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Investigation of atmospheric aerosol with multiwavelength lidar

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Results of investigation of particle size distribution of atmospheric aerosol are presented. The data were collected by means of multiwavelength lidar. A method of analysis of the lidar data without using the lidar ratio is described.

Keywords: atmospheric aerosol, lidar, particle size distribution.

1. Introduction

Recently a particular interest has been given to the investigation of properties of atmospheric aerosol. Such information are very important for physics and chemistry of the atmosphere [1], climate modeling, long period weather forecast, undestanding mechanism of cloud formation [2], and the aerosol influence on living organisms [3, 4]. In these fields the remote investigation by means of the lidar technique is widely used. Especially important is the investigation with the lidars working simultaneously at several wavelengths since it can provide the information about size distribution of aerosol particles [5–8].

2. Theoretical background

The common approach to retrieving the aerosol particle size distribution $n(z, R)$ from the lidar data (z is distance from the lidar and R – the particle radius) consists in determination of total scattering extinction coefficient α or the backscattering coefficient β for each lidar wavelength λ. Assuming that the aerosol consists of spherical droplets and that the light emitted by the lidar is not absorbed by atmospheric gases these coefficients can be related to $n(z, R)$ function by following equations:

$$\alpha(z, \lambda) = \int_0^\infty Q^E(\lambda, R)\pi R^2 n(z, R)\mathrm{d}R$$
$$\beta(z, \lambda) = \int_0^\infty Q^B(\lambda, R)\pi R^2 n(z, R)\mathrm{d}R \tag{1}$$

Efficiencies of scattering $Q^E(R, \lambda)$ and $Q^B(R, \lambda)$ can be determined using, *e.g.*, Mie theory [9]. The coefficients α or β that are calculated using the Eq. (1) might be compared with the analogical coefficients that are achieved by solving the set of lidar equations. These equations describe the signals $S(z, \lambda)$ registered by the lidar receiver at each distance z for each wavelength λ:

$$L(z, \lambda) = S(z, \lambda)z^2 = A(\lambda)\beta(z, \lambda)\exp\left[-2\int_0^z \alpha(x, \lambda)\mathrm{d}x\right] \tag{2}$$

where $A(\lambda)$ denotes the apparatus constant and $L(z, \lambda)$ – so called the range corrected lidar signal. Here the single scattering regime is assumed.

In order to solve the lidar equation one has to assume some relation between α and β [9]. In this respect the commonly used approach, known as the lidar ratio, was first suggested by CURCIO and KNESTRICK [10]:

$$\beta(z, \lambda) = C\alpha^k(z, \lambda) \tag{3}$$

where k is the number. Using this relation each lidar equation can be inversed which provides the extinction coefficient expressed by the formula [10]:

$$\alpha(z, \lambda) = \frac{\left[\dfrac{L(z, \lambda)}{L(z_F, \lambda)}\right]^{\frac{1}{k}}}{\dfrac{1}{\alpha(z_F, \lambda)} + \dfrac{2}{k}\displaystyle\int_z^{z_F}\left[\dfrac{L(x, \lambda)}{L(z_F, \lambda)}\right]^{\frac{1}{k}}\mathrm{d}x} \tag{4}$$

From that and (3) the β-coefficient can be found. According to the so called Klett method [11], this requires the knowledge of the extinction coefficient $\alpha(z_F,\lambda)$ at reference distance z_F. CURCIO and KNESTRICK [10] have experimentally evaluated $k = 0.66$, however many authors have reported different values [12]. TWOMEY and HOWELL [13] examined the lidar ratio (3) basing on Mie theory. They found the linear relation between α and β at various size distributions of the scattering particles. They also concluded that in general such relation could not be unique one, and that the linear relation between the backscattering and the extinction coefficients is evident only if the white light is used instead of the monochromatic light (that is applied in lidars). Modification of lidar ratio by use of $C = C(z, \lambda)$ which leads to so called Klett–Fernald method [14, 15], does not eliminates the misdoubts about these solutions.

Our approach to this problem consists in substitution of relations (1) to the equation (2). Consequently only the function $n(z, R)$ remains unknown in the system of lidar equations. This function can be found by minimization technique, when the set of experimental data $S(z, \lambda)$ is substituted to the left side of the Eq. (2).

