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Partitioning between aerosol and NO₂ absorption in the UV spectral region

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ABSTRACT

A significant database of simultaneous measurements of NO₂ column amounts and aerosol optical properties has recently become available that permits partitioning between aerosol and gaseous absorption. The aerosol column absorption optical thickness, (AAOT) was inferred from the measurements of global and diffuse atmospheric transmittances by a UV-Multifilter Rotating Shadowband radiometer (UV-MFRSR), calibrated using AERONET CIMEL sun-sky radiometers. The NO₂ column amounts were measured using a double-Brewer MK III spectrometer (#171) operated in direct-sun mode using a new 6-wavelength retrieval algorithm. Ancillary measurements of column particle size distribution and refractive index in the visible wavelengths (by AERONET sun-sky almucantar inversions), ozone (by Brewer) and surface pressure constrained the forward radiative transfer model input, so that a unique solution for AAOT was obtained in each UV-MFRSR spectral channel. In fall-winter months with typically dry conditions and low aerosol loadings, the NO₂ absorption represented a significant source of error in aerosol absorption measurements. This was confirmed by UV-MFRSR AAOT retrievals at 325nm, where the NO₂ absorption cross-section is only half the value at 368nm. Thus, the NO₂ correction not only reduces AAOTs obtained from traditional aerosol remote sensing techniques (shadowband or Cimel sunphotometer), but also is capable of changing the spectral dependence of aerosol absorption, which could result in an incorrect interpretation of aerosol composition. To further confirm these findings, a new UV-MFRSR instrument was modified by adding a 440 nm channel to provide spectral overlap with AERONET AAOT inversions in the visible wavelengths.

Keywords: NO₂ absorption, aerosol absorption, single scattering albedo, CIMEL sun photometer, AERONET network, UV multi-filter rotating shadow band radiometer, diffuse fraction measurements

1. INTRODUCTION

Improvements in our ability to model tropospheric photochemistry related to the production of boundary layer smog relies on better knowledge of aerosol absorption properties, trace-gas absorption, and accurate measurements of UV irradiance¹⁻¹². We have recently shown that aerosol measurements made by a standard UV Multifilter Rotating Shadowband Radiometer (UV-MFRSR) and CIMEL sun and sky photometer can be combined to extend the determination of aerosol absorption properties from the visible into the UV¹³⁻¹⁴. Because of the neglect of NO₂ absorption, the uncertainties of individual inversions of aerosol UV absorption under typical urban pollution conditions are currently unknown and require validation and correction using independent NO₂ measurements¹⁵. The possibility of deriving NO₂ column amounts from traditional filter sun-photometer measurements is still debated in the literature¹⁶⁻¹⁹ and spectroscopic measurements are hoped to provide best solution²⁰⁻²². Recently, a significant database of NO₂ column amounts has become available from a Brewer MK III spectrometer (#171) operated in direct-sun mode using a new 6-wavelength retrieval algorithm²².

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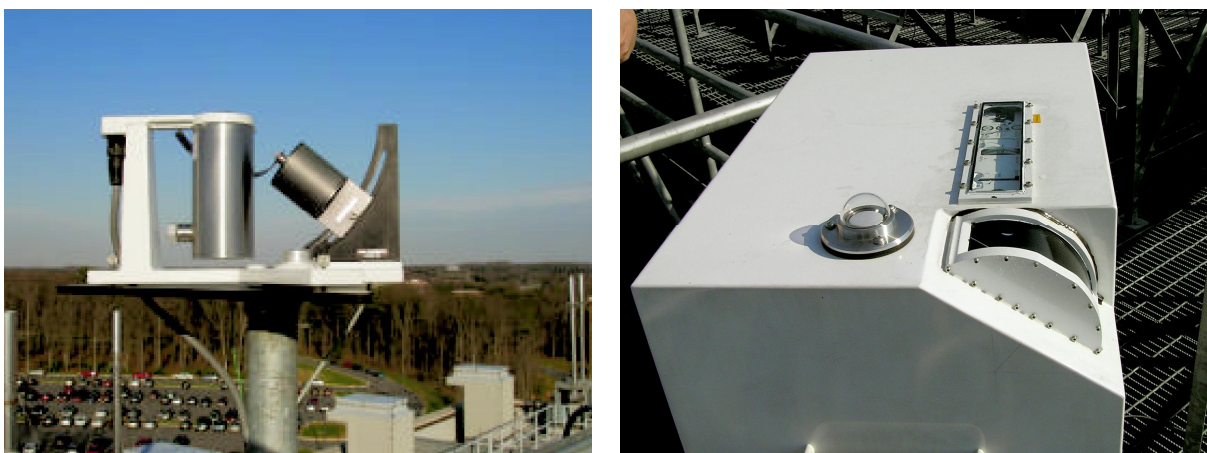


