



Aerosol variability in the Río de la Plata estuary: Multimodal analysis of four years of AERONET observations

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Abstract. We analyze four years (2020–2023) of AERONET observations from Montevideo_FING, an urban–estuarine site at the Río de la Plata, to characterize aerosol optical properties and to assess whether inversion-retrieved particle-volume size distributions (PVSDs) are better represented by a multimodal than by a standard bimodal form. The record has a median aerosol optical depth at 500 nm (AOD₅₀₀) of 0.067 and weak total-load seasonality, but the fine–coarse balance varies markedly: the fine mode fraction (FMF) increases from 0.42 in summer to 0.72 in winter, and wind-resolved analysis shows stronger fine mode contribution under northerly and westerly flow than under southerly and easterly flow.

In the almucantar inversion subset, a modified Gaussian Mixture Model (mGMM), adapted here for inversion-retrieved 22-bin PVSDs, classifies 68% of cases as multimodal, indicating that strict bimodality is not the dominant state under inversion-qualified conditions. In a spectral-consistency test based on Mie calculations and the Ångström exponent of AOD, the median signed relative difference decreases from 17% with the standard bimodal PVSD representation to 3.4% with the multimodal representation obtained with mGMM.

In a multisite comparison, Montevideo occupies an intermediate position between marine and urban–continental reference ranges, consistent with a mixed coastal–urban setting. TROPOMI NO₂ columns are also higher in the overland quadrants, providing independent contextual support for stronger lower-tropospheric anthropogenic influence under overland flow. Overall, the 2020–2023 record shows a clear seasonal and wind-dependent redistribution between fine and coarse aerosol contributions.

1 Introduction

Atmospheric aerosols significantly impact the Earth's radiative balance, contribute to cloud formation, and affect air quality. Despite decades of research, they remain the largest source of uncertainty in climate forcing assessments. Recent research estimates aerosol effective radiative forcing between -1.6 and -0.6 W m⁻² (68% confidence), with aerosol–cloud interactions dominating this uncertainty (Bellouin et al., 2020). Reducing this uncertainty requires sustained, high-quality observations across diverse environments to constrain aerosol type-dependent optical properties (Dubovik et al., 2002). The spectral quantification of aerosol optical properties through standardized networks is essential for evaluating satellite retrievals and atmospheric models at global scales.



The Aerosol Robotic Network (AERONET) program, operated by NASA, has provided standardized sun photometer observations of aerosol optical depth (AOD) and retrievals of microphysical properties at hundreds of sites worldwide since the 1990s (Holben et al., 1998, 2001; Giles et al., 2019). The Montevideo station (34.918° S, 56.167° W) samples an urban–estuarine environment at the Río de la Plata, where coastal circulations and synoptic flow modulate marine influence, local emissions, and episodic regional smoke (Meccia et al., 2013; Berri and Dezzutti, 2020; D’Angelo et al., 2023; Osorio et al., 2024). The Montevideo region usually exhibits low-AOD conditions, but these are intermittently perturbed by biomass-burning intrusions (Cúneo et al., 2022). This combination of marine influence, urban emissions, and generally low-AOD conditions makes Montevideo a useful site for characterizing aerosol optical and microphysical variability in a mixed coastal-urban environment.

A central challenge in such mixed environments is the description of aerosol particle-volume size distributions (PVSDs) inferred from sky-radiance/direct-sun AERONET inversions. AERONET also provides a simplified bimodal representation by fitting lognormal PVSDs to fine mode and coarse-mode partitions of the full 22-bin PVSD. In coastal–urban mixtures, the sum of these two lognormal components may underrepresent additional submicron structure as marine and continental influences coexist and evolve. To address this limitation, Taylor et al. (2014) proposed a Gaussian Mixture Model (GMM) approach.

In this work, we present a four-year characterization (2020–2023) of aerosol optical parameters in Montevideo using AERONET direct-sun and inversion products. We present a modified Gaussian Mixture Model (mGMM) that involves multimodal fits to the original 22-bin AERONET PVSD: our goals are to demonstrate that substantial fitting improvements can be obtained relative to the bimodal simplification and that those modal fits can be related to certain physical/environmental features. Both optical parameters and mGMM-derived modal PVSDs are analyzed as a function of season and wind direction; for the latter analysis, we apply a boundary layer height (BLH)-weighted approach that emphasizes lower-layer flow as a practical proxy for air-mass origin. Mie calculations are used to transform and compare mGMM modal PVSDs and the AERONET bimodal PVSD against measured 500 nm AERONET AODs and Ångström exponents. Independent TROPOMI NO₂ observations are employed as supporting context for lower-tropospheric pollution.

2 Data

2.1 AERONET

2.1.1 Network overview

AERONET is an international network of ground-based sun photometers/sky radiometers operated by NASA, providing standardized observations of aerosol optical depth and retrievals of optical and microphysical properties on regional to global scales (Holben et al., 1998, 2001). The network employs standardized instrumentation, calibration, and data processing protocols across all sites, enabling direct intercomparison of results. Instruments acquire direct-sun and sky-radiance data; the combination of spectral AODs and angular sky-radiance measurements enables the retrieval of particle size distributions and refractive indices (Dubovik et al., 2002). All data undergo multi-level quality assurance, with Level 2.0 products incorporat-



ing cloud screening and post-field calibration (Holben et al., 2001). This standardization places Montevideo observations in a global context.

2.1.2 Montevideo

The Montevideo AERONET station operates a CIMEL CE 318-TS sun photometer/sky radiometer on the rooftop of the Faculty of Engineering of the Universidad de la República (34.918° S, 56.167° W) at 71 m a.s.l., with a field of view toward the estuary. Its urban-estuarine location places the station under the influence of Río de la Plata estuary breezes, which modulate the coastal boundary layer. Northerly winds facilitate the transport of continental aerosols.



(a)



(b)

Figure 1. (a) Location of the AERONET photometer in Montevideo. Base map: Google Satellite imagery; map data © Google. (b) View of the monitoring station located at the roof of the Facultad de Ingeniería in Montevideo.

The wind regime of the Río de la Plata is characterized by frequent easterly winds that transport humid air from the river and the Atlantic Ocean towards the city, thus favoring the presence of marine aerosols in the coastal boundary layer. The immediate urban environment introduces anthropogenic sources such as the harbor of Montevideo and the oil refinery, which are located approximately 4 and 7.5 km northwest of the AERONET station, respectively. Besides local mixing, the station records regional transport: northerly and northwesterly winds carry dust and continental organic matter, and in austral spring, abrupt AOD increases associated with biomass burning plumes from inland Uruguay, Brazil, and Paraguay are frequent (Osorio et al., 2024).

2.1.3 AERONET Photometer

The CIMEL CE 318-TS photometer, the standard instrument of the network, acquires data in nine spectral bands (340, 380, 440, 500, 670, 870, 935, 1020, and 1640 nm) with nominal filter bandwidths of 2–4 nm in the UV channels and ~10 nm in the visible and near-infrared channels (Giles et al., 2019). The 935 nm channel is used to retrieve the water vapor column, while



the 1640 nm short-wave infrared channel yields important information on intermediate-sized coarse-mode particles (O'Neill et al., 2008) and, together with the 1020 nm filter, is used to verify the data consistency of the instrument (Giles et al., 2019).

75 The instrument performs autonomous measurement sequences of direct and diffuse solar radiation in almucantar and hybrid geometry (combining azimuthal and zenithal scanning). This provides spectral AOD (Version 3 Level 2.0) and derived parameters such as the Ångström exponent, fine mode fraction, size distribution, and complex refractive index (Holben et al., 1998; Dubovik et al., 2000, 2002; Giles et al., 2019). The typical accuracy of Level 2.0 AOD is 0.01–0.02 in the visible, which is a direct consequence of periodic calibrations and systematic corrections.

