A portable reflected-sunlight spectrometer for measuring atmospheric CO_2 and CH_4: Accounting for aerosols

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1 Reflected-Sunlight Measurements

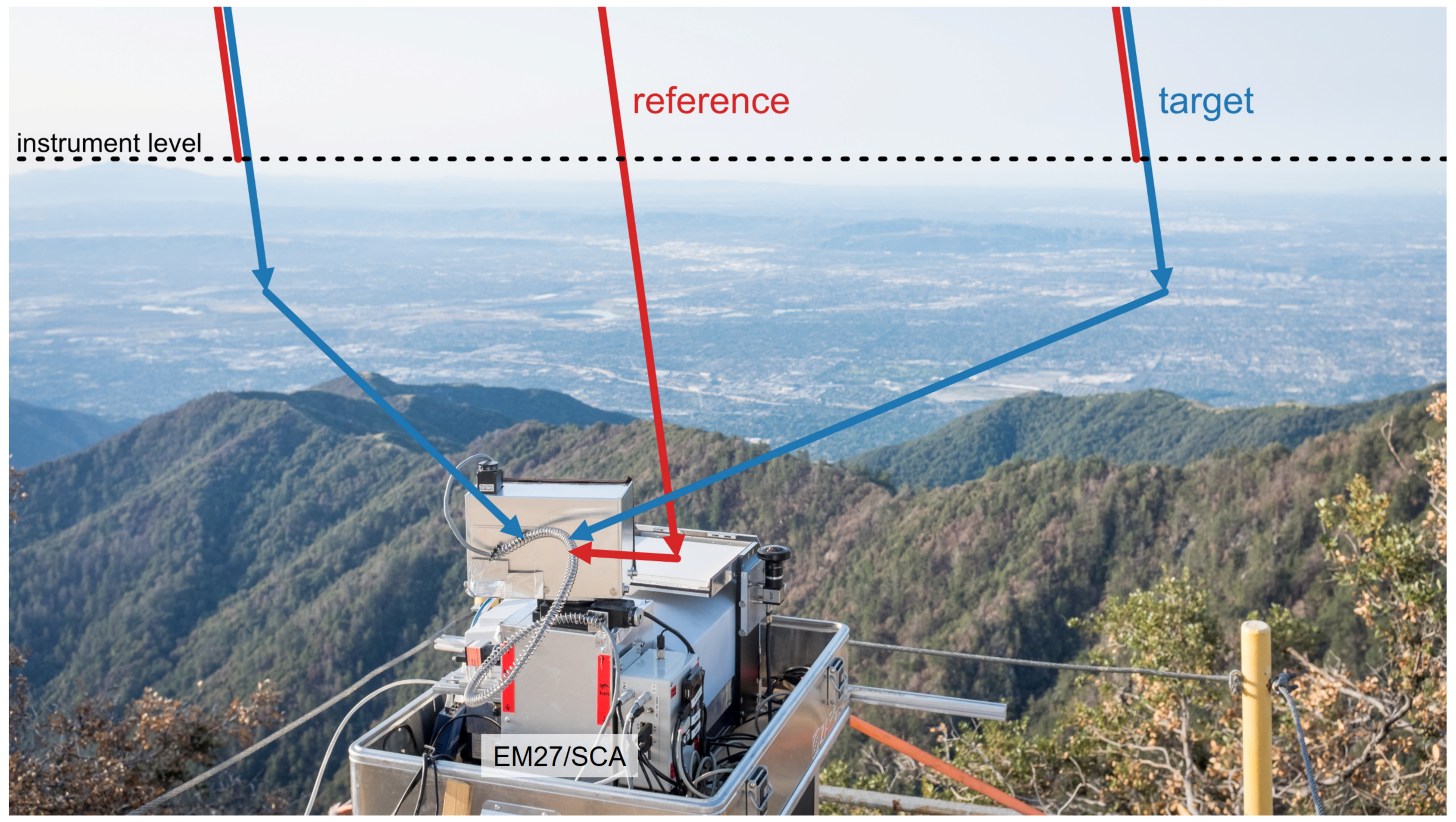

Figure 1: The EM27/SCA FTS (Löw et al. 2023) placed on Mt. Wilson ***observing the Los Angeles basin****. The instrument collects sunlight reflected by ground targets in the basin (blue light path). A reflector plate at the instrument is observed to constrain the above instrument column (red light path).*

