al.* [9], to

determine $n_c(r)$ from spectral measurements of $\tau_{ae}(\lambda)$, let $n_c(r) = h(r) \times f(r)$, where $h(r)$ is rapidly varying function of r , while $f(r)$ is more slowly varying. So, the equation (1) becomes:

$$\begin{aligned} \tau_{ae}(\lambda) &= \int_{r_a}^{r_b} \pi r^2 \times Q_{ext}(r, \lambda, m) \times h(r) \times f(r) \times dr \\ &= \sum_{j=1}^q \int_{r_j}^{r_{j+1}} \pi r^2 \times Q_{ext}(r, \lambda, m) \times h(r) \times f(r) \times dr \end{aligned} \tag{2}$$

Here $r_1 = r_a$ and $r_b = r_{q+1}$ and r_1, r_2, \dots, r_{q+1} are the $q+1$ boundaries of q coarse intervals of integration. If $f(r)$ is constant within each coarse interval. Thus the Fredholm equation is replaced by simultaneous equations

$$\bar{g} = \hat{A} \bar{f} + \bar{\varepsilon} \tag{3}$$

where,

$$\begin{aligned} g_i &= \tau_{ae}(\lambda_i) & i &= 1, 2, \dots, p \\ A_{ij} &= \int_{r_j}^{r_{j+1}} \pi r^2 \times Q_{ext}(r, \lambda_i, m) \times h(r) \times dr & j &= 1, 2, \dots, p \\ f_j &= f(\bar{r}_j) \end{aligned} \tag{4}$$

The elements ε_i of the unknown vector $\bar{\varepsilon}$ represent the deviations between measurement (g_i) and theoretical estimate ($\sum_j A_{ij} \times f_j$). These deviations arise from measurement and quadrature (integration) errors and due to uncertainties of the kernel function $\pi r^2 \times Q_{ext}(r, \lambda, m)$.

\bar{r}_j are the midpoints of coarse intervals. Note, the solution of equation (3) is obtained in logarithmic scale with respect to particle radius r .

If $h(r)$ takes the form of a Junge size distribution [28],

$$h(r) = r^{-(v^*+1)}$$

With this substitution, King *et al.* [9] apply a quadrature method and a minimization procedure leading to the solution vector

$$f = \left(A^T \times C^{-1} \times A + \gamma H \right)^{-1} \times A^T \times C^{-1} \times g \tag{5}$$

where C is the measurement covariance matrix; γ is a nonnegative Lagrange multiplier (choice of the Lagrange multiplier is discussed by King [29]); H is a smoothing matrix; A is the matrix representation of the kernel function.

The smoothing matrix H is defined by Twomey [30] as:

$$H = \begin{bmatrix} 1 & -2 & 1 & 0 & 0 & & \\ -2 & 5 & -4 & 1 & 0 & 0 & \\ 1 & -4 & 6 & -4 & 1 & 0 & \\ 0 & 1 & -4 & 6 & -4 & 1 & 0 \\ & & & \ddots & & & \\ & & & & \ddots & & \\ & & & & & 0 & 1 & -4 & 5 & -2 \\ & & & & & & 0 & 1 & -2 & 1 \end{bmatrix} \quad (6)$$

The measurement covariance matrix C is diagonal with elements given by $C_{ij} = \sigma_{\tau_M}^2 \times (\lambda_i) \times \delta_{ij}$, where δ_{ij} is Kronecker delta function [31].

Two important advantages can result from the assumption $n_c(r) = h(r) \times f(r)$. Firstly, when $h(r)$ represents size distribution $n_c(r)$ exactly, the solution vector \vec{f} will have all components equal to one. Secondly, the smoothing constraint guaranties minimum curvature of \vec{f} on a linear scale and it works better when $f_j = f(\bar{r}_j)$ are almost constants.

The columnar size distribution $n_c(r)$ varies over some orders of magnitude and has an explicitly large curvature, thus it is difficult to constraint.