80 2.2 Satellite

As an independent indicator of lower-tropospheric anthropogenic influence, tropospheric nitrogen dioxide (NO₂) columns derived from the TROPOMI instrument onboard Sentinel-5 Precursor were used (Veefkind et al., 2012; Fioletov et al., 2022). The offline NO₂ product (OFFL) employs algorithms that combine ultraviolet-visible spectrometry with TM5-MP model fields to separate tropospheric and stratospheric fractions and improve surface reflectance correction (van Geffen et al., 2022). The
85 analyses are based on the COPERNICUS/S5P/OFFL/L3_NO2 collection from Google Earth Engine, with reprojection to 0.01°, quality filtering (qa_value ≥ 0.75), and daily and seasonal aggregation over the site surroundings (Google Earth Engine Team, 2025).

2.3 Meteorological Data

2.3.1 ERA5 Reanalysis from ECMWF

90 For wind analysis we used hourly vertical profiles from the ERA5 reanalysis (C3S/ECMWF) at 0.25° × 0.25° horizontal resolution (Hersbach et al., 2020; Copernicus Climate Change Service, 2025). We extracted the horizontal wind components (u , v) and geopotential (z) at multiple isobaric levels near the surface. The geopotential is converted to height above mean sea level (MSL) as $Z = z/g_0$ (m a.s.l.), where $g_0 = 9.81 \text{ m s}^{-2}$. The analysis uses the consolidated ERA5 record covering the 2020–2023 period.

95 2.3.2 CAMS Composition Reanalysis

We used the CAMS global composition reanalysis (EAC4) to obtain boundary layer height (BLH). This reanalysis is produced with the ECMWF Integrated Forecasting System (IFS) model (Inness et al., 2019; Copernicus Atmosphere Monitoring Service, 2025). EAC4 provides 3-hourly fields at an approximate horizontal resolution of ~0.7° (~80 km). The 3-hourly BLH values were spatially interpolated to the site location using inverse-distance weighting from the four nearest grid cells.

100 To match ERA5 hourly profiles, the BLH time series was then resampled using nearest-neighbor assignment in time. The resulting BLH (m above ground level) defines the mixing layer depth used for vertical wind averaging.



3 Methods

3.1 AERONET data products

The aerosol optical depth at 500 nm (τ_{500} , AOD_{500}) was used as the primary reference for baseline AOD conditions and intercomparison (Holben et al., 1998). We calculated the Ångström exponent (Ångström, 1929) using the standard AERONET algorithm (AOD values measured at 440 and 870 nm). These results are compared in Section 4.3.1 with the analogous (440 to 870 nm) linear slope in log-log space applied to Mie-derived spectra to quantify the consistency of the spectral slope.

3.1.1 Spectral Deconvolution Algorithm (SDA) Products

The Spectral Deconvolution Algorithm (SDA) provides a fine/coarse partition of spectral AOD (O'Neill et al., 2003). We use the SDA-derived (500 nm) fine and coarse AOD, τ_f and τ_c , and their contribution to the total AOD:

$$\tau_a = \tau_f + \tau_c. \quad (1)$$

The fine mode fraction (FMF) is given by:

$$FMF = \frac{\tau_f}{\tau_a}. \quad (2)$$

These quantities, derived from the SDA (FMF, τ_f , and τ_c), will be used in Section 4.1 to describe the balance of fine and coarse aerosols, as well as their seasonal and wind-direction dependence. Since the FMF is an optical partition of the SDA (based on AOD) and the $C_{v,f}/C_v$ ratio is a volume fraction derived from sky-radiance inversion, they are related proxies of fine-coarse balance and are not interchangeable quantities.

3.1.2 Periodicity analysis (Lomb–Scargle)

To assess periodicities in the irregularly sampled AERONET time series, we computed normalized Lomb–Scargle periodograms (Lomb, 1976; Scargle, 1982) using MATLAB's *plomb* function (Signal Processing Toolbox). This analysis was applied to AOD_{500} , FMF, and the inversion-derived $C_{v,f}/C_v$ series. Prior to analysis, non-finite values and missing timestamps were removed. Periodograms were evaluated on a grid of trial periods between 1 and 2000 days. The dominant period T_{dom} was defined as the period of maximum normalized power. The annual-band power fraction, B_{ann} , was defined as

$$B_{ann} = \frac{\int_{f \in \mathcal{A}_{ann}} P_N(f) df}{\int_{f \in \mathcal{A}_{tot}} P_N(f) df}, \quad (3)$$

where $P_N(f)$ is the normalized Lomb–Scargle power, $\mathcal{A}_{ann} = [0.9f_0, 1.1f_0]$ with $f_0 = 1/365 \text{ day}^{-1}$, and $\mathcal{A}_{tot} = [1/2000, 1] \text{ day}^{-1}$ (the full analyzed range). Thus, B_{ann} is a dimensionless spectral-power fraction that measures how concentrated the normalized periodogram power is near the annual frequency within the analyzed 1–2000 day period range. The same band definition was used for all reported series.



3.1.3 Size distribution retrieval

130 Almuqantar sky-radiance inversions provide the column-integrated particle-volume size distribution (PVSD), $dV/d\ln r$ (Dubovik et al., 2002). The standard bimodal representation derived from the AERONET inversion product partitions the retrieved PVSD into a fine (f) and a coarse (c) mode using a separation radius, r_s , defined as the local minimum within $r \in [0.439, 0.992] \mu\text{m}$ (O'Neill et al., 2023). Once r_s is found, the distribution is divided into two intervals ($r < r_s$ and $r > r_s$). The microphysical retrieval parameters characterizing these modes (particle-volume concentrations $C_{v,f}, C_{v,c}$; mode-radius parameters $r_{v,f}, r_{v,c}$; and standard deviations σ_f, σ_c) are provided as part of the AERONET Version 3 inversion product (AERONET Team, 2020; Sinyuk et al., 2020). For the modal analyses in Sect. 4.2, we use the AERONET Version 3 Level 2.0 almuqantar inversion record, selected under the standard inversion geometry and radiance-quality constraints. We use this derived bimodal representation as a baseline for comparison with the mGMM approach presented in Sect. 3.2.

3.1.4 Complex refractive index

140 AERONET retrieval parameters include the complex refractive index $m(\lambda) = n(\lambda) + i\kappa(\lambda)$ at four wavelengths (440, 675, 870, and 1020 nm) (Dubovik et al., 2002). Due to the higher uncertainty of these retrievals under the frequent low AOD conditions in Montevideo, Level 1.5 refractive indices are used as inputs for internal (site-specific) spectral-consistency assessment.

3.2 Modified Gaussian Mixture Model (mGMM) implementation

To describe the modal structure of the PVSD, we implemented a modified Gaussian Mixture Model (mGMM). The workflow is applied independently to each AERONET inversion PVSD, so the number of modes and their parameters are selected case by case rather than imposed as a fixed structure. Relative to the Gaussian Mixture Model (GMM) framework proposed by Taylor et al. (2014), the modified workflow used here updates the fitting and mode-selection procedure to improve statistical consistency and reproducibility, while preserving the native 22-bin radius grid of the AERONET inversion product. The reader is referred to Taylor et al. (2014) for the base formalism and the discussion of specialized GMM parameters.

150 The volume size distribution $y = dV/d\ln r$ is modeled as a sum of Gaussian modes:

$$\hat{y}(x) = \sum_{i=1}^n a_i \exp \left[- \left(\frac{x - b_i}{c_i} \right)^2 \right], \quad (4)$$

where each mode i has an amplitude $a_i > 0$, centered at b_i , and width $c_i > 0$.

