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5 fine-mode and coarse-mode dust is requested (Kok, 2011; Zhang et al., 2013). Fine and coarse dust particles influence the Earth's radiation budget, cloud processes, and environmental conditions in a different way (Nabat et al., 2012; Mahowald et al., 2014). The optical properties and radiative impact are widely controlled by coarse-mode dust particles. However, 20–25% of the dust-related optical depth is caused by fine-mode dust according to Aerosol Robotic Network (AERONET) sun/sky photometer observations (see Sect. 3). Regarding the influence on cloud processes, coarse dust particles belong to the most favorable cloud condensation and ice nuclei (DeMott et al., 2010). Fine-mode dust particles, on the other hand side, can have a significant impact on air quality, defined in PM (particulate matter) aerosol levels and even may sometimes dominate PM_{1,0} (particles with diameters < 1.0 μm) observation at sites close to deserts such as Cyprus. As an example, on 1 April 2013 the 500 nm aerosol particle optical thickness (AOT) increased from 1 to 4 between 08:00 and 12:00 UTC over Cyprus during a rather strong Saharan dust outbreak. These large AOT values indicate fine-mode dust mass concentrations of 35–140 μg m⁻³ in the tropospheric column up to 4–5 km height which is 10–15% of the total dust mass concentration.

10 In this contribution we present to our knowledge the first attempt to use empirical knowledge on the light depolarizing properties of fine-mode and coarse-mode dust (Sakai et al., 2010) in the interpretation of polarization lidar observations with the goal to separate fine-mode dust, coarse-mode dust, and remaining non-dust aerosol components. So far, polarization lidars are used to identify dust and to separate backscatter and extinction coefficients of dust and non-dust aerosol (e.g., Sugimoto and Lee, 2006; Nishizawa et al., 2007; Tesche et al., 2009, 2011; Groß et al., 2011; Ansmann et al., 2011a, 2012). The most important parameter in these studies is the so-called particle linear depolarization ratio. The laser transmits linearly polarized laser pulses and the receiving unit detects the parallel- and cross-polarized signal components with respect to the plane of laser polarization. The ratio of calibrated cross-polarized to parallel-polarized signal yields the volume linear depolarization ratio from which the particle depolarization ratio can be computed (e.g., Tesche et al., 2009).

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model. The irregularly shaped dust particles are assumed to be spheroidal dust particles. This approach works well in the case of the analysis of pure sun/sky photometer data (Dubovik et al., 2006). However, it is shown by Wagner et al. (2013) that this particle shape model introduces significant uncertainties in the LIRIC aerosol products when applied to lidar backscatter returns, i.e., to light scattering information for a scattering angle of exactly 180° . Products of the LIRIC data analysis are height profiles of particle backscatter and extinction coefficients at the three wavelengths, and particle volume and mass concentration profiles separately for fine-mode and coarse-mode particles. GARRLiC is an extended version of LIRIC and pursues an even deeper synergy of lidar and radiometer data in the retrievals. To apply the LIRIC and GARRLiC methods, lidar and photometer observation have to be performed simultaneously. Thus, cloudfree conditions are required.

In contrast, the POLIPHON approach is designed to explicitly avoid the use of a particles shape model and also a strong dependence on photometer observations. POLIPHON is applicable even at cloudy conditions which often occur during dust outbreaks. The technique is based on measured 180° light-depolarization characteristics for dust aerosol particles. This approach, originally developed for the separation of non-dust and dust fractions (here denoted as one-step POLIPHON method) is extended to allow even a separation of fine-mode dust from coarse-mode dust (two-step POLIPHON technique). The comparably simple and robust method for the retrieval of optical properties, volume, and mass concentrations is outlined in Sect. 4. POLIPHON belongs to the family of well established lidar aerosol-typing methods which built on empirically gathered optical aerosol properties. Examples of observations and applications are presented by Groß et al. (2013) and Burton et al. (2014).

In Sect. 2, the lidar and photometer instruments are briefly described. Section 3 presents an overview of typical optical properties of desert dust in terms of fine-mode and coarse-mode dust characteristics to emphasize the need for the development of more sophisticated fine and coarse dust separation techniques. The extended two-step POLIPHON methodology is outlined in Sect. 4. The new retrieval scheme is then

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The SAMUM-1 conversion factors in Fig. 4 are in good agreement with simulations of Barnaba and Gobbi (2004). Based on several thousands of realistic combinations of particle number concentration, size distribution, and refractive index characteristics, the volume-to-extinction ratio for dust size distributions dominated by supermicron dust particles typically ranges from $0.6\text{--}0.9 \times 10^{-6}$ m for 532 nm wavelength. The maximum value is 1.0×10^{-6} m for very large dust particles. For submicron-dust-dominated particle ensembles, the conversion factors are between 0.25×10^{-6} m and 0.4×10^{-6} m.

4 Two-step POLIPHON method

4.1 Theoretical background

The new two-step POLIPHON method uses the same separation technique as the one-step approach. The latter method is described in detail by Tesche et al. (2009) for a two-aerosol component mixture of desert dust and biomass burning smoke. We briefly introduce the one-step approach and use the notation of Tesche et al. (2009).

The procedure to separate dust-related and smoke-related (or more general non-dust-related) profiles of backscattering starts from the equation for the particle depolarization ratio

$$\delta_p = \frac{\beta_{nd}^\perp + \beta_d^\perp}{\beta_{nd}^\parallel + \beta_d^\parallel}. \quad (1)$$

β^\perp and β^\parallel are so-called cross and parallel-polarized particle backscatter coefficients which can in principle be computed from the lidar return signals detected with the cross-polarized and parallel-polarized signal channels. The indices d and nd denote dust and non-dust aerosol components, respectively. The sum of all four backscatter contributions in Eq. (1) yields the total particle backscatter coefficient β_p . The overall particle backscatter coefficient β_p is calculated in the way described in Sect. 2.1.

As shown by Tesche et al. (2009) the particle depolarization ratio can be expressed by

$$\delta_p = \frac{\beta_{nd}\delta_{nd}(1 + \delta_d) + \beta_d\delta_d(1 + \delta_{nd})}{\beta_{nd}(1 + \delta_d) + \beta_d(1 + \delta_{nd})} \quad (2)$$

with the dust and non-dust depolarization ratios δ_d and δ_{nd} , respectively. After substituting β_{nd} by $\beta_p - \beta_d$, we solve the resulting equations to obtain a solution for β_d :

$$\beta_d = \beta_p \frac{(\delta_p - \delta_{nd})(1 + \delta_d)}{(\delta_d - \delta_{nd})(1 + \delta_p)} \quad \text{for } \delta_p > \delta_{nd}, \quad (3)$$

$$\beta_d = 0 \quad \text{for } \delta_p \leq \delta_{nd}. \quad (4)$$

The non-dust particle backscatter coefficient is then obtained from $\beta_p - \beta_d$.

In the computation after Eqs. (1)–(3) we need to estimate the non-dust depolarization ratio δ_{nd} and the dust depolarization ratio δ_d . The particle depolarization ratio for Saharan dust of $\delta_d = 0.31 \pm 0.03$ (Freudenthaler et al., 2009; Groß et al., 2011) is in good agreement with the one for Asian dust (Sugimoto et al., 2003; Shimizu et al., 2004). The non-dust-related depolarization ratio δ_{nd} may vary from 0.015–0.15 according to the literature. The values accumulate around 0.05 according to published values (Murayama et al., 1999; Murayama et al., 2004; Fiebig et al., 2002; Sugimoto et al., 2003; Müller et al., 2005, 2007; Sugimoto and Lee, 2006; Chen et al., 2007; Heese and Wiegner, 2008).

In Fig. 5, the one-step and the two-step methods are illustrated. In our two-step approach we now introduce three types of aerosols: non-dust particles causing a particle linear depolarization ratio of $\delta_{nd} = 0.05$, fine-mode dust causing a depolarization ratio of $\delta_{df} = 0.16$, and coarse-mode desert dust causing a particle depolarization ratio of $\delta_{dc} = 0.39$. Our focus is on lofted free tropospheric aerosol. We assume the absence of coarse marine particles here so that spherical particles exclusively belong to the fine mode particle fraction.

