Columnar aerosol size distribution function obtained by inversion of spectral optical depth measurements for Zanjan area

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ABSTRACT

We are reporting the calculated values of columnar aerosol size distribution function for atmosphere of Zanjan, a city in Northwest Iran (36.7 N, 48.6 E). Ground-based measurements of the total optical depth of the Zanjan atmosphere at 440 nm, 670 nm, 870 nm and 1020 nm are recorded using a Cimel CE318-2 sunphotometer during the time period of October 2006 to September 2008. The spectral aerosol optical depth has been obtained by subtraction of molecular optical depth from the total optical depth for each wavelength channel. Also the Ångström exponent, α, is determined by a logarithmic fit to the aerosol optical depth when it is plotted versus the logarithm of the wavelength. Daily average of the measured aerosol optical depth and α values has been implemented in an inversion algorithm for calculation of the columnar aerosol size distribution function. In this algorithm, the aerosols are considered as spheres of different size and refractive index of 1.45. As a result, we found that during the measurements usually more than 50% of the aerosol volume concentrations were belong to those with radius of $> 1\ \mu\mathrm{m}$, 40% to those with radius of $< 0.5\ \mu\mathrm{m}$ and the rest 10% have radii between the mentioned sizes. Considering these results, aerosols in the coarse mode are the most dominant ones. We believe this related to Zanjan geographical location when it is located in a dry area and subjected to frequent dust winds. Also the urban anthropogenic aerosols have a considerable share in the aerosol content of the atmosphere.

1. INTRODUCTION

Zanjan a city in Northwest Iran located at 36.7 N, 48.6 E and 1800 m above the mean sea level, frequently experiences dust storms in end of spring and beginning of autumn. Dust sources like the Tigris and Euphrates basin has a major role in the aerosol content of this region [1]. Other resources like anthropogenic aerosols and dust from the Qom Lake are other minor contaminants in the atmosphere of this area [2]. Here we are reporting the size distribution function for the atmospheric aerosols of these area that have been calculated by an inversion algorithm applied to the aerosol optical depth (AOD) of the atmosphere recorded in the time period of October 2006 to September 2008. The AOD values have been retrieved from the data recorded by a Cimel CE318-2 sunphotometer (SPM) in the sun mode.

The atmospheric aerosol size distribution plays an important role in climate changes [3,4]. So it is necessary to determine the size distribution function of the aerosol particles, $n(r)$, in a mathematical formalism. The relationship between the size of atmospheric aerosol particles and the wavelength dependence of the extinction coefficient was first suggested by Ångström in 1929 [5]. Since then the size distribution began to be retrieved by extinction measurements [3]. In 1969 Yamamoto and Tanaka were the first to apply a numerical inversion algorithm to spectral measurements of extinction coefficient in order to determine an aerosol size distribution [5,6]. We also use the same method to determine $n(r)$ for the atmosphere of Zanjan in the mentioned time period. Our results show, more than 50% of the aerosol volume concentrations were belong to those with radius of $> 1\ \mu\mathrm{m}$, 40% to those with radius of $< 0.5\ \mu\mathrm{m}$ and the rest 10% have radii between the mentioned sizes.

2. METHOD

The attenuation of solar radiation passing through the atmosphere is given by Bouguer-Beer-Lambert law,

$$I_\lambda = I_{0\lambda}\exp[-m\tau_{\mathrm{tot}}(\lambda)], \tag{1}$$

Where I_λ is the observed spectral direct-beam irradiance at wavelength λ, $I_{0\lambda}$ is the extraterrestrial solar spectrum corrected for the actual sun-earth distance, m is the optical air mass and $\tau_{\mathrm{tot}}(\lambda)$ is the wavelength-dependent total optical depth (TOD) [7]. TOD for each recording of the SPM in the sun mode can be calculated from Eq. (1). After subtracting the mean daily molecular optical depth (MOD) from the daily-averaged TOD, the aerosol optical depth (AOD) will be obtained. AOD or $\tau_a(\lambda)$**,** like TOD is calculated for four wavelengths, 440 nm, 670 nm, 870 nm and 1020 nm. Ångström empirical formula is written as,

