

Classification of aerosol properties derived from AERONET direct sun data

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Abstract. Aerosol spectral measurements by sunphotometers can be characterized by three independent pieces of information: 1) the optical thickness (AOT), a measure of the column aerosol concentration, 2) the optical thickness average spectral dependence, given by the Angstrom exponent (α), and 3) the spectral curvature of α ($\delta\alpha$). We propose a simple graphical method to visually convert (α , $\delta\alpha$) to the contribution of fine aerosol to the AOT and the size of the fine aerosols. This information can be used to track mixtures of pollution aerosol with dust, to distinguish aerosol growth from cloud contamination and to observe aerosol humidification. The graphical method is applied to the analysis of yearly records at 8 sites in 3 continents, characterized by different levels of pollution, biomass burning and mineral dust concentrations. Results depict the dominance of fine mode aerosols in driving the AOT at polluted sites. In stable meteorological conditions, we see an increase in the size of the fine aerosol as the pollution stagnates and increases in optical thickness. Coexistence of coarse and fine particles is evidenced at the polluted sites downwind of arid regions.

1 Introduction

The aerosol optical thickness at wavelength λ ($AOT(\lambda) \equiv \tau_\lambda$) is a standard parameter measured by sunphotometers like the ones operating in the AERONET (Holben et al., 1998, <http://aeronet.gsfc.nasa.gov>). τ_λ represents the extinction of radiation of wavelength λ that results from the presence of atmospheric aerosols. The Angström exponent α represents

the slope of the wavelength dependence of the AOT in logarithmic coordinates (Angstrom, 1929):

$$\alpha(\lambda_1, \lambda_2) = -\ln(\tau_{\lambda_2}/\tau_{\lambda_1})/\ln(\lambda_2/\lambda_1) \quad (1)$$

In the solar spectrum, α is a good indicator of the size of the atmospheric particles determining the AOT: $\alpha > 1$ are mainly determined by fine mode, submicron aerosols, while $\alpha < 1$ are largely determined by coarse, supermicron particles (e.g., Kaufman et al., 1994). The Angstrom parameter is commonly employed in operational sunphotometry. However, α alone does not provide unambiguous information on the relative weight of coarse and fine modes in determining the AOT. Large fine mode particles can have the same α as mixtures of coarse mode and small fine mode ones. We shall demonstrate this with examples in the next section.

Several authors have discussed how the spectral variation of the Angstrom exponent can provide further information about the aerosol size distribution (King et al., 1978; Nakajima et al., 1986; Kaufman, 1993; Eck et al., 1999; O'Neill et al., 2001a, b, 2003; Schuster et al., 2006). Kaufman (1993) pointed-out that negative values of the difference $\delta\alpha = \alpha(440, 613) - \alpha(440, 1003)$ indicate the dominance of fine mode aerosols, while positive differences indicate the effect of two separate particle modes. Eck et al. (1999) have shown how in the wavelength range 380–870 nm, α can increase by a factor of 2–5 as wavelength increases for biomass burning and urban aerosols, while remaining constant or decreasing in the presence of mineral dust. O'Neill et al. (2001a) demonstrated that an Angström exponent-based separation of coarse from fine mode contribution to AOT is feasible in part because of the coarse mode AOT spectral variation being approximately neutral. Schuster et al. (2006) addressed the link between Angstrom exponent curvature and the ratio between fine and total aerosol volume.

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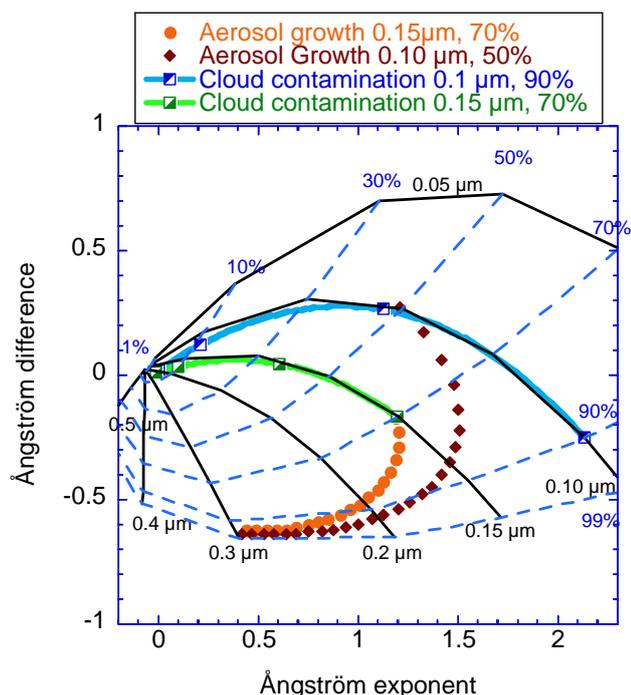


Fig. 1. Simulations of the classification of the aerosol properties as a function of the Angström exponent α (440, 870) and the difference $\delta\alpha = \alpha(440, 675) - \alpha(675, 870)$, for bimodal, lognormal size distributions with refractive index $m = 1.4 - 0.001i$. The black solid lines are each for a fixed size of the fine mode R_f and the dashed blue lines for a fixed fraction contribution η of the fine mode to the AOT at 675 nm. Split squares represent the effects of a cloud contamination of 0, 50, 90 and 99% in the AOT of two grid points: 1) $\eta = 70\%$, $R_f = 0.15$ (bright green line) and 2) $\eta = 90\%$, $R_f = 0.1$ (turquoise line). This contamination results in a departure from the original grid points along the constant R_f lines and towards the origin. Conversely, hydration of the aerosol fine mode (two starting conditions simulated: 1) $\eta = 50\%$, $R_f = 0.1$ (brown diamonds), and 2) $\eta = 70\%$, $R_f = 0.15$ (orange circles)) is accompanied by a movement towards the origin along the opposite direction, with concurrent increase in R_f and η .

Here we build upon the concept of Kaufman (1993) to propose a new, straight-forward graphical framework that, on the basis of three spectral AOT observations, allows to: 1) infer aerosol fine mode size (R_f) and fractional contribution to total AOT (η), and 2) separate AOT increases due to aerosol humidification from AOT increases due to the addition of coarse particles as in the case of cloud contamination. To this end, we classify the aerosols in a new space, AOT vs. $\delta\alpha$

vs. α . This space is invariant to the bulk AOT namely, the space is invariant to changes in AOT for a given size distribution. Any AOT will be in the same point if the fine and coarse modes stays the same. This space is sensitive to the balance between the fine and coarse modes and therefore is ideal to separate processes related to aerosol fine size from extinction fraction. Cloud contamination will enhance the weight of the coarse mode while humidification will increase the fine mode. Overall, this method provides an additional tool to interpret in terms of size-dependent properties, direct-sun observations of aerosol spectral extinction.

4 Conclusions

A graphical framework to classify aerosol properties using direct-sun sunphotometer observations has been presented. The method relies on the combined analysis of the Angstrom exponent α and of its spectral curvature $d\alpha/d\lambda$ here represented by $\delta\alpha = \alpha(440, 675) - \alpha(675, 870)$. Plotting data in these coordinates was shown to allow for inference of aerosol fine mode size and fractional contribution to total AOT. Adding information on AOT to the plot then permits to separate AOT growth by aerosol humidification and/or coagulation (aging) from AOT growth by inclusion of coarse mode particles or cloud contamination. Application to AERONET climatological data from three continents allowed to identify various aerosol properties peculiar to these locations. Data from Beijing (China) and Kanpur (India) confirmed these locations to be affected by superposition of dust, and high pollution conditions. At both sites it is pollution haze (fine mode aerosols) that generates the largest aerosol loads, reaching AOT levels >2 in Beijing. The method allowed for easy identification of the mineral dust fingerprint in the Rome (Italy) data, while confirming the absence of a significant contribution of dust at the Ispra site, located just 400 km North. Growing haze appears in this framework as a branching along higher fine mode radius and extinction fraction. Conversely, locations where biomass burning represents the main source of aerosols showed limited hygroscopic growth of fine mode particles, and a rather focused range of fine mode size ($dR_f \sim 0.05 \mu\text{m}$). In these cases, some cloud contamination was observed at the two sites of Alta Floresta (Brazil) and Mongu (Zambia). Overall, the analysis scheme provides an additional, versatile tool to characterize aerosol properties and to explore the important aerosol-cloud border region by means of easily accessible, direct sun photometric observations.