SIMPLE METHOD OF AEROSOL PARTICLE SIZE DISTRIBUTION RETRIEVING FROM MULTIWAVELENGTH LIDAR SIGNALS

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ABSTRACT

An improved lidar retrieval of the aerosol particle size distribution (APSD) is presented. A predefined APSD function with few free parameters is directly substituted into the lidar equations. The minimization technique allows to find the parameters which provide the best fit of APSD comparing theoretically generated signals with the experimental ones. The method does not require former knowledge of the lidar ratio. The approach was tested on typical APSD presented by Seinfeld and Pandis (1997). For our purpose these distributions were approximated with two mode combination of lognormal functions. With a lidar working at five wavelengths in UV – near IR spectral range a satisfactory retrieval of synthetic APSD is possible for the particles within the range 100 – 5000 *nm*.

INTRODUCTION

Among many methods of the aerosol investigation the optical remote studies play very important role. Lidars working simultaneously at several wavelengths are often used for these observations. This technique provide opportunity for remote investigation of aerosol particle size distribution (APSD). Such studies are possible due to different properties of the light scattering by particles of different sizes at various wavelengths (λ). A common approach to the retrieving of APSD from the lidar signals consists in determination of total scattering coefficients α_λ and/or the backscattering coefficient β_λ. Both coefficients can be related to APSD function *n(z,r)* by following equations (Seinfeld and Pandis, 1997):

$$\alpha_\lambda(\mathrm{z}) = \int_0^\infty Q_\lambda^E(r)\pi r^2 n(z,r)dr$$

$$\beta_\lambda(\mathrm{z}) = \int_0^\infty Q_\lambda^B(r)\pi r^2 n(z,r)dr\,, \tag{1}$$

where z is a distance from the lidar, r - the particle radius, while Q_λ^E and Q_λ^B denote the efficiencies of total extinction and backscattering, respectively. These efficiencies can be calculated using *e.q.* Mie theory (Bohren and Huffman, 1999), when spherical shape of the particles is assumed.

On the other hand α_λ and β_λ can be found from lidar signals $S_\lambda(z)$ described by lidar equations. For each wavelength:

$$S_\lambda(z) = \frac{A_\lambda}{z^2}\beta_\lambda(z)\exp\left(-2\int_{z_0}^{z}\alpha_\lambda(y)dy\right). \tag{2}$$

where A_λ are the apparatus constants. In each single equation (2) both α_λ and β_λ are unknown, therefore in order to retrieve them an additional information is necessary. The relation - so called *lidar ratio* - is usually used (Klett, 1981). For the white light it takes the form:

$$q_L = \frac{\beta_\lambda}{\alpha_\lambda^k}\,, \tag{3}$$

where k is a number.

Deriving APSD from the scattering coefficients (1) is a classic example of inverse ill-posed problem, typical in earth sciences. Solution of Fredholm integral equations of 1st kind is required. Various inversion techniques have been proposed for this purpose. Successful approach to this problem needs some additional assumptions about the solution, like a smoothness or/and a positivity. This can be obtained with a predefined form of *n(z,r)* distribution, like histogram or Junge function, log-normal function or linear combination of these functions with different parameters (Seinfeld and Pandis, 1997). In such a case the solution is reduced to finding these parameters.

Such approach was presented by Herman *et al* (1971). They fit Junge distribution to signals from bistatic lidar. Rajeev and Parameswaran (1998) have shown two iterative methods of APSD determination: with assumed Junge distribution or without any assumed shape, taking for calculations an arbitrary lidar ratio. Heitzenberg *et al* (1998) proposed randomized minimization search technique (iterative least square algorithm) to derive an assumed histogram distribution. The research group from ITR applied the inversion with regularization for deriving both APSD and the refractive index using the signals registered with their aerosol and Raman lidar (Müller *et al*, 1999). They developed the technique based on Tichonov regularization (Veselovskii *et al*, 2002, 2004). In recent years the eigenvalue analysis was applied for the lidar data inversion (Veselovskii *et al*, 2005). Detailed review of different approaches to APSD retrieval was done by Böckman (2001). Her hybrid method presented in this paper was later applied for experimental data (Böckmann *et al*, 2005). As the result APSD, the refractive index and single scattering albedo was retrieved.