$$\tau_a(\lambda) = \beta\lambda^{-\alpha}, \tag{2}$$

Where β is the turbidity coefficient and α is the Ångström exponent reflecting the aerosol size distribution [3]. The Ångström exponent is determined by a logarithmic fit to the AOD when it is plotted versus the logarithm of the wavelength. The obtained AODs at three wavelength channels, 440 nm, 670 nm and 870 nm have been used to retrieve $\boldsymbol{\alpha}$ [8]. Assuming a spherical shape and refractive index of m', the AOD and aerosol size distribution function relate as,

$$\tau_a(\lambda) = \int_{r=r_{\min}}^{r=r_{\max}} \int_{z=0}^{z=\infty} \pi r^2 Q_{\mathrm{ext}}(r,\lambda,m')n'(r,z)dzdr, \tag{3}$$

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Where $n'(r,z)$ is the height-dependent aerosol number density in the radius range r to $r+dr$; and $Q_{\text{ext}}(r,\lambda,m')$ the extinction efficiency factor from the Mie theory [5]. Since all measured quantities by SPM are summed over the atmospheric column we replace $n'(r,z)$ with,

$$n(r) = \int_{z=0}^{z=\infty} n'(r,z)dz\,, \tag{4}$$

Where $n(r)$ is the unknown columnar aerosol size distribution, i.e., the number of particles per unit of area per the unit radius interval**,** in a vertical column through the atmosphere [5]. Considering Eq. (4), Eq. (3) can be written as,

$$\tau_a(\lambda) = \int_{r=r_{\min}}^{r=r_{\max}} \pi r^2 Q_{\text{ext}}(r,\lambda,m')n(r)dr\,, \tag{5}$$

Since the AOD can be experimentally obtained, one can retrieve the size distribution by the inversion of the AOD measurements through Eq. (5) [5]. In this work, the aerosols are considered as spheres of different sizes from $r_{\min} = 5\text{ nm}$ to $r_{\max} = 2\,\mu\text{m}$ and refractive index of $m' = 1.45$. In order to find the exact value of $n(r)$, a continuous spectra of $\tau_a(\lambda)$ is needed. In other words infinite number of equations like Eq. (5), for continuous spectra of the light wavelength is required. But we are measuring the AOD only on four wavelength channels. So we can determine $n(r)$ only for four radius values. Therefore Eq. (5) can be written as,

$$\tau_a(\lambda) = \sum_{j=1}^{4} \int_{r=r_j}^{r=r_{j+1}} \pi r^2 Q_{\text{ext}}(r,\lambda,m')n(r)dr\,, \tag{6}$$

When the integration limits are $r_1 = r_{\min} = 5\text{ nm}$, $r_2 = 0.5\,\mu\text{m}$, $r_3 = 1\,\mu\text{m}$, $r_4 = 1.5\,\mu\text{m}$ and $r_5 = r_{\max} = 2\,\mu\text{m}$. We let $n(r) = h(r)f(r)$, where $h(r)$ is a rapid varying function of r and $f(r)$ is more slowly varying. $h(r)$ has the form of a Junge size distribution [5],

$$h(r) = r^{-(\alpha+3)}\,, \tag{7}$$

Also we consider all the particles that their radiuses are in the range of r_j to r_{j+1} have the same $f(r) = f_j$ and Eq. (6) becomes,

$$\tau_a(\lambda) = \sum_{j=1}^{4} f_j \int_{r=r_j}^{r=r_{j+1}} \pi r^2 Q_{\text{ext}}(r,\lambda,m')r^{-(\alpha+3)}dr\,, \tag{8}$$

And for an specified wavelength, λ_i**,** Eq. (8) changes to,

$$\tau_a(\lambda_i) = \sum_{j=1}^{4} f_j \int_{r=r_j}^{r=r_{j+1}} \pi r^2 Q_{\text{ext}}(r,\lambda_i,m')r^{-(\alpha+3)}dr\,, \tag{9}$$

