Supporting information for:

Constraining aerosol vertical profile in the boundary layer using hyperspectral measurements of oxygen absorption

Zhao-Cheng Zeng[1], Vijay Natraj[2], Feng Xu[2], Thomas J. Pongetti[2], Run-Lie Shia[1], Eric A. Kort[3], Geoffrey C. Toon[2], Stanley P. Sander[2], and Yuk L. Yung[1,2]

[1] Division of Geological and Planetary Sciences, California Institute of Technology

[2] Jet Propulsion Laboratory, California Institute of Technology

[3] Department of Climate and Space Sciences and Engineering, University of Michigan

Correspondence to zcz@gps.caltech.edu

Contents:

Text S1 to S7

Figure S1 to S5

Text S1: AOD extrapolation using Ångström exponent law

The AERONET site at Caltech makes measurements of total AOD, from which aerosol optical properties including single scattering albedo (SSA) and phase function can be retrieved. The wavelength range covered by AERONET-Caltech measurements ranges from 340 to 1020 nm. The AOD value in the O_2 $^1\Delta$ band at 1.27 μm can be estimated using the Ångström exponent law (Seinfeld and Pandis, 2006; Zhang et al., 2015):

$$\frac{\tau}{\tau_0} = \left(\frac{\lambda}{\lambda_0}\right)^{-k} \quad (1)$$

where λ and τ are the wavelength and the corresponding AOD to be interpolated, respectively; λ_0 and τ_0 are the reference wavelength and the corresponding AOD from AERONET, respectively; and k is the Ångström exponent. The k value is obtained by applying linear regression (using the logarithmic form of Equation (1)) to the AERONET AOD measurements at six different wavelengths (340, 380, 440, 500, 870, and 1020 nm).

Text S2: Calculation of aerosol layer height (ALH)

The ALH, which is the center of mass of the scatterers, is calculated in a similar way to Xu et al. (2017) and Koffi et al. (2012):

$$ALH_{MiniMPL} = \frac{\sum_{i=1}^{n} \beta_i \cdot Z_i}{\sum_{i=1}^{n} \beta_i} \quad (2)$$

β_i and Z_i are, respectively, the backscatter signal and the height at level i.

Text S3: GFIT and 2S-ESS models

Gas absorption coefficients and ray paths are computed using the GFIT model (Sen et al., 1996). GFIT has been used extensively for quantitative analysis of solar absorption spectra of the Earth's atmosphere, including the ATMOS shuttle spectra (Irion et al., 2002) and ground based TCCON spectra (Wunch et al., 2011). Surface pressure and atmospheric pressure profiles, which are associated with oxygen vertical distribution, are obtained from the NCEP–NCAR reanalysis dataset (Kalnay et al., 1996) on a daily basis. Details of the atmospheric profiles of trace gas volume mixing ratio, pressure and temperature used in GFIT are described in Fu et al. (2014).

The 2S-ESS model performs an exact computation of the single scattering using all moments of the phase function, while the multiply scattered radiation is calculated using the two-stream approximation. This model has been used for greenhouse gas (GHG) remote sensing in several previous studies (Xi et al., 2015; Zhang et al., 2015, 2016; Zeng et al., 2017). Aerosol optical properties, including SSA and phase function, are taken from AERONET measurements at Caltech, as mentioned in Section 2.2. The total AOD value used in the model is optimized to match the CLARS radiance measurement, as described in Section 3.2.

Text S4: Fitting of sorted spectra

To minimize the impact of data noise on the comparison, we fit the sorted spectra using Equation (3), which is formulated to quantify the spectral shape:

$$f(x) = a_1 - a_2 * (1 - x) - a_3 * \exp(-a_4 * x) \tag{3}$$

where a_1 is the largest radiance at the continuum level; a_2, a_3, and a_4 are parameters to be fitted. x is the sorted channel number, ranging from 1 to 3982, and normalized to be between 0 and 1 when doing the fitting. Assuming the absorption lines are well resolved, then the exponential part of the formula, based on the Beer-Lambert extinction law, approximates the oxygen line by line and collision-induced absorptions. The linear part of the formula is used to provide a first order approximation of the continuum shape (e.g. continuum tilt) and the variation of the instrument response across the window that are not accounted by the exponential part. Even when the spectral absorption lines are not fully resolved, we found this formula well capture the spectral shape. The spectral data are filtered by excluding anomalous data more than 1.5 standard deviations away from the mean and the nonlinear fit is then implemented using a standard least squares regression. The fitting results are shown in Figure S5(c).