Specifically, we (i) use a least-squares (L2-norm) objective to fit each candidate curve, minimizing the sum of squared residuals (SSR), and select the number of modes using the resulting adjusted coefficient of determination and a Fisher-z significance criterion; (ii) fit directly on the native 22-bin grid from the AERONET inversion product, with interpolation used only for visualization; and (iii) apply a dedicated fine mode acceptance criterion so localized submicron structure is not missed by a purely global statistic (explained in detail below).

For the steps involving the fine mode described below, note that, for simplicity's sake, we use the fixed historical cutoff radius $r_{\text{cut}} = 0.6 \mu\text{m}$, rather than the minimum currently employed in the AERONET Version 3 inversion to define r_s for each



size distribution (Sect. 3.1.3). This fixed cutoff is used later to identify candidate fine modes, evaluate localized improvements in the fine radius range, and compute the mGMM fine mode volume fraction.

Fitting procedure. For each candidate number of modes n , the parameters (a_i, b_i, c_i) are fitted to the native 22-bin AERONET PVSD using MATLAB's *lsqcurvefit*. The fit minimizes the sum of squared residuals (SSR) and is formulated as a bounded non-linear least-squares problem, with modal centers kept within the measured $\ln r$ range and amplitudes and widths constrained to positive values ($a_i > 0$ and $c_i > 0$):

$$\text{SSR} = \sum_{k=1}^N [y_k - \hat{y}(x_k)]^2. \quad (5)$$

Goodness of fit is evaluated using the adjusted coefficient of determination:

$$R_{\text{adj}}^2 = 1 - (1 - R^2) \frac{N-1}{N-p}, \quad \text{where } R^2 = 1 - \frac{\text{SSR}}{\text{SST}}, \quad \text{SST} = \sum_{k=1}^N (y_k - \bar{y})^2. \quad (6)$$

Here N is the number of radius bins used in each fit (typically 22), and $p = 3n$ is the number of fitted parameters (amplitude, center, and width for each of the n modes).

Initial conditions. Initial parameter values are assigned directly from the observed PVSD and the residual structure of the previous fit. For $n = 1$ (Eq. 4), the amplitude (a_1) is initialized to $\max(y)$, the center (b_1) to the location of the distribution maximum in $\ln r$ space, and the width (c_1) to a default value $c_0 = 0.5$. For $n > 1$, the optimized coefficients from the $(n-1)$ -mode fit are retained and the new mode is initialized at the location of the maximum positive residual after setting negative residuals to zero. All candidate widths are subject to a global upper bound; for candidates initialized in the fine-radius band ($r_{v,i} < r_{\text{cut}}$), a tighter upper-width bound is applied to discourage an unrealistically broad submicron component extending into the coarse branch.

Mode selection. For each PVSD, models are evaluated sequentially from $n = 1$ up to a maximum of 6, stopping when $R_{\text{adj}}^2 \geq 0.99$ or when $n = 6$ is reached. This upper limit allows additional structure beyond the standard bimodal representation while limiting overfitting. An additional mode is retained if the Fisher-z statistic (Fisher, 1921)

$$F(\rho) = \frac{1}{2} \ln \left(\frac{1+\rho}{1-\rho} \right), \quad \rho = \sqrt{R_{\text{adj}}^2}, \quad (7)$$

shows a statistically significant improvement from the n -mode to the $(n+1)$ -mode fit at significance level $\alpha = 0.05$ (5% test threshold). To avoid missing small submicron components that contribute only slightly to global fit statistics, candidate fine-mode components ($r_{v,i} < r_{\text{cut}}$) are also retained when they produce a localized improvement in the fine-radius band ($r < r_{\text{cut}}$), $\Delta R_{\text{fine}}^2 \geq 0.005$. Here ΔR_{fine}^2 is computed as the increase in the usual (non-adjusted) coefficient of determination (R^2) evaluated only over the fine-band bins, whereas the global stopping criterion uses adjusted R_{adj}^2 . After the sequential selection, we apply an additional fine-band refinement: if the fine-band fit remains weak ($R_{\text{fine}}^2 < 0.90$), we attempt to add a single extra fine mode, constrained to $r < r_{\text{cut}}$, and retain it only if it improves R_{fine}^2 without degrading the global R_{adj}^2 .

Physical parameters. Once the optimal number of modes is determined, the Gaussian coefficients (a_i, b_i, c_i) are converted to physical lognormal parameters (Taylor et al., 2014). The center b_i is the location of the mode in logarithmic radius space.



We report its radius as the volume-weighted particle-radius mean of the fitted mode, $r_{v,i} = \exp(b_i)$. The width c_i determines the spread of the mode; the corresponding standard deviation in logarithmic-radius space is $\sigma_i = c_i/\sqrt{2}$, and the geometric standard deviation is $\sigma_{g,i} = \exp(\sigma_i)$. The particle-volume concentration of each mode, $C_{v,i}$, is obtained by integrating the Gaussian function over $\ln r$ space:

$$C_{v,i} = \int_{-\infty}^{\infty} a_i \exp \left[- \left(\frac{x - b_i}{c_i} \right)^2 \right] dx = a_i c_i \sqrt{\pi}. \quad (8)$$

Overall:

$$r_{v,i} = \exp(b_i), \quad \sigma_i = \frac{c_i}{\sqrt{2}}, \quad \sigma_{g,i} = \exp(\sigma_i), \quad C_{v,i} = a_i c_i \sqrt{\pi}. \quad (9)$$

These parameters are directly comparable with the bimodal AERONET outputs described in Section 3.1.3. Modes with negligible volume contribution ($C_{v,i}/C_v < 0.05$) are filtered out from the final results in an attempt to minimize the impact of numerical artifacts.

From the retained modal volumes, we define the mGMM fine-mode volume fraction as

$$\frac{C_{v,f}}{C_v} = \frac{\sum_{i:r_{v,i} < r_{\text{cut}}} C_{v,i}}{C_v}, \quad (10)$$

where $C_v = \sum_i C_{v,i}$ and $r_{\text{cut}} = 0.6 \mu\text{m}$. Although this fine-to-total volume ratio derived from the inversion is related to the FMF derived from SDA, the two quantities are neither equivalent nor interchangeable.

Volume conservation. To ensure that the fitted distribution preserves the total aerosol particle-volume concentration, modal volumes are computed from the analytical Gaussian integrals rather than from the interpolated plotting curve. The fitted amplitudes are then rescaled by $(C_{v,\text{data}}/C_{v,\text{fit}})$, where $C_v = \int y(x) dx$.

3.3 Spectral extrapolation for Mie calculations

To compute $\text{AOD}_{\text{Mie}}(\lambda)$ from the retrieved modal distributions, we apply Mie scattering theory using AERONET-derived complex refractive indices (Section 3.1.4); the total $\text{AOD}_{\text{Mie}}(\lambda)$ is obtained as the sum of the modal $\text{AOD}_{\text{Mie}}(\lambda)$ contributions. This provides an internal site-specific consistency assessment by comparing the Mie-derived $\text{AE}_{440-870}$ with the corresponding AE from direct-sun observations. The refractive-index treatment used in these calculations is described below.

Real part.

The dispersion of n is represented with a Cauchy equation (Barker et al., 2023; Shepherd et al., 2018):

$$n(\lambda) = A + \frac{B}{\lambda^2} + \frac{C}{\lambda^4}, \quad (11)$$

where λ is wavelength in micrometers. With only four AERONET wavelengths, fitting the three Cauchy coefficients (A , B , C) leaves one residual degree of freedom, making the estimates more sensitive to measurement noise than a constrained two-parameter fit. To improve robustness, we set $C = 0$ and fit A and B by least squares over 440–1020 nm, following Shepherd et al. (2018).