Considering the four mentioned wavelengths, Eq. (9) can be written as a matrix equation,

$$T = AF + \varepsilon\,, \tag{10}$$

Where $\boldsymbol{T}$ is a columnar matrix whose elements are $T_i = \tau_a(\lambda_i)$, $i = 1,2,3,4$, and representing the AOD value at λ_i. $\boldsymbol{F}$ is a columnar matrix and its elements are, $F_j = f(\bar{r}_j), j = 1,2,3,4$ and $\bar{r}_j = (r_j + r_{j+1})/2$ is the midpoint of the two coarse values of r_j and r_{j+1}**.** $\boldsymbol{A}$ is a square matrix and its elements are representing by, $A_{ij} = \int_{r=r_j}^{r=r_{j+1}} \pi r^2 Q_{\text{ext}}(r,\lambda_i,m')r^{-(\alpha+3)}dr$, where $\boldsymbol{\epsilon}$ is an unknown error vector whose elements $\boldsymbol{\epsilon_i}$ represent the deviation between measurement (T_i) and its theoretical value $\left(\sum_{j=1}^{4} A_{ij}F_j\right)$ [5]. The conventional approach to such a problem is to use the least square techniques and then Eq. (10) can be written as,

$$F = (A^T A)^{-1} A^T T\,, \tag{11}$$

Where $\boldsymbol{A^T}$ is transpose of $\boldsymbol{A}$ [9]. Many studies have pointed out that the solution derived from Eq. (11) is unstable because it is under constraint. The constrained linear inversion solution to solve this problem derived by Phillips (1962) and Twomey (1963) [9]. This solution has the following form,

$$F = (A^T A + \gamma H)^{-1} A^T T\,, \tag{12}$$

Where $\boldsymbol{H}$ is a (4×4) matrix given by,

$$H = \begin{bmatrix} 0.75 & -0.25 & -0.25 & -0.25 \\ -0.25 & 0.75 & -0.25 & -0.25 \\ -0.25 & -0.25 & 0.75 & -0.25 \\ -0.25 & -0.25 & -0.25 & 0.75 \end{bmatrix}, \tag{13}$$

And γ is a non-negative multiplier [9]. In performing the inversion described above, it is necessary to select a value for γ, but referring to Eq. (12), $\gamma_{\text{rel}} = (\gamma \boldsymbol{H})_{11}/(\boldsymbol{A^T A})_{11}$ is the dominant term. A minimum value for γ_{rel} should be chosen in a way that all elements of $\boldsymbol{F}$ are positive (negative values are not physical solution) [5].

3. AEROSOL SIZE DISTRIBUTION RESULTS

The method that described in the previous section has been applied to the recorded data by our SPM during the time period of October 2006 to September 2008. We choose 97 days of this time period when the sky was sunny during the measurements. The values of $n(r)$ have been determined for the midpoints of the coarse radius intervals, $\bar{r}_1 = 0.25\,\mu\text{m}$, $\bar{r}_2 = 0.75\,\mu\text{m}$, $\bar{r}_3 = 1.25\,\mu\text{m}$ and $\bar{r}_4 = 1.75\,\mu\text{m}$. These are representative of very fine, fine, coarse and very coarse aerosols respectively. So we are defining four classes for aerosol sizes. Always amount of very fine aerosols are more than other aerosol sizes. Also one can say for about 90% of the days, the amount of coarse aerosols are more than fine ones. For these days the Ångström exponent has values between 0.2 and 1.2 $(0.2 \le \alpha \le 1.2)$. For example see Fig. 1.

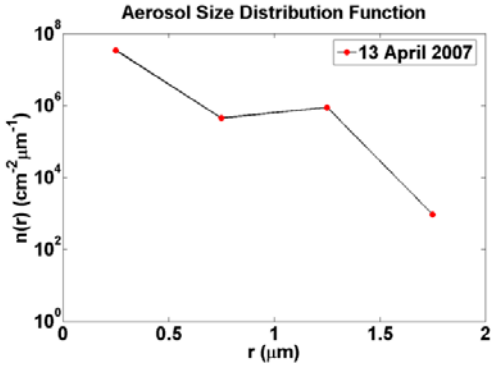

Figure 1. *Aerosol size distribution on April 13, 2007,* $(\alpha = 0.54, \tau_a(440\text{ nm}) = 0.22)$.

For rest of the days, amount of fine aerosols are more than coarse aerosols and $\alpha \geq 1.2$. Fig. 2 shows a sample of such days.

