Angstrom exponent and bimodal aerosol size distributions

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[1] Power laws have long been used to describe the spectral dependence of aerosol extinction, and the wavelength exponent of the aerosol extinction law is commonly referred to as the Angstrom exponent. The Angstrom exponent is often used as a qualitative indicator of aerosol particle size, with values greater than 2 indicating small particles associated with combustion byproducts, and values less than 1 indicating large particles like sea salt and dust. In this study, we investigate the relationship between the Angstrom exponent and the mode parameters of bimodal aerosol size distributions using Mie theory calculations and Aerosol Robotic Network (AERONET) retrievals. We find that Angstrom exponents based upon seven wavelengths (0.34, 0.38, 0.44, 0.5, 0.67, 0.87, and 1.02 μm) are sensitive to the volume fraction of aerosols with radii less then 0.6 μm but not to the fine mode effective radius. The Angstrom exponent is also known to vary with wavelength, which is commonly referred to as curvature; we show how the spectral curvature can provide additional information about aerosol size distributions for intermediate values of the Angstrom exponent. Curvature also has a significant effect on the conclusions that can be drawn about two-wavelength Angstrom exponents; long wavelengths (0.67, 0.87 μm) are sensitive to fine mode volume fraction of aerosols but not fine mode effective radius, while short wavelengths (0.38, 0.44 μm) are sensitive to the fine mode effective radius but not the fine mode volume fraction.

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1. Introduction

[2] Knowledge of the aerosol optical thickness (AOT) throughout much of the shortwave spectral region (~0.3–5 μm) is necessary to compute the shortwave aerosol radiative forcing at the surface and the top of the atmosphere. The AOT is easily measured in discrete spectral intervals with Sun photometers located at the surface, but gas and water vapor absorption prevent the measurement of AOT at all wavelengths of interest. This difficulty is easily circumvented because [*Angstrom*, 1929] noted that the spectral dependence of extinction by particles may be approximated as a power law relationship:

$$\tau(\lambda) = \tau_1 \lambda^{-\alpha}, \tag{1}$$

where $\tau(\lambda)$ is the aerosol optical thickness (AOT) at the wavelength λ, τ_1 is the approximated AOT at a wavelength of 1 μm (sometimes called the turbidity coefficient, as per [*Angstrom*, 1964]), and α has come to be widely known as the Angstrom exponent.

[3] In addition to being a useful tool for extrapolating AOT throughout the shortwave spectral region, the value of the Angstrom exponent is also a qualitative indicator of aerosol particle size [*Angstrom*, 1929]; values of $\alpha \lesssim 1$ indicate size distributions dominated by coarse mode aerosols (radii $\gtrsim$0.5 μm) that are typically associated with dust and sea salt, and values of $\alpha \gtrsim 2$ indicate size distributions dominated by fine mode aerosols (radii $\lesssim$0.5 μm) that are usually associated with urban pollution and biomass burning [*Eck et al.*, 1999; *Westphal and Toon*, 1991]. *Kaufman et al.* [1994] demonstrated that the Angstrom exponent can be a good indicator of the fraction of small particles with radii r = 0.057–0.21 μm relative to larger particles with radii r = 1.8–4 μm for tropospheric aerosols.

[4] Since the Angstrom exponent is easily measured using automated surface Sun photometry [*Holben et al.*, 1998] and is becoming increasingly accessible to satellite retrievals [*Nakajima and Higurashi*, 1998; *Higurashi and Nakajima*, 1999; *Deuzé et al.*, 2000; *Ignatov and Stowe*, 2002; *Jeong et al.*, 2005], the true utility of this parameter lies in its empirical relationship to the aerosol size distribution. For instance, α has been used to characterize the maritime aerosol component at island sites [*Kaufman et al.*, 2001; *Smirnov et al.*, 2002, 2003], biomass burning aerosols in South America and Africa [*Dubovik et al.*, 1998; *Reid et al.*, 1999; *Eck et al.*, 2001b, 2003], and urban aerosols in Asia [*Eck et al.*, 2001a]. Measurements also indicate that the Angstrom exponent varies with wavelength, and that the spectral curvature of the Angstrom exponent contains useful information about the aerosol size

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distribution [*King and Byrne*, 1976; *King et al.*, 1978; *Eck et al.*, 1999, 2001a, 2001b, 2003; *Kaufman*, 1993; *O'Neill and Royer*, 1993; *O'Neill et al.*, 2001a, 2001b, 2003; *Villevalde et al.*, 1994].