Figure 1 UV-MFRSR shadowband radiometer, part of USDA UV Monitoring and Research Program network (left) and Modified Double Brewer spectrometer (right), were continuously run side by side at NASA GSFC site in Greenbelt, Maryland (lat=39.03 deg N, long=76.88 deg W), in 2002-2004. The instruments are part 4-instrument cluster at NASA Laboratory for atmospheres, Atmospheric Chemistry and Dynamics branch for use in determining trace gas amounts, aerosol column amounts, aerosol optical properties, and UV irradiance.

This paper describes first results of using Brewer NO₂ measurements²² to partition between aerosol and gaseous absorption in the UVA wavelengths (325-368nm). Section 2 describes briefly the data and measuring techniques. Section 3 discusses aerosol single scattering albedo results with and without NO₂ correction and section 4 outlines future plans expanding aerosol absorption measurements into the visible wavelengths using modified UV-VIS MFRSR instrument.

2. DATA SETS

To address the lack of spectral aerosol and gaseous absorption measurements in UV, a suite of four complementary ground-based instruments was established during 2002-2005 and is currently in use at Goddard Space Flight Center in Greenbelt, Maryland. The result has been a self-consistent set of aerosol property measurements for a period of more than one year obtained by combining the CIMEL (NASA AERONET program), UV-MFRSR (USDA UV Monitoring and Research Program), and the NO₂ and ozone measurements obtained every 20 minutes from a co-located Brewer spectrometer. Figure 1 shows location of UV-MFRSR (left panel) and Brewer (right panel) instruments on an elevated platform at the roof of the building. Here we briefly discuss data products and measuring techniques from each instrument.

NO₂ measurements: The Brewer instrument (Figure 1, right) is a double monochromator (282.5 to 363.5 nm with 0.55 nm resolution) similar to those that are widely used to measure ozone and UV radiation reaching the Earth's surface²⁰. We have operated this Brewer since 2000 and have established its high calibration accuracy, good instrument stability, low spectral stray light, and high spectral resolution. Since 2002 we have developed techniques to measure trace gases (NO₂, SO₂, and O₃) from Brewer direct-sun observations²². The 20-minute NO₂ measurements (available since 2003) were used to partition between aerosol and gaseous absorption described in this paper.

Aerosol UV absorption measurements: The primary data set consists of 3-minute measurements of diffuse and total irradiance collected with the UV-MFRSR instrument²³ (optical head #271) from USDA UVB Monitoring and Research Network, UVMRP²⁴. A single measurement cycle consisted of measuring total horizontal irradiance (no sun blocking) following by 3 irradiance measurements with different positions of the shadow band blocking the sun and sky radiance on each side of the sun (at 9°). All spectral channels were measured within one second by 7 separate solid-state detectors with interference filters sharing a common Teflon diffuser.