220 **Imaginary part.**

For the imaginary part, we assume that $\kappa(\lambda)$ is constant over the wavelengths used in the Mie calculations. Spectral consistency between the Mie-derived and measured AOD is evaluated in Section 4.3.1 through $AE_{440-870}$.

3.4 Wind data processing up to the BLH

We would argue that vertical wind profile data should be weighted by characteristic aerosol vertical profiles so that statements
225 about wind direction are more optically relevant in terms of the impact of aerosols. Aerosols tend to be more abundant near the ground while decreasing with height in the first tens to hundreds of meters, both in urban and coastal environments (Quang et al., 2012; Zauli Sajani et al., 2018; Blanchard et al., 1984).

Using ERA5 wind profiles and CAMS boundary layer height (BLH), we compute BLH-weighted horizontal wind components that emphasize the lower mixing layer while preserving information from the full layer depth.

230 From these weighted components, we derive hourly wind direction and speed and define an equivalent wind height, Z_{eq} , to characterize the representative altitude of the weighted flow. This provides a compact and physically interpretable wind proxy for the aerosol analyses that follow. Full implementation details (vertical reference, level selection, weighting kernel, temporal smoothing, and Z_{eq} statistics) are provided in Appendix A.

Wind direction classification. To investigate the origin and observed aerosol distribution in Montevideo, we assume that
235 wind direction is a crucial factor. To this end, we conducted several tests. We found that dividing the wind rose into four 90° quadrants according to the following criteria allowed us to identify characteristic size patterns: The northern quadrant (NQ) corresponds to the wind direction from the 315° to 45° window, the eastern quadrant (EQ) from 45° to 135°, the southern quadrant (SQ) is defined from 135° to 225°, and the western (WQ) is defined from 225° to 315°. The geographical context of the selected quadrants is shown in Figure 2. This cardinal orientation is motivated by Montevideo's coastal location.
240 Air arriving from SQ and EQ sectors traverses the Río de la Plata estuary and the adjacent Atlantic Ocean before reaching the site, whereas NQ and WQ winds more often reflect continental flow from inland regions. Under this geometry-based framework, SQ and EQ are more likely to reflect marine/estuarine influence (O'Dowd and De Leeuw, 2007; Smirnov et al., 2012; Blanchard et al., 1984), whereas NQ and WQ are more likely to reflect continental influence. These associations provide a practical transport proxy but do not imply unique source attribution for individual events. The classification is supported by
245 the systematic differences in FMF and $C_{v,f}/C_v$ between the SQ+EQ and NQ+WQ groupings (Section 4.1.2). Accordingly, we use NQ+WQ as a continental grouping and SQ+EQ as a marine (marine/estuarine-influenced) grouping in subsequent sections. For AOD derived from direct observations of the sun and derived products (FMF, τ_f , τ_c), the larger sample size permits finer binning (e.g., 16 sectors of 22.5°) in some figures. By contrast, inversion products based on almucantar inversions (e.g., $C_{v,f}/C_v$) have a smaller sample size; with a 16-sector seasonal stratification, the median count is ~ 30 cases per sector-
250 season, which can lead to noisy directional medians. Therefore, for inversion-based analyses we use wind-direction quadrants and the two combined groupings (NQ+WQ and SQ+EQ).



Figure 2. Geographic context of the Montevideo wind-direction framework, including the location of Buenos Aires. Quadrants follow fixed 90° sectors: NQ (315–45°), EQ (45–135°), SQ (135–225°), and WQ (225–315°). Base map: ArcGIS World Imagery, hosted by Esri (Sources: Esri, DigitalGlobe, GeoEye, i-cubed, USDA FSA, USGS, AEX, Getmapping, Aerogrid, IGN, IGP, swisstopo, and the GIS User Community | Powered by Esri).

4 Results and discussion

The results cover the measurement period 2020–2023 at the Montevideo AERONET site (Version 3 Level 2.0 data from March 2020 through November 14, 2023). Montevideo’s location at the urban–estuarine interface creates an aerosol environment
 255 shaped by competing continental and marine influences. We organize our analysis in four stages: first, we characterize the aerosol optical parameters using standard AERONET products; second, we apply the mGMM methodology to characterize the modal structure and compare with other AERONET sites; third, we compare the mGMM performance with the standard bimodal model derived from the AERONET inversion parameters and with the original implementation of Taylor et al. (2014); finally, we provide independent support using satellite NO₂ observations.

260 4.1 AOD, FMF, τ_f , τ_c from direct sun observations

We examine the temporal and directional dependence of direct-sun optical parameters to establish a seasonal baseline.

4.1.1 Temporal and seasonal variability

Monthly AOD medians at 500 nm for the four-year period are shown in Figure 3a, with seasonal distributions summarized in Figure 3b. The lowest seasonal median occurs in autumn ($\tau_{500} = 0.059$) and the highest in summer ($\tau_{500} = 0.070$), giving
 265 a seasonal-median span of $\Delta = 0.012$ (17.4% relative to the annual median, $\tau_{500} = 0.067$). This seasonal shift is modest compared with episodic high-AOD events in the right tail of the record.

The 99th percentile reaches $\tau_{500} = 0.283$, consistent with occasional smoke intrusions from biomass-burning events. Biomass burning in the Amazon and central Brazil produces regional-scale aerosol enhancements in South America (Moreira et al., 2017), and specific intrusions reaching Montevideo have been documented (Osorio et al., 2024). Montevideo’s median AOD

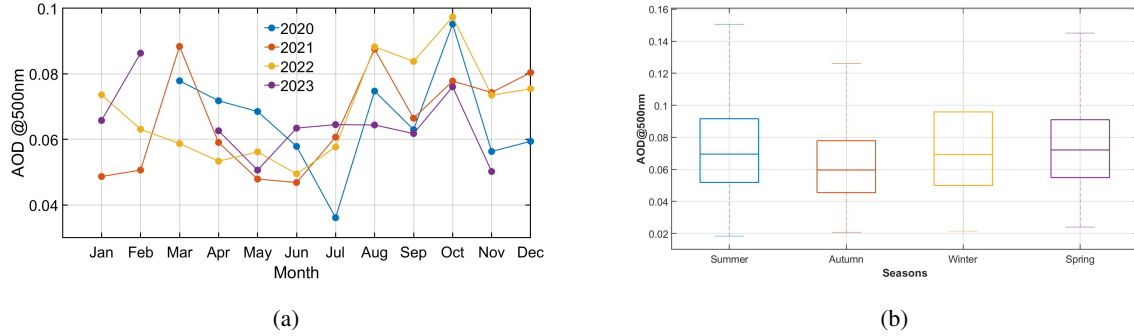


Figure 3. (a) Monthly AOD medians at 500 nm, differentiated by year to show seasonal variability. Temporary instrumentation and maintenance problems explain the missing months. (b) Seasonal boxplots of monthly AOD medians for the four-year analyzed period.

270 ($\tau_{500} = 0.067$) is consistent with the relatively low-AOD conditions reported for the Buenos Aires AERONET site, although the published Buenos Aires statistic is a long-term mean and is therefore not directly equivalent to the Montevideo median (Cúneo et al., 2022). In contrast, FMF shows a larger seasonal contrast in monthly medians (Figure 4a). The highest seasonal median occurs in winter ($FMF = 0.72$) and the lowest in summer ($FMF = 0.42$), giving a seasonal-median span of $\Delta = 0.30$ (58.0% relative to the annual median).