Another solution to the problem of lidar data analysis was proposed by Kusiel and Zolotov (1997, 2003). They developed mean ordinates method. They assumed APSD function as combination of several log-normal functions. Using these distributions, the optical characteristics (like α_λ and β_λ), were calculated and compared with those measured by lidar technique. Then the mean ordinates over those solutions were calculated and model closest to the mean ordinates was taken as the most probable solution. The mean ordinates method was used for inverting the horizontal lidar data.

A different approach was presented by Ligon *et al* (1996, 2000). In order to shorten the calculation time they used the Monte Carlo method of approximation of APSD.

Certainty of retrieving the APSD was experimentally tested by Joshiyama *et al* (1996). They measured optical parameters of artificial aerosol with bistatic lidar and compared the results with mathematical model.

As mentioned above in all these methods APSD is derived from α_λ and/or β_λ coefficients (1), with the use of lidar ratio (3). The lidar ratio was first suggested by Curcio and Knestrick (1966). They have experimentally evaluated k to be equal to *0.66,* while Fenn (1966) reported different values. Then Twomey and Howell (1965) found the linear relation between α_λ and β_λ basing on Mie theory and various size distributions of particles. They also concluded that in general such relation could not be a unique one, and that the linear correlation between the backscattering and the extinction coefficients is evident only for the white light. Good linearity is reported for clouds where a multiple light scattering takes place (O'Connor *et al*, 2004). It is not clear, however, whether the linear lidar ratio can be used for the monochromatic laser radiation. Analysis of large data-set of lidar returns of EARLINET shows about 40 % variability of $\beta_\lambda/\alpha_\lambda$ ratio for aerosols in boundary layer (Pappalardo, 2005). Therefore the value of the lidar ratio is often guessed or assumed (Landulfo *et al*, 2003; Iwasaka *et al*, 2003).

The coefficients α_λ and β_λ that are necessary for APSD inversion can be found using the lidar signal inversion technique by Klett (1981) and Fernald (1984). However in this case the aerosol parameters in the reference point must be known. When the vertical profiling is performed the reference point is usually selected at high altitudes, where the aerosol concentration is negligible and the molecular lidar ratio: $\beta_\lambda/\alpha_\lambda = 3/8\pi$ can be used. Then the backward solving of the lidar equation is applied. In case of clear sky the measurement of total optical thickness by sun-photometers allows to deduce the lidar ratio (G. Karasiński *et al*, 2007), which is usually considered constant with height. Such solutions are not applicable in many experimental situations, *e.q.* for aerosol layer under the cloud cover. Some problems can be also overcame when common measurement by aerosol and Raman lidars is performed, however the Raman signal registration is short-distant and is not well applicable for multiwalelength lidars.

In this paper an approach to the problem of APSD determination is proposed. It does not require the lidar ratio knowledge. Predefined functions $n(z,r)$ are substituted directly to equations describing the lidar signals (2). The experimental estimates of α_λ and/or β_λ values are not needed. Application of the minimization technique allows to derive the best fit of APSD by comparison the artificially generated signals with the lidar returns.

DESCRIPTION OF THE METHOD

In a first step equations (2) representing the registered signals $S_\lambda(z)$, should be converted to so called range corrected form:

$$L_\lambda(z) = S_\lambda(z)\cdot z^2 = A_\lambda \beta_\lambda(z)\exp\left[-2\int_{z_0}^{z}\alpha_\lambda(x)dx\right]\cdot \quad (4)$$

Due to digitization the lidar signals are quantitized *e.q.* in space with the interval of dz. For further analysis the ratio of the signals $L_\lambda(z_l)$ at distance z_l and at its neighbour distance $z_{l+1}=z_l+dz$ is taken:

$$\frac{L_\lambda(z_{l+1})}{L_\lambda(z_l)} = \frac{\beta_\lambda(z_{l+1})}{\beta_\lambda(z_l)}\exp\left[-2\alpha_\lambda(z_{l+1})dz\right]\cdot \quad (5)$$

