Day and night-time retrieval of vertical and columnar aerosol properties using GRASP with sun photometer and lidar measurements during an episode of Canadian wildfires smoke transported over the Atlantic.

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Introduction

Aerosol particles significantly impacts climate and air quality, making their accurate characterization essential. The GRASP algorithm (Generalized Retrieval of Atmosphere and Surface Properties, Dubovik et al., 2021) allows the use of sun-lunar-sky photometers and multi-wavelength Lidar data to obtain aerosol microphysical and optical properties in the atmospheric column and vertically. In this manner, this work presents an inversion strategy that integrates aerosol optical depth (AOD) and sky radiance data from the AERONET station at Izaña Atmospheric Observatory, along with Range Corrected Signals (RCS) at 532nm and 808nm from the CE376 CIMEL lidar into the GRASP algorithm. As a result, vertical profiles of extinction, backscattering, volume concentration, and Angstrom exponent are obtained, along with the single scattering albedo and the aerosol size distribution for an event that combined Saharan dust with smoke from wildfires in Canada, on June 30, 2023. The obtained aerosol properties are compared with other methods such as AERONET (Sinyuk et al., 2020) and a modified two-wavelength Klett inversion method implemented specifically for the CE376 lidar (Sanchez-Barrero et al., 2024). This study aims to lay the groundwork for future research combining CE376 lidar and sun-lunar-sky photometers with GRASP inversion algorithm, by presenting preliminary results characterizing the aerosols present, and analyzing their key differences from established inversion methods.

Methodology

We used Level 1.5 AOD and raw sky radiance data from both Almucantar and Hybrid scan methods at 440, 675, 870, and 1020 nm from AERONET, along with RCS signals at 532 and 808 nm obtained from the CE376 lidar. The AOD and sky radiance data were filtered and processed following the methodology of Roman et al. (2018). The RCS data were denoised using a wavelet transform with local thresholding, as described by Mao (2012), and then further filtered and processed using the Roman et al. (2018) approach. Following this, GRASP retrievals were performed if for each scan scenario:

1. The number of sky radiance points for each wavelength is greater or equal to 10.

2. For each wavelength there is at least one point within the scattering angles ranges: $\geq 3.2^{\circ}$ to 6° , $\geq 6^{\circ}$ to 30° , $\geq 30^{\circ}$ to 78° , and $\geq 78^{\circ}$.

3. A valid AOD measurement must be available within ±16min for each wavelength.

Specifically, the lidar+photometer inversion, formerly known as GARRLiC (Generalized Aerosol Retrieval from Radiometer and Lidar Combined data) and now integrated into the GRASP algorithm (Dubovik et al., 2021), was employed. In this case, with RCS signals at 532 nm and 808 nm, the inversion strategy was configured assuming a bimodal size distribution, comprising 10 logarithmically spaced triangular intervals for the fine mode and 15 for the coarse mode, covering a total radius range from 0.05 µm to 15 µm.

Additionally, real and imaginary refractive indices, lidar ratio, and columnar single scattering albedo (SSA) were treated as distinct for each mode and allowed to vary with the available wavelengths (440, 523, 675, 808, 870, and 1020 nm). The fraction of spherical particles was also retrieved. The vertical aerosol distribution was assumed to vary with altitude and differ between the fine and coarse modes.

This two-mode inversion enabled the retrieval of vertical profiles for various aerosol volume concentration for each mode, and Angstrom exponent, as well as backscattering, extinction, for each wavelength and mode. Finally, the errors associated with the inversion of each product retrieved through GRASP were calculated, following the methodology of Herrera et al. (2022).

0.030-AERONET **(b)** (a) \rightarrow GRASP Coarse GRASP Fine ---- GRASP total 0.950.025 $\left[\mu m^{3}/\mu m^{2}\right]$ 0.90[... M.W. [...] 0.85 dV(r)/dln(r) 0.01 0.01 SSA 0.800.005AERONET \rightarrow GRASP Coarse GRASP Fine \rightarrow GRASP total 0.000 10.05321020 0.00.11.0100 440 675 808 870 Radius $[\mu m]$ Wavelength [nm]

Results

Regarding the size distributions obtained by GRASP and AERONET (Figure 1a), both show strong overall agreement, with the AERONET distribution falling within the error range of the GRASP results. The most notable difference is observed in the coarse mode (blue), where AERONET indicates a higher concentration at the mode's peak. Both size distributions reveal the presence of a dominant coarse mode along with a significant fine mode, which is shifted towards larger radii—a characteristic typically observed in long-range transported boreal biomass burning aerosols.