We want to retrieve columnar aerosol size distribution $n_c(r)$ over the radii interval $[r_a, r_b]$. This is a typical inverseproblem and we can solve it using an iteration process. This means we have to start with some zero-approximation $h^{(0)}(r)$ of the rapidly varying multiplier in $n_c(r) = h(r) \times f(r)$ and use it to evaluate a first-order approximation of solution vector $\vec{f}^{(1)}$ with aid of (5).

Then we have to utilize $\vec{f}^{(1)}$ and obtain some reasonable first order approximation $h^{(1)}(r)$, which better represents the size distribution than the initially assumed weighting function. The first-order weighting function is then substituted back into (4) from which a second-order $\vec{f}^{(2)}$ is obtained through (5). This iterative procedure is continued until a stable result is obtained [32].

4. CLASSIFICATION OF EXPERIMENTAL DATA AND EXPECTED SOLUTION TYPE

In most cases the columnar size distributions can be classified in terms of three different types of distributions, although gradations between two different types are occasionally observed making this classification somewhat arbitrary [9, 29].

The Figure 1 illustrate the classification of the measured AOTs and the corresponding expected solution types I, II and III of distributions in the same figures 1.-a-, -b- and -c- respectively.

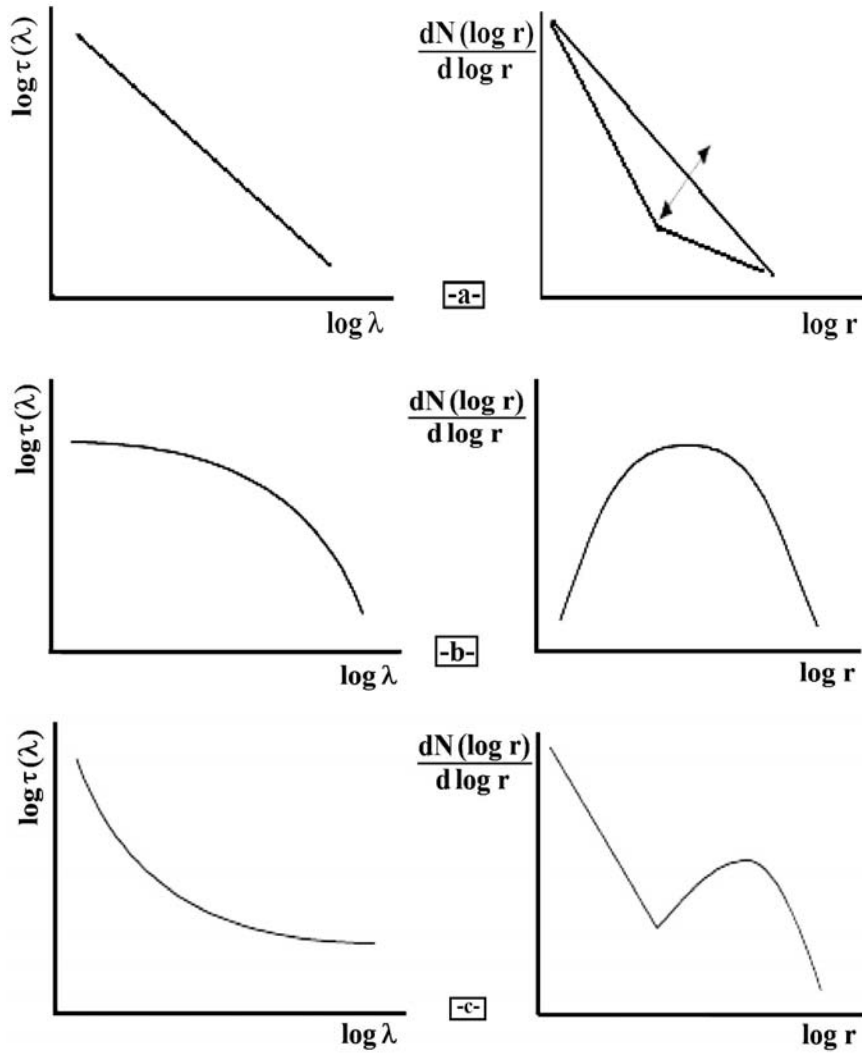


Fig. 1: Measured AOTs and corresponding columnar size distributions
 -a- Type I; -b- Type II; -c- Type III [9]