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The basic equation of our two-step retrieval scheme is

$$\delta_p = \frac{\beta_{nd}^\perp + \beta_{df}^\perp + \beta_{dc}^\perp}{\beta_{nd}^\parallel + \beta_{df}^\parallel + \beta_{dc}^\parallel}. \quad (5)$$

In each of the two steps, two kinds of aerosols are separated (see Fig. 5). In the first round we start from

$$\delta_p = \frac{\beta_{pf}^\perp + \beta_{dc}^\perp}{\beta_{pf}^\parallel + \beta_{dc}^\parallel}. \quad (6)$$

Index pf indicates fine-mode particles as a whole, i.e., spherical as well as non-spherical sub-micrometer particles with radii of ≤ 500 nm. Analog to the step from Eq. (1) to Eq. (3), here we obtain for the coarse dust backscatter coefficient

$$\beta_{dc} = \beta_p \frac{(\delta_p - \delta_{pf, \max})(1 + \delta_{dc})}{(\delta_{dc} - \delta_{pf, \max})(1 + \delta_p)} \quad \text{for } \delta_p > \delta_{pf, \max}, \quad (7)$$

$$\beta_{dc} = 0 \quad \text{for } \delta_p \leq \delta_{pf, \max}. \quad (8)$$

We assume $\delta_{pf, \max} = 0.12$ and $\delta_{dc} = 0.39$ in Eq. (7) to determine the coarse-mode contribution β_{dc} to β_p . Such a fine-mode characterizing depolarization ratio of $\delta_{pf, \max} = 0.12$ instead of 0.16 for fine dust after Sakai et al. (2010) assumes that the fine-mode always includes a certain fraction (here about 25%) of anthropogenic haze and/or biomass burning smoke. All δ_p values between $\delta_{pf, \max} = 0.12$ and $\delta_{dc} = 0.39$ indicate mixtures of coarse dust and fine (spherical and non-spherical) particles (see Fig. 5).

We recommend to generally assume a 25% contribution of fine spherical particle to the overall fine-mode fraction (in the polluted Northern Hemisphere) when pronounced dust layers are detected and the two-step method is going to be applied. Even for strong desert dust outbreak plumes, $\delta_{pf, \max} = 0.16$ (assuming that only pure fine dust

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contributes to FMF) in the first round is probably too large. During situations with traces of soil or desert dust in lofted aerosol layers, indicated by low depolarization ratios of about 0.05–0.10, the two-step method will only deliver backscatter coefficients for fine-mode particles when using $\delta_{\text{pf, max}} = 0.12$. Then, the solutions of the one-step and two-step methods may be compared. The range of fine and coarse dust profiles obtained with the two methods may be used as the range of possible solutions in terms of fine and coarse dust backscatter coefficients.

Before we can start the second round, i.e., the separation of fine spherical from fine non-spherical particles) we have to remove the optical effects of coarse-mode dust from the total particle backscatter coefficient and the particle depolarization ratio. The profile of the fine-mode-related backscatter coefficient is given by

$$\beta_{\text{pf}} = \beta_{\text{p}} - \beta_{\text{dc}}. \quad (9)$$

Regarding the removal of the coarse-mode depolarization effect, one may use Eq. (11) of Tesche et al. (2009), here in the form of

$$\delta_{\text{pf}} = \frac{\beta_{\text{dc}}(\delta_{\text{p}} - \delta_{\text{dc}}) + \beta_{\text{pf}}\delta_{\text{p}}(1 + \delta_{\text{dc}})}{\beta_{\text{dc}}(\delta_{\text{dc}} - \delta_{\text{p}}) + \beta_{\text{pf}}(1 + \delta_{\text{dc}})}. \quad (10)$$

However, it can be shown that solving of Eq. (10) is equivalent to the simple setting according to

$$\delta_{\text{pf}} = \delta_{\text{p}} \quad \text{for } \delta_{\text{p}} \leq 0.12, \quad (11)$$

$$\delta_{\text{pf}} = 0.12 \quad \text{for } \delta_{\text{p}} > 0.12 \quad (12)$$

in our case with $\delta_{\text{pf, max}} = 0.12$. The profile for δ_{pf} after Eqs. (11) and (12) corresponds to the fine-mode backscatter profile after Eq. (9). An example for the coarse-dust-corrected profiles of the particle backscatter coefficient and depolarization ratio is shown in Sect. 4.

In the second round, we set the maximum fine-mode dust depolarization ratio (upper boundary of possible fine-mode depolarization ratios) to $\delta_{df} = 0.16$ as suggested by Sakai et al. (2010). Since the maximum depolarization value is still assumed to be 0.12, the assumption implies again that the fine-mode dust fraction is of the order of 75% and the residual part consists of haze and smoke particles.

The second round starts from

$$\delta_{pf} = \frac{\beta_{nd}^{\perp} + \beta_{df}^{\perp}}{\beta_{nd}^{\parallel} + \beta_{df}^{\parallel}}. \quad (13)$$

Analog to the step from Eqs. (6) to (7), now we obtain

$$\beta_{df} = \beta_{pf} \frac{(\delta_{pf} - \delta_{nd})(1 + \delta_{df})}{(\delta_{df} - \delta_{nd})(1 + \delta_{pf})} \quad \text{for } \delta_p > \delta_{nd}, \quad (14)$$

$$\beta_{df} = 0 \quad \text{for } \delta_p \leq \delta_{nd}. \quad (15)$$

which can be solved by assuming that the non-dust depolarization ratio is $\delta_{nd} = 0.05$ and the pure fine-mode dust depolarization ratio is $\delta_{df} = 0.16$ (see Fig. 5). Finally, we obtain the fine-mode backscatter coefficient for the remaining spherical particles,

$$\beta_{nd} = \beta_{pf} - \beta_{df}. \quad (16)$$

By using characteristic lidar ratios S_{df} , S_{dc} , and S_{nd} in Table 1 (for the Cyprus area in this study), we can convert the retrieved backscatter profiles β_{df} , β_{dc} , and β_{nd} , into respective particle extinction coefficient profiles for the three resolved aerosol components.

In the final step of the two-step POLIPHON retrieval, the set of particle backscatter and extinction coefficients are converted into particle volume and mass concentrations. The mass concentrations m_{df} , m_{dc} , and m_{nd} for fine dust, coarse dust, and non-dust

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particles, respectively, can be obtained from the backscatter coefficients β_{df} , β_{dc} , and β_{nd} by using the following relationships (Ansmann et al., 2011a, 2012):

$$m_{\text{df}} = \rho_{\text{d}}(v_{\text{df}}/\tau_{\text{df}})\beta_{\text{df}}S_{\text{df}}, \quad (17)$$

$$m_{\text{dc}} = \rho_{\text{d}}(v_{\text{dc}}/\tau_{\text{dc}})\beta_{\text{dc}}S_{\text{dc}}. \quad (18)$$

$$5 \quad m_{\text{nd}} = \rho_{\text{nd}}(v_{\text{nd}}/\tau_{\text{nd}})\beta_{\text{nd}}S_{\text{nd}}. \quad (19)$$

The particle densities ρ_{d} and ρ_{nd} are assumed to be 2.6 g m^{-3} and 1.5 g m^{-3} , respectively (Ansmann et al., 2012). The conversion factors v_m/τ_m with column particle volume concentration v_m and corresponding optical thickness τ_m for aerosol component m are obtained from photometer observations as shown in Sect. 3. An extended discussion on the range of observed conversion factors is given by Ansmann et al. (2012). As mentioned in Sect. 3, typical conversion factors are $0.6\text{--}0.9 \times 10^{-6} \text{ m}$ for supermicron dust, $0.25\text{--}0.4 \times 10^{-6} \text{ m}$ for submicron dust, and around $0.18 \times 10^{-6} \text{ m}$ for anthropogenic fine-mode aerosol.

15 4.2 Consistency check: POLIPHON vs. AERONET results

Because of the numerous assumptions and thus high degree of freedom in this two-step retrieval, we use AERONET observations as constraints to check the quality of the POLIPHON backscatter profiles. Goal is to check to what extend our results and the made assumptions are in consistency with the column values of aerosol optical properties as retrieved from accompanying sun/sky photometer observations.

The AERONET parameters useful for comparison are the aerosol particle optical thickness AOT_{A} , the Ångström exponent AE_{A} , and fine-mode fraction FMF_{A} . The respective lidar-derived quantities are AOT_{L} , AE_{L} , and FMF_{L} , which are calculated from the backscatter coefficient profiles in the planetary boundary layer (PBL) and the free troposphere (FT) and the parameters listed in Table 1. We distinguish local PBL aerosol particles and FT particles after long-range transport.