The reflected-sunlight measurement geometry has several advantages:

- The measurements are more sensitive to concentrations close to the ground, due to the horizontal path component, and therefore ideal for emission monitoring.
- Spectra can be recorded independent of the sun's position, allowing for a flexible choice of observation targets.
- Together with atmospheric scattered sunlight spectra insights into atmospheric scattering processes can be gained.

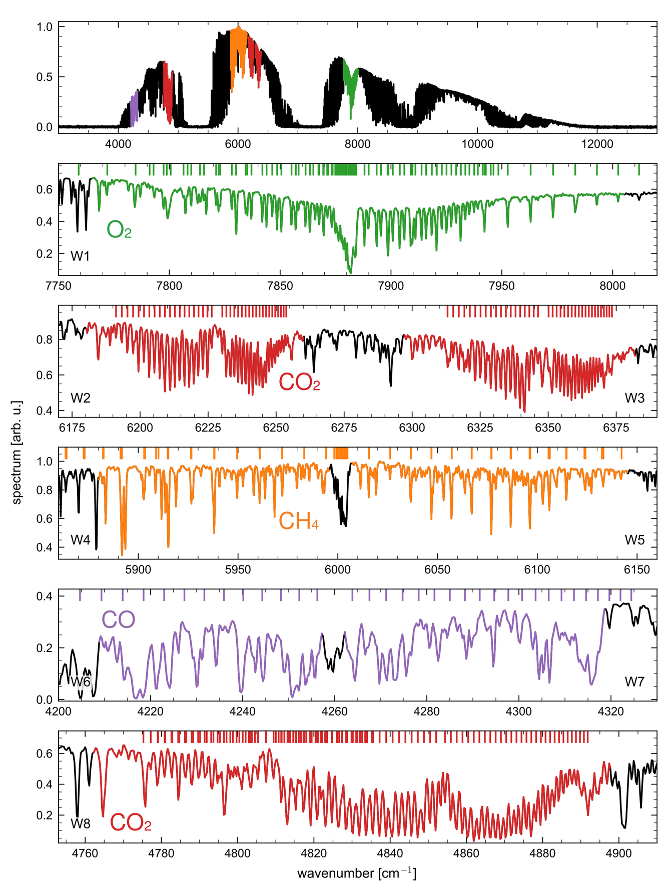

Figure 2: Typical ***reflected-sunlight spectrum*** *of the Baldwin Park (BP) target at around 15:50 LT (at SZA 46.6). The top panel shows the full spectrum. The panels below show the sections from which absorber column densities are retrieved.*

