ESTIMATION OF MASS CONCENTRATION PROFILES FOR 2-COMPONENTS EXTERNAL MIXTURES OF AEROSOLS, BASED ON MULTIWAVELENGTH DEPOLARIZATION LIDAR

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ABSTRACT

Multiwavelength depolarization Raman lidar measurements are used in this study to quantitatively separate high depolarization and low depolarization component's contribution to the 532nm backscatter coefficient, as well as to the total mass concentration. Linear volume depolarization ratio was computed using the relative amplification factor and the system-dependent molecular depolarization. The low depolarizing component was considered as urban with a LR of 80 sr and a mean δ_{part} of 3%, while for the high depolarizing component, a LR of 40 sr and a mean δpart of 35% was assumed.

OPAC classification and conversion factors were used to calculate mass concentration. We found that calibrated depolarization measurements are critical to distinguish between smoke-reach aerosol during the winter and dust-reach aerosol during the summer, as well as between elevated aerosol layers with different origins. Good agreement was found between lidar retrievals and DREAM forecasts in cases of Saharan dust intrusions.

1. INTRODUCTION

The atmosphere contains particles with an infinite variety of shapes, sizes and refractive indexes. Shape, size distribution and composition of aerosol particles influence their scattering characteristics and thus the radiative balance. Measurement of the atmospheric aerosols properties is complicated due to the composition and size of these particles [1]. Atmospheric layers often contain a mixture of different aerosol types, with completely different scattering and absorption properties.

The diffuser shape may be identified by measuring the depolarization of the initial plane polarized laser light. The depolarization products are used to distinguish between spherical (e.g. water droplets, with low depolarization ratios) or nonspherical (e.g. ice crystals, with high depolarization ratios) particles [2].

Tesche et al. [3] used the 532-nm particle depolarization ratio to quantitatively separate dust and non-dust contributions to the 532-nm backscatter coefficient. Basically, the method can be applied for any mixture of low and high-depolarizing aerosols, with apriori knowledge of the mean optical characteristics of the 2 assumed components. The method requires high accuracy input data, therefore accurate estimation of the backscatter and particle depolarization profile, as well as complementary information on the type of components.

Calculation of the backscatter coefficient profile can be done with a good accuracy even for simple backscatter lidars, where a lidar ratio has to be assumed along with the calibration in far range [4]. Particle depolarization ratio instead, is very sensitive to the instrument's function and to the calibration procedure. Several sources of systematic errors have to be considered for a non-ideal lidar system: beam purity (i.e. polarization), p-s separation, filters performances (i.e. bandwidths, stability vs. temperature), imperfection of the optical path (i.e. angle of incidence on the filter) [5]. These are difficult to estimate independently. Freudenthaler et al. [6] shows that depolarization observations can be realized highly accurate.

Multiwavelength depolarization Raman lidar measurements were done in Magurele (lat. 44.35 N, long 26.03 E, 90 m asl), 6 Km far from Bucharest, in a pre-urban area.

2. METHODOLOGY

In our study, we used depolarization lidar data to derive mass concentration for low and high-depolarizing components of aerosols. The linear particle depolarization ratio can be used to separate the contribution of each aerosols component from the total aerosol backscatter, using several apriori known parameters, such as lidar ratio and linear particle depolarization for each of the two classes.

2.1. Calibrated depolarization

The linear volume depolarization ratio δ^V is defined as the ratio of the cross–polarized lidar return signal to the parallel-polarized backscatter signal:

$$\delta^V(Z) = \frac{\beta_\perp^{total}(Z)}{\beta_\parallel^{total}(Z)} = \frac{P_\perp(Z)}{P_\parallel(Z)} \qquad (1)$$

For an ideal system, this would be equal to the ratio of the received signals in cross and parallel channels. For a non-ideal system, a calibration function taking into account the whole system depolarization effects and the differential detection on the two channels has to be considered. This function includes contribution from the

imperfect separation of the two polarization components by the polarizing beamsplitter cube (PBC), misalignment of the PBC with respect to the plane of polarization of the emitted laser beam, as well as the impurity of the laser beam [6]. It cannot be estimated for the whole range, but it has to be assumed constant and computed for a region free of aerosols (i.e. molecular contribution only). The choice of a proper calibration region, as well as of the molecular depolarization (as detected through the optoelectronic line) influences significantly the result.