This form allows to omit the apparatus constants A_λ, which are usually unknown. Left hand side of (5) describe the experimental signals, while the right hand side can be calculated from Mie equations (1) when $n(z,r)$ is assumed. Using a minimization technique with the cost function:

$$\chi^2(z_l) = \sum_\lambda \left\{\frac{L_\lambda(z_{l+1})}{L_\lambda(z_l)} - \frac{\beta_\lambda(z_{l+1})}{\beta_\lambda(z_l)}\exp\left[-2\alpha_\lambda(z_{l+1})dz\right]\right\}^2 \quad (6)$$

allows to find APSD.

In order to fit the size distribution the predefined form is necessary. Sum of modes: $n(r,z) = \sum_{j=1}^{K} n_j(r,z)$ $(K = 1,2)$ is usually used. Each mode is described by the log-normal function:

$$n_j(r,z) = \frac{C_j(z)}{\sqrt{2\pi}\cdot\log\sigma_j(z)}\cdot \cdot\frac{1}{r}\cdot\exp\left\{-\frac{\left[\log r - \log R_j(z)\right]^2}{2\cdot\log^2\sigma_j(z)}\right\}, \quad (7)$$

where R_j denotes the modal radius, C_j - concentration of aerosol in a given mode, and σ_j - width.

TEST OF THE METHOD

The approach was tested with the synthetic size distributions after Seinfeld and Pandis (1997). They described several typical APSD with three mode lognormal functions. We assumed a uniform spatial distribution of aerosol. Particles are characterized by the refraction coefficients of water. That allowed to find the scattering coefficients (1). In order to simulate a typical experiment with multiwavelength lidar, the synthetic range corrected lidar signals $L_\lambda(z_l)$ for five wavelengths (1064, 800, 532, 375 and 355 *nm*) were calculated by means of formulas (2). Using these signals the reconstruction of the left hand side of (5) was possible.

Our initial test consisted in approximation of APSD by single lognormal function. Such approach is used by some researchers (Hess *et all*, 1998). A matrix of lognormal functions $n_{RC\sigma}(r,z_0)$ was constructed. Each element of the matrix was expressed by equation (7). In order to cover ranges of parameters predicted by Seinfeld and Pandis (1997), the matrices were generated for the modal radiuses R in the range 5 to 650 *nm*, the particle concentrations C changing from 0.01 to 3500 cm^{-3} and widths σ varying from 1.7 to 7. Using these functions and the equation (1) the matrices of coefficients $\alpha_{RC\sigma\lambda}$ and $\beta_{RC\sigma\lambda}$ were calculated for each wavelength. Integration for particle radiuses from 1 *nm* to 10 *μm* was performed. Then with pairs of elements from $\alpha_{RC\sigma\lambda}$ and $\beta_{RC\sigma\lambda}$ matrices the right hand side of (5) were constructed. For each pair the value of $\chi^2_{RC\sigma}$ was determined by minimization technique. Cost function (6) was applied. The optimal distribution $n(r,z_0)$ was found as the arithmetic mean of all analyzed cases of weights $1/\chi^2_{RC\sigma}$.

Results of these investigation are presented in Fig 1. A good approximation of the assumed APSD by single mode lognormal function was found only for *free troposphere* aerosol (Fig. 1a), due to a specific shape of its distribution. In other cases this approximation was not satisfactory. For some particle radius ranges (like *remote continental*, r≈400-700 *nm*) the discrepancies between the assumed and fited distributions reached two orders of magnitude. For *polar* APSD (Fig. 1c), the approximation is quite good for the particles of radius larger than 300 *nm*, but it is not acceptable for smaller ones. In all cases for r<100 *nm* the approximation is poor.

Much better approximation of APSD can be achieved with two mode lognormal distribution. In such case matrices of $\alpha_{RC\sigma\lambda}$ and $\beta_{RC\sigma\lambda}$ coefficients were prepared for each mode separately: for the accumulation mode modal radiuses R were in a range 5 – 200 *nm* and concentrations C changed from 20 to 3500 cm^{-3}; for the coarse mode R from 200 to 2500 *nm* and C from 0.01 to 20 cm^{-3} were used. For both modes the widths of the functions changed from 1.7 to 7.