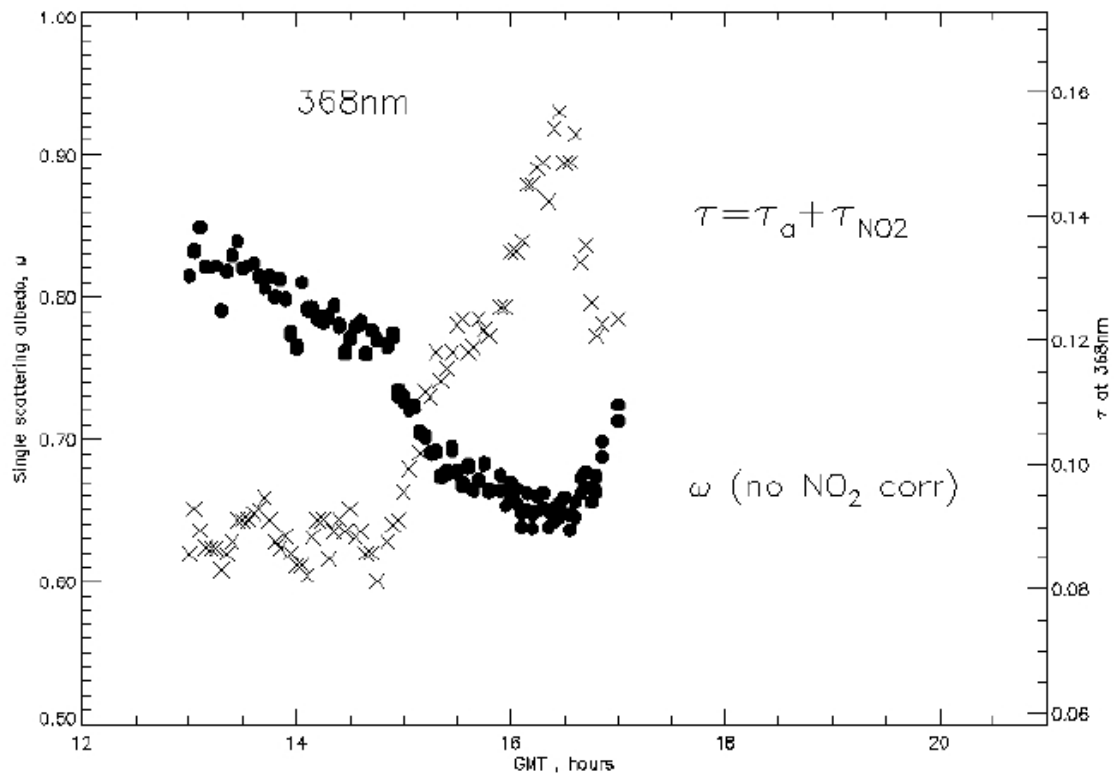


Figure 2 Standard UV-MFRSR retrievals of extinction optical thickness, τ (crosses with right axis scale) and single scattering albedo, ω (black circles with left axis scale) retrievals at 368nm performed at NASA GSFC site on November 10 2003. The retrieval error bars (1σ τ error ~ 0.01 and 1σ ω error ~ 0.06) are not shown for clarity. The actual solar zenith angle was used in retrieval for each 3-min UV-MFRSR measurement. The retrieval assumptions were: surface albedo 0.02, zero NO₂ amount, Brewer measured total ozone, boundary layer aerosol profile and AERONET inverted particle size distribution within ± 60 min of each CIMEL almucantar measurement.

The complete shadowing cycle takes ~ 10 sec and was repeated every 3 minutes throughout the day without averaging of the data. The raw data (voltages) were automatically transmitted every night (via dedicated telephone modem) to the USDA UVMRP processing center at the Colorado State University (Fort Collins, CO) for voltage corrections and further processing. The standard UVMRP calibration procedure differs from that used in our experiments, where we used only cosine corrected voltages calibrated on-site against co-located reference AERONET sunphotometers. This calibration method yields more accurate measurements of aerosol extinction optical thickness τ_a and diffuse and direct atmospheric transmittances¹³.