Text S5: Retrieval using look-up tables (LUTs)

As shown in Figure S5, two LUTs are built to successively retrieve the total AOD and effective ALH. The total AOD is retrieved using the observed CLARS-level reflectance at the continuum level (Figure S5(b)). On the other hand, the reflectance in the intermediate absorption window is used to retrieve the effective ALH (Figure S5(d)). Using the retrieved effective ALH, the geometric thickness (GT) of the aerosol layer can be derived from the empirical correlation as shown in Figure 1(b). As described in Section 2.3, the GT of the aerosol layer in this study is defined as the ratio of the integrated total aerosol loading (represented by NRB) over all different levels to the maximum aerosol loading.

The retrieved profile in Figure 2(f) is reconstructed by assuming a Gaussian distribution. The mean (μ) of this distribution is the retrieved effective ALH, while the standard deviation (σ) is calculated in the following way. An aerosol vertical profile following the Gaussian distribution is given by:

$$f(x) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(\frac{-(x-\mu)^2}{2\sigma^2}\right) \tag{3}$$

where x is the height and $f(x)$ is the aerosol vertical profile. The maximum value of the profile is $\frac{1}{\sigma\sqrt{2\pi}}$ when $x = \mu$. Since the integral of the Gaussian distribution $f(x)$ is unity, the GT of this profile, defined as the ratio of integrated $f(x)$ to the maximum value $\frac{1}{\sigma\sqrt{2\pi}}$, is $\sigma\sqrt{2\pi}$. As a result, $\sigma = \mathrm{GT}/\sqrt{2\pi}$. Using the retrieved μ and calculated σ, the aerosol vertical profile can be constructed as shown in Figure 2(f).

Text S6: Phase function and SSA from satellite observations and model simulations

Knowledge of the aerosol phase function and SSA are important caveats in applying the proposed algorithm. These parameters can be obtained using AERONET measurements. However, in the absence of AERONET data, satellite observations and/or model simulations can also be employed to characterize them. For example, the phase function can be retrieved using MISR (Diner et al., 2005) with its multi-angle capability, while SSA can be retrieved from several different instruments and simulations by global chemical models with improving accuracy (e.g., Jethva et al., 2014; Kinne et al., 2003). On the other hand, ALH is much less constrained (higher uncertainty in retrievals) by current measurements or model simulations. Therefore, the proposed algorithm has the potential to be applied on a global scale (including regions without AERONET measurements) to derive aerosol parameters that are currently unavailable.

Text S7: Calculation of surface albedo from CLARS-FTS measurements

One of the advantages of the CLARS geometry is that the surface albedo (shown in Figure S2(b)) can be calculated by dividing SVO-observed (incident sunlight) by LABS-observed (reflected sunlight) radiance on clear days using measurements at continuum wavelengths where gas absorption can be ignored. These derived surface albedos are used in the 2S-ESS RT model. In this study, the assumed surface albedos between 0.15 and 0.20 are typical values for urban settings such as those in Los Angeles. For bright surfaces such as deserts, the accuracy of this method needs further investigation. Conceptually, if the surface reflectance is large, then the relative contribution from aerosol to the total observed radiance is small. With smaller contribution from aerosol scattering, the look-up tables in Figures S5(a) and (c) will have smaller spectral variability for different AOD and ALH scenarios. As a result, the smaller spectral variability will lead to a larger uncertainty in retrievals. Wang et al. (2014) and Ding et al. (2016) have shown that, for bright surfaces, the sensitivity of radiance to ALH decreases. They recommend polarimetric measurements to improve sensitivity.