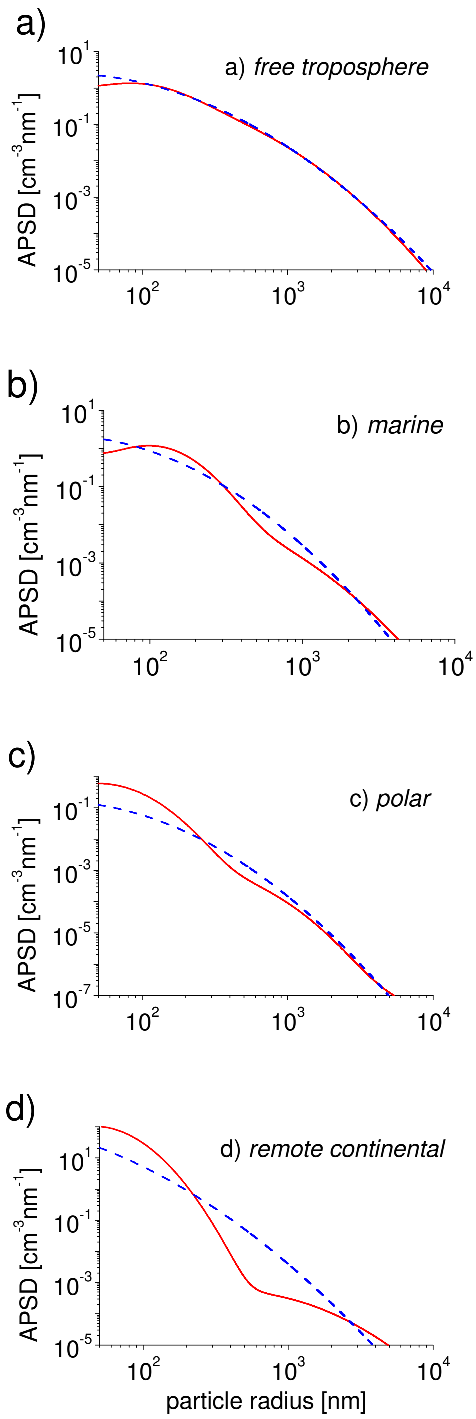

Fig. 1. Reconstruction of APSD with single mode lognormal function. Continuous lines - assumed APSD, dashed lines - their approximation.

When the two mode approximation is applied the systematic search of these matrixes needs the calculation of the cost function (6) for billions of cases. For this reason the Monte Carlo method, probing about 0.1% of all the cases was used.

Similarly to the single mode approximation, the value of $\chi^2_{RC\sigma}$ (6) for each probe was determined and the optimal distribution $n(r,z_0)$ was found as the arithmetic mean of the results with weights $1/\chi^2_{RC\sigma}$.

The fits are presented in Fig 2(a-d). In this case the quite good approximation was received for all considered aerosols. For the particle radiuses beginning from 50 *nm* (i.e. within the range larger than for single mode approximation) the discrepancy between assumed and retrieved distribution is smaller than 20 %. For the particle radiuses smaller than 50 *nm* (except for special cases, like *polar* aerosol – Fig. 2c) the fit is poor. We believe that it is due to a weak contribution of small particles to the light scattering.

EVALUATION OF PARTICLE REFRACTION INDEX

Precise fit of two mode log-normal function to the APSD provides the opportunity to evaluate the refraction index of aerosol particles. In order to verify its value a following numerical experiment was performed. *Marine* aerosol at certain distance z_0 *was* considered. The right hand side of equation (5) was generated for the refraction coefficient of water (Segelstein, 1981) as well as for the refraction coefficient of sea salt (Volz, 1972). Then the matrices of $\alpha_{RC\sigma\lambda}$ and $\beta_{RC\sigma\lambda}$ coefficients were calculated.