5. RESULTS

The method for determining the columnar aerosol size distribution $n_c(r)$ described in the section 3 has been carried out at Tamanrasset and Blida since 2006 from aerosol optical depth measurements by AERONET (Aerosol Robotic Network) at five and eight different wavelengths ranging between 0.340 and 1.640 μm .

In our study, all inversions were performed assuming the complex refractive index of the aerosol particles was wavelength and size independent and given by $m = 1.45 - 0.00i$ as in King *et al.* [9].

In lieu of $n_c(r)$ or, equivalently, dN/dr , the size distribution results are presented in terms of $dN/d\log r$, representing the number of particles per unit area per unit log radius interval in a vertical column through the atmosphere.

5.1 In Tamanrasset

From the inspection of results for 192 different days at Tamanrasset, we derive three examples:

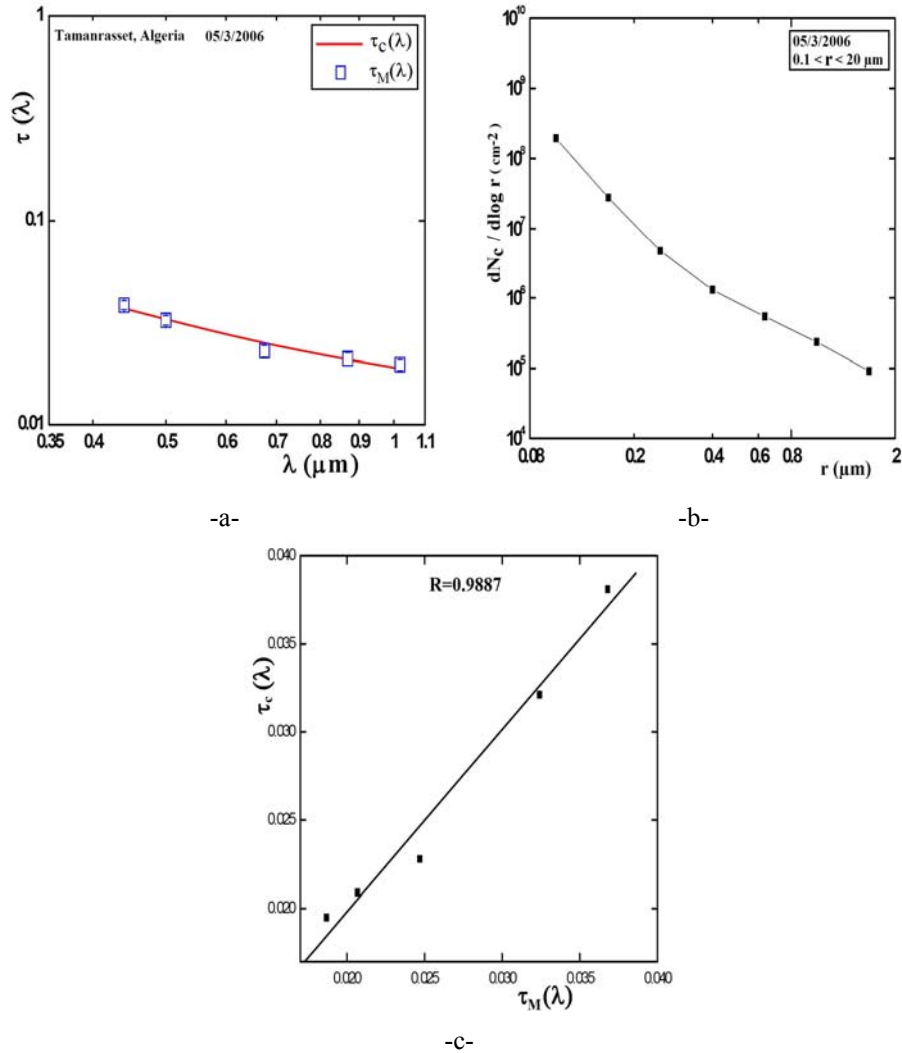


Fig. 2: Measured and retrieved AOT and corresponding columnar size distribution for 05 Mars 2006, (-a-, -b- and -c-)

Fig. 2 illustrates the spectral optical depth measurements for 05 Mars 2006, corresponding size distribution and the linear fit between optical depth measurements

and calculations, this day represents an example of the first type (Type I) for which the measured AOTs nearly follow Ångström's formula given by equation,

