THREE WAVELENGTHS LIDAR MEASUREMENTS FOR ATMOSPHERIC AEROSOL CHARACTERIZATION

O. Cavalieri(1), M. R. Perrone(1), F. De Tomasi(1), G. P. Gobbi(2)

(1)CNISM, Dipartimento di Fisica, Università del Salento, Lecce (Italy), perrone@le.infn.it
(2)Istituto di Scienze dell'Atmosfera e del Clima, ISAC-CNR, Rome (Italy), g.gobbi@isac.cnr.it

ABSTRACT

Three wavelengths (355 nm, 532 nm and 1064 nm) lidar measurements have been used to characterize the dependence on altitude of the aerosol size distribution by the combined analysis of the extinction Ångstrom coefficient α(355 nm, 1064 nm) and the Angstrom exponent difference δα = α(355 nm, 532 nm) - α(532 nm, 1064 nm). Lidar measurements were performed at the Physics Department of Universita' del Salento (Lecce), in south eastern Italy. This monitoring site is affected by air masses from the Atlantic, continental Europe, the Mediterranean Sea, and northern Africa. As a consequence, mixed advection patterns leading to aerosol properties quite dependent on altitude are dominant. Results on lidar measurements performed on 28 July, 2011 are presented and discussed as a case study.

1. INTRODUCTION

Atmospheric aerosols play an important role in controlling the Earth's climate. In the attempts to quantify such an impact, main difficulties arise from the large spatial and temporal variability of aerosol concentrations and related physical, optical, and chemical properties. Many efforts have been undertaken to develop new methodologies and/or to define new parameters to characterize aerosol properties. The Ångstrom exponent (α) that is calculated from the spectral dependence of the aerosol optical depth (AOD) is commonly used as a good indicator of the dominant size of the atmospheric aerosol. Submicron accumulation-mode particles are characterized by α > 1 whereas super micron particles are characterized by α < 1. More specifically, α values near zero (or even negative) correspond to coarse mode particles such as sea spray and desert dust. However, α cannot provide information on the relative contribution of coarse and fine mode particles if different aerosol types are present in the air column. Several studies based on sun photometer measurements have shown that the spectral variation of α can provide additional information about the aerosol size distribution, as it is summarized in [1]. A straight-forward graphical framework to classify aerosol properties using dirct-sun sunphotometer observations was introduced in [2]. The method relies on the combined analysis of the Ångstrom exponent and its spectral curvature δα/δλ and it has been shown that δα versus α scatterplots can allow inferring the aerosol fine mode size (R_f) and the fine-mode fractional contribution to the total AOD. Direct-sun sunphotometer observations from the AERONET [3] were used in [2] to demonstrate the feasibility of the introduced classification scheme. More specifically, α was calculated from the AOD values at 440 nm and 870 nm (α(440, 870)) and the Ångstrom exponent difference δα = α(440, 675)- α(675,870) was defined as a measure of the Angstrom exponent curvature δα/δλ. The method applied to AERONET climatological data from three continents has allowed to identify various aerosol properties peculiar to these locations.
Three wavelengths (355 nm, 532 nm and 1064 nm) lidar measurements have been used in this study to retrieve vertical resolved α-δα values and infer the dependence on altitude of the aerosol size distribution accordingly to [2]. A short overview of the lidar system is provided in Section 2. Lidar measurements and analytical results are discussed in Section 3. Summary and conclusion are presented in Section 4.

2. LIDAR SYSTEM AND OPERATION

The ground-based lidar system at the Physics Department of Universita' del Salento (Lecce) in south eastern Italy (40.4°N; 18.1°E), operates within the European Aerosol Research LIdar NETwork (EARLINET) since May 2000 [4]. The lidar system nowadays employs an f/4 Newtonian telescope with a 30-cm-diameter mirror and a frequency-tripled Nd-YAG laser delivering pulses at 355, 532, and 1064 nm, respectively, at a repetition rate of 30 Hz. It is estimated to achieve full overlap between 0.5 – 1.0 km above the ground level. The lidar system has been designed to derive vertical profiles of the aerosol backscatter (β(z)) coefficient at 355 nm, 532 nm and 1064 nm, respectively and of the volume depolarization ratio (DR(z)) at 355 nm during day time measurements. Backscatter lidar measurements suffer from a need to assume an aerosol-to-backscatter ratio LR (also referred to as the aerosol-lidar ratio) to solve the ill-posed problem of the lidar equation and extract aerosol extinction and backscatter coefficient profiles. Altitude independent lidar ratios have been used in this study to retrieve β(z) profiles. More specifically, LR values were chosen in order to obtain vertically integrated AODs equal (within experimental uncertainties) to corresponding AODs retrieved from sun photometer measurements collocated in space and time. Notice that an AERONET sun/sky photometer operates at the lidar site since the year 2003. The statistical uncertainties of β(z) values were calculated from the error propagation law by assuming a Poisson noise on the lidar signals. Extinction coefficient (ε(z)) profiles were calculated from β(z) profiles in accordance with the following relationship ε(z) = LR x β(z). Then, aerosol optical depths were retrieved from ε(z) profiles by assuming that ε(z) values do not vary with altitude below the height where the lidar system is estimated to achieve full overlap. This height (0.5 – 1.0 km AGL) is of the order of the planetary boundary layer (PBL) height. So, we can assume that aerosol particles are well mixed within the PBL. The volume depolarization ratio is the ratio of cross-polarized to parallel-polarized backscatter coefficient and under our experimental conditions, takes a value of 0.0045 in a pure molecular atmosphere and higher values in presence of non spherical particles such as desert dust particles. More details on the lidar system and data analysis are reported in [5].

3. RESULTS AND DISCUSSION

The lidar measurements performed on 28 July, 2011 from 12:30 UTC until 13:00 UTC are analyzed in this section. This day was characterized by a mixed advection pattern in accordance with the seven-day analytical back trajectories from the Hybrid Single Particle Lagrangian Integrated Trajectory Model (HYSPLIT) [6]. Figure 1 shows that air masses from north western Africa and the Mediterranean Sea were advected at the lidar site up to ~ 3 km above the ground level (AGL). While, air masses from the Atlantic and Western Europe were advected from 3 km up to ~ 5 km AGL.

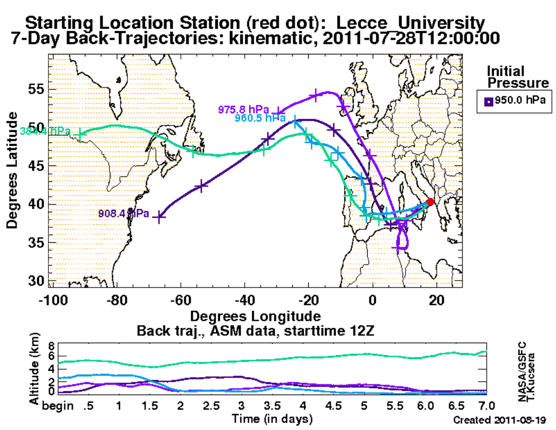

Figure 1. Seven-day analytical back trajectories from HYSPLIT for 28 July 2011 at 12:00 UTC.

The backscatter coefficient profiles retrieved from lidar measurements at 355, 532, and 1064 nm, respectively are plotted in Fig. 2. An altitude independent lidar

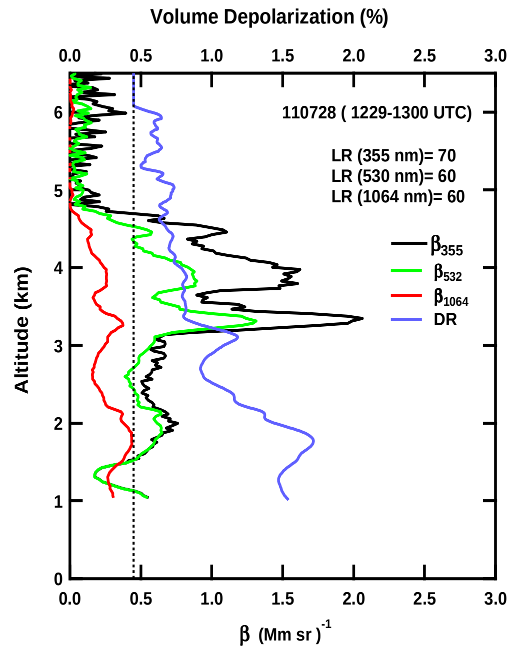

Figure 2. Backscatter coefficient profiles at 355 nm (β_{355}), 532 nm (β_{532}), and 1064 nm (β_{1064}) retrieved from lidar measurements performed on 28 July, 2011 from 12:29 UTC till 13:00 UTC. The blue line represents the volume depolarization vertical profile.

ratio LR = 70 sr was used to retrieve the β(z) profiles at 355 nm. While, an altitude independent lidar ratio LR = 60 sr was used to retrieve the β(z) profiles at 532 and 1064 nm, respectively. AODs calculated from extinction lidar profiles were equal to 0.31, 0.21, and 0.07 at 355, 532, and 1064 nm, respectively. The blue line in Fig. 2 shows the DR(z) profile. DR(z) percentage values vary within 1-1.7% up to about 3 km AGL and take values within the 0.6-0.9% range from 3 up to 6 km AGL. The higher DR(z) values up to about 3 km AGL are likely due to the contribution of desert dust particles advected from north western Africa and/or to sea-salt particles from the Mediterranean Sea, in accordance with Fig. 1. While, the smaller DR(z) values retrieved at altitudes larger than 3 km indicate that the contribution of spherical particles, as those due to anthropogenic pollution, was larger at these altitudes. The air masses from the Atlantic which have crossed Western Europe before getting to the lidar site have likely been responsible for the advection of anthropogenic pollution to the lidar site (Fig. 1). Extinction coefficients averaged over 0.5 km altitude range were used in this study to calculated Ångstrom exponents for different lidar wavelength pairs. Fig. 3 (black line) shows the α(z) profile calculated from the ε(z) profiles at 355 nm and 1064 nm. α(z) varies with altitude since the dominant size of the atmospheric particles varies with altitude as the DR(z) profile has also suggested. More specifically, α(z) varies within 0.18-1.1 and 1.6-1.9 from 1 up to 3.0 km and from 3 km up to 4.5 km AGL, respectively. These results indicate that large particles as those due to sea spray and/or desert dust are dominant up to ~ 3 km AGL. While, α values retrieved above 3 km, which are larger than 1.5, indicate a dominating presence of fine-mode particles such as smoke or urban aerosol. The Ångstrom exponent difference δα = α(355 nm, 532 nm) - α(532 nm, 1064 nm) has been used in this study as a measure of the Ångstrom exponent curvature δα/δλ. Fig. 3 (red line) shows the vertical profile of δα which varies with altitude from -0.48 up to 0.35.

According to [2], a classification framework was built to infer aerosol fine mode size (R_f) and fractional contribution to total AOD (η) from the scatterplot of the δα versus α data points retrieved from lidar measurements. To this end, by Mie calculations we determined reference points corresponding to bimodal size distributions characterized by a variety of fine mode (R_f) and coarse mode (R_c) modal radii combined to lead to prescribed fractions (η) of the fine mode to total AOD at 553 nm. The used real (n) and imaginary

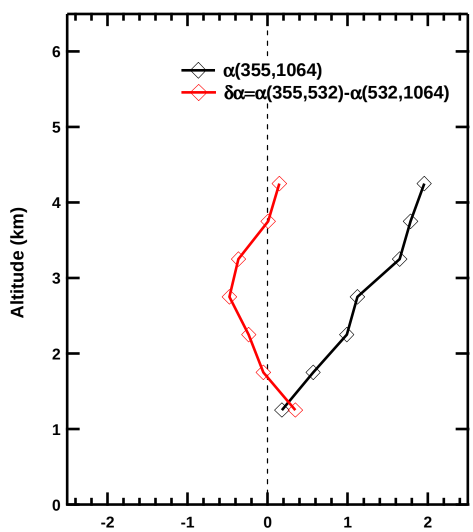

Figure 3. Vertical profile of α(z) calculated from the extinction profiles at 355 nm and 1064 nm and of the Ångstrom exponent difference: δα = α(355 nm, 532 nm) - α(532 nm, 1064 nm).

(k) refractive index values at the lidar wavelengths are reported in Tab. 1. These values were retrieved from AERONET climatological data for the Lecce lidar site. Grid points relevant to Mie calculations plotted in the α-δα coordinates are given in Fig. 4 in addition to the experimentally determined α-δα values (red dots). δα and α values are equal to 0.35 and 0.18, respectively in the aerosol layer from 1.0 up to -1.5 km AGL. Then, we observe from Fig. 4 that the aerosol particles located in that layer are characterized by a bimodal size distribution with fine mode modal radius $R_f \cong 0.1$ µm. The fine mode fraction η takes a value slightly

Table 1. Real (n) and imaginary (k) refractive index values used in the simulations of the aerosol classification scheme

Wavelength (nm)	n	k
355	1.442	0.0080
532	1.455	0.0047
1064	1.460	0.0024

smaller than 30% in that layer. The contribution of fine mode particles increases with altitude as α values, which increase with altitude, reveal (Fig. 3). More specifically, Fig. 4 shows that η which is slightly larger than 50% (α = 0.57, δα = -0.05) within the 1.5-2.0 km altitude range, increases up to about 80% (α = 1.1, δα = -0.48) within the 2.5- 3 km altitude range, and takes values close to 90% above 3 km from the ground level. Aerosol particles located from about 3 up to 4.5 km AGL are also characterized by bimodal size distributions with smaller fine mode modal radii (0.05 µm $<R_f$ 0.12 µm). These results are supported by both the analytical back trajectories and the volume depolarization ratio profile and indicate that on that day the contribution of desert dust and/or sea-salt particles advected over south eastern Italy decreased with altitude. While, the contribution of fine mode particles has increased with altitude.

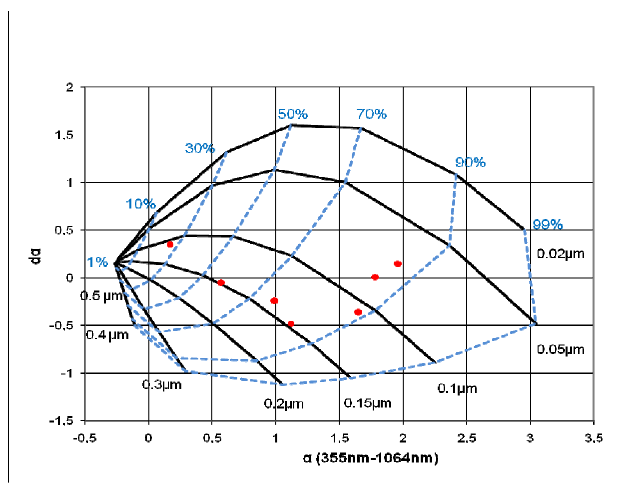

Figure 4. Simulations of the classification of the aerosol properties according to [2]. Black lines refer to selected R_f values. Blue dotted line represent selected η values. The red dots represent the data points plotted in Fig. 3.

It is also worth noting from Fig. 4 that the aerosol particles located up to about 3 km AGL were characterized by bimodal size distributions having the fine mode modal radius R_f within the 0.1-0.15 μm range.

4. SUMMARY AND CONCLUSION

The straight-forward graphical framework to classify aerosol properties developed in [2] has for the first time been applied to three wavelength lidar measurements to infer altitude resolved parameters of bimodal size distributions representing atmospheric particles. The methodology has been applied to lidar measurements performed on 28 July, 2011 from 12:30 UTC until 13:00 UTC over Lecce. Air masses from western Africa and the Mediterranean Sea were mainly advected on this day over the lidar site up to about 3 km AGL. While, air masses from the Atlantic, which have crossed north western Europe before getting to the lidar site, were advected from 3 up to about 5 km AGL. So, aerosol microphysical properties were quite dependent on altitude on this day, as the volume depolarization vertical profile retrieved from lidar measurements has also indicated. It has been shown that the scatterplot of $\delta\alpha$ versus α data points retrieved from lidar measurements has allowed to infer the dependence on altitude of the main parameters (η and R_f) which characterize aerosol size distributions. These results indicate that applying the introduced classification scheme to three-wavelength lidar observations has good potential for providing an altitude-resolved size characterization of atmospheric aerosols.

5. ACKNOWLEDGMENTS

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