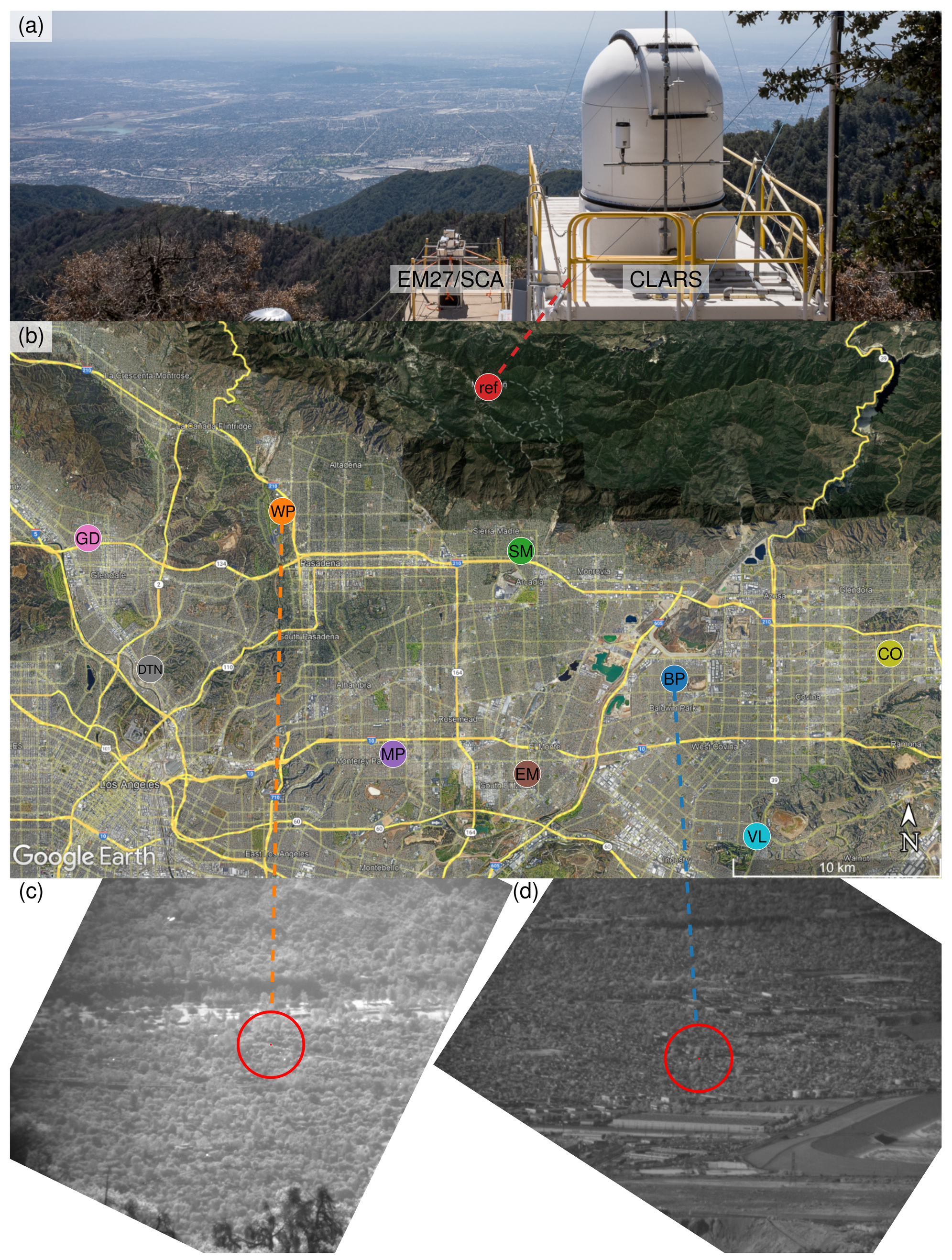

Figure 3: The EM27/SCA and the CLARS-FTS (Fu et al. 2014) side-by-side on Mt. Wilson (ref) observing nine ***ground scattering targets*** *(other colored markers) in the LA Basin. The lower panels show FOV camera images of the West Pasadena (WP, c) and Baldwin Park (BP, d) targets. The instrument FOV is marked with a red circle.*

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Take-Home Message

- Aerosol scattering causes shortening of the effective light path in reflected-sun measurements.
- The effect does not cancel in XCO_2 (or XCH_4) due to wavelength dependency of scattering.
- Full physics retrieval which retrieves also aerosol properties can mitigate the bias.