$$\tau_M(\lambda) = \beta \times \lambda^{-\alpha}$$

The observed Mie optical depths and corresponding standard deviations are shown in the left portion of the figure while the size distributions obtained by inverting these data is shown in the right portion.

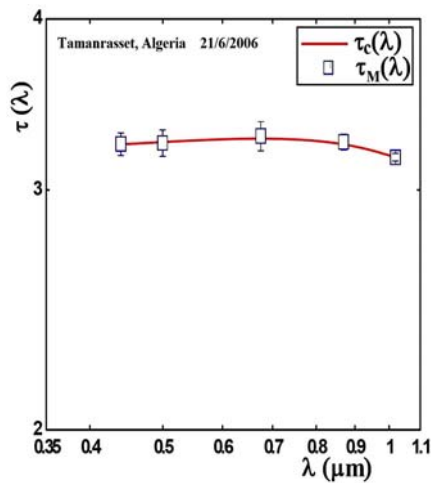
$\tau_c(\lambda)$ in the left portion indicates how the inverted size distributions are able to reproduce the AOTs measurements (i.e., the direct problem $g = A \times f$). The aerosol size distributions illustrated in the same figure can be best described as constructing Junge or two slopes type of distributions.

On the same figure, we consider the linear fit between $\tau_M(\lambda)$ and $\tau_c(\lambda)$, we see that the correlation is good with a correlation coefficient $R = 0.9887$.

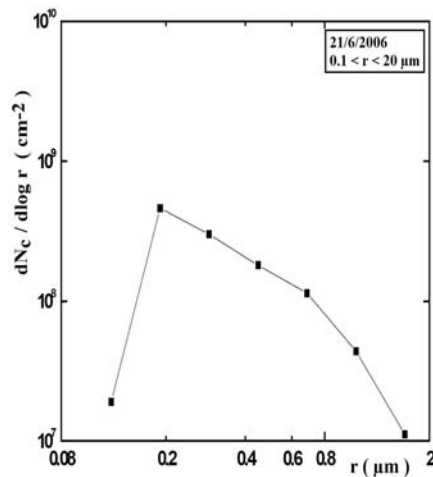
For the data of 21 June, when measured AOTs exhibit small negative curvature (Fig.3) the solutions tend to be monodisperse (Type II). This is not unexpected because the tendency for negative curvature suggests an absence of both small and large particles.

The optical depth measurements which produce this second class of distributions are typically very large. An important modification is the case when experimental data constitute two overlapping monodisperse distributions (we can see this case on Blida results presented below).