The aerosol absorption was inferred by fitting UV-MFRSR measured transmittances with the forward radiative transfer model with Mie scattering code incorporated. Ancillary measurements included τ_a , aerosol effective particle size distribution and refractive index in the visible wavelengths (by a CIMEL sun-sky radiometers, NASA AERONET network)²⁵, column ozone and surface pressure. The measurements constrained the model input, so that a unique solution for column average imaginary part of effective refractive index, k , was obtained independently in each UV-MFRSR spectral channel¹⁴. The inferred k values combined with the AERONET particle size distribution data allowed calculation of the column average single scattering albedo, ω .

3. RESULTS

Previous aerosol absorption retrievals both in UV¹⁴ and visible wavelengths²⁵ were performed assuming zero NO₂ amounts. A sample day of standard UV-MFRSR aerosol retrievals is shown in Figure 2. The entire day was completely cloud-free with high-pressure dry weather and clear visibility. Aerosol loading was extremely low in the morning ($\tau_{368} \sim 0.07$) until approximately 10am local time (15UT in Figure 2), when sudden increase was observed in τ_{368} data, which lasted for ~ 1.5 hours. After the maximal value $\tau_{368} \sim 0.16$ was reached at 11.5 local time, τ_{368} started to decline. Unfortunately, UV-MFRSR data were not available in the afternoon due to the shadowing (sun tracking) problem on that day (the problem was solved for subsequent days). However, in the morning UV-MFRSR τ_a data agreed well with interpolated and pressure corrected AERONET (version 1) τ measurements (root mean square differences ~ 0.004). Such good agreement was achieved because of the way the UV-MFRSR was calibrated¹³ and because both datasets were consistently neglecting NO₂ and in effect reporting the sum of aerosol extinction, τ_a , plus NO₂ gaseous absorption optical thickness: $\tau = \tau_a + \tau_{NO_2}$. If no correction is done in the presence of elevated NO₂ amounts, the retrieved single scattering albedo, ω , (Figure 2) is lower than the true aerosol single scattering albedo, ω_a defined as ratio of aerosol scattering and extinction optical thickness: $\omega_a = \tau_a^{scat} / \tau_a$. This can be illustrated qualitatively assuming a homogeneous aerosol-NO₂ mixture and neglecting molecular (Rayleigh) scattering:

$$\omega(noNO_2corr) = \frac{\omega_a \tau_a}{\tau_a + \tau_{NO_2}} \quad (1)$$

It is clear that retrieval of ω_a is only possible if NO₂ absorption optical thickness, τ_{NO_2} is known from *a-priori* measurements. In this work we use independent column NO₂ measurements by co-located Brewer spectrometer²² to correct ω retrievals in 3 longer UVA channels of the UV-MFRSR (Table 1).

Table 1 UV-MFRSR #271 spectral band model ¹⁾

Nominal band wavelength, nm ²⁾	299.845	305.497	311.575	317.730	325.592	332.654	368.011
	UVB channels				UVA channels		
λ_{eff} ³⁾	300.397	305.726	311.706	317.779	325.687	332.636	367.963
λ_{rad} ⁴⁾	300.063	305.313	311.753	317.986	325.808	332.208	367.956
τ_R ⁵⁾	1.216	1.128	1.031	0.947	0.854	0.786	0.5105
τ_{O_3} ⁶⁾	3.335	1.55	0.681	0.292	0.095	0.020	0.00007
τ_a ⁷⁾	0.123	0.121	0.118	0.116	0.113	0.111	0.1
τ_{NO_2} ⁸⁾	0.0034	0.0043	0.0052	0.0061	0.0074	0.0084	0.0148