In order to solve the problem for each lidar equation we calculate the ratio of the lidar signals for two distances: z_F and z_{F+l}. Here the discrete form of the signals that are provided by lidar digitisers was applied. We achieved the following formula:

$$\frac{L(z_{F+l}, \lambda)}{L(z_F, \lambda)} = \frac{\beta(z_{F+l}, \lambda)}{\beta(z_F, \lambda)} \exp\left[-2\int_{z_F}^{z_{F+l}} \alpha(x, \lambda)\mathrm{d}x\right] \tag{5}$$

where l denotes index of point in the space with respect to the reference distance z_F. Then the integration range was divided as follows:

$$\frac{L(z_{F+l}, \lambda)}{L(z_F, \lambda)} \beta(z_F, \lambda) = \beta(z_{F+l}, \lambda)\exp\left[-2\int_{z_F}^{z_{F+l-1}} \alpha(x, \lambda)\mathrm{d}x - 2\int_{z_{F+l-1}}^{z_{F+l}} \alpha(x, \lambda)\mathrm{d}x\right] \tag{6}$$

Using the trapezoidal approximation of the integral one obtains:

$$\frac{L(z_{F+l}, \lambda)}{L(z_F, \lambda)} \beta(z_F, \lambda)\exp\left[2\int_{z_F}^{z_{F+l-1}} \alpha(x, \lambda)\mathrm{d}x\right] =$$

$$= \beta(z_{F+l}, \lambda)\exp\left\{-2\left[\alpha(z_{F+l-1}, \lambda) + \alpha(z_{F+l}, \lambda)\right]\frac{\Delta z}{2}\right\} \tag{7}$$

where Δz denotes the distance between digitisation points. Then the formulas (1) for coefficients α and β are substituted to the right side of the equation:

$$\frac{L(z_{F+l}, \lambda)}{L(z_F, \lambda)} \beta(z_F, \lambda)\exp\left[\alpha(z_{F+l-1}, \lambda)\Delta z + 2\int_{z_F}^{z_{F+l-1}} \alpha(x, \lambda)\mathrm{d}x\right] =$$

$$= \pi\int R^2 n(z_{F+l}, R)Q^B(R, \lambda)\mathrm{d}R \exp\left[-\pi\Delta z\int R^2 n(z_{F+l}, R)Q^E(R, \lambda)\mathrm{d}R\right] \tag{8}$$

The right side of this equation depends on the $n(z, R)$ function only. Usually the distance between the digitisation points is of several meters so one can assume that in the transparent atmosphere the extinction coefficient does not change too much within Δz distance. Therefore in the first step of the iteration ($l = 1$) one can assume that $\alpha(z_F, \lambda) \approx \alpha(z_{F\pm1}, \lambda)$. Then the Eq. (8) takes the form:

$$\frac{L(z_{F\pm l}, \lambda)}{L(z_F, \lambda)}\beta(z_F, \lambda) =$$

$$= \pi\int R^2 n(z_{F\pm1}, R)Q^B(R, \lambda)\mathrm{d}R \exp\left[-2\pi\Delta z\int R^2 n(z_{F\pm1}, R)Q^E(R, \lambda)\mathrm{d}R\right] \tag{9}$$

It provides opportunity to fit the $n(z_{F\pm1}, R)$ function. Then using Eq. (1) value of $\alpha(z_{F\pm1}, \lambda)$ function can be found. It can be substituted again to the Eq. (8), which can be solved for $n(z_{F\pm2}, R)$. In this way the consecutive iteration of the Eq. (8) provides opportunity to find the distance-dependent distribution of aerosol particle sizes $n(z, R)$. As it was already shown by KLETT [11] the better solution is achieved when the reference point z_F is chosen at a high altitude, where the aerosol concentration is low and the scattering properties of the atmosphere are mainly determined by scattering of light on the air molecules. In this case $\alpha(z_F, \lambda)$ and $\beta(z_F, \lambda)$ are well known since they are determined by Rayleigh theory and the barometric formula [16]. Then the lidar equation should be solved in backward direction ($l < 0$) so the Eq. (8) takes the form:

$$\frac{L(z_{F+l}, \lambda)}{L(z_F, \lambda)}\beta(z_F, \lambda)\exp\left[\alpha(z_{F+l+1}, \lambda)\Delta z + 2\int_{z_F}^{z_{F+l+1}}\alpha(x, \lambda)\mathrm{d}x\right] =$$

$$= \pi\int R^2 n(z_{F+l}, R)Q^B(R, \lambda)\mathrm{d}R \exp\left[-\pi\Delta z\int R^2 n(z_{F+l}, R)Q^E(R, \lambda)\mathrm{d}R\right] \tag{10}$$