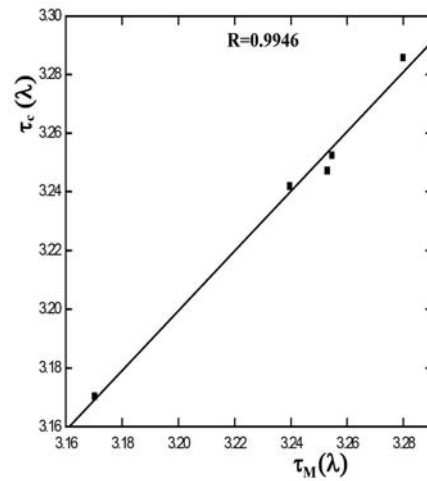
For the correlation between the observed Mie optical depths $\tau_M(\lambda)$ and the reproduced AOTs $\tau_c(\lambda)$ is always good and $R = 0.9946$.



-a-



-b-

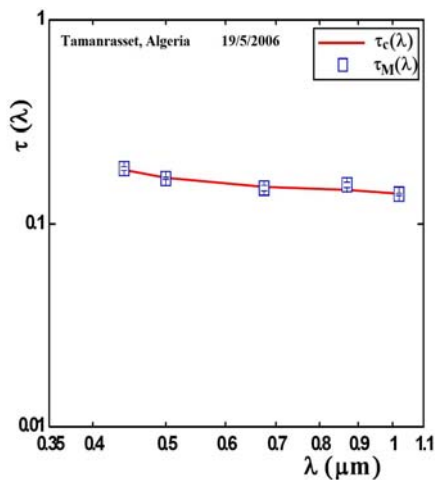


-c-

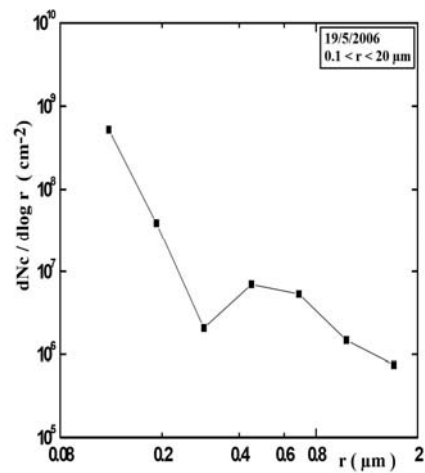
Fig. 3: Measured and retrieved AOT and corresponding columnar size distribution for 21 June 2006, (-a-, -b- and -c-)

The most interesting distribution type is one for which the AOTs intermediate between these of type I and type II. In this case $\tau_M(\lambda)$ tends generally to have positive curvature.

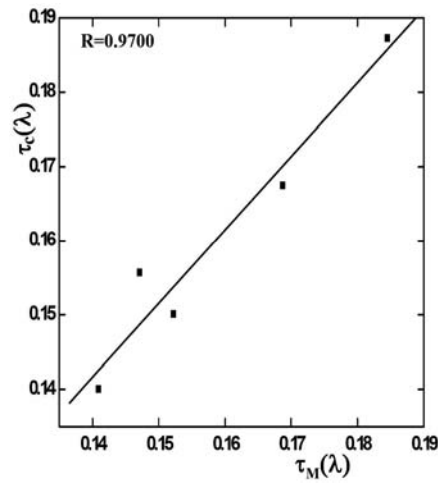
An example of this type (Type III) is illustrated in Fig. 4 for the data of 19 May. The solution is usually a bi-modal distribution and the correlation coefficient between the observed and the reproduced AOTs is $R = 0.9700$.



-a-



-b-



-c-

Fig. 4: Measured and retrieved AOT and corresponding columnar size distribution for 19 May 2006, (-a-, -b- and -c-)

5.2 In Blida

The inspection of plots of $\log \tau(\lambda)$ vs. $\log \tau_M(\lambda)$ (Fig. 5) reveals the presence of at least two kinds of aerosols with possible bi-modal and Junge or tow slopes distributions because plots on May to November have positive curvature and all other curves nearly follow Ångström’s formula. The retrieved columnar number distributions (Fig. 5) are quite interesting.