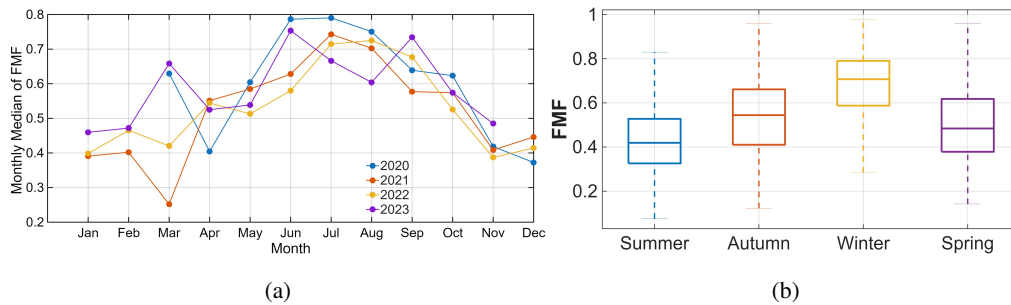


Figure 4. Monthly FMF medians for the four-year analyzed period. (a) Monthly medians by year. (b) Seasonal boxplots of monthly FMF medians at 500 nm.

275 To assess annual periodicity, we use the normalized Lomb-Scargle periodograms in Figure 5. For FMF (Figure 5a), the dominant period is $T_{\text{dom}} = 367.3$ days and the annual peak is narrower and more distinct. For AOD (Figure 5b), the dominant period is $T_{\text{dom}} = 439.0$ days and spectral power is distributed across annual and subannual frequencies. Annual organization is quantified by the annual-band power fraction, B_{ann} (Eq. 3): $B_{\text{ann}} = 0.00393$ (0.393%) for AOD and $B_{\text{ann}} = 0.07439$ (7.439%) for FMF. Thus, the annual-band fraction in FMF is 18.94 times that in AOD, indicating a more concentrated annual component

280 in FMF than in AOD.

Because FMF is an intensive, bounded parameter, we also examined the extensive-parameter variations of τ_f and τ_c (Figure 6) to determine how the seasonal fine-coarse redistribution appears in optical depth. τ_f increases in winter ($\tau_{f,\text{winter}} =$

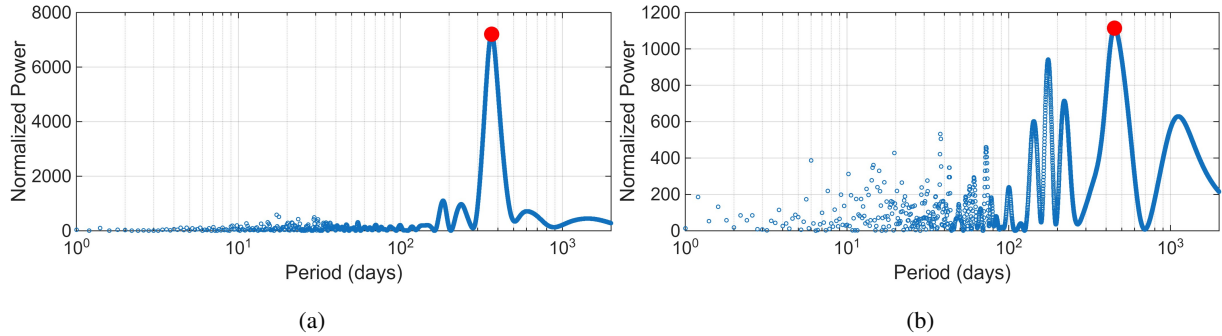


Figure 5. Normalized Lomb–Scargle periodograms for FMF (a) and AOD (b), computed for the full record. The red marker indicates the dominant peak (T_{dom}).

0.043) and decreases in summer ($\tau_{f,\text{summer}} = 0.026$), giving a seasonal-median span of $\Delta = 0.017$ (54.1% relative to its annual median). Conversely, τ_c reaches its seasonal minimum in winter ($\tau_c = 0.022$) and its seasonal maximum in summer ($\tau_c = 0.038$), yielding a comparable seasonal-median span of $\Delta = 0.016$ (51.0% relative to its annual median).

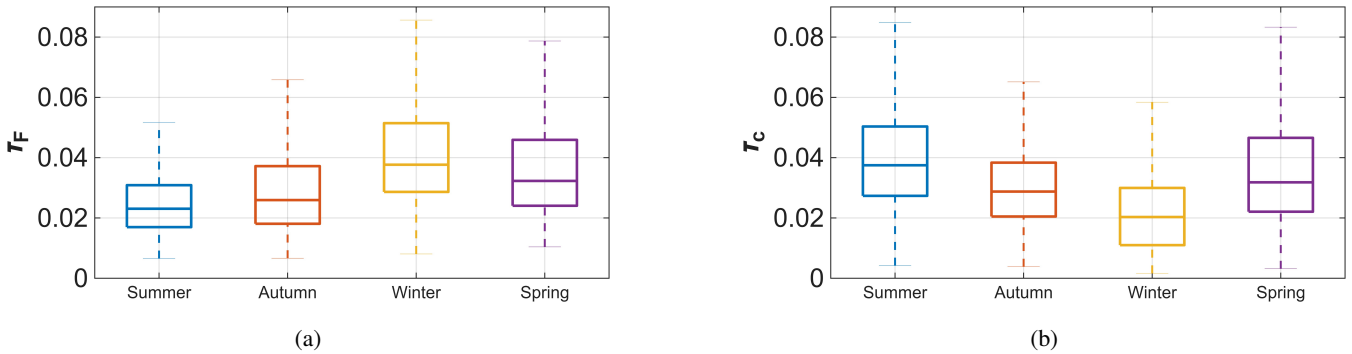


Figure 6. Seasonal boxplots of the fine mode AOD, τ_f (a), and the coarse-mode AOD, τ_c (b), at 500 nm.

Seasonal medians therefore indicate that the annual variability is expressed more strongly in the intensive parameter FMF than in the extensive parameter AOD. This contrast is consistent with the Lomb–Scargle result (B_{ann} higher for FMF) and with the opposite seasonal behavior of τ_f and τ_c , which have comparable relative amplitudes but opposite seasonal phase. Together, these results indicate that the annual cycle is primarily characterized by shifts in the AOD fine/coarse balance (captured by FMF, Figure 4b) rather than by major swings in total AOD (Figure 3b). The next section assesses whether this redistribution also depends on wind direction.



4.1.2 Wind-direction dependence

Using the BLH-based wind treatment described in Section 3.4 (see also Fig. 2), we examine the wind-direction dependence of the direct-sun SDA-derived optical parameters (τ_f , τ_c , and FMF). We first present median values for sixteen contiguous 22.5° sectors spanning 360°. The large-volume archive of direct-sun AOD measurements supports this sector-resolved view. We then use the four-quadrant grouping to summarize the main directional contrast in FMF.

τ_f is highest for northerly and westerly directions (NQ and WQ; panel (a) of Figure 7). This directional pattern is consistent with continental influence and provides transport-regime context for the following analysis. Beyond the regional biomass-burning intrusions documented for Montevideo (Osorio et al., 2024), NQ/WQ flow may also reflect broader continental and urban influence, including possible transport from the Buenos Aires metropolitan area (Instituto Nacional de Estadística y Censos, 2024).