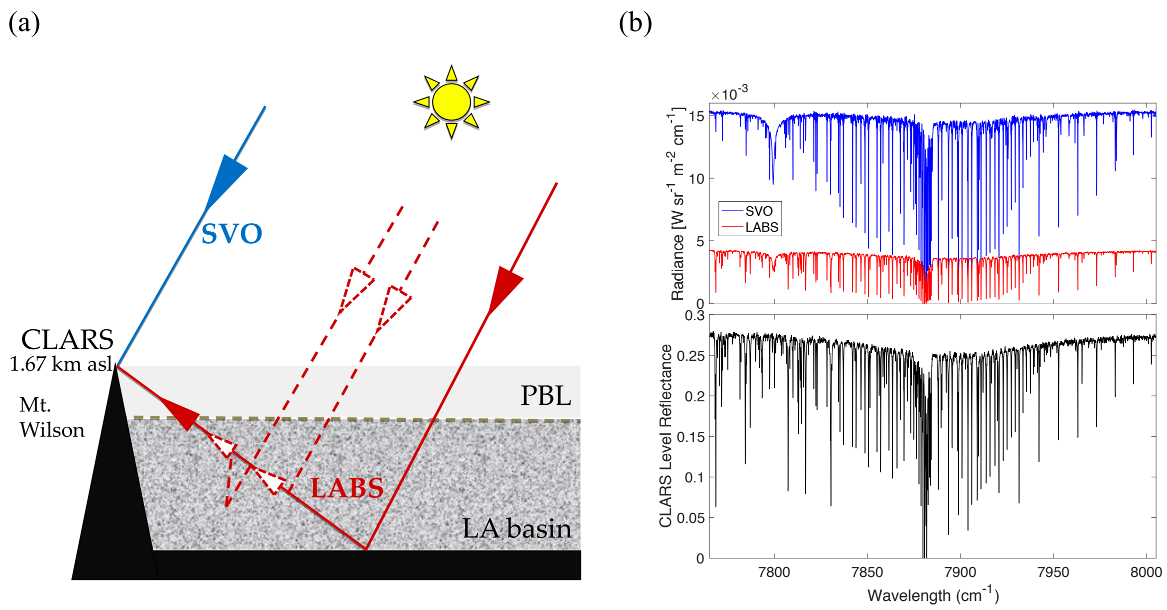

Figure S1. (a) Schematic figure of CLARS observation over the Los Angeles basin. CLARS has two modes of operation: the Los Angeles Basin Survey mode (LABS; in solid red) and the Spectralon Viewing Observation mode (SVO; in blue). An example of light path changes due to aerosol scattering along the path from the basin to the mountain top is illustrated (single and multiple scattering in dotted red); (b) Examples of CLARS-FTS measurements in the oxygen band at 1.27 µm. The top panel shows the observed radiance from SVO (blue) and LABS (red) modes, where the LABS measurements are acquired over the West Pasadena surface target. These measurements are made at 14:00 h on September 17, 2013 with a solar zenith angle of 46.43°. The bottom panel shows the CLARS level reflectance, which is the ratio of the LABS and SVO radiances shown in the top panel.

(a)

(b)