Based on calibrated volume depolarization ratio, the linear particle depolarization ratio is further computed as:

$$\delta^{part} = \frac{\beta_{\perp}^{part}}{\beta_{\parallel}^{part}} = \frac{\left(1+\delta^{mol}\right)\cdot \delta^{V} \cdot R - \left(1+\delta^{V}\right)\cdot \delta^{mol}}{\left(1+\delta^{mol}\right)\cdot R - \left(1+\delta^{V}\right)} \quad (2)$$

where R is the backscatter ratio:

$$R = \frac{\beta^{mol} + \beta^{part}}{\beta^{mol}} \quad (3)$$

The system-dependent molecular depolarization δ^{mol} can be estimated for the aerosol-free regions considering the measured system function.

2.2. Separation of low and high depolarizing components

Real aerosol in the atmosphere is always a mixture of different components from different sources and processes. The number of combinations is unlimited, as both the chemical composition, and their proportions vary in time and space. Nevertheless, certain classifications are possible and generally used to distinguish for example between continental, maritime, smoke and dust.

Tesche et al. [3] developed a method to separate the optical properties of desert dust and biomass burning particles as a function of height in the mixed dust and smoke plumes. They used a multiwavelength aerosol Raman lidar to compute the backscatter coefficient profile without any assumption on the lidar ratio, and a depolarization lidar to separate the contribution of dust and smoke to the total backscatter. Certain assumptions on the mean particle depolarization ratio of pure dust and that of the pure smoke were used in this study. These were picked up from the SAMUM-1 campaign [7] and from literature: $\delta_{dust} = 0.27...0.35$, and $\delta_{smoke} = 0.02...0.15$ at 532 nm.

We used this method in a more general way to separate low and high depolarizing components of mixed aerosol, based on:

$$\beta_{high}^{part} = \beta_{total}^{part} \cdot \frac{\left(\delta_{total}^{part} - \delta_{low}^{part}\right)\cdot\left(1+\delta_{high}^{part}\right)}{\left(\delta_{high}^{part} - \delta_{low}^{part}\right)\cdot\left(1+\delta_{total}^{part}\right)}, \quad (4)$$

$$\beta_{high}^{part} + \beta_{low}^{part} = \beta_{total}^{part}$$

where the high depolarizing component can be mineral or volcanic dust, and the low depolarizing component can be smoke, biomass burning or urban with a high content of soot. Only statistical errors propagated through the calculus chain were considered to estimate the accuracy of the retrieved component's backscatter.

2.3. Calculation of extinction-to mass concentration conversion factors

OPAC (Optical Properties of Aerosols and Clouds) provides optical properties in the solar and terrestrial spectral range of atmospheric particulate matter, calculated on the basis of the microphysical data (size distribution and spectral refractive index) under the assumption of spherical particles.

Aerosols are modeled as components, each of them representative for a certain origin, that is, an internal mixture of all chemical substances that have a similar origin. These components may be externally mixed to form aerosol types, which mean that there is no physical or chemical interaction between particles of different components. Main characteristics of each component are given in [8]. Various combinations of components and various humidity values can be used. The software package also allows calculation of derived optical properties like mass extinction coefficients and Ångström coefficients. Mixing in OPAC is based on the particle number densities of the components. Custom aerosols can be defined, by selecting up to 5 components and providing number density for each of them.

OPAC was used to compute mass-extinction ratio, asymmetry parameter and lidar ratio for various aerosol types, in the default and custom mode. Mineral dust and volcanic dust were considered as high-depolarization components of the mixed aerosol layers, while urban with high soot content was considered as low-depolarization component. Volcanic dust was obtained from OPAC's classification of mineral dust, by adding sulfates in variable proportions. All parameters were computed for dry (0% relative humidity) conditions in the troposphere, and for humid (50% relative humidity) conditions in the Planetary Boundary Layer (PBL).

3. RESULTS AND DISCUSSIONS

The dependence of the mass-extinction ratio η to the aerosol type and humidity conform OPAC is shown in Fig.1. We considered an increased proportion of soot for the urban-type aerosol, as well as of the mineral transported and sulfates for mineral dust and volcanic dust respectively, in order to test the sensitivity of the output parameters. It can be seen from the graphs below, that the variation of the mineral transported component does not influence the behavior of the output parameters, which are not sensitive to the humidity either. On the other hand, the proportion of soot in the internal mixture of urban aerosol, and the humidity, have an important effect. The same applies for volcanic

dust. Considering dry conditions for the tropospheric layers, and a high content of soot (83%) internally mixed with water soluble (16.99%) and insoluble (0.01%) components, we obtained for the urban aerosol $\eta = 0.9 \pm 0.1$ g/m^2. Similarly, for the volcanic dust with (30%) sulfates, (60%) water soluble and (10%) mineral components, we obtained $\eta = 1.3 \pm 0.1$ g/m^2. For the mineral dust, we assumed a significant component of mineral transported, which led to: $\eta = 2.6 \pm 0.05$ g/m^2.These values are similar to recent studies in literature, e.g. [9]
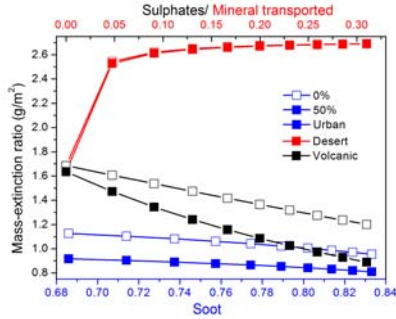

Figure 1. Variation of mass-extinction ratio η with the proportion of the critical component (soot for urban, mineral transported for mineral dust and sulfates for volcanic dust) for 0% (empty markers) and 50% (plain markers) relative humidity.
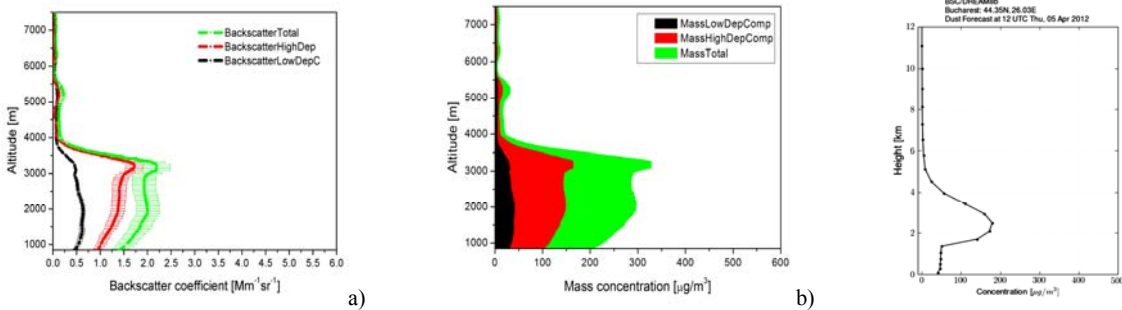
Values obtained by direct modeling were used first to separate the contribution of the low depolarizing (urban) and high depolarizing (mineral and volcanic dust) components to the total backscatter, as described above. Considering the values of asymmetry parameter g and lidar ratio *LR* obtained for mineral and volcanic dust, which are very similar, no distinction was made at this stage between the two. Fig. 2a presents profiles obtained for April 5th, 2012, when dense and persistent Saharan dust air masses were present at our station. HYSPLIT backtrajectories show that layers at 1.5, 2.5 and 4.0 Km are all originating from different Saharan regions. With this confirmation, we can assume for the next step that the high-depolarizing component of the mixed aerosol is typical mineral dust, and using the mass-extinction ratio for this aerosol type, we obtain mass concentration profiles (Fig. 2b). It can be seen that the concentration of low-depolarization particles is almost constant with altitude (black in Fig.2b), while the concentration of dust particles is significant up to 4 Km (red in Fig.2b). Dust REgional Atmospheric Model (DREAM) also pointed a strong transport of Saharan dust to Europe for this period. Forecasted dust concentration profile at Bucharest station is shown in Fig. 2c. Profiles from DREAM and lidar agree very well from 2 Km, reaching up to 0.2 mg/m^3. In the PBL, DREAM underestimates the concentration of dust, which is due to superposition of dust produced by local sources.

a)   b)   c)

Figure 2. April 5th, 2012: Separation of a) backscatter coefficient, and b) mass concentration profiles for a case of Saharan dust mixed with urban aerosols; c) Dust mass concentration profile forecasted by DREAM

A different case was measured on April 21st, 2010, when volcanic dust from Iceland was transported in the troposphere, and it was mixed with continental aerosols. For this case, differences between the parameters for mineral and volcanic dust, although not important for the separation of the component's backscatter, are important when converting from extinction to mass concentration. Fig. 3 presents the profiles obtained for the high depolarizing component and the total mass concentration, when applying η for mineral dust ($\eta = 2.6 \pm 0.05$ g/m^2) and for volcanic dust ($\eta = 1.3 \pm 0.1$ g/m^2) with 30% content of sulfates. The profiles for low-depolarizing component are the same for both cases (not shown).
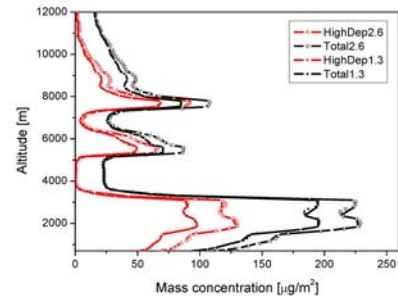

Figure 3. Comparison in terms of total (black) and high depolarizing component (red) mass concentration, for different compositions (triangles: mineral dust; dots: volcanic dust)

Differences between the 2 cases considered are approx. 34% for the high-depolarizing component's mass concentration, and approx. 20% for the total (Fig.3). It is therefore important for quantitative retrievals to make distinction between different types of aerosols and apply the correct conversion factor. However, averaged values can be used for similar aerosols when a rapid estimation of the mass concentration is needed, because the method is less sensitive to the aerosol composition than to the lidar inversion errors: calibration of the backscatter and depolarization, assumptions on lidar ratio (when elastic backscatter is used).

4. CONCLUSIONS

Our study refers to a combination of measured and modeled data, used to calculate mass concentration profiles for mixed aerosols. The mixture considered consists of 2 components with significantly different depolarization properties, e.g. smoke + mineral dust. We demonstrated that the retrieval of mass concentration profiles from multiwavelength depolarization Raman lidar is possible with sufficient accuracy, but dependent on an appropriate calibration of the depolarization and careful selection of the mass-extinction conversion factors.

Variations of the proportions of the internally mixed components for different aerosol classes and their effect on critical parameters such as mass-extinction ratio, lidar ratio and asymmetry parameter were studied for dry (free troposphere) and humid (PBL) conditions. Simulations showed that for dry conditions in the upper layers, the mass-extinction ratio decreases with the proportion of soot for urban-type aerosol (1.15 to 0.9 $\pm$ 0.1 g/m^2), increases sharply with the content of mineral transported (1.7 to 2.6 $\pm$ 0.05 g/m^2), and decreases slowly with the content of sulfates for volcanic dust aerosols (1.7 to 1.3 $\pm$ 0.1 g/m^2). Properties of the mineral dust are much less sensitive to an increase in humidity than those of the urban and volcanic dust aerosols, for which different values of the optical parameters have to be used bellow and above PBL.

Several cases were analyzed and mass concentration profiles were retrieved for mineral dust mixed with urban, and volcanic dust mixed with urban. Good agreement was found between our retrievals and DREAM forecasts in cases of Saharan dust intrusions. The sensitivity of the retrieval with respect to the apriori estimation of aerosol type was tested on a case of volcanic dust. Differences in the total mass concentration obtained when assuming mineral dust and when assuming volcanic dust was found to be less than 20%, which is acceptable for certain applications, such as aviation safety.

Main advantage of the method described is that is simple and fast, providing sufficient information for real time assessment. It is based on the hypothesis that any mixture in the atmosphere can be described as a combination of low-depolarizing and high-depolarizing particles of a particular type. Within the same category, the uncertainty in the estimation of particle depolarization and mass-extinction ratio is sufficiently low (<35%), therefore not critical for the calculation of the mass concentration. Nevertheless, the accuracy is also related to the usual calibration errors (backscatter, particle depolarization), which have to be considered.

6. REFERENCES

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