Aerosol ultraviolet absorption experiment (2002 to 2004), part 2: absorption optical thickness, refractive index, and single scattering albedo

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Abstract. Compared to the visible spectral region, very little is known about aerosol absorption in the UV. Without such information it is impossible to quantify the causes of the observed discrepancy between modeled and measured UV irradiances and photolysis rates. We report results of a 17-month aerosol column absorption monitoring experiment conducted in Greenbelt, Maryland, where the imaginary part of effective refractive index k was inferred from the measurements of direct and diffuse atmospheric transmittances by a UV-multifilter rotating shadowband radiometer [UV-MFRSR, U.S. Department of Agriculture (USDA) UV-B Monitoring and Research Network]. Colocated ancillary measurements of aerosol effective particle size distribution and refractive index in the visible wavelengths [by CIMEL sun-sky radiometers, National Aeronautics and Space Administration (NASA) Aerosol Robotic Network (AERONET)], column ozone, surface pressure, and albedo constrain the forward radiative transfer model input, so that a unique solution for k is obtained independently in each UV-MFRSR spectral channel. Inferred values of k are systematically larger in the UV than in the visible wavelengths. The inferred k values enable calculation of the single scattering albedo ω, which is compared with AERONET inversions in the visible wavelengths. On cloud-free days with high aerosol loadings [$\tau_{\text{ext}}(440) > 0.4$], ω is systematically lower at 368 nm ($\langle\omega_{368}\rangle = 0.94$) than at 440 nm ($\langle\omega_{440}\rangle = 0.96$), however, the mean ω differences (0.02) are within expected uncertainties of ω retrievals ($\sim$0.03). The inferred ω is even lower at shorter UV wavelengths ($\langle\omega_{325}\rangle \sim \langle\omega_{332}\rangle = 0.92$), which might suggest the presence of selectively UV absorbing aerosols. We also find that ω decreases with decrease in aerosol loading. This could be due to real changes in the average aerosol composition between summer and winter months at the Goddard Space Flight Center (GSFC) site. Combing measurements of τ_{ext} and ω, the seasonal dependence of the aerosol absorption optical thickness, $\tau_{\text{abs}} = \tau_{\text{ext}}(1-\omega)$ is derived in the UV with an uncertainty of 0.01 to 0.02, limited by the accuracy of UV-MFRSR measurement and calibration. The τ_{abs} has a pronounced seasonal dependence with maximum values $\sim$0.1 occurring in summer hazy conditions and $<$0.02 in the winter and fall seasons, when aerosol loadings are small. The measured τ_{abs} is sufficient to explain both the magnitude and seasonal dependence of the bias in satellite estimates of surface UV irradiance previously seen with ground-based UV measurements.
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Subject terms: ultraviolet radiation; aerosol absorption; single scattering albedo; CIMEL sunphotometer; AERONET network; ultraviolet multifilter rotating shadowband radiometer; diffuse fraction measurements.

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1 Introduction

Aerosols in the boundary layer can significantly change air quality either directly or by affecting the rate of tropospheric ozone (urban smog) formation.[1–4] Scattering by aerosols increases the actinic flux and the rates of photochemical reactions in the upper parts of the planetary boundary layer,[1–6] while aerosol absorption reduces the amount of UV radiation available for chemical reactions within and below the aerosol layer.[1–5] Therefore, without accurate knowledge of aerosol UV absorption (or single-scattering albedo) the magnitude and even the sign of the aerosol effect on tropospheric photochemistry remain highly uncertain.[1–6] By the same reasoning, the boundary layer aerosol absorption uncertainty remains a serious obstacle in satellite estimation of biologically harmful UV

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irradiance at the surface.[7–13] Although it is well known that iron oxides in desert dust[14–16] and soot produced by fossil fuel burning and urban transportation[17–21] strongly absorb UV radiation, properties of other potential UV absorbers, e.g., organic and nitrated and aromatic aerosols,[22,23] are poorly known. In addition, different aerosol components are often mixed in an atmospheric column downwind from urban regions.[24] This makes it difficult to quantify aerosol effects on UV irradiance and photolysis rates from the models. On the other hand, ground-based passive remote sensing techniques enable estimation of column aerosol absorption without prior knowledge of aerosol composition. The techniques are based on nearly simultaneous measurements of direct sun irradiance and diffuse sky radiance[25–28] or irradiance,[29–34] from which column average absorption can be inferred (with aircraft measurements providing vertically resolved information[21,24]). The multiyear mean column aerosol absorption climatology in the visible has been established for several sites using CIMEL almucantar inversions[25–27] from the global Aerosol Robotic Network (AERONET) network,[35,36] but these inversions are not yet available at UV wavelengths. Ground-based remote measurements of aerosol UV absorption were also demonstrated,[37–42] but these retrievals have not been validated. Neither technique has yet enabled deriving seasonal aerosol absorption climatology.