2 Non-Scattering Retrievals

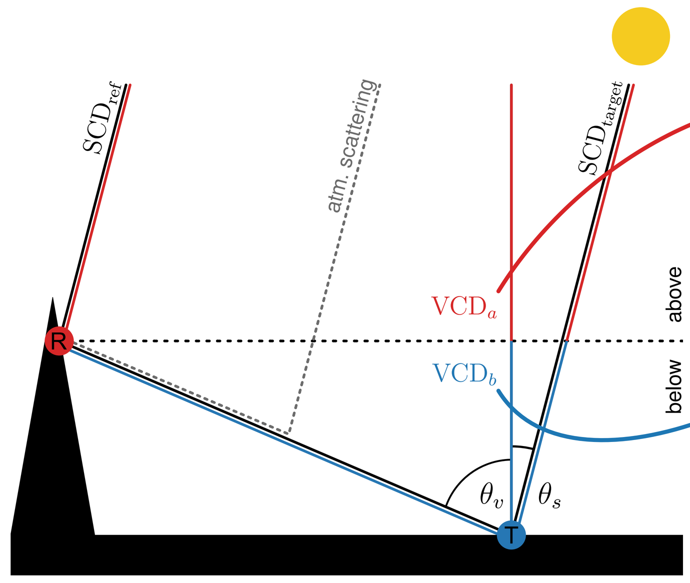

Figure 4: ***Observational geometry*** *of reflected-sun measurements. The partial VCD_a (red) is retrieved from reflector measurements. The VCD_b (blue) below instrument level is derived from the difference of target (T) and reflector (R) measurements.*

Our retrieval algorithm is based on the RemoTeC radiative transfer and retrieval algorithm, previously employed for solar backscatter satellite measurements.

We retrieve SCDs[1] of O_2, CO_2, CH_4 and CO from the absorption spectra and convert them to partial VCD[2] above and below instrument level (Fig. 4).

$$VCD_a = \frac{SCD_{ref}}{AMF_a}, \quad VCD_b = \frac{SCD_{target} - SCD_{ref}}{AMF_b}$$

$$AMF_a = \frac{1}{\cos(\theta_s)}, \quad AMF_b = \frac{1}{\cos(\theta_s)} + \frac{1}{\cos(\theta_v)}$$

The comparison of partial VCD retrieved from EM27/SCA and CLARS-FTS spectra show good agreement (Fig. 5).

The main variability in the partial is caused by aerosol-induced shortening of the effective light path.

Aerosol scattering is the main error in the non-scattering retrieval due to the long horizontal light path and the shallow viewing angle.

The ratio between retrieved and geometric SCD quantifies the influence of aerosol scattering (Fig. 6).

The light path shortening increases with smaller scattering angle and with higher AOD[3].

The ratio between the GHG and the O_2 column (XGHG) does not cancel the scattering error fully due to the wavelength dependency of scattering (black in Fig. 8).

[1] SCD: slant column density
[2] VCD: vertical column density
[3] AOD: Aerosol optical depth

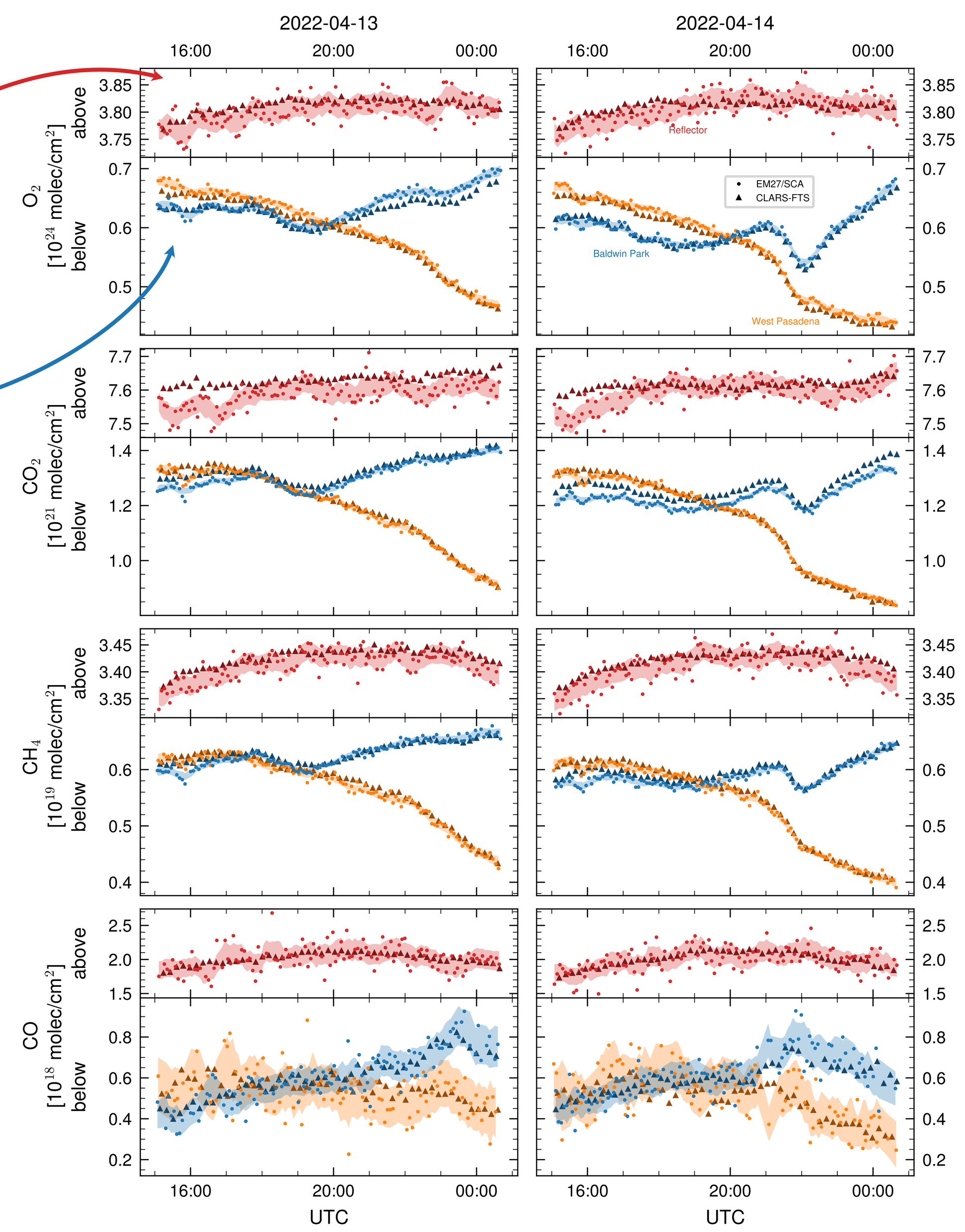

Figure 5: ***Time series*** *of the VCD_a (above instrument level) and VCD_b (below instrument level) of O_2, CO_2, CH_4, and CO (top tow to bottom row) measured by the EM27/SCA (dots) and the CLARS-FTS (triangles). VCD_a is calculated from reflector measurements (red). The VCD_b is calculated from the difference between measurements of the WP (orange) and BP (blue) targets and the reflector. The EM27/SCA precision error is displayed as shading around the rolling average.*

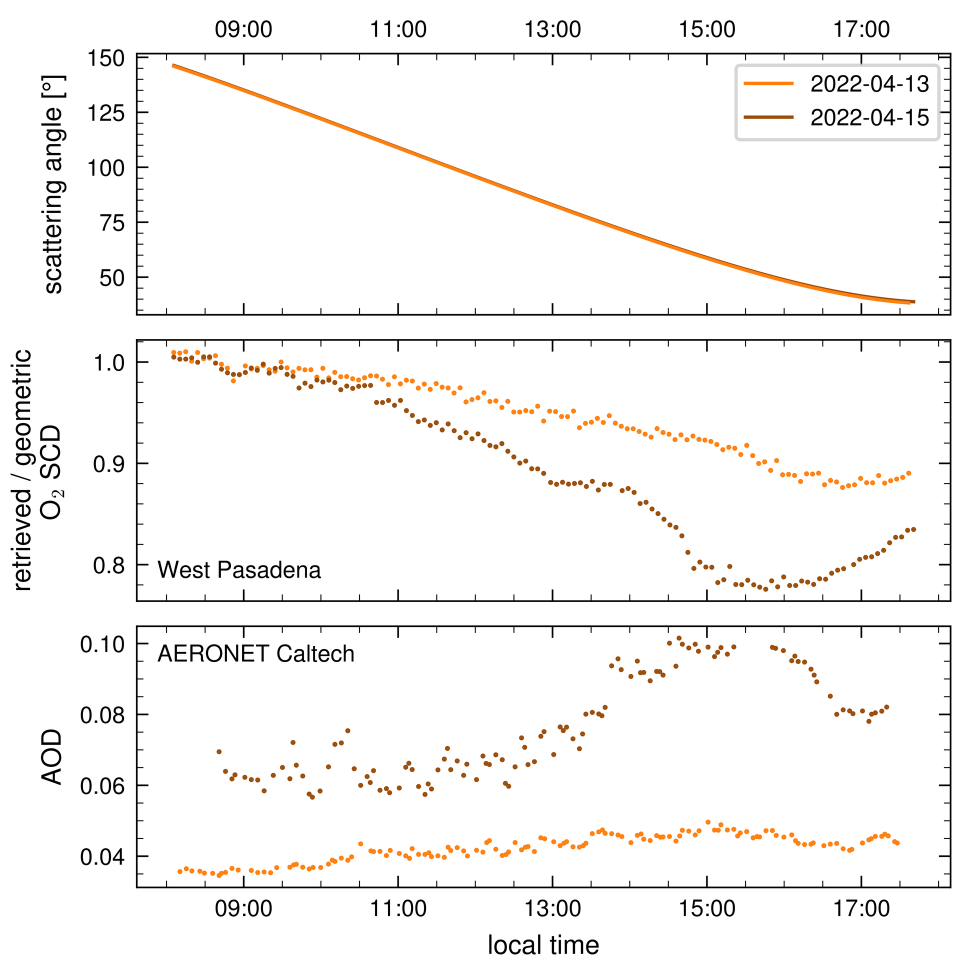

Figure 6: ***Impact of aerosol scattering*** *on the retrieved column densities. The ratio of retrieved to geometric O_2 slant column density (middle) quantifies the change in effective light path. Smaller scattering angle (top) and higher aerosol optical depth (bottom).*

3 Accounting for Aerosols

We include aerosol scattering below instrument level in our retrieval.

Aerosol parameters (size, layer height, abundance) are simultaneously retrieved with the absorber columns.

The optical properties are computed from the aerosol parameters.

O_2 is no longer retrieved in favor of the aerosol information.

The VCDb retrieved with the scattering retrieval do not show the strong light path shortening effect (Fig. 7).

The XGHG is calculated from the retrieved SCD and the a priori O_2 SCD.

The scattering-induced high bias is mitigated by explicitly including aerosols in the retrieval (Fig. 8)

- Non-scattering retrieved XCO_2 is larger towards low retrieved/geometric O_2, despite using retrieved O_2 for the XCO_2
- Scattering retrieved XCO_2 shows a much shallower slope.

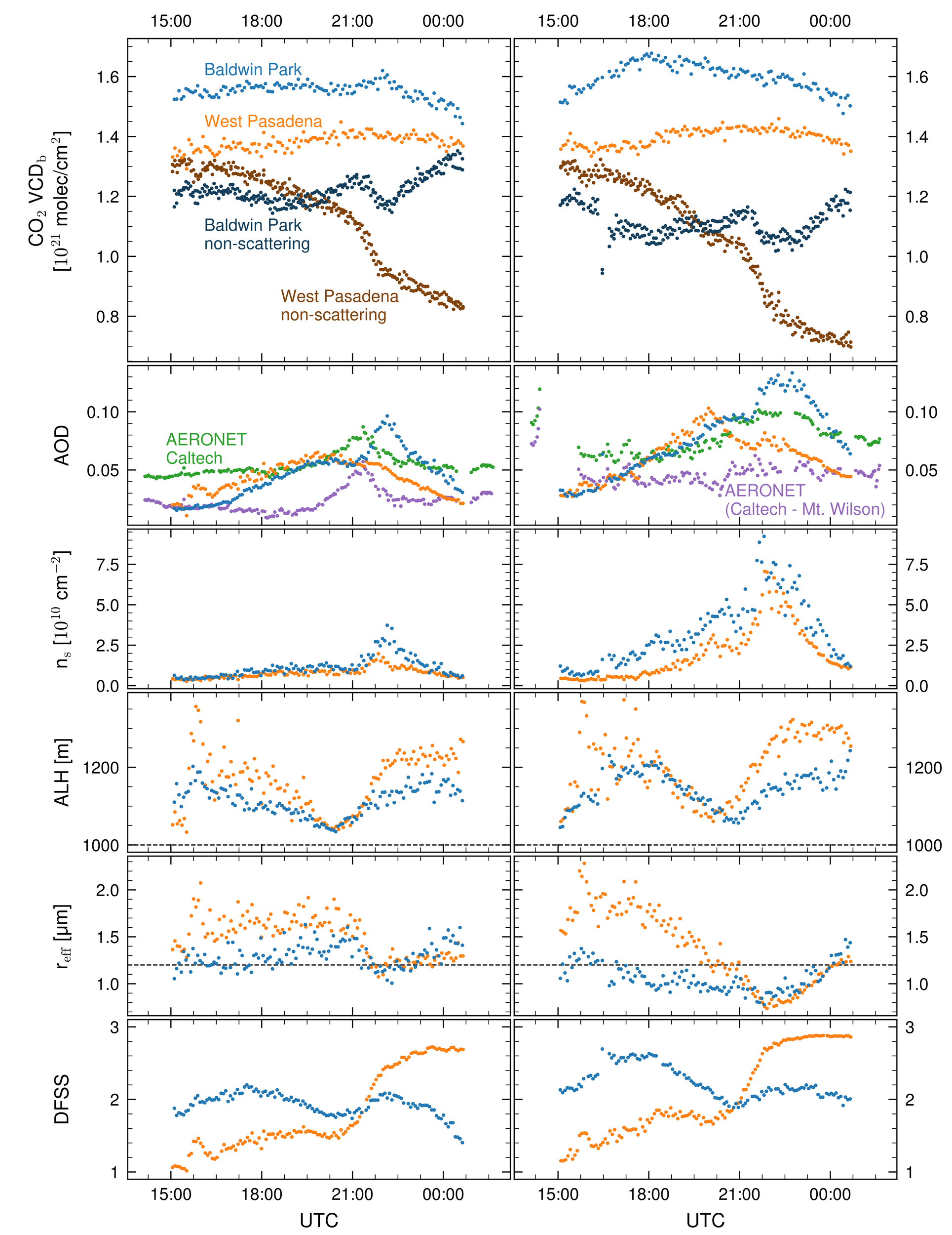

Figure 7: Scattering retrieval results for VCD_b and aerosol parameters.

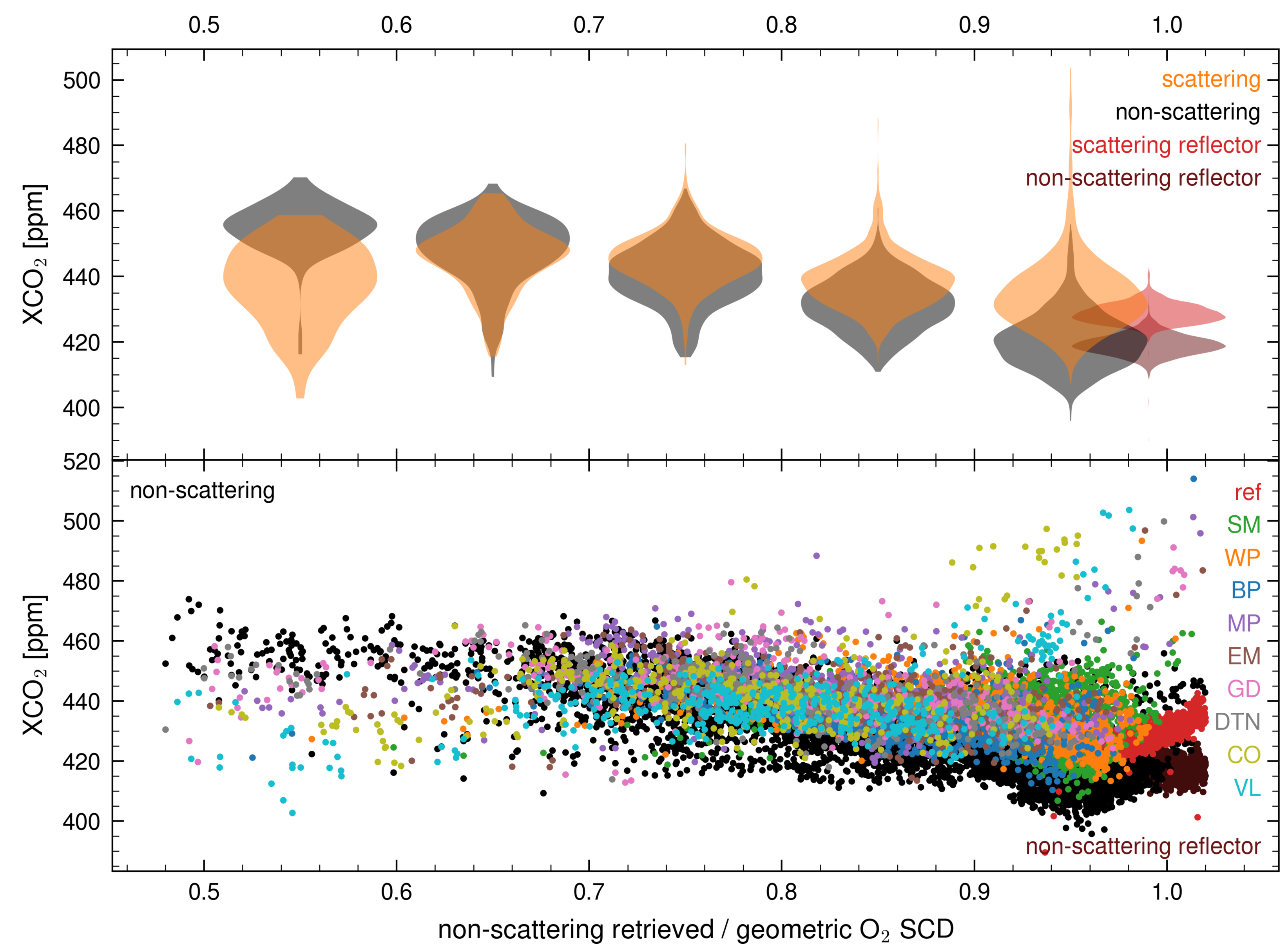

Figure 8: Scattering-induced bias in XCO_2 is much smaller for the scattering retrieval (colors) than for the non-scattering retrieval, despite using the retrieved O_2 SCD only in the non-scattering case. The upper panel shows the distribution for 0.1 wide O_2 ratio intervals.

References

Fu et al. (2014), *Near-infrared remote sensing of Los Angeles trace gas distributions from a mountaintop site*, Atmos. Meas. Tech., 7, 713–729, https://doi.org/10.5194/amt-7-713-2014

Löw et al. (2023), *A portable reflected-sunlight spectrometer for CO_2 and CH_4*, Atmos. Meas. Tech., 16, 5125–5144, https://doi.org/10.5194/amt-16-5125-2023

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