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We define the lidar-derived optical depth for a given aerosol type m (nd: $m = 1$, df: $m = 2$, dc: $m = 3$) and layer l (planetary boundary layer, PBL: $l = 1$, free troposphere, FT: $l = 2$) as follows:

$$\tau_{m,l} = S_{m,l} \int_{z_{l,\text{bot}}}^{z_{l,\text{top}}} \beta_{m,l}(z) dz. \quad (20)$$

Table 1 provides an overview of all input parameters. Lidar ratios for the PBL (lowest 300–450 m of the troposphere) are found around 30 sr because of the marine influence and around 60–80 sr in the FT from the CUT–AERONET long-term observations (2010–2014).

Because we compare in Sect. 4 solutions obtained with the one-step approach (nd: $m = 1$, d: $m = 2$ after Tesche et al., 2009) and the two-step method we introduce the parameter M with $M = 2$ in the case of the one-step method and $M = 3$ in the case of the two-step method. Now the total particle optical depth AOT_L derived from the lidar observations can be written as:

$$\tau_L = \sum_{m=1}^M \sum_{l=1}^2 \tau_{m,l}. \quad (21)$$

The lidar-derived column Ångström exponent AE_L is given by

$$\alpha_L = \frac{\sum_{m=1}^M \sum_{l=1}^2 \alpha_{m,l} \tau_{m,l}}{\sum_{m=1}^M \sum_{l=1}^2 \tau_{m,l}} \quad (22)$$

with characteristic Ångström exponents in Table 1.

Typical Ångström exponents in Table 1 for the free troposphere and boundary-layer aerosol over Cyprus are obtained from the long-term AERONET–EARLINET studies (2010–2014). The pure dust Ångström exponents are from the SAMUM-1 campaign.

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The fine-mode fraction FMF_L is computed from the lidar data as follows:

$$f_L = \frac{\sum_{m=1}^{M-1} \sum_{l=1}^2 \tau_{m,l}}{\sum_{m=1}^M \sum_{l=1}^2 \tau_{m,l}}. \quad (23)$$

4.3 Retrieval uncertainties

Uncertainties in the separation of the backscatter coefficients of spherical particles and fine and coarse dust particles are caused by four sources: (a) uncertainties in the computation of the basic products, i.e., of the particle depolarization ratios and backscatter coefficients, (b) uncertainties in the assumptions on characteristic depolarization ratios for fine dust and coarse dust, (c) uncertainties in the assumption of the contribution of haze and smoke particles to the free tropospheric aerosol, and (d) uncertainties in the input parameters in Table 1. According to the error discussions by Tesche et al. (2009) and Mamouri et al. (2013) the overall uncertainty in the separation of the backscatter coefficients for the different aerosol types is of the order 20–40 %. The uncertainty in the retrieved mass concentration profiles may be of the order of 50 %. However, as the good agreement and consistency between the lidar and AERONET photometer observations in the next section indicate, the retrieval uncertainties are usually much lower (of the order of 25 % or even less). In this first feasibility study on the potential of polarization lidar to provide detailed insight into fine-mode and coarse-mode dust optical and microphysical properties we avoid to present error bars in the next section to keep the figures simple and to facilitate the discussion.

Another error source arises from a potential interference by other non-spherical aerosol types. The lidar/photometer data analysis and interpretation must be always accompanied by extended backward trajectory analysis and atmospheric transport model simulations to be sure that soil or desert dust is the only aerosol component that significantly depolarises backscattered laser light. In 2010, volcanic dust emitted by the Icelandic volcano Eyjafjallajökull and Saharan dust occurred simultaneously over wide areas of southeastern Europe and complicated the aerosol lidar data analysis

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supermicron particles from deserts in the Middle East and northern Africa reach Limassol and partly mix with this fine-mode soil dust from the north. One of such events is discussed in this section.

Figure 6 shows the arrival of an extended desert dust layer over Limassol in the late morning of 28 September 2011. Before, a northerly airflow from Turkey prevailed according to the backward trajectories in Fig. 7. The air mass transport changed from northerly advection on 26–27 September 2011 to more complex features in the regional aerosol transport resulting from a major Arabian dust outbreak on 28–29 September 2011.

The particle size distributions in Fig. 8 (taken from the AERONET data base, level 2.0 data) show a bimodal shape with a strong increase of the coarse-mode fraction when the dust outbreak arrived. The weak coarse mode on 26–27 September 2011 (blue lines in Fig. 8) consists most likely of marine particles as Fig. 9 suggests. In this figure, pure marine size distributions as observed over Barbados with AERONET photometers during a field campaign (see AERONET site of Barbados–SALTRACE, located at the west coast of Barbados, at the Caribbean Institute for Meteorology and Hydrology, CIMH) and at Ragged Point (east coast of Barbados) are compared with the observations over Limassol on 26–27 September 2011. As can be seen, the coarse modes over Limassol and Barbados are rather similar. Thus the marine aerosol impact may fully explain the occurrence of the coarse mode in the volume size distributions of the Cyprus AERONET site on 26–27 September 2011. Marine particles are confined to heights of 300–450 m during the period studied here.

Typical marine AOTs are of the order of 0.04–0.06 at 500 nm with a fine-mode contribution of 40–50 % as can be seen in Fig. 9 for the Barbados cases. If we subtract a potential fine-mode marine contribution of 0.025–0.03 and a similar urban-haze contribution from the observed fine-mode AOT of 0.172, about 0.1–0.12 is left for the fine-mode aerosol in the free troposphere on 26–27 September 2011. The depolarization ratio profile in Fig. 10 shows that this fine-mode aerosol produces significantly enhanced depolarization ratios around 10–15 %. A considerable fraction of the free tropospheric

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fine-mode aerosol must therefore be dust. Since the aerosol crosses populated and industrialized areas in Turkey and further to the north, we must assume that a mixture of fine dust and other (spherical) aerosol components (urban haze, fire smoke) was present and lowered the overall particle depolarization ratio. As can be seen in Fig. 10, the depolarization ratio went up to almost 0.35 on 28 September 2011, when the Arabian dust arrived.

An overview of the AERONET photometer observations from 26–30 September is presented in Fig. 11. The 500 nm AOT_A increased to values around 0.7 during the desert dust outbreak in the morning of 29 September 2011. At the same time, the 500 nm FMF_A dropped to values of 0.25. During the fine-mode dust days (26–27 September), FMF_A was high with values > 0.9 . Later on the values from 0.4 and 0.7 indicated mixed aerosols. The Ångström exponent AE_A was around 1.8 during the fine-mode dust days and dropped to values of 0.5–1.0 when the major dust outbreak dominated the aerosol conditions over Limassol. The minimum value of $AE_A = 0.25$ was observed in the early morning of 29 September 2011.

5.2 Retrieval of fine-mode and coarse-mode backscatter coefficients

Figures 12–15 show examples of applications of the new two-step method in terms of the basic quantities, the backscatter coefficients. For comparison, the particle backscatter coefficients obtained with the one-step method are shown in the central panels of these figures, assuming typical depolarization ratios of 0.05 and 0.31 for dust and non-dust particles (see Fig. 5). In the case of the two-step method, the characteristic particle depolarization ratios are 0.05, 0.16, and 0.39 for spherical particles, fine dust, and coarse dust, respectively. As outlined in Sect. 4, it is assumed that roughly 25% of the fine particles in the free troposphere are of anthropogenic origin (urban haze and biomass burning smoke).

The aerosol conditions as observed before the arrival of the major dust storm are given in Fig. 12. The free tropospheric aerosol layer extended from about 350 to 3500 m height. The particle depolarization ratio range from 0.1–0.14 in the main aerosol layer

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so that the good agreement with FMF_A is a clear sign that our two-step approach worked successful. Especially for 26–29 September period a very good agreement of the AERONET and the two-step-method results is obtained. At the end of our observational period (30 September 2011), when the backward trajectories (not presented) showed a complex air mass transport structure with prevailing westerly winds, both methods are no longer in good agreement with the AERONET results because of the undefined conditions on mixing of marine, urban smoke, and dust aerosols.