To validate column aerosol absorption retrievals in the UV and produce a long-term seasonal data set of aerosol column absorption optical thickness τ_{abs} a UV multifilter rotating shadowband radiometer[43,44] (UV-MFRSR, Yankee Environmental Systems, Turners Falls, Massachusetts) and a rotating triad of sun-sky CIMEL radiometers [reference instruments of the National Aeronautics and Space Administration (NASA) AERONET network[35,36]] were run side-by-side continuously for 17 months at the NASA Goddard Space Flight Center in Greenbelt, Maryland. A previous paper[45] showed that the AERONET data could be used for UV-MFRSR daily on-site calibration and accurate measurements of τ_{ext} at three UV-A wavelength channels. The essential advantage of the schadowband technique[29–34] is that calibration obtained for direct-sun voltage can be directly applied to obtain diffuse atmospheric transmittance.[43,45] The transmittance combined with accurate τ_{ext} data and a radiative transfer model enables the aerosol absorption and single scattering albedo retrievals described in this paper. The paper is organized as follows. Section 2 describes briefly the data sets used in this study. Section 3 provides a description of the UV-MFRSR inversion technique implementation, and Sec. 4 discusses aerosol absorption results and comparisons with AERONET inversion data in the visible wavelengths. Section 5 discusses application of the aerosol UV absorption optical thickness data to explain the bias in satellite surface-UV estimates. A sensitivity study and accuracy assessments of the aerosol UV absorption retrievals are discussed in the appendix (Sec. 7).

6 Conclusions

First, it was demonstrated that an advantage of the shadowband technique in measuring aerosol absorption is that the accurate irradiance calibration can be established by calibrating the direct sun component and comparing with sunphotometers such as AERONET CIMELs. The shadowband method is complementary to the AERONET almucantar retrieval of aerosol single scattering albedo,[25–27] because the retrievals are more reliable at low solar zenith angles. Therefore, combined use of both instruments enables us to derive the complete diurnal cycle of aerosol absorption.

Second, there are specific advantages in measuring aerosol absorption in UV that lead us to believe that ω retrieval results can be used down to $\tau_{ext}\sim 0.2$: (1) the measured accuracy of AERONET reference instruments in the UV with additional pressure and true ozone corrections could be made better than the previously estimated value of ~ 0.01 at 340 nm;[35,36,47] (2) the surface albedo is much smaller in the UV than in the visible spectral region and does not affect the aerosol retrievals as much; (3) τ_{ext} in UV is larger (for the same aerosol mass) than in the visible spectral range; (4) careful characterization of the UV-MFRSR instrument, correction for known systematic errors, monitoring of instrument performance via daily CIMEL intercomparisons, and characterizing atmospheric conditions; (5) stability and repeatability of individual ω retrievals; and (6) ancillary and redundant aerosol measurements available at GSFC site. Indeed, measurement redundancy and instrument intercomparisons were key factors in helping to increase the accuracy of aerosol absorption measurements.

Third, inferred values of the effective UV imaginary refractive index were first used for comparisons of aerosol single scattering albedo ω at 325, 332, and 368 nm and with AERONET retrievals[25] at 440 nm, ω_{440}. The measured differences in absorption between 368 and 440 nm might suggest the presence of selectively UV absorbing aerosols[22,23] or interference from gases other than ozone. However, the differences might also be caused by uncorrected systematic instrumental effects or absolute calibration uncertainties of sky radiances ($\sim$5% for almucantar technique[26]). Continuing colocated measurements at the GFSC location is important to improve the comparison statistics, but conducting these measurements at different sites with varying background aerosol conditions is also desirable.

Fourth, using all cases for cloud-free days, we derived the diurnal and seasonal dependence of aerosol absorption optical thickness τ_{abs} in the UV wavelengths. The expected accuracy of τ_{abs} retrieval from UV-MFRSR measurements is $\sim$0.01 to 0.02, limited by the UV-MFRSR measured accuracy and calibration (V_0). The variability in aerosol size distribution and real refractive index becomes comparable to the measured uncertainties only for large aerosol loadings ($\tau_{ext}>0.5$). The τ_{abs} values show a pronounced seasonal dependence of τ_{ext} with maximum values $\tau_{abs}\sim 0.1$ occurring in summer hazy conditions[5] and <0.02 in winter and fall seasons, when aerosol loadings are small.

Finally, it was found that ω decreases with decreases in τ_{ext}. This could be due to real changes in the average aerosol composition between summer and winter months at the GSFC site. Obviously, continuation of UV-MFRSR measurements at the GSFC site with an enhanced unit (adding 440-nm channel) is important to increase confidence in reported data.