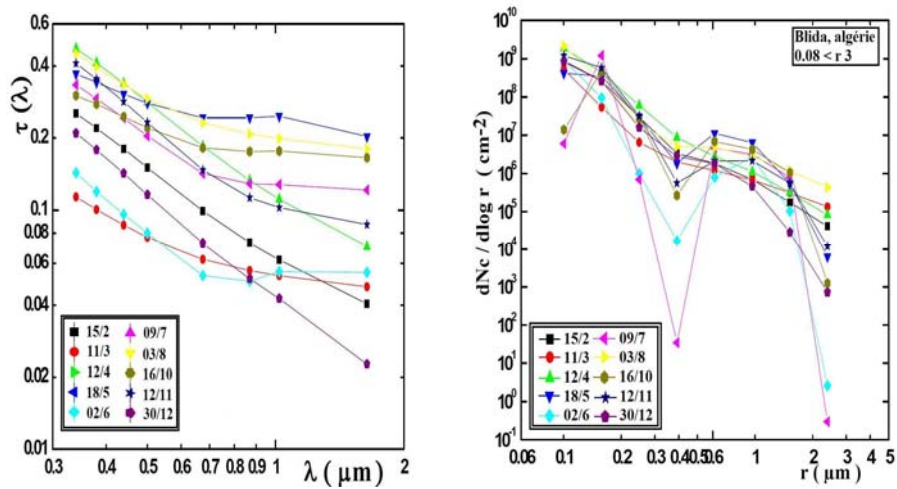


Fig. 5: Measured and retrieved AOT and corresponding columnar size distribution during 2006 in Blida

For 15 February, 11 March, 12 April and 30 December are constituted by aerosols with quite similar properties and distributions are Junge-type. The 18 May, 02 June, 09 July, 16 October and 12 November aerosols ensemble has all bi-modal distributions with a large amount of small particles, deep minimum at $0.3870 \mu\text{m}$ and maximum at $0.9718 \mu\text{m}$.

Distributions of 03 August have similar but smoother shape – minimum at $0.3870 \mu\text{m}$ is much shallower and about six orders more particles are present at largest radii compared with 09 July.

The data case of $\tau(\lambda)M$ for 18 May has small negative curvature at the four longest wavelengths, and the corresponding size distribution could equally well be categorized as a type II (monodisperse) distribution composed by two monodisperse which could be fine ash and water droplets, thus making classification according to three distinct types somewhat arbitrary [9].

6. CONCLUSION

In this paper, we investigate linear optimization method for the solution of the atmospheric aerosol particle size distribution function retrieval problem from spectral measurements of the particulate (Mie) optical depth at wavelengths region between 0.340 and $1.640 \mu\text{m}$.

We first described this method of solution developed by King in 1978 and classified experimental data and expected solution type. Then, we applied the proposed method on Tamanrasset site southern Algeria for 192 days during 2006.

When, the columnar aerosol size distributions have been determined from data delivered by AERONET network at five wavelengths 0.440 , 0.500 , 0.675 , 0.870 and $1.020 \mu\text{m}$. The optical depth measurements and corresponding aerosol size distributions are illustrated in Fig. 2-4 for a few of these days. Our results are in agreements with those found by King in Tucson, Arizona.

His results generally indicate (at least for $r \geq 0.1 \mu\text{m}$) that the aerosol size distribution on a particular day can be represented either as a Junge distribution (type I), a relatively monodisperse distribution such as a log-normal or gamma distribution (type II), or as a two-component system consisting of a combination of both of these types (type III).

Finally, we want to apply the method on Blida site northern Algeria from spectral data at eight wavelengths 0.340 , 0.380 , 0.440 , 0.500 , 0.675 , 0.870 , 1.020 , $1.640 \mu\text{m}$, when the retrieved columnar number distributions are quite interesting, what is represented on Fig. 5.

The presence of at least two kinds of aerosols is very clear with bi-modal (Type III) and Junge-type (Type I) distributions. The type I distributions occur mainly in the winter and early spring months and the type III occur throughout the late spring to late summer months.