Number of wavelengths that are usually used in multiwavelength lidars changes from 3 till 7 so using the minimization procedure the particle size distribution cannot be found in details. One of the commonly used solutions is assumption that the function $n(z, R)$ can be expressed as a sum of modes, *i.e.*, $n(z, R) = \sum_i f_i(z, R)$. Usually the modes are described by lognormal function:

$$f_i(z, R) = \frac{1}{R}\frac{N_{0i}(z)}{\sqrt{2\pi}\ln\mu_i(z)}\exp\left\{\frac{\left[\ln R - \ln r_{mi}(z)\right]^2}{\ln^2\mu_i(z)}\right\} \tag{11}$$

where $N_{0i}(z)$, $r_{mi}(z)$ and $\mu_i(z)$ are parameters of the function that are found due to the minimization procedure. The distribution of real particle sizes can be well approximated by three mode approach, where each mode is responsible for fine coarse and large particles, respectively [1].

The construction of our mobile multiwavelength lidar was described elsewhere [17]. Briefly, in the optical sender pulsed Ti:Sa laser (with II harmonic) and pulsed Nd:YAG laser (with II and III harmonics) are installed. The beams were sent vertically to the atmosphere. Energies of the lasers pulses are about 100 mJ, while their repetition rates are about 10 Hz. The radiation scattered backward in the atmosphere is collected by a Newtonian telescope whith the main mirror of diameter about 400 mm and focal length 1200 mm. The light is directed to the polychromator. For each wavelength the signal is registered in a separate channel. The signals are converted by 12–bits A/D converters (50 MHz) installed in the PC. The system is controlled by software written in Lab View [17].

3. Experiment

Investigation of the atmospheric aerosol by means of multiwavelength lidar was performed during campaign, which took place in 31st August–4th September of 2004 in Wrocław (Poland). The lidar was located close to the city centre. The results are presented in Fig. 1**a**. One observe high backscattering coefficient in the boundary layer. Its altitude reached about 2200 m. The backscattering coefficient that was registered above this altitude is comparable to the value that is achieved from Rayleigh theory of the light scattering for air molecules [9]. It means that above the boundary layer containment of the aerosols in the air was negligible. The lognormal distribution of the aerosol particles retrieved using the lidar signals registered at wavelengths of 355, 532, 770 and 1064 nm as a function of altitude is shown in Fig. 1**b**. For the aerosol

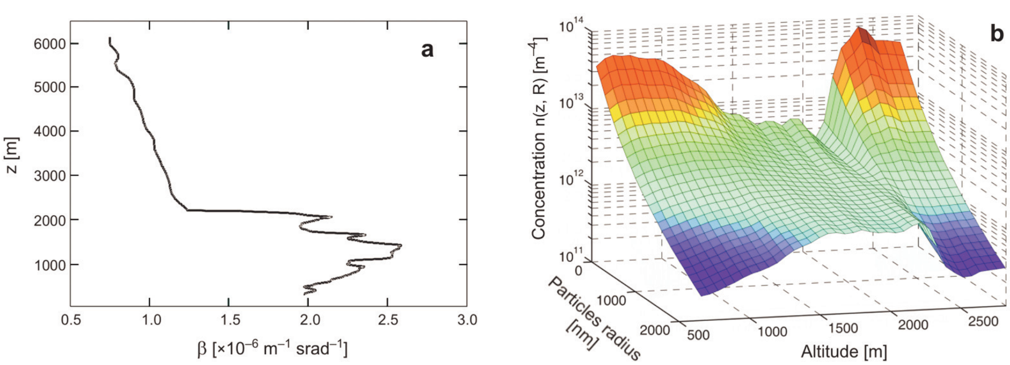

Fig. 1. Results of measurements done in Wrocław (4.09.2004, 00.57): backward scattering coefficient as a function of altitude z (**a**), distribution of aerosol particles radius R as a function of altitude (**b**).