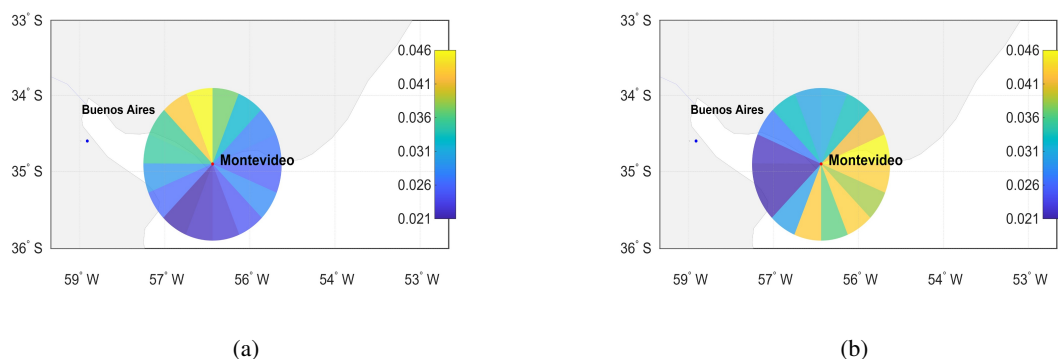


Figure 7. Fine-mode AOD, τ_f (a), and coarse-mode AOD, τ_c (b), by wind direction, grouped into sixteen 22.5° sectors (all direct-sun observations).

Turning to the coarse mode, panel (b) of Figure 7 shows higher τ_c in easterly sectors. This behavior is consistent with greater marine/estuarine influence in those sectors (Smirnov et al., 2002; O’Dowd and De Leeuw, 2007), although coarse-mode aerosol can include mixed contributions. To examine the fine–coarse balance, we analyze FMF, calculated according to the description given in Section 3.1.1.

The sector-resolved FMF map (Figure 8) shows higher FMF in northerly and westerly sectors and lower FMF in southerly and easterly sectors. This ordering is summarized with the four directional quadrants defined in Section 3.4. In Figure 9a, the NQ and WQ cumulative distributions lie close together and are shifted toward higher FMF than SQ and EQ. In Figure 9b, the quadrant summaries preserve the same ordering, with NQ and WQ generally above SQ and EQ. The quadrant groups are used here as transport-regime proxies rather than as unique source attributions.

Accordingly, we retain NQ+WQ and SQ+EQ as the working wind-regime groups for the remainder of the analysis.

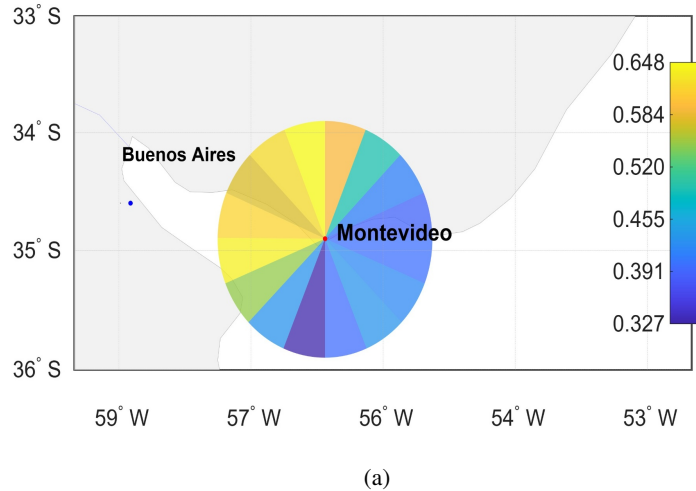


Figure 8. FMF by wind direction, grouped into sixteen 22.5° sectors (all direct-sun observations).

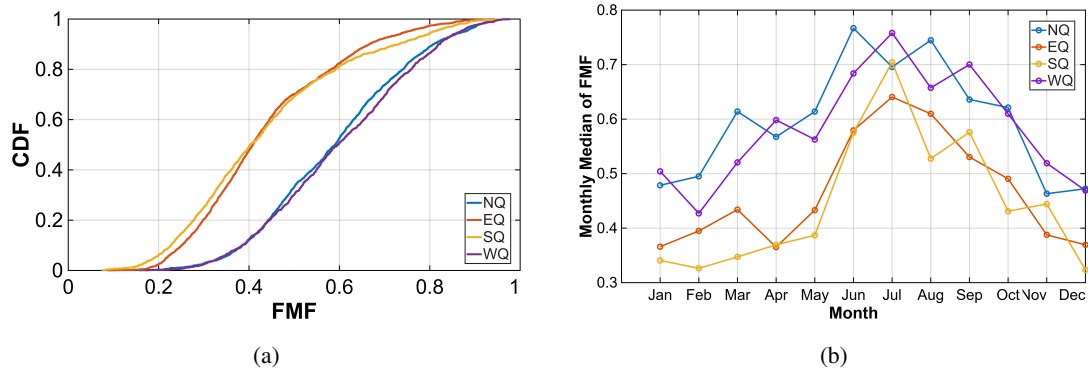


Figure 9. FMF by wind-direction quadrant. Panel (a) shows cumulative distributions; panel (b) shows monthly medians by quadrant. Quadrants correspond to North (NQ), East (EQ), South (SQ), and West (WQ).

4.2 mGMM results and multisite comparison

To characterize the modal distribution, we apply the mGMM approach to the column-integrated particle-volume size distributions (PVSDs) retrieved from AERONET Version 3 Level 2.0 almucantar inversions. These retrievals require the standard almucantar solar geometry ($50^\circ \leq \theta_0 \leq 80^\circ$) and sufficient sky-radiance information for stable inversions (Dubovik et al., 2002). The Montevideo inversion statistics reported below are based on this record ($N = 2322$), while the multisite comparisons use analogous Level 2.0 inversion records for each reference site.

Here we use the mGMM fine-mode volume fraction, $C_{v,f}/C_v$ (Eq. 10), as an inversion-based indicator of fine-coarse volume balance. FMF is an optical ratio derived from direct-sun products, whereas $C_{v,f}/C_v$ is derived from mGMM modes



320 fitted to the inversion PVSDs. The two quantities describe related but distinct aspects of the fine–coarse partition. We use $C_{v,f}/C_v$ as an approximate consistency check on the seasonal and wind-regime contrasts identified in Section 4.1.2.

The section is organized as follows. We first quantify the prevalence of multimodal configurations in the Montevideo inversion record (Section 4.2.1). We then examine whether the inversion-derived fine–coarse volume balance shows seasonal and wind-regime contrasts analogous to those identified in the direct-sun optical parameters (Sections 4.1.1 and 4.1.2) and in
325 the inversion subset (Section 4.2.2). Next, we place Montevideo in context by comparing its fine–coarse volume balance and retrieved modal radii, expressed as mGMM $r_{v,i}$ values, with those of maritime and urban/continental reference sites using the same mGMM analysis (Section 4.2.3). Model comparisons and spectral consistency analyses are presented in Section 4.3, while the satellite NO₂ sensitivity checks are presented in Section 4.2.3 and the results are summarized in Section 4.4.

4.2.1 Modal structure overview

330 In the first stage, we examined the prevalence of different modal configurations by applying the mGMM algorithm to the 4-year AERONET modal retrievals. Figure 10 presents the frequency histogram of mode combinations for all valid observations.

The distribution reveals that 2.4% of valid observations are unimodal ($K=1$), 29.5% present a strictly bimodal structure ($K=2$), and 68.0% are best described with more than two modes ($K \geq 3$) while 56.5% of all cases are specifically classified as three-mode ($K=3$). These choices are made by the mGMM process for each particle-size distribution that was examined;
335 therefore, the number of choices equals the 2322 particle-size distributions that were subject to mGMM characterization. These results indicate that approximately 68% of the observations depart from a strict bimodal assumption.

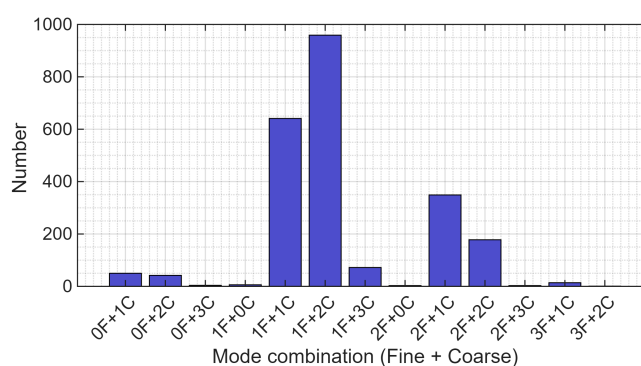


Figure 10. Frequency histogram of mode combinations retrieved by the mGMM algorithm for the 4-year AERONET inversion record. Each bar represents a different modal structure: 1F+1C (one fine + one coarse mode), 2F+1C (two fine + one coarse mode), 1F+2C (one fine + two coarse modes), etc.