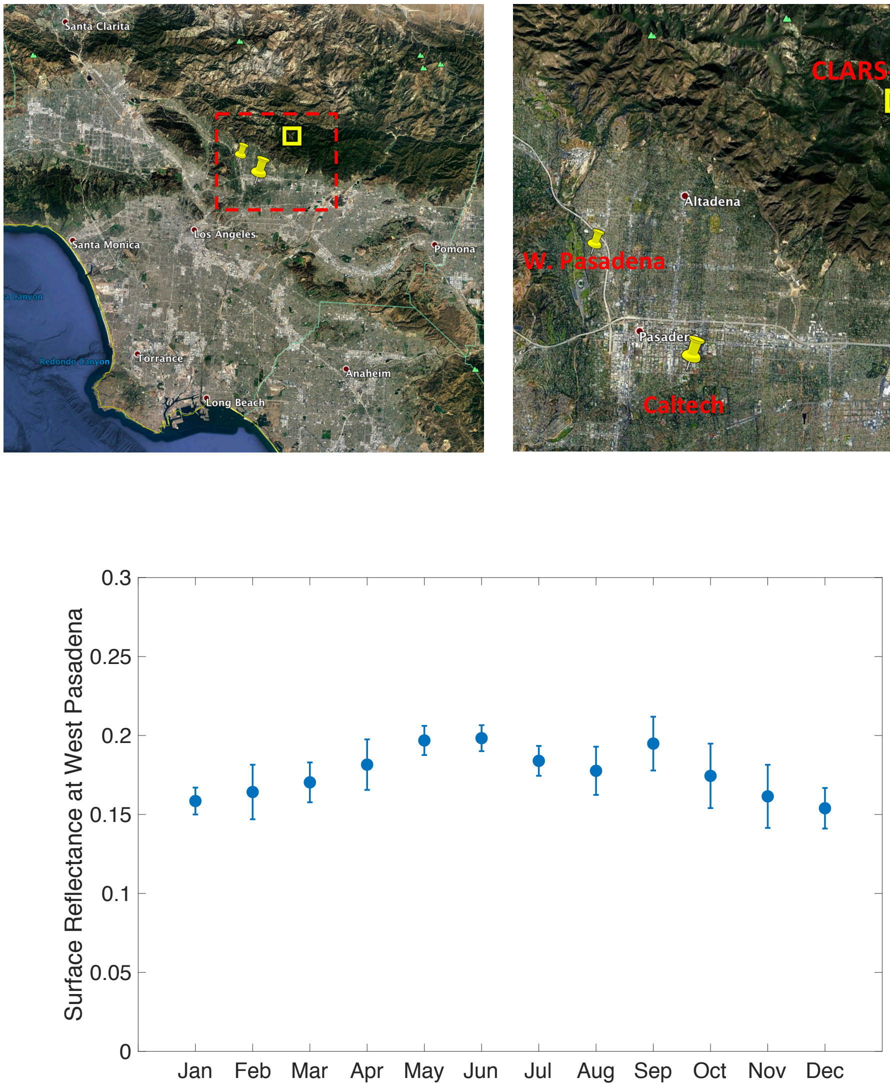

Figure S2. (a) Locations of the CLARS FTS instrument, the West Pasadena surface target, and Caltech (where the AERONET and MiniMPL instruments are located). The horizontal distance from the West Pasadena surface reflection point to Caltech is about 5 km, and that from CLARS-FTS to both West Pasadena and Caltech is about 11 km; (b) Monthly averaged surface reflectance at 1.24 μm at West Pasadena. The surface albedo at a particular surface target can be estimated by dividing SVO-observed (incident sunlight) by LABS-observed (reflected sunlight) radiance on relatively clean days using continuum wavelengths in the 1.24 μm spectral region where gas and aerosol extinction can be ignored. A scale factor is derived using the 2S-ESS RT model to correct for small effects from aerosol scattering using, the AOD and aerosol optical properties obtained from the AERONET instrument at Caltech. The error bars (one standard deviation) indicate the uncertainty in the surface albedo estimates.

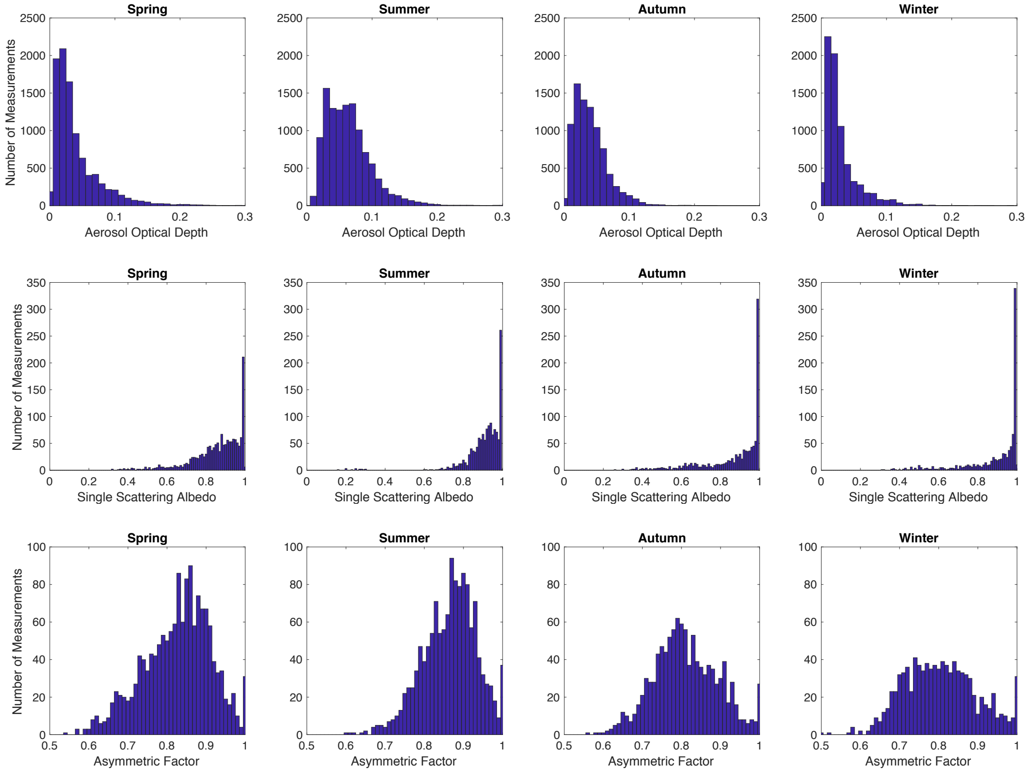

Figure S3. Monthly histograms of (top) aerosol optical depth; (middle) single scattering albedo; and (bottom) asymmetry parameter obtained from AERONET measurements at Caltech from 2011 to 2017.

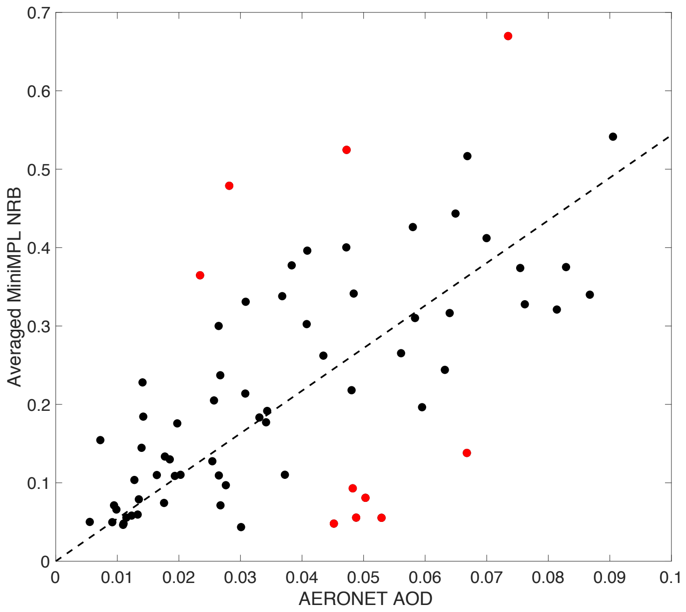

Figure S4. Correlation plot between averaged MiniMPL normalized relative backscatter signal and AERONET AOD at 1.27 µm. Measurements that deviate by more than 1.5 standard deviations from the mean (red dots) are excluded from Figure 3. The reason for the large differences may be the inhomogeneous spatial distribution of aerosols.

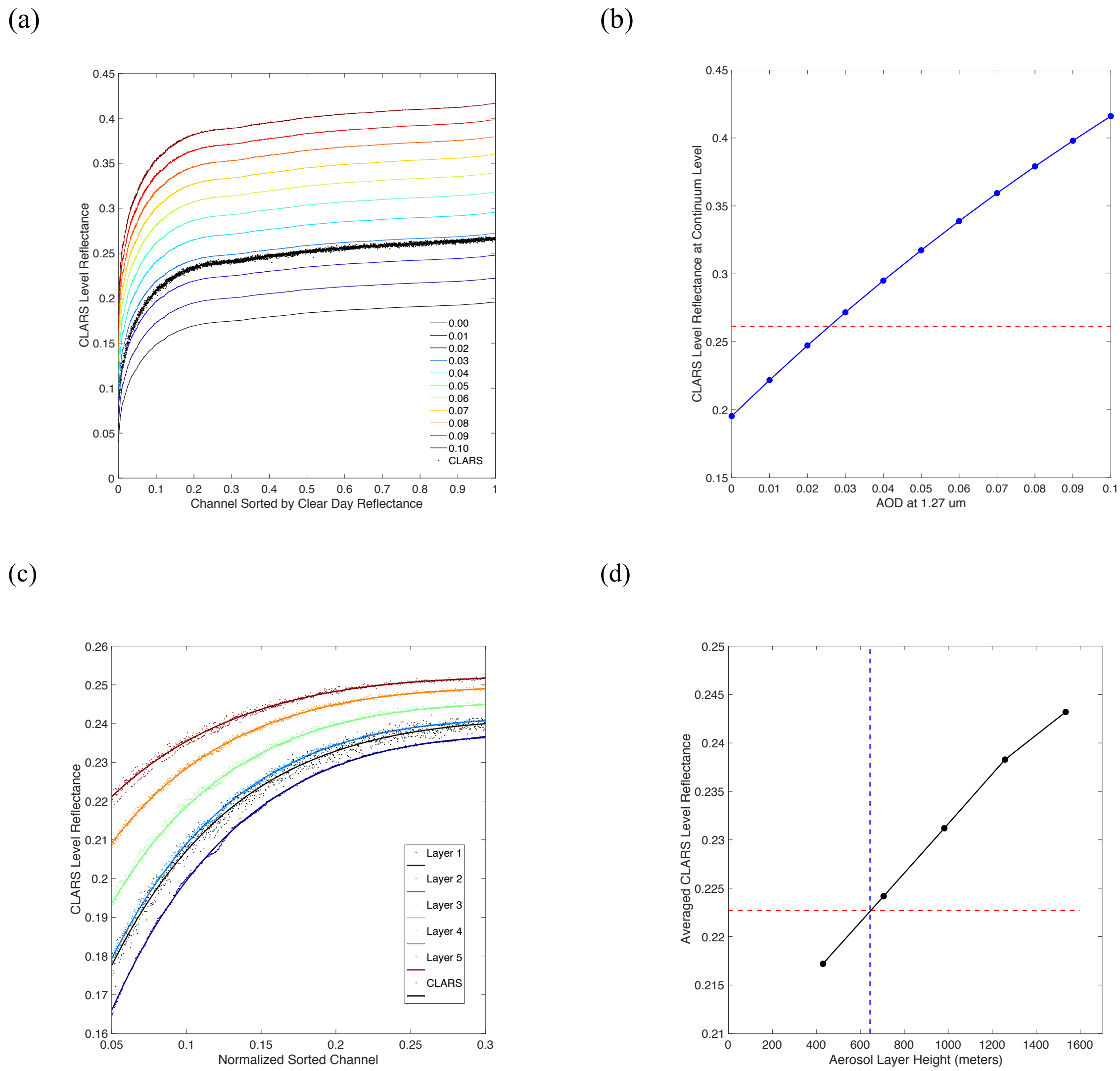

Figure S5. Examples of retrievals algorithms based on look up tables. The retrieval process can be separated into two steps. First, retrieve total AOD by constructing a look up table of simulated spectra using different values of total AOD, as shown in (a) and calculating the reflectance at the continuum level (the highest reflectance value), as shown in (b). In practice, to minimize uncertainty, the mean of the highest 50 reflectance values is used as the continuum level reflectance. Here, the aerosol is assumed to be vertically well-mixed. Second, retrieve the effective ALH after retrieving total AOD. The total AOD is uniformly partitioned into each of the five layers in the RT model, the simulated spectra are fitted using Equation (3) and finally compared with CLARS measurements, as shown in (c). In this analysis, the intermediate absorption band window (values between 0.05 and 0.3 of normalized sorted channel value), which shows the largest sensitivity to aerosol vertical structure, is used. Different metrics can be used to quantify the difference in reflectance between model simulations and measurements. Here, we use the mean value of reflectance over the intermediate absorption window calculated by averaging all CLARS level reflectance values, and build the look up table, as shown in (d). The dotted red line corresponds to the mean reflectance value of the CLARS measurement. The dotted blue line indicates the retrieved effective aerosol layer height.