The fiting procedure was repeated two times: ones within the broad range of parameters (as described in previous chapter) and then in a narrow range of the parameters, around that determined in the previous step. In this case the matrices of $\alpha_{RC\sigma\lambda}$ and $\beta_{RC\sigma\lambda}$ coefficients were calculated again with high

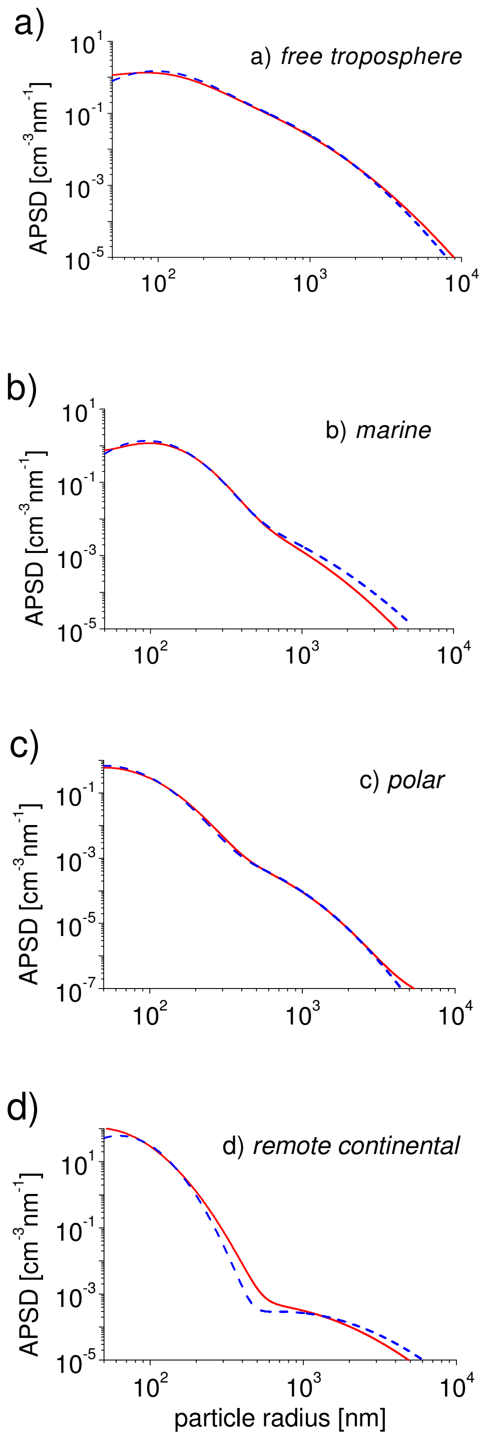

Fig. 2. Reconstruction of APSD with two-mode lognormal function. Continuous lines - assumed APSD, dashed lines - their approximation.

precision for water and for sea salt refraction indexes. Both cases were compared with the lidar signals which also were calculated for the water and for the sea salt. For all considered cases values of the cost function were found. Results of such procedure are shown in Fig. 4(a-d).

The lowest values of χ^2 were obtained for the case when the refractive index assumed for the signal generation coincides with the refractive index that was used in the fit. On the contrary, for the signals with water refractive index and the matrices calculated for the sea salt refractive index the value of χ^2 was about 15 – 20 times larger. This indicates that search for the minimum of χ^2 - parameter versus the refraction index provides the opportunity to determine the optimal refraction index and, in turn, to evaluate the chemical composition of aerosol particles.

EXPERIMET

We applied this method for analysis of measurements performed during the campaign in Warsaw, (Poland) in July 2006. The experiment was done with our multiwavelength lidar (Ernst *et al*, 2003, Chudzyński *et al*, 2006). Its sender generated five wavelength (1064, 782, 532, 391, 355 *nm*).

Example of results, i.e. the effective radius of aerosol particles r_{eff} under the base of cloud as a function of the altitude, is presented in Fig. 4. The registration was done 26th of July 2006 at 11:45 UTC. At this time the sky was covered by sparse cumulus clouds of bases at 1.8 *km* altitude. The effective radius was calculated using the retrieved APSD and the formula:

$$r_{eff}(z) = \frac{\int r^3 n(r,z)dr}{3\int r^2 n(r,z)dr}. \quad (8)$$

The refraction index of water was assumed. As one can see at low altitudes, up to 1.65 *km*, the effective particle radius r_{eff} is uniform. Its mean value is about 180 *nm*.

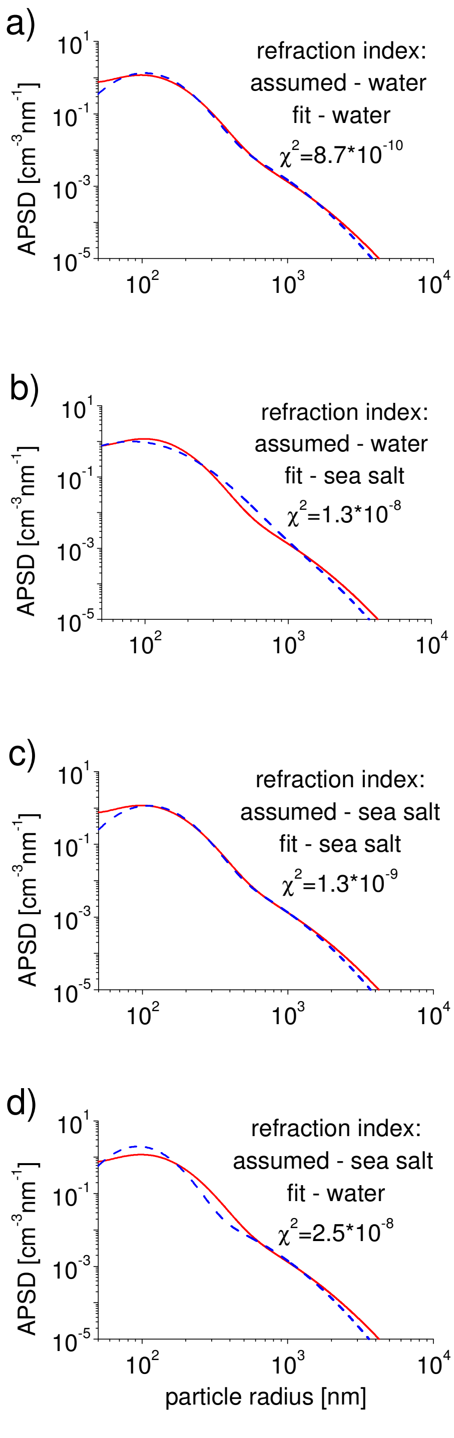

Fig. 3. Illustration of method of the refraction coefficient evaluation (marine aerosol). Continuous lines - assumed APSD, dashed lines - their two - mode approximation.

A fast increase of r_{eff} up to the value of 1200 *nm* is observed starting from about 1.65 *km*, i.e. about 150 *m* below the could base.

More detailed description of investigation of aerosol properties in vicinity of clouds is presented by Jagodnicka *et al* (2008).

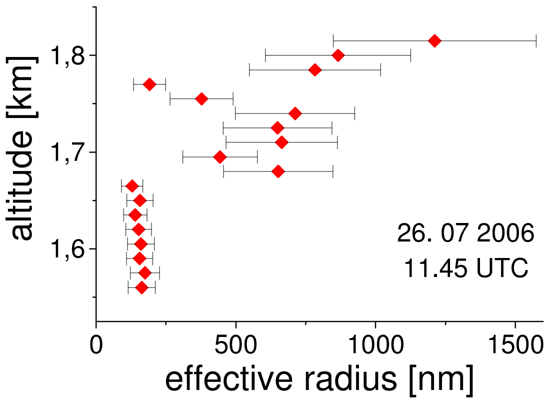

Fig. 4. Effective radius of aerosol particles as a function of the altitude. Measurement done under the base of cumulus (26.06.2006, Warsaw, Poland).

CONCLUSION.

Simple method of aerosol particle size distribution retrieving from lidar signals was presented. Due to application of direct fiting of APSD to the lidar signals this technique does not need knowledge of lidar ratio. Therefore our method can be successfully used when retrieving the aerosol scattering coefficients is difficult, for example under the clouds. To our knowledge this is an unique method providing opportunity to determine the APSD as a function of distance from the lidar.

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