REFERENCES

- [1] S. Twomey, '*Atmospheric Aerosols*', Developments in Atmospheric Science, Vol. 7, Elsevier Scientific Pub. Co., 302 pages, 1977.
- [2] C.E. Junge, '*Air Chemistry and Radioactivity*', Academic Press, 382 pages, 1963.

- [3] G.F. Bohren and D.R. Huffman, 'Absorption and Scattering of Light by Small Particles', Wiley, 1983.
- [4] C.N. Davies, '*Size Distribution of Atmospheric Particles*', Journal of Aerosol Science, Vol. 5, N°3, pp. 293 – 300, 1974.
- [5] Y.F. Wang and C.C. Yang, '*Regularizing Active Set Method for Retrieval Of Atmospheric Aerosol Particle Size Distribution Function*', Journal of Optical Society of America A, Vol. 25, pp. 348 - 356, 2008.
- [6] G. Tonna, T. Nakajima and R. Rao, '*Aerosol Features Retrieved from Aureole Data: A Simulation Study Concerning a Turbid Atmosphere*', Applied Optics, Vol. 34, N°21, pp. 4486 - 4499, 1995.
- [7] Y.F. Wang, S. Fan, X. Feng, G.J. Yan and Y.N. Guan, '*Regularized inversion method for retrieval of aerosol particle size distribution function in $W^{l,2}$ space*', Applied Optics, Vol. 45, N°28, pp. 7456 - 7467, 2006.
- [8] P.H. McMurry, '*A Review of Atmospheric Aerosol Measurements*', Atmospheric Environment, Vol. 34, pp. 1959 - 1999, 2000.
- [9] M.D. King, D.M. Byrne, B.M. Herman and J.A. Reagan, '*Aerosol Size Distributions Obtained by Inversion of Spectral Optical Depth Measurements*', Journal of the Atmospheric Sciences, Vol. 35, N°11, pp. 2153 - 2167, 1978.
- [10] A. Ben-David, B.M. Herman and J.A. Reagan, '*Inverse Problem and the Pseudoempirical Orthogonal Function Method of Solution. 2. Use*', Applied Optics, Vol. 27, N°7, pp. 1243 - 1254, 1988.
- [11] T. Nakajima, G. Tonna, R. Rao, P. Boi, Y. Kaufman and B. Holben, '*Use of Sky Brightness Measurements from Ground for Remote Sensing of Particulate Polydispersions*', Applied Optics, Vol. 35, N°15, pp. 2672 - 2686, 1996.
- [12] O.V. Dubovik, T.V. Lapyonok and S.L. Oshchepkov, '*Improved Technique for Data Inversion: Optical Sizing of Multicomponent Aerosols*', Applied Optics, Vol. 34, N°36, pp. 8422 - 8436, 1995.
- [13] D.L. Phillips, '*A Technique for the Numerical Solution of Certain Integral Equations of the First Kind*', Journal of the Association for Computing Machinery, Vol. 9, N°1, pp. 84 – 97, 1962.
- [14] S. Twomey, '*On the Numerical Solution of Fredholm Integral Equations of the First Kind by the Inversion of the Linear System Produced by Quadrature*', Journal of the Association for Computing Machinery, Vol. 10, N°1, pp. 97 – 101, 1963.
- [15] G.E. Shaw, '*Inversion of Optical Scattering and Spectral Extinction Measurements to Recover Aerosol Size Spectra*', Applied Optics, Vol. 18, N°7, pp. 988 – 993, 1979.
- [16] K.S. Shifrin and L.G. Zolotov, '*Spectral Attenuation and Aerosol Particle Size Distribution*', Applied Optics, Vol. 35, N°12, pp. 2114 – 2124, 1996.
- [17] C. Böckmann, '*Hybrid Regularization Method for the Ill-posed Inversion of Multiwavelength Lidar Data in the Retrieval of Aerosol Size Distributions*', Applied Optics, Vol. 40, N°9, pp. 1329 – 1342, 2001.
- [18] S. Twomey, '*Comparison of Constrained Linear Inversion and An Iterative Nonlinear Algorithm Applied to the Indirect Estimation of Particle Size Distributions*', Journal of Computational Physics, Vol. 18, N°2, pp. 188 – 200, 1975.
- [19] Y.F. Wang, '*Computational Methods for Inverse Problems and Their Applications*', Higher Education Press, Beijing, 2007.
- [20] M.T. Chahine, '*Inverse Problems in Radiation Transfer: Determination of Atmospheric Parameters*', Journal of the Atmospheric Sciences, Vol. 27, N°6, pp. 960 – 967, 1970.