5.4 Particle extinction and mass concentration profiles

Finally, Fig. 18 provides an example for the computation of the particle extinction coefficients and mass concentrations from the particle backscatter coefficients. These retrievals complete the POLIPHON data analysis. For each aerosol component (fine spherical, fine dust, coarse dust), extinction and mass profiles are presented. The results for the lowermost 300 m have to be interpreted with caution because all profiles rely on the assumption of a linear increase of the backscatter coefficient from 300 m towards the ground.

As can be seen in Fig. 18, fine-mode dust significantly contributes to the total particle extinction coefficient in the free troposphere as demonstrated in Sect. 3. Extinction values for fine-mode dust are of the order of $30\text{--}50\text{ Mm}^{-1}$.

The mass concentration profiles show that coarse dust mostly contributes to particle mass in the free troposphere. Maximum values of close to $1000\text{ }\mu\text{g m}^{-3}$ are found at 1.2 km height. Fine dust mass concentrations are around $40\text{ }\mu\text{g m}^{-3}$ in the free troposphere and $50\text{--}100\text{ }\mu\text{g m}^{-3}$ close to the ground. The fine spherical particles show much less mass concentrations of $15\text{--}20\text{ }\mu\text{g m}^{-3}$ in the PBL and $10\text{--}15\text{ }\mu\text{g m}^{-3}$ in the free troposphere. Thus, fine dust can dominate $PM_{1.0}$ levels at ground during dust outbreak situations.

6 Conclusions

The separation of profiles of fine dust and coarse dust optical properties by means of the polarization lidar technique has been proposed for the first time. The presented new lidar method is an extension of the traditional method applied to distinguish non-spherical and spherical particles. Now, fine-mode and coarse-mode dust profiles in terms of particle backscatter and extinction coefficients, volume and mass concentrations can be derived. A feasibility study based on complex aerosol observations with EARLINET lidar and AERONET sun/sky photometer observations over Limassol, Cyprus, demonstrated the applicability and usefulness of the new two-step POLIPHON method. Good agreement with AERONET column aerosol observations was found. Such a step forward in the application of polarization lidar technique provides important new insight into dust properties for atmospheric and environmental research.

It should be emphasized that the developed one-wavelength polarization method as presented here is only one of several potential ways to retrieve fine and coarse-mode dust profiles. The new retrieval technique requires a considerably number of assumptions. Nevertheless, the advantage of simple lidars is that they are usually robust and favorable for long-term monitoring efforts.

In the next step, multiwavelength polarization lidars, providing depolarization ratios at two wavelengths (Sugimoto and Lee, 2006; Groß et al., 2011; Kanitz et al., 2014) or even three wavelengths (Ansmann et al., 2014; Müller et al., 2014), with additional Raman or high-spectral-resolution channels for extinction and backscattering profiling, are best candidates for an almost unambiguous separation of fine and coarse dust profiles. Based on the measured wavelength dependence of light depolarization, backscattering and extinction coefficients, three to four aerosol types (marine, haze and smoke, fine dust, coarse dust) which all have different characteristics in terms of particle depolarization ratio and wavelength dependence of backscattering and extinction, can then be discriminated.

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Table 1. Overview of all input parameters used in our study. The particle lidar ratios and the PBL and FT Ångström exponents for non-dust aerosol are based on a careful analysis of the AERONET data in combination with the EARLINET data set for 2010–2014 with and without lofted aerosol layers in the FT over Limassol. SAMUM-1 observations provided the dust Ångström exponents. The depolarization ratios are taken from the literature. The indices nd, df, dc, d as used in the retrieval in Sect. 4 denote non-spherical particles, fine-mode dust, coarse-mode dust, and total (fine and coarse) dust, respectively.

Parameter	Symbol	Value	Source/reference
Particle lidar ratio (marine)		20 sr	Groß et al. (2011)
Particle lidar ratio (dust)	S_{df}, S_{dc}	35–40 sr	Mamouri et al. (2013)
Particle lidar ratio (urban haze, smoke)		50–70 sr	CUT, climatology
Particle lidar ratio (PBL, spherical)	$S_{nd}(\text{PBL})$	30 sr	CUT, climatology
Particle lidar ratio (FT, spherical)	$S_{nd}(\text{FT})$	60–80 sr	CUT, climatology
Ångström Exponent (PBL, spherical)	$\alpha_{nd}(\text{PBL})$	0.5–1.5	CUT, climatology
Ångström Exponent (FT, spherical)	$\alpha_{nd}(\text{FT})$	2.0	CUT, climatology
Ångström Exponent (fine dust)	α_{df}	1.5	SAMUM-1, Fig. 3
Ångström Exponent (coarse dust)	α_{dc}	−0.2	SAMUM-1, Fig. 3
Ångström Exponent (total dust)	α_d	0.25	SAMUM-1, Fig. 3
Particle linear depolarization ratio (spherical)	δ_{nd}	0.05	Müller et al. (2007)
Particle linear depolarization ratio (fine dust)	δ_{df}	0.16	Sakai et al. (2010)
Particle linear depolarization ratio (coarse dust)	δ_{dc}	0.39	Sakai et al. (2010)
Particle linear depolarization ratio (total dust)	δ_d	0.31	Freudenthaler et al. (2009)

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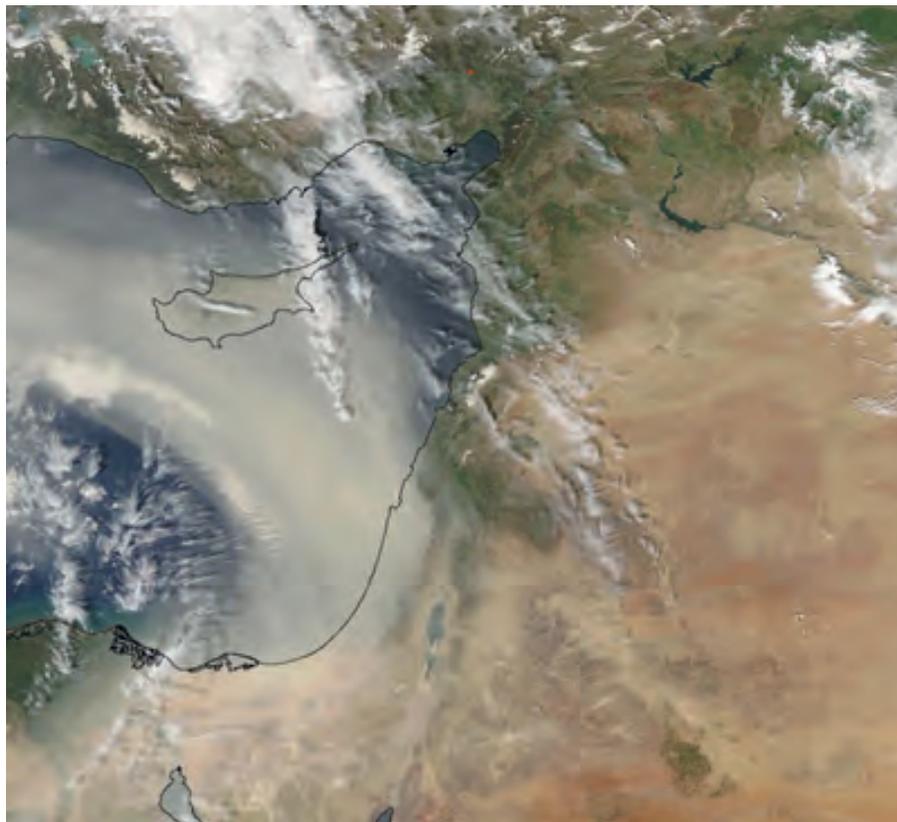



Figure 1. Major Saharan dust outbreak moving northward and striking Cyprus on 1 April 2013 (AQUA-MODIS image, 10:40 UTC).