The complexity of the event is evident in the SSA curves (Figure 1b). While there is generally strong agreement between AERONET and GRASP for the fine mode at shorter wavelengths and for the total SSA in longer ones, with the AERONET curve remaining within GRASP's error margins, the coarse mode shows lower values with larger errors at shorter wavelengths. This can be attributed to the medium aerosol load (AOD at 440 nm around 0.19) and the reduced sensitivity to the coarse mode, which is optically less dominant in this scenario. In terms of the curves' behavior, both the fine and coarse modes obtained by GRASP, display the typical characteristics associated with the aerosols involved in the event, namely biomass burning and Saharan dust.



Figure 1: Volume size distributions obtained by GRASP (total and by mode) with error (shaded) and AERONET (a), and single scattering albedo (SSA) obtained by GRASP (total and by mode) with error (shaded) and AERONET (b) at wavelengths of 440, 532, 675, 808, 870, and 1020 nm for June 30, 2023, at 10:13:36 UTC.

Examining the vertical profiles of aerosol volume concentration (Figure 2a) and Angström exponent (Figure 2b), these clearly show the vertical structure of the aerosols present in the event and their concentration. The fine mode (in red) exhibits a smoother vertical distribution with lower concentrations compared to the coarse mode (in blue), which shows more pronounced peaks. Both figures put in perspective how the main layer of smoke, situated between 3500–4000 m a.s.l. with maximum concentrations of 20 µm³/cm³, is enshrouded by two distinct layers of Saharan dust. These layers have varying concentrations, with the most dense (approximately 45 µm³/cm³) located over the smoke layer, roughly between 3700–4400 m a.s.l. Additionally, the coarse mode concentration shows a notable peak near the surface around 3000 m a.s.l., which may be suggesting the presence of heavier, dust-laden particles closer to the ground.



Concentration profiles for each **−**: 4500 ∃ 4500 mode with error (shaded) (a), and [m]북. 4000 Height 4000 Ångstrom exponent profile at wavelengths of 35003500 532 and 808 nm (b) obtained 3000 -3000 through GRASP for June 30, 2023, at 10:13:36 UTC. 2500 25000.51.01.5 $CV \left[\mu m^3/cm^3\right]$ α [U.A.]

Finally, looking at the total extinction profiles obtained from GRASP and the modified Klett method (referred to as the Klett method hereafter) in Figure 3a, GRASP generally produces lower values for both wavelengths (532 and 808 nm), though the Klett profiles remain within the error range of GRASP. These differences likely stem from the distinct calculation methods used in the two inversions.

For the backscatter profiles (Figure 3b), there is better agreement at 532 nm, with GRASP yielding lower values than the Klett method, though still within the GRASP error margins. However, at 808 nm, GRASP shows higher values than the Klett method, marking a more significant discrepancy between the two approaches. As mentioned earlier, these differences may stem from variations in methodologies, such as the calculation of the lidar ratio or reduced sensitivity due to the aerosol load.

Klett method (dashed) at wavelengths of 532 and 808 nm for June 30, 2023, at 10:13:36 UTC.

Conclusions

The integration of the CE376 CIMEL lidar with sun-sky-lunar photometer data within the GRASP inversion algorithm has demonstrated significant potential for retrieving detailed aerosol properties, including the size distribution, SSA, and volume concentration profiles. By incorporating RCS signals at 532 nm and 808 nm, the lidar provides essential vertical information that, when combined with AERONET AOD and sky radiance data, enables accurate characterization of both fine and coarse aerosol modes. This study successfully showcased the inversion strategy's ability to effectively capture aerosol properties during complex events, such as the June 30, 2023 occurrence, which involved a mixture of Saharan dust and Canadian wildfire smoke.

The aerosol properties obtained through GRASP were compared with AERONET and the modified Klett method, revealing strong overall agreement. However, some differences such as in backscattering profiles and SSA indicate areas that could benefit from further refinement. This study highlights the CE376 lidar's potential as a valuable tool for future research focused on enhancing vertical aerosol characterization using the GRASP algorithm.

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