- [21] F. Ferri, A. Bassini, and E. Paganini, 'Modified Version of the Chahine Algorithm to Invert Spectral Extinction Data for Particle Sizing', *Applied Optics*, Vol. 34, N°25, pp. 5829 – 5839, 1995.
- [22] C. Böckmann and A. Kirsche, 'Iterative Regularization Method for Lidar Remote Sensing', *Computer Physics Communications*, Vol. 174, N°8, pp. 607 – 615, 2006.
- [23] Y.F. Wang, S.F. Fan and X. Feng, 'Retrieval of the Aerosol Particle Size Distribution Function by Incorporating a Priori Information', *Journal of Aerosol Science*, Vol. 38, N°8, pp. 885 – 901, 2007.
- [24] A. Angstrom, 'On the Atmospheric Transmission of Sun Radiation and on Dust in the Air', *Geografiska Annaler*, Vol. 11, pp. 156 - 166, 1929.
- [25] P.H. Wang, M.P. McCormick, T.J. Swissler, M.T. Osborn, W.H. Fuller and G.K. Yue, 'Inference of Stratospheric Aerosol Composition and Size Distribution from SAGE II Satellite Measurements', *Journal of Geophysical Research*, Vol. 94, N°D6, pp. 8435 - 8446, 1989.
- [26] G.K. Yue, M.P. McCormick and W.P. Chu, 'Retrieval of Composition and Size Distribution of Stratospheric Aerosols with the SAGE II Satellite Experiment', *Journal of Atmospheric and Oceanic Technology*, Vol. 3, N°3, pp. 371 - 380, 1986.
- [27] G.P. Box, K.M. Sealey and M.A. Box, 'Inversion of Mie Extinction Measurements Using Analytic Eigenfunction Theory', *Journal of the Atmospheric Sciences*, Vol. 49, N°22, pp. 2074 - 2081, 1992.
- [28] C.E. Junge, 'The Size Distribution and Aging of Natural Aerosols as Determined From Electrical and Optical Data on the Atmosphere', *Journal of Meteorology*, Vol. 12, N°1, pp. 13 - 25, 1955.
- [29] M.D. King, 'Sensitivity of Constrained Linear Inversion to the Selection of the Lagrange Multiplier', *Journal of the Atmospheric Sciences*, Vol. 39, N°6, pp. 1356 - 1369, 1982.
- [30] S. Twomey, 'On the Numerical Solution of the Fredholm Integral Equations of the First Kind by the Inversion of the Linear System Produced by Quadrature', *Journal of the Association Computing Machinery*, Vol. 10, N°1, pp. 97 - 101, 1963.
- [31] M.D. King and D.M. Byrne, 'A Method for Inferring Total Ozone Content from the Spectral Variation of Total Optical Depth Obtained with a Solar Radiometer', *Journal of the Atmospheric Sciences*, Vol. 33, N°11, pp. 2242 – 2251, 1976.
- [32] B.M. Herman, S.R. Browning and J.A. Reagan, 'Determination of Aerosol Size Distributions from Lidar Measurements', *Journal of the Atmospheric Sciences*, Vol. 28, N°5, pp. 763 - 771, 1971.