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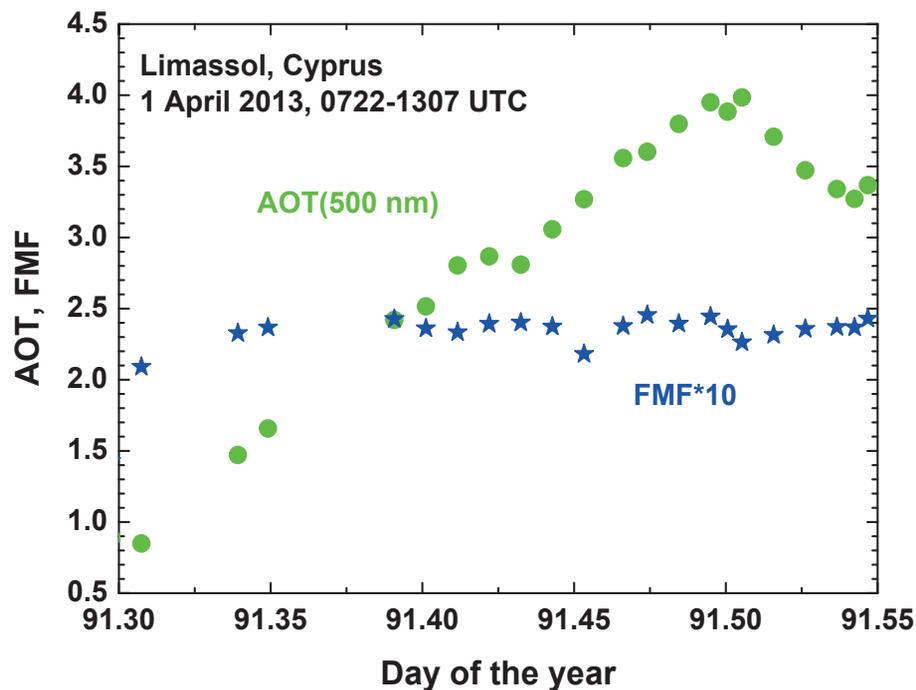


Figure 2. 500 nm aerosol particle optical thickness (AOT, green circles) and fine-mode fraction (FMF multiplied by a factor of 10, blue stars), observed with AERONET sun/sky photometer on 1 April 2013. Peak AOT values reached 4.0 around noon. The dust-dominated FMF was always close to 0.24.

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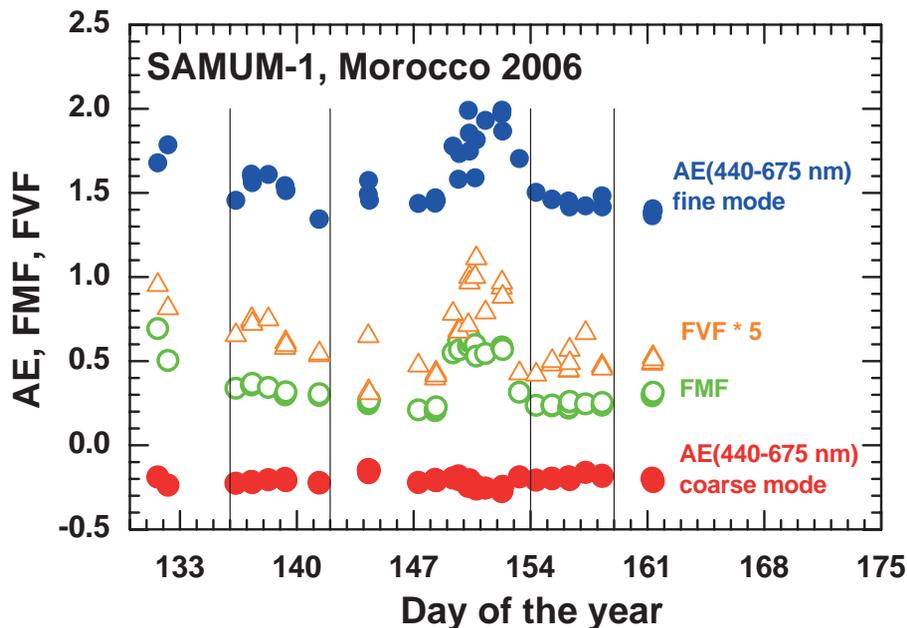


Figure 3. SAMUM-1 pure Saharan dust Ångström exponent AE (440–675 wavelength range) for fine mode (blue circles) and coarse mode (red circles), fine-mode 500 nm AOT fraction FMF (open green circles), and fine-mode particle volume fraction FVF (multiplied by a factor of 5, open orange triangles) observed with AERONET sun/sky photometer. The measurements were performed at Ouarzazate (30.9° N, 6.9° W), very close to the Sahara in southeastern Morocco. Pure dust episodes occurred from 136–142 (16–21 May 2006) and from 154–159 (3–7 June 2006), indicated by vertical lines).

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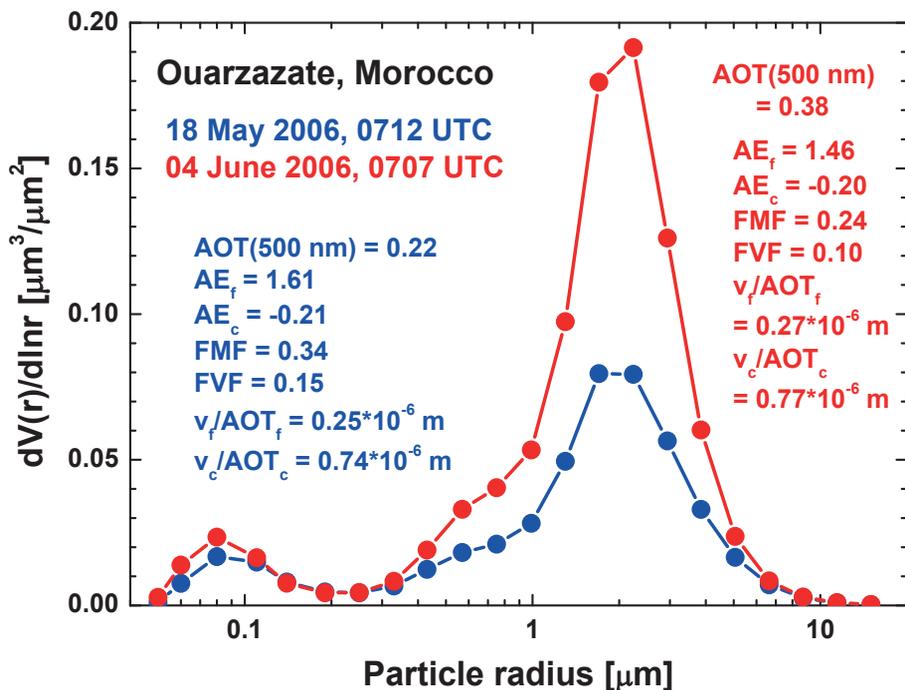


Figure 4. Column-integrated particle volume concentration as a function of particle radius (22 radius intervals) of pure Saharan dust observed with AERONET photometer over Ouarzazate, southern Morocco, during SAMUM-1 in the morning of 18 May 2006 (blue) and 4 June 2006 (red). Values for 500 nm AOT, fine-mode and coarse-mode Ångström exponents AE_f and AE_c , fine-mode fraction FMF, and fine-mode particle volume fraction FVF are given in addition. The conversion factors v_f/AOT_f and v_c/AOT_c for fine-mode and coarse-mode dust with volume fractions v_f and v_c , respectively, are used in the conversion of the lidar-derived optical properties into volume and mass concentrations by means of the POLIPHON method (see Sect. 4).

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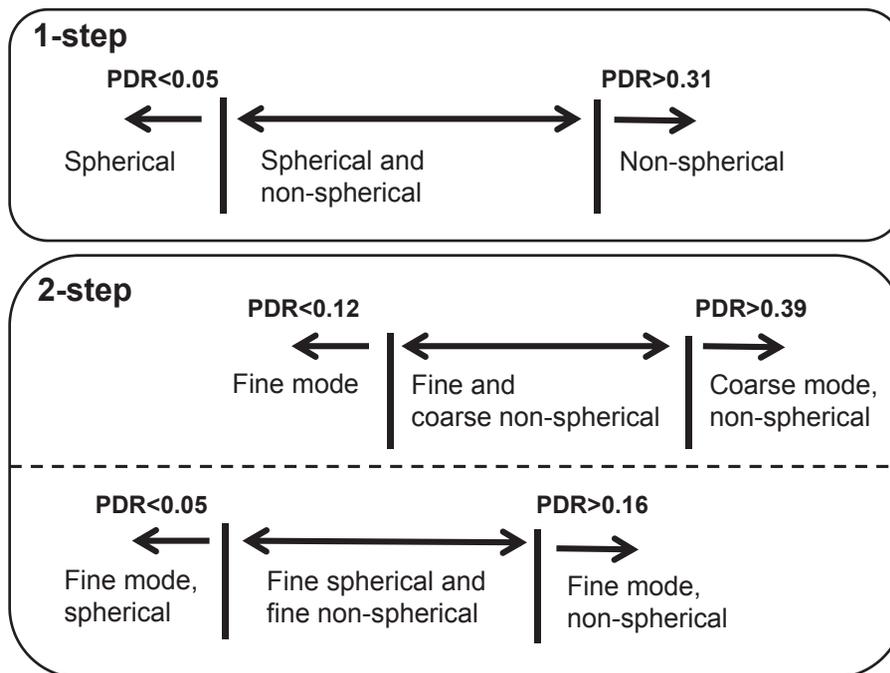


Figure 5. Illustration of the one-step and two-step methods to separate spherical particles from non-spherical dust particles (1-step method) and spherical particles (fine mode), fine dust, and coarse dust particles (two-step method) by means of particle depolarization ratio PDR.