1) Example is given for standard pressure, 350DU column ozone amount, 1DU NO₂ column amount and $\tau_a = 0.1$ at 368nm (Angstrom parameter $\alpha=1$); 2) Spectral response functions were measured by NOAA Central UV Calibration Facility in air. All wavelengths are shifted to vacuum wavelength scale¹³; 3) Channel weighted effective wavelength at the bottom of atmosphere (neglecting NO₂); 4) Equivalent monochromatic wavelength for direct irradiance at the bottom of atmosphere¹³; 5) Rayleigh scattering coefficients were based on the work by Bates²⁶; 6) The high spectral resolution (~ 0.05 nm) ozone absorption coefficients are based on the laboratory measurements of Bass and Paur²⁷; 7) Nominal aerosol model with $\tau_a(368)=0.1$ and Angstrom parameter $\alpha=1$; 8) τ_{NO_2} was calculated assuming 1 DU NO₂ column amount using high-resolution NO₂ cross-section²⁸, averaged over UV-MFRSR slit functions and weighted by solar spectrum.

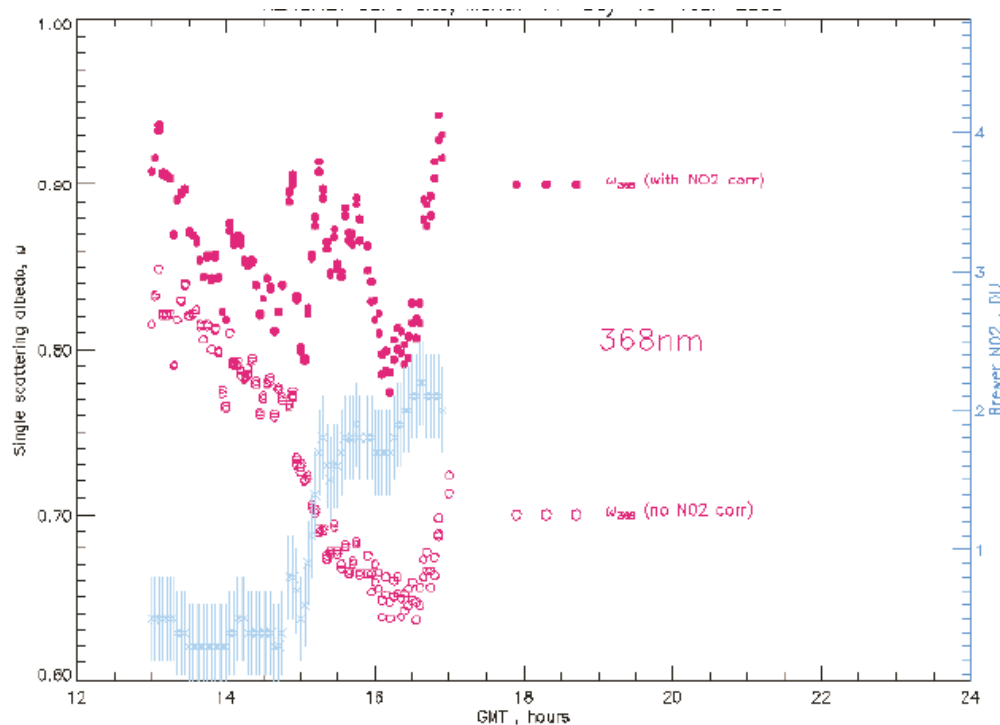


Figure 3. Brewer column NO_2 measurements (with error bars with right axis scale) and UV-MFRSR retrievals of aerosol single scattering albedo before (open circles) and after (filled circles) NO_2 correction applied.

Because of multiple Rayleigh scattering one should not simply invert equation (1) to calculate ω_a even if τ_{NO_2} and τ_a are known. Doing so would underestimate ω correction. Instead, full aerosol inversions should be performed with modified input to the forward multiple scattering radiative transfer (RT) model as follows:

- 1) The Brewer column NO_2 measurements were time interpolated to the 3 minute UV-MFRSR measurements (Figure 3) and τ_{NO_2} values were calculated in each UVA channel using slit-average NO_2 cross sections (τ_{NO_2} is 3 times smaller in the UVB wavelengths Table 1).
- 2) The aerosol extinction optical thickness was decreased by subtracting τ_{NO_2} from the measured total extinction optical thickness (after Rayleigh and ozone corrections): $\tau_a = \tau - \tau_{\text{NO}_2}$
- 3) The τ_{NO_2} was added back to the forward radiative transfer model as gaseous absorption. The NO_2 vertical distribution was assumed in the boundary layer. Steps (2) to (3) preserve total extinction optical thickness, but change partition between aerosol and gaseous extinction.
- 4) Inversions of effective column aerosol absorption were performed as previously described.¹⁴

Figure 3 shows Brewer retrieved NO_2 column amounts as well as ω retrievals with and without NO_2 correction at UV-MFRSR longest UVA channel (368nm). In the morning NO_2 amounts were small $\sim 0.5\text{DU}$, which is comparable to the accuracy of the Brewer NO_2 data²². The UV-MFRSR ω retrievals were within ω error bars (~ 0.06) on that day. However, starting at 10am, the NO_2 amount jumped up to $\sim 2\text{DU}$. The aerosol τ_{368} started increasing at the same time reaching $\tau \sim 0.16$ before noon. However, only 30% of the total τ increase can be attributed to NO_2 pollution ($\Delta\tau_{\text{NO}_2} \sim 0.03$). We interpreted this event as local pollution plume, which moved over site before noon. The plume consisted of both elevated NO_2 and aerosol amounts.

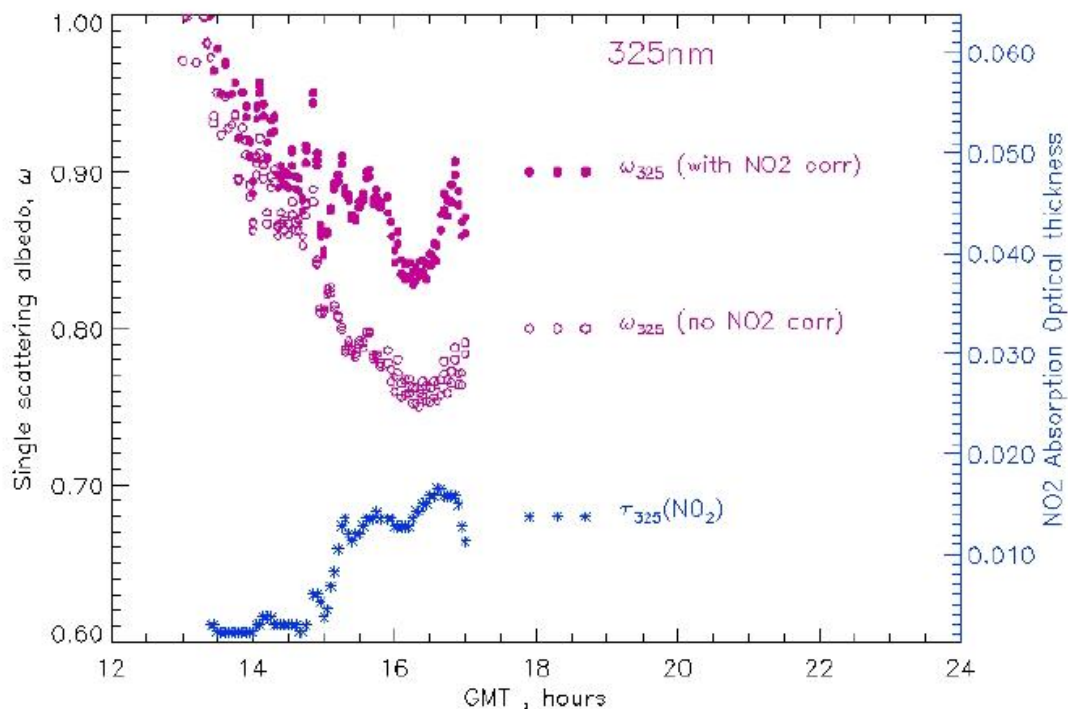


Figure 4 Brewer τ_{NO_2} measurements at 325nm (derived from column NO_2 measurements shown in Figure 3) are shown with stars symbols and right axis scale. UV-MFRSR retrievals of ω are shown before (open circles) and after (filled circles) NO_2 correction with left axis scale.