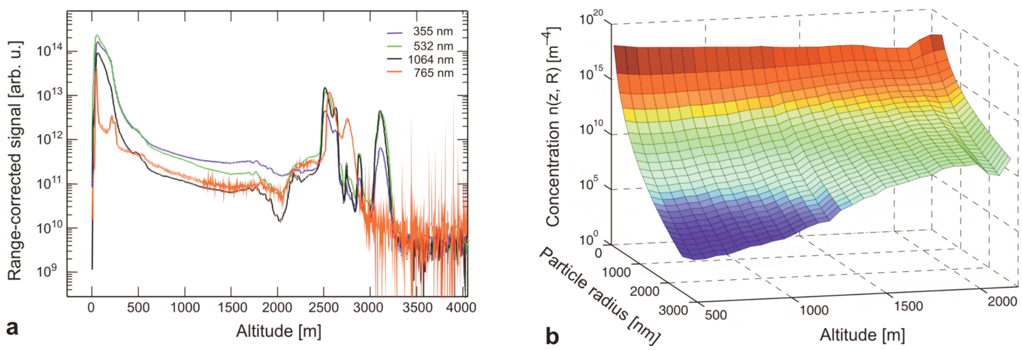

Fig. 2. Results of measurements done in Warsaw (27.04.2005, 22.00): the lidar signals $S(z, \lambda)$ as a function of altitude z for different wavelengths (**a**), distribution of aerosol particles radius R as a function of altitude (**b**).

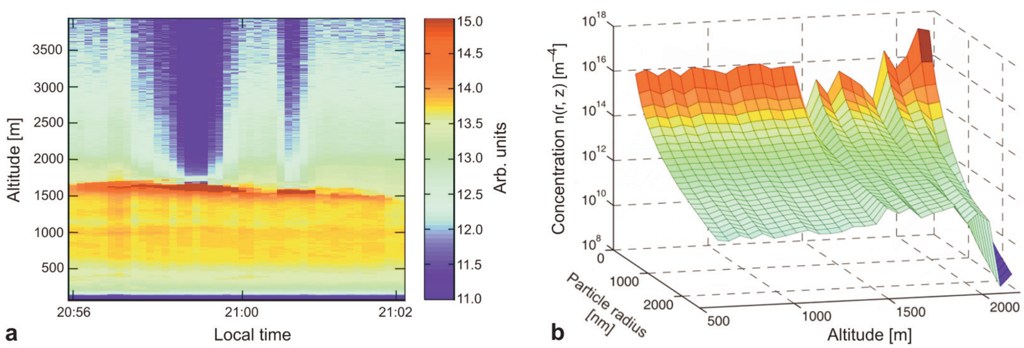

Fig. 3. Results of measurements done in Warsaw (10.05.2005): range corrected lidar signal $L(z, \lambda)$ as a function of altitude z for at 532 nm (**a**), distribution of aerosol particles radius R as a function of altitude (**b**).

located at low altitudes the particle concentration decreases quickly with the radius increase, but for the altitudes above 1200 m up to about 2000 m the number density of small particles decreases with the altitude increase while that of large particles rises. This can be explained as a condensation of water droplets that takes place at the top of the boundary layer. However this process is weak and a cloud was not formed. At the top of the boundary layer a fast decrease of $n(z, R)$ function is observed which suggests that it this region the condensation does not take place.

A similar lidar campaign was organized in spring of 2005 in Warsaw. The signals registered in 24th April are shown in Fig. 2**a**. The maximum of the signals that are observed at the altitude of about 2500 m acknowledges the existence of the cloud (*altostratus*). The aerosol particle size distribution is shown in Fig. 2**b**. One can see that the number density of large particles rises with the altitude – the closer is the distance to the cloud the larger the observed mean radius of the particles.

The next observation was performed in 10th May. In Fig. 3**a** using the colour scale the range corrected lidar signal registered at 532 nm as a function of time and altitude is presented. At the top of the boundary layer (1500–1700 m) the clouds were present sporadically. Up to the altitude about 1400 m the aerosol size distribution (Fig. 3**b**) was approximately homogeneous, which suggests that the well stabilized boundary layer was present in the atmosphere. Above this altitude $n(z, R)$ function becomes crimped. It suggests stratification of the atmosphere, which at the altitude of about 1600 m transforms to the cloud layer mentioned above. The decrease of aerosol size distribution at higher altitudes (above 1800 m) is rather due to numerical artefact.

4. Conclusions

In conclusion, we presented original results of measurements of particle size distribution of atmospheric aerosol registered by means of multiwavelength lidar as well as the original method of these data retrieving from the lidar signals. These techniques are still under development in order to provide more regular data that can serve for studies of different phenomena in the atmosphere.

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