[5] The focus of this paper is to explore the relationship between the spectral dependence of extinction and the size distribution of atmospheric aerosols. We begin by illustrating the sensitivity of α to the median radius of monomodal aerosol size distributions, using multiwavelength Mie calculations of $\tau(\lambda)$ for 38 monomodal lognormal aerosol size distributions. Then we explore the relationship between α and 45 bimodal lognormal aerosol size distributions, demonstrating that α is more sensitive to the fine mode volume fraction than the fine mode median radius. Next, we apply the same technique to explore the information content in the wavelength dependence of α (i.e., the curvature of α). Finally, we discuss application of the Angstrom exponent and the spectral curvature for setting limits on the possible size distributions associated with aerosol optical depth measurements.

[6] Before proceeding further, however, a word about the origination of the term "Angstrom exponent" is in order. The term "Angstrom exponent" originates from an early treatise by Anders Angstrom that provides the first documentation of equation (1) currently available in English [*Angstrom*, 1929]. However, that article cites an even earlier laboratory study whereby Lundholm documented the power law relationship for the absorption of thin powders at Uppsala in 1912, prompting at least one author to point out that we are honoring the wrong person [*Bohren*, 1989]. Unfortunately, the Lundholm citation of *Angstrom* [1929] is obscure and perhaps incorrect, as our library staff was unable to locate it. Compounding matters even further, it is not even clear whether Lundholm or Lindholm documented the power law in 1912, as *Angstrom* [1929] uses both forms of spelling. Our library staff did find references to a F. Lindholm at Uppsala in *Science Abstracts*, but none of the abstracts mention the spectral dependence of particulate extinction. Subsequent references to Lundholm and Lindholm on this topic have not appeared in the atmospheric literature, and Angstrom claimed full credit for introducing the methods of evaluating atmospheric turbidity parameters in later articles [*Angstrom*, 1930, 1961, 1964]. So who should get the credit for originating and promoting equation (1)? After presenting the discussion above, we maintain the standard nomenclature. Angstrom admittedly did not originate the empirical power law, but he did publish at least four articles documenting the relationship between α and particle size; meanwhile, we are unable to obtain any public documentation of equation (1) by Lundholm or Lindholm, making it impossible for us to fairly evaluate this person's contribution.

9. Conclusions

[41] We have discussed the relationship between polydisperse aerosol size distributions and the spectral dependence of the aerosol optical thickness, using both Mie calculations and AERONET aerosol retrievals. We began by showing the expected inverse relationship between the Angstrom exponent and the effective radius for synthetic monomodal aerosol size distributions.

[42] Next, we discussed the sensitivity of the Angstrom exponent and curvature to the details associated with bimodal aerosol size distributions, focusing specifically on the fine mode effective radius and the fine mode fraction of aerosols. We demonstrated that calculations of the Angstrom exponent for seven wavelengths (0.34, 0.38, 0.44, 0.50, 0.67, 0.87, and 1.02 μm) are sensitive to the fraction of aerosols in the fine mode (V_{fine}/V_{total}), but not to the fine mode effective radius ($R_{eff}(fine)$). Nonetheless, this multiwavelength Angstrom exponent is not a rigorous indicator of V_{fine}. Rather, AERONET retrievals at approximately 50 sites indicate that $\alpha \gtrsim 2$ correspond to fine mode fractions of $V_{fine} \gtrsim 0.5$, and $\alpha \lesssim 1$ correspond to fine mode fractions of $V_{fine} \lesssim 0.5$. The same AERONET data set indicates that intermediate values of α correspond to fine mode fractions between 0.2 and 0.85, but improved estimates of the fine mode aerosol fractions can be obtained by considering the spectral curvature. For instance, if $\alpha = 1.5$, then curvatures of $a_2 \lesssim -0.3$ indicate fine mode fractions of $0.5 \lesssim V_{fine} \lesssim 0.85$, and $a_2 \gtrsim +0.3$ indicate fine mode fractions of $0.2 \lesssim V_{fine} \lesssim 0.5$. Although the curvature is useful for estimating the fine mode fraction of aerosols at intermediate values of α, it provides no additional information when $\alpha \lesssim 1$ and $\alpha \gtrsim 2$.

[43] Spectral curvature of aerosol extinction also plays a significant role when the Angstrom exponent is calculated with only two wavelengths. Angstrom exponents calculated from longer wavelength pairs (λ = 670, 870 μm) are sensitive to the fine mode fraction of aerosols but not the fine mode effective radius; conversely, shorter wavelength pairs (λ = 380, 440 μm) are sensitive to the fine mode effective radius but not the fine mode fraction [see also *Eck et al.*, 1999, 2001b; *Kaufman*, 1993]. Hence it is important to consider the wavelength pair used to calculate the Angstrom exponent when making qualitative assessments about the corresponding aerosol size distributions.