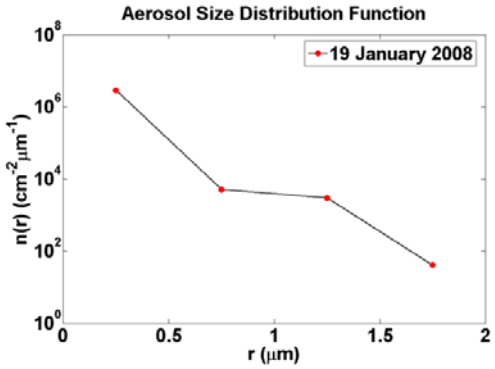

Figure 2. *Aerosol size distribution on January 19, 2008,* $(\alpha = 2.22, \tau_a(440\text{ nm}) = 0.05)$.

Also in approximately 10% of the days, amount of very coarse aerosols are considerable. We have $\alpha \leq 0.2$ for these days. For example for a very dusty day on July 3, and one day after it on July 4, 2008, the amounts of very coarse particles are noticeable. (Fig. 3).

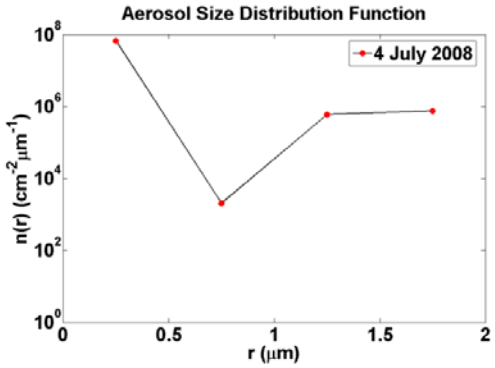

Figure 3. *Aerosol size distribution on July 4, 2008, one day after a very dusty day,* $(\alpha = 0.09, \tau_a(440\text{ nm}) = 0.36)$.

As it can be seen in Figures 1-3, the Ångström exponent determines relative population of aerosols in the four mentioned class sizes and the AOD is related to the total population of aerosols in all sizes. (Fig. 4,5).

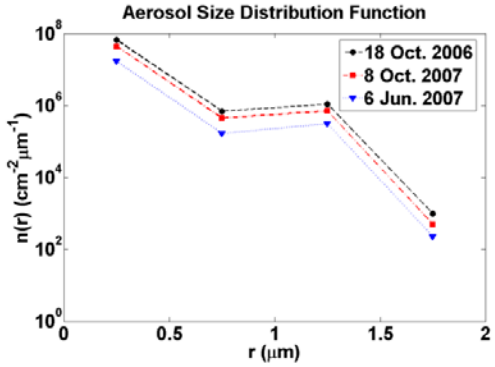

Figure 4. *Aerosol size distribution for 3 days with* $\alpha = 0.71$, $\tau_a(440\text{ nm}) = 0.11$, 0.27, *and* 0.41 *respectively.*

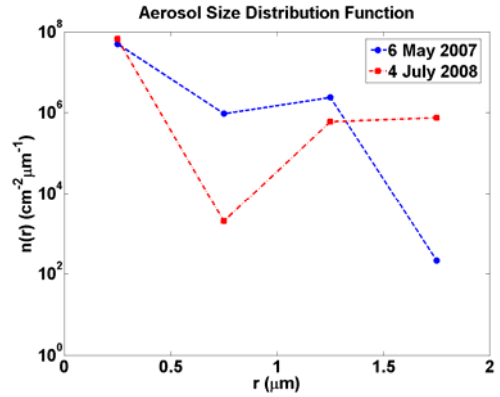

Figure 5. *Aerosol size distribution for May 6, 2007, and July 4, 2008, when* $\tau_a(440\text{ nm}) = 0.36$, *for both days and* $\alpha = 0.22$, *and* 0.09 *respectively.*

Considering the volume concentration of the aerosols, we find that during the measurements usually more than 50% of the aerosol volume concentrations are belong to those with radius of $> 1\ \mu\text{m}$, 40% to those with radius of $< 0.5\ \mu\text{m}$ and the rest 10% have radii between the mentioned sizes. Considering these results, aerosols in the coarse mode are the most dominant ones. We believe this related to Zanjan geographical location when it is located in a dry area and subjected to frequent dust winds. Also the urban anthropogenic aerosols have a considerable share in the aerosol content of the atmosphere.

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