The diurnal dependence of the true (NO_2 corrected) aerosol single scattering albedo, ω_a , is also shown in Figure 3. As expected, accounting for NO_2 absorption increased retrieved ω_a values proportionally NO_2 amounts with maximal $\Delta\omega < 0.15$. The NO_2 correction was twice as large as other uncertainties in UV-MFRSR ω_a inversions for these conditions (~ 0.06)¹⁴. The NO_2 absorption is therefore a primary source of error in ω_a estimations under low aerosol loading at urban and sub-urban sites where elevated levels of tropospheric NO_2 are expected from local traffic emissions. At GSFC site in Greenbelt low aerosol loadings are typical in the Fall-Winter months under dry and cold weather conditions.

To confirm our findings, the aerosol absorption measurements were repeated at 325nm, with and without NO_2 correction (Figure 4). The NO_2 absorption cross-section at 325nm is about half its value at 368nm (Table 1). As expected, the NO_2 correction has increased retrieved value of ω , but the effect was roughly half of that at 368nm.

The interesting question to consider is whether additional aerosol in the pollution plume had the same composition as the background aerosol? Before NO_2 correction the aerosol absorption appeared substantially larger at 368nm than at 325nm: $\Delta\alpha(\text{no } \text{NO}_2 \text{ corr}) \sim 0.1$. However, after correction applied ω spectral differences were greatly reduced by half: $\Delta\alpha(\text{with } \text{NO}_2 \text{ corr}) \sim 0.05$. Therefore, the NO_2 correction not only reduces τ_a obtained from traditional aerosol remote sensing techniques (shadowband or Cimel sunphotometer), but also is capable of changing the ω spectral dependence, which could result in an incorrect interpretation of aerosol composition.

Qualitatively, the NO₂ correction to ω retrievals is determined by the ratio of NO₂ and aerosol extinction optical thickness at particular wavelength (equation 1):

$$\omega_a = \omega(noNO_2corr) \cdot \left[1 + \frac{\tau_{NO_2}}{\tau_a} \right] \quad (2)$$

In polluted conditions (large τ_a) the correction is small even for significant NO₂ amounts. On the other hand, in clean environments (small τ_a) the NO₂ effect can be significant even for relatively low NO₂ amounts. At GSFC site in Greenbelt, Maryland we expect larger NO₂ corrections in Fall-Winter months, while corrections should be minimal in Summer time.

4. FUTURE WORK

The first UV-MFRSR co-located comparisons with AERONET almucantar data²⁵ in 2002 – 2004 showed 50% larger aerosol absorption in the UV wavelengths than in the visible range (at 440nm)¹⁴. However, the lack of spectrally overlapping measurements with AERONET visible inversions currently prevents attribution of such differences either to differences between techniques, instrumentation/calibration, or presence of larger UV absorption in urban aerosols (organic carbon) and/or gases. In this study we have demonstrated that NO₂ correction applied to UV-MFRSR aerosol absorption retrievals increases the derived value of aerosol single scattering albedo in proportion to the ratio of NO₂ and aerosol optical thickness. However, since the NO₂ cross section is roughly the same at 368nm and 440nm, the NO₂ correction cannot explain these differences if applied consistently to both UV-MFRSR¹⁴ and AERONET²⁵ inversions. To address this issue, we have purchased a new UV-MFRSR unit and have modified it by replacing the 300nm channel to match AERONET CIMEL 440 nm channel. When in operation at GSFC late this summer, the new modified UV-VIS-MFRSR unit will provide spectral overlap with the CIMEL visible wavelength inversions of aerosol absorption (single-scattering albedo) and answer the above uncertainty.

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