To further visualize the dominant aerosol populations, Figure 11 presents a histogram of all mGMM-fitted mode radii ($r_{v,i}$). Peak ranges were defined as the two adjacent log-space bins with the highest counts around each modal maximum.

The distribution of mGMM-fitted mode radii derived from the PVSD exhibits a relative minimum near $r_{v,i} \approx 0.6 \mu\text{m}$,
340 indicating low recurrence of modal centers at the fine–coarse boundary. This pattern is consistent with the adopted mGMM

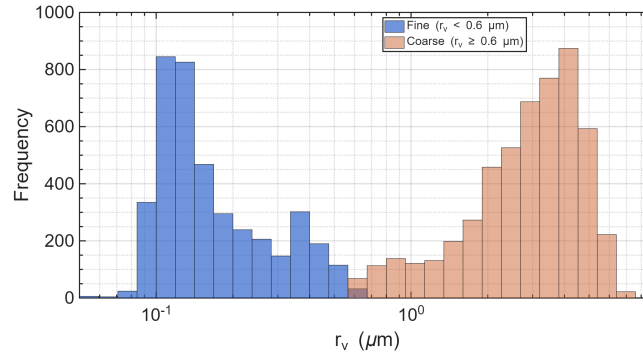


Figure 11. Histogram of all mGMM-fitted mode radii ($r_{v,i}$). Two primary peaks are visible, with a fine-mode maximum in the 0.10–0.14 μm range and a coarse-mode maximum in the 3.2–4.5 μm range.

cutoff radius r_{cut} and supports using $C_{v,f}/C_v$ as the fine mode volumetric fraction in the subsequent seasonal and directional analyses. Once the modal structure of the aerosol size distribution has been characterized, we now examine whether this translates into consistent wind-direction-dependent shifts in the inversion-derived fine/coarse volume fraction.

4.2.2 Seasonality and consistency with wind-direction

345 As shown in Section 4.1.2, the direct-sun products FMF, τ_f , and τ_c show a directional separation in Montevideo: τ_f and FMF are higher under NQ and WQ winds, whereas FMF is lower under SQ and EQ winds and τ_c is higher in easterly sectors. To test directional contrasts associated with modal retrievals, we evaluate whether this separation has an analogous representation in the mGMM retrievals, using the mGMM-derived fine-mode volume fraction, $C_{v,f}/C_v$. FMF is an optical ratio derived from direct-sun observations, whereas $C_{v,f}/C_v$ is a volume ratio derived from inversion modes; the two quantities are related
350 proxies for the fine–coarse balance, but they are not interchangeable (O’Neill et al., 2023).

To quantify the periodicity, we calculated a normalized Lomb–Scargle periodogram for the inversion-derived $C_{v,f}/C_v$ sequence (period range: 1–2000 days). The dominant peak occurs at $T_{\text{dom}} \approx 379$ days, while the power at 365 days remains close to this maximum (difference $\sim 5\%$; Figure 12). The annual-band power fraction is $B_{\text{ann}} = 2.220\%$ (Eq. 3), higher than the AOD and lower than the FMF in the direct-sun analysis of Section 4.1.1. Thus, $C_{v,f}/C_v$ falls between AOD and FMF in
355 annual-band power fraction, while retaining a distinct annual-scale peak. The inversion record is smaller and more irregular than the direct-sun record (inversions: $N = 2322$ over 554 days; direct-sun: $N = 53045$ over 900 days). The lower B_{ann} value relative to FMF may therefore partly reflect this sampling difference, so these comparisons of periodicity metrics should be interpreted with caution.

Seasonal statistics reinforce this annual-scale behavior (Figure 13): the lowest median occurs in summer ($C_{v,f}/C_v = 0.144$),
360 the highest in winter (0.346), and autumn/spring remain intermediate (0.224 and 0.217). This seasonal ordering is consistent with a shift from more coarse-mode dominated conditions in summer to more fine-mode dominated conditions in winter, in line with the FMF annual cycle described in Section 4.1.

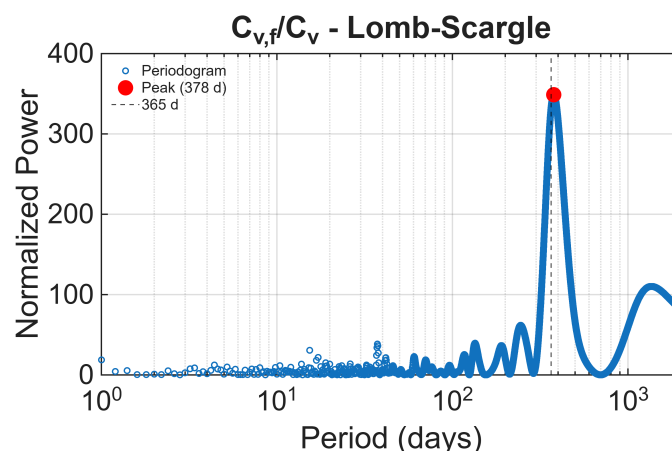


Figure 12. Normalized Lomb–Scargle periodogram for the mGMM-derived fine mode volume fraction ($C_{v,f}/C_v$), computed from the raw inversion record. The red marker denotes the dominant peak ($T_{\text{dom}} \approx 379$ days), and the dashed vertical line marks the 365-day annual period. The power at 365 days remains close to the maximum, consistent with a broad annual-scale peak.

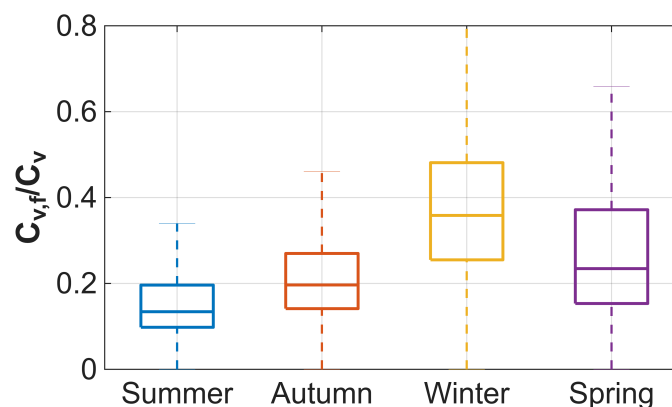
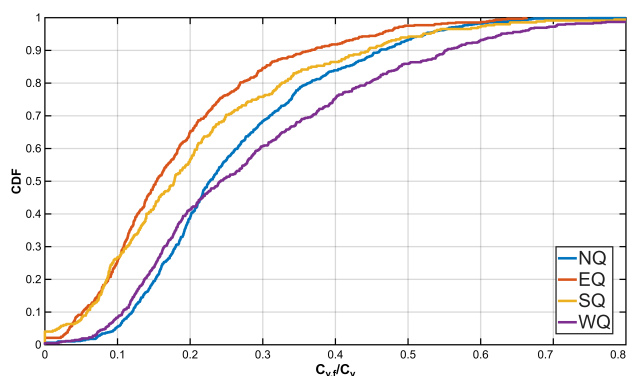
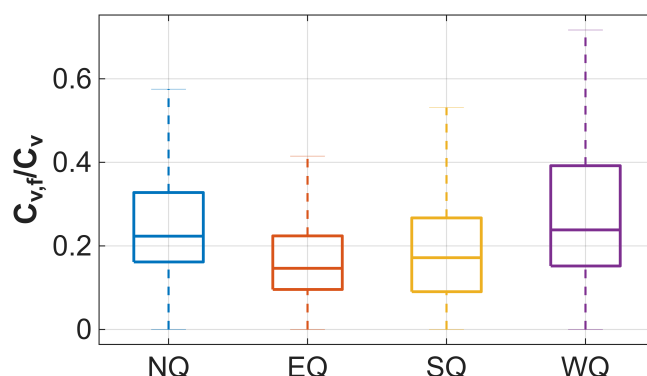


Figure 13. Seasonal variability of the mGMM-derived fine mode volume fraction ($C_{v,f}/C_v$). Box plots show median (horizontal line), interquartile range (box), and whiskers ($1.5 \times \text{IQR}$), with the observed shift from lower summer values to higher winter values.