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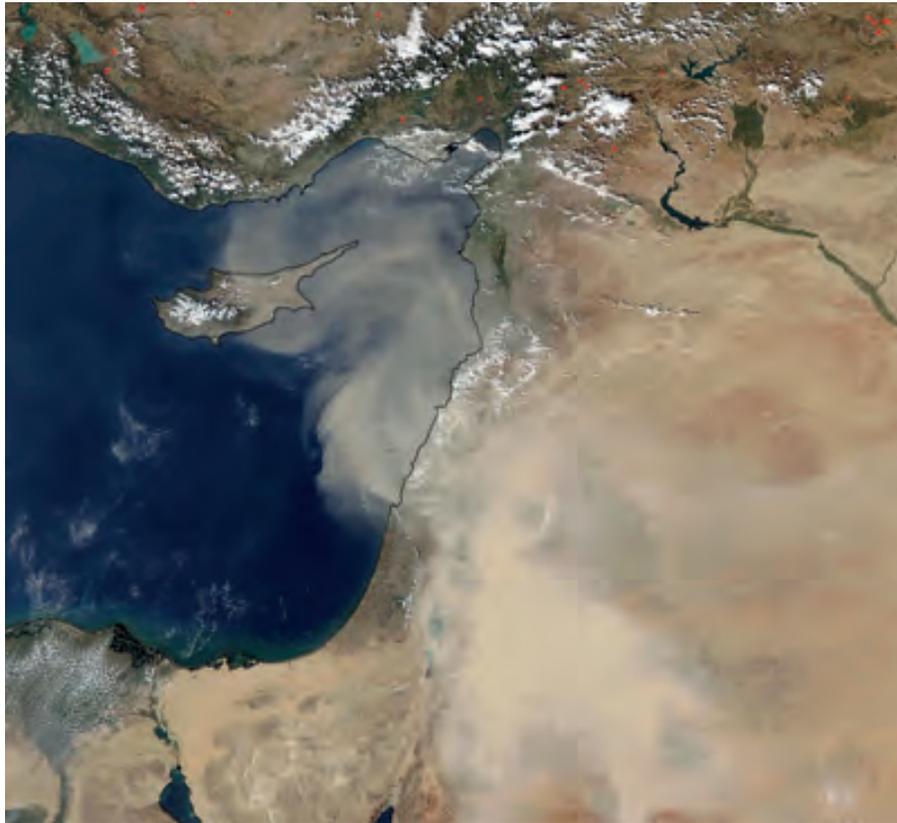


Figure 6. Dust outbreak from deserts in the Middle East reaching Cyprus on 28 September 2011 (TERRA-MODIS image, 08:30 UTC).

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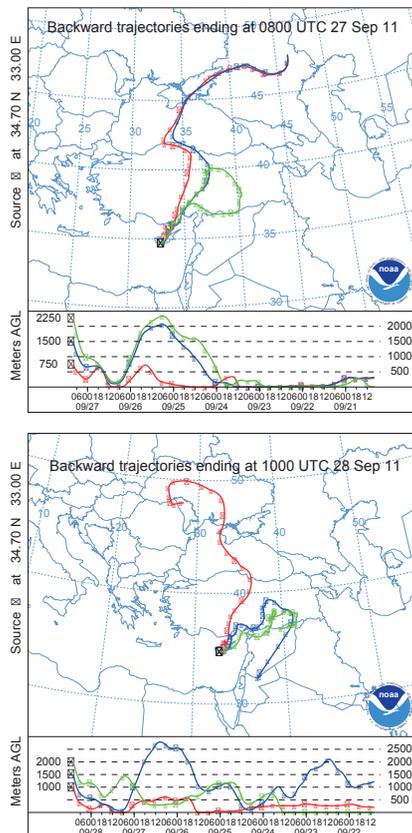
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Figure 7. Seven-day backward trajectories arriving at Limassol, Cyprus, at different height levels (red, blue, green) on 27 September 2011, 08:00 UTC (top) and 28 September 2011, 10:00 UTC (bottom). Calculations are performed with HYSPLIT (HYbrid Single-Particle Lagrangian Integrated Trajectory) Model. Access via NOAA ARL READY Website (<http://www.arl.noaa.gov/HYSPLIT.php>).

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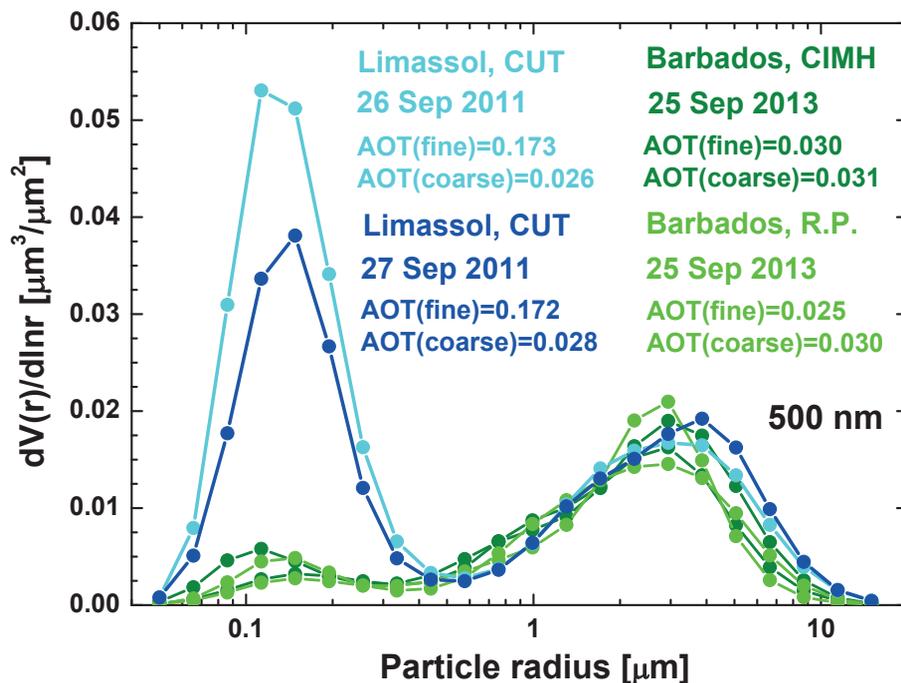


Figure 9. Column-integrated particle volume size distribution for pure marine conditions derived from AERONET observations at Barbados (CIMH and AERONET station Ragged Point, R. P.) on 25 September 2013 and for mixed aerosol conditions at the coastal AERONET station CUT, Limassol (measured at 07:41 UTC on 26 and 27 September 2011). The coarse mode at Limassol can be related to coarse marine particles. The pronounced fine mode is mostly related to urban haze in the boundary layer and fine-mode dust in the free troposphere.

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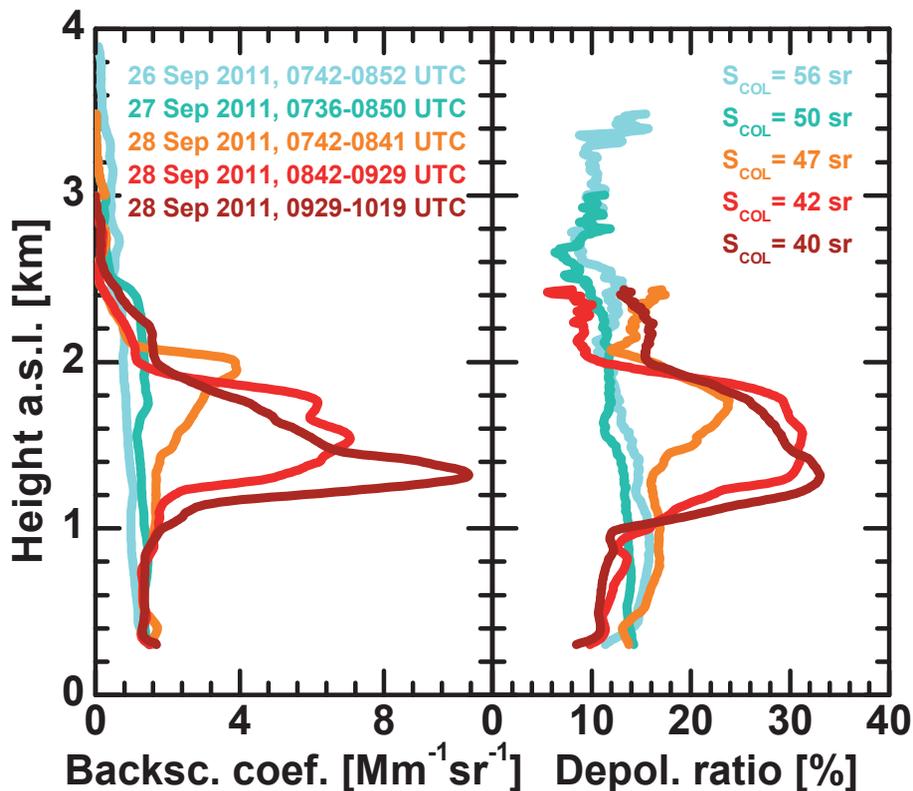



Figure 10. 532 nm particle backscatter coefficient (left) and particle linear depolarization ratio (right) in the free troposphere during a northerly airflow with fine-mode dust (26–27 September 2011) and during the arrival of a strong Arabian dust outbreak in the morning of 28 September 2011. The column lidar ratios S_{col} for the entire vertical aerosol column are derived from combined photometer-lidar data analysis (Mamouri et al., 2013). 60–70 min of lidar return signals are averaged.

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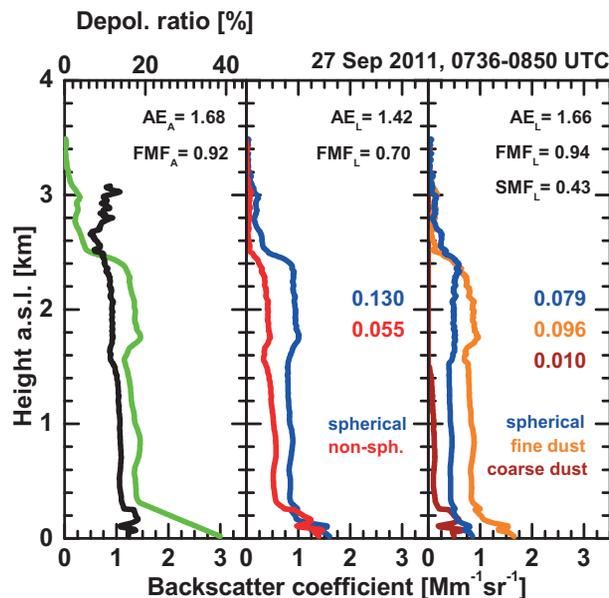


Figure 12. (Left) 532 nm particle backscatter coefficient (green) and particle linear depolarization ratio (black), (center) particle backscatter coefficient for non-spherical and spherical particles (1-step method), and (right) particle backscatter coefficient for spherical particles, fine and coarse dust (2-step method) observed on 29 September 2011, 07:36–08:50 UTC. 74 min of lidar return signals are thus averaged. The corresponding AOTs for the different aerosol components are given as colored numbers, computed from the backscatter profiles multiplied by appropriate lidar ratios as given in Table 1. AE, FMF, and SMF denote Ångström exponent, fine-mode fraction, and spherical mode fraction, respectively. Index A indicates AERONET values, index L lidar-derived results obtained by means of Eqs. (22) and (23). In the central panel $FMF_L = SMF_L$. For the lowest 300 m a linear increase of the total backscatter coefficient from 300 m height to the surface (see left panel) is assumed.

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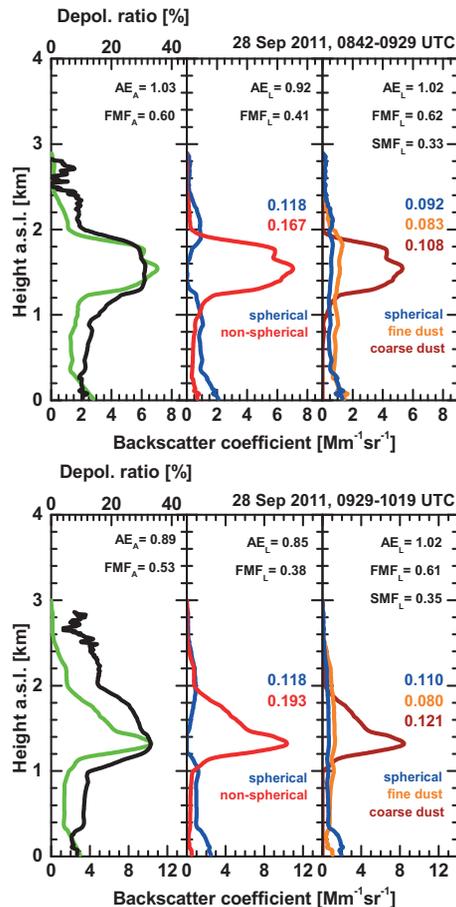


Figure 13. Same as Fig. 12, except for two periods on (top) 28 September 2011, 08:42–09:29 UTC (47 min signal average) and (bottom) 28 September 2011, 09:29–10:19 UTC (50 min signal average).

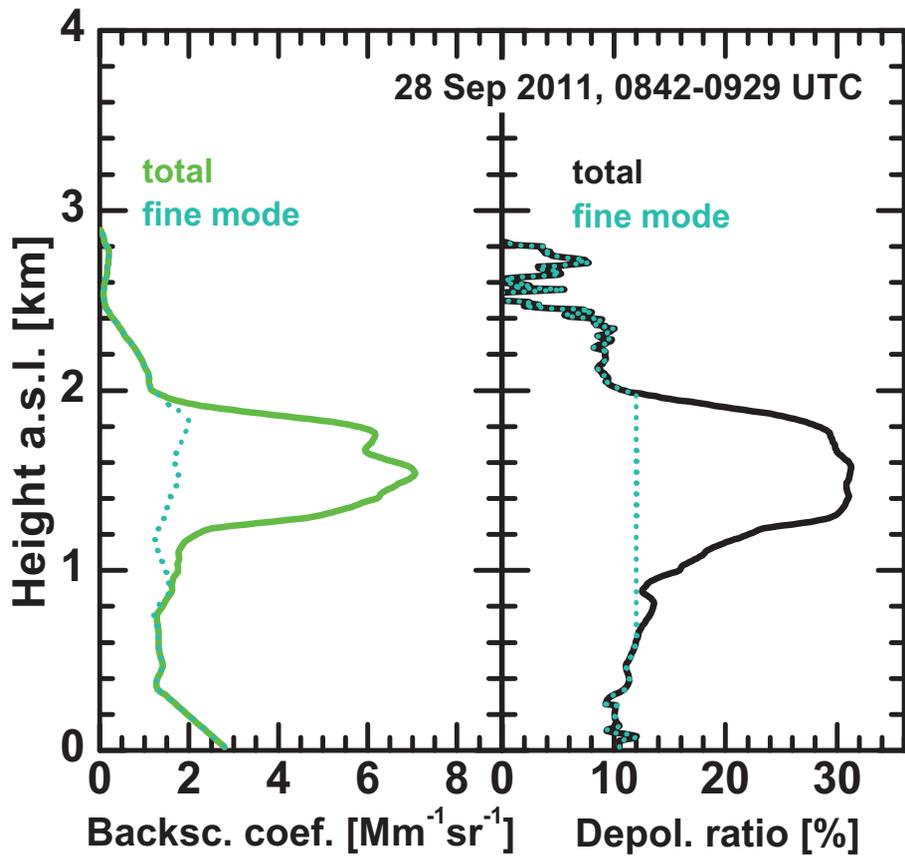


Figure 14. Total particle backscatter coefficient (green, left) and particle depolarization ratio (black, right) and respective fine-mode (fine spherical particles and fine dust) contributions (blue dashed, obtained after step 1 of the 2-step data analysis). The fine-mode profiles are used as input for step 2 in the 2-step approach (see Fig. 5).

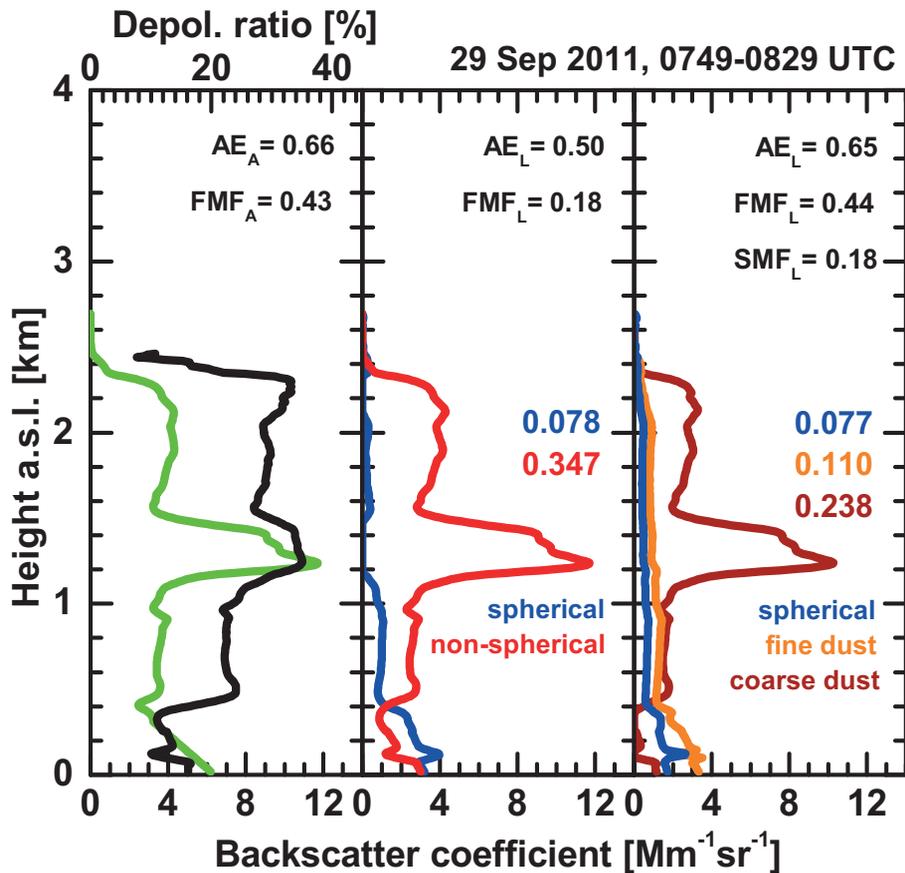


Figure 15. Same as Fig. 12, except for a morning measurement on 29 September 2011, 07:49–08:29 UTC (40 min signal average).

Fine and coarse dust profiling

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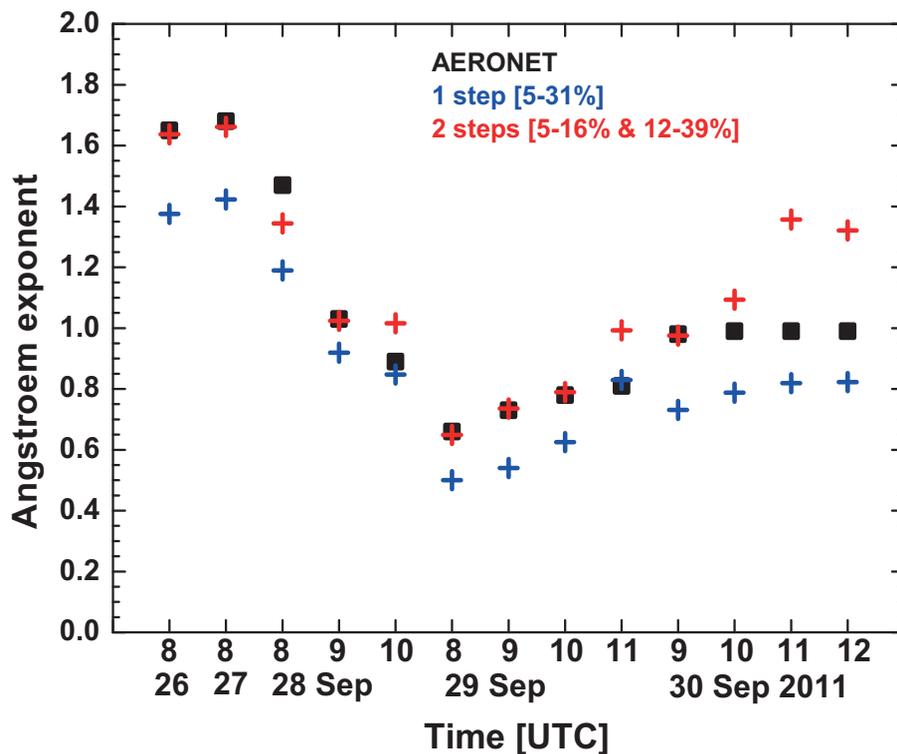


Figure 16. Comparison of Ångström exponents (AE_A vs. AE_L) derived from AERONET observations (black squares) and retrieved from the lidar measurements (blue crosses, 1-step method, red crosses, 2-step method) for the entire measurement period from 26–30 September 2011.

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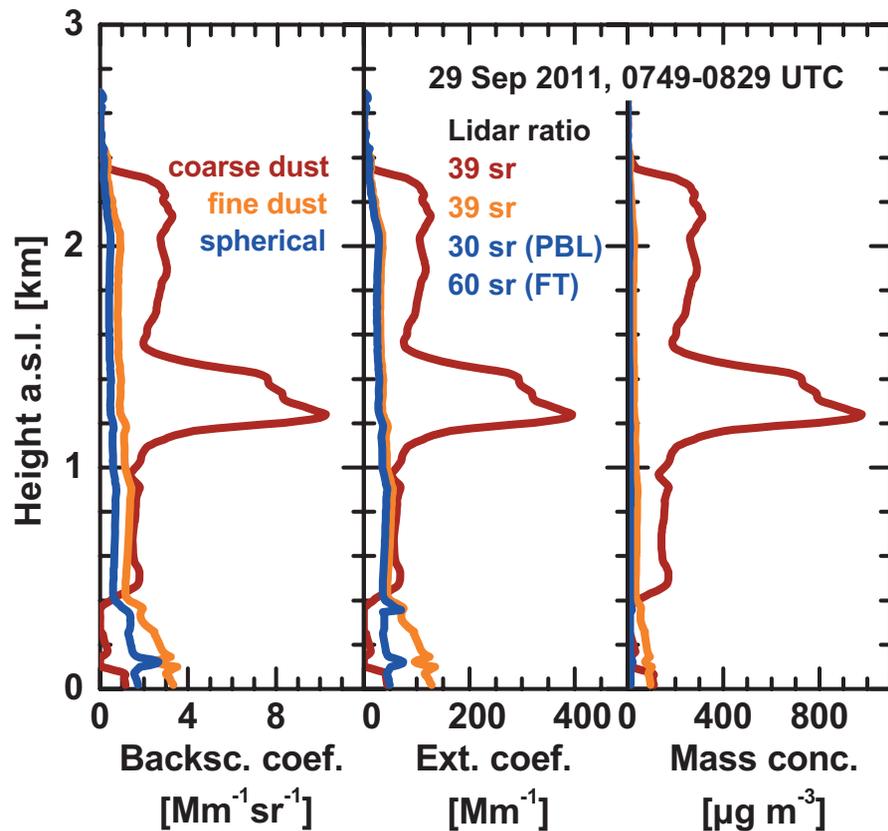



Figure 18. 532 nm particle backscatter and extinction coefficients, and particle mass concentrations computed from the backscatter coefficients in Fig. 15 by means of the two-step method, the lidar-ratio information in Table 1, and volume-to-extinction conversion factors as discussed in Sects. 3 and 4.