Directional stratification shows the same contrast (Figure 14): EQ and SQ are systematically shifted toward lower $C_{v,f}/C_v$ than NQ and WQ, as seen in both cumulative distributions and quadrant box plots. This directional ranking is consistent with the FMF wind-direction behavior reported in Section 4.1.2, where offshore, marine/estuarine-influenced flow (SQ and EQ) is associated with lower FMF than continental flow (NQ and WQ). Taken together, the inversion-derived $C_{v,f}/C_v$ pattern exhibits a seasonal and directional contrast in a manner that is analogous to the FMF behavior. This agreement between related proxies is consistent with modulation of the aerosol fine–coarse balance across transport regimes, without implying strict metric equivalence or unique source attribution.



(a) CDF by wind quadrant (NQ, EQ, SQ, WQ).



(b) Box plots by wind quadrant (NQ, EQ, SQ, WQ).

Figure 14. Wind-direction dependence of $C_{v,f}/C_v$ using the same quadrant framework as the FMF analysis. Panel (a) shows empirical cumulative distributions; panel (b) shows box-plot summaries by quadrant.

4.2.3 Multisite modal comparison

We next compare the Montevideo wind–season $C_{v,f}/C_v$ statistics with selected sites. We apply the same mGMM algorithm introduced in Section 3.2 to Version 3 Level 2.0 inversions from 2020–2023 for the reference sites. The marine set includes AERONET sites Ragged Point, Cape San Juan, ARM Graciosa, and Amsterdam Island; the urban and continental set includes São Paulo, Santiago de Chile, Buenos Aires, and Beijing. Within the subset definitions used here, SQ and EQ summer conditions provide the marine/estuarine-influenced comparison case, whereas NQ and WQ winter conditions provide the higher-fine-fraction winter case. Figure 15 shows the geographic distribution of the sites. For the reference sites, we use the full 2020–2023 Level 2.0 record. We compare the sites using the fine-mode volume fraction and the median of all volume-weighted particle-radius means for the coarse modes (derived from the two-coarse-mode mGMM fits). The selected reference-site medians provide the marine and urban/continental comparison ranges used to position Montevideo in the multisite analysis. The Low/High NO_2 values (Table 1 and 2) are descriptive sensitivity subsets based on the lower/upper quartile of the filtered 2020–2023 daily TROPOMI NO_2 record. For the low- NO_2 case, Q25 is computed from the full filtered daily NO_2 column distribution, and the subset then retains Montevideo inversions whose matched daily column satisfies $\text{NO}_2 \leq \text{Q25}$ and whose winds fall within SQ or EQ. The high- NO_2 subset is defined analogously, using $\text{NO}_2 \geq \text{Q75}$ and retaining NQ or WQ winds. Their satellite context is discussed in Section 4.4.



Figure 15. Geographic location of the AERONET marine and urban reference sites used in the multisite comparison. Blue circles: marine reference sites; red squares: urban reference sites; green star: Montevideo (MVD, study site). Base map: Esri World Imagery (Sources: Esri, DigitalGlobe, GeoEye, i-cubed, USDA FSA, USGS, AEX, Getmapping, Aerogrid, IGN, IGP, swisstopo, and the GIS User Community | Powered by Esri).

385 Tables 1 and 2 include a comparison of the fine-mode volume fraction ($C_{v,f}/C_v$, Eq. 10), which summarizes the fine-coarse
balance. At the reference sites, marine medians are systematically lower than urban ones, consistent with a marine aerosol
regime with a larger relative coarse-mode contribution, as described by Smirnov et al. (2002). Montevideo lies between these
selected-site ranges: the full record remains intermediate, the SQ and EQ summer subset enters the marine range, and the NQ
and WQ winter subset shifts toward the urban range. We next compare the corresponding coarse-mode r_v statistics using the
390 same reference-site framework.

Table 1. Comparison of fine-mode volume fraction ($C_{v,f}/C_v$) between Montevideo and marine reference sites. Entries include [Q25, Q75]
and sample size N using AERONET Version 3 Level 2.0 inversions (2020–2023). MVD rows include the full record (MVD Total), SQ and
EQ summer (DJF), and the descriptive low- NO_2 subset within SQ and EQ ($\text{NO}_2 \leq \text{Q25}$).

Site	N	$C_{v,f}/C_v$ [Q25, Q75]
Ragged Point	256	0.138 [0.056, 0.205]
Cape San Juan	535	0.101 [0.040, 0.169]
ARM Graciosa	617	0.175 [0.091, 0.270]
Amsterdam Is.	116	0.126 [0.027, 0.307]
MVD Total	2322	0.218 [0.139, 0.343]
MVD SQ and EQ Summer	304	0.123 [0.092, 0.200]
MVD Low NO_2 (SQ and EQ)	147	0.098 [0.068, 0.155]



Table 2. Comparison of fine-mode volume fraction ($C_{v,f}/C_v$) between Montevideo and urban reference sites. Entries include [Q25, Q75] and sample size N using AERONET Version 3 Level 2.0 inversions (2020–2023). Montevideo rows include the full record (MVD Total), NQ and WQ winter (JJA), and the descriptive high- NO_2 subset within NQ and WQ ($\text{NO}_2 \geq \text{Q75}$).

Site	N	$C_{v,f}/C_v$ [Q25, Q75]
São Paulo	163	0.448 [0.348, 0.522]
Santiago	919	0.264 [0.204, 0.363]
Buenos Aires	1631	0.281 [0.177, 0.391]
Beijing	1233	0.266 [0.199, 0.418]
MVD Total	2322	0.218 [0.139, 0.343]
MVD NQ and WQ Winter	315	0.353 [0.239, 0.487]
MVD High NO_2 (NQ and WQ)	428	0.238 [0.172, 0.350]

The radius comparison first separates cases in which the mGMM retrieves one coarse mode ($N_c = 1$) from those with two coarse modes ($N_c = 2$). This split is useful in Montevideo because the two-coarse branch appears in 1180 of 2322 inversions (50.8%) for which the small- and large-coarse modes are separated by about $2.5 \mu\text{m}$ in median r_v . All reported r_v values are volume-weighted particle-radius means from mGMM fits to AERONET almucantar inversion PVSDs.

Table 3. Coarse-mode volume-weighted particle-radius means for MVD and the marine reference sites. Entries include [Q25, Q75] using Level 2.0 data. Here N denotes the number of inversions with exactly one coarse mode ($n_c = 1$) or exactly two coarse modes ($n_c = 2$); cases with $n_c \neq 1, 2$ are excluded. The Montevideo low- NO_2 row uses the Q25 definition stated above and is a descriptive sensitivity subset. The corresponding urban/continental reference-site comparison is given in Table 4.

Site	Single Coarse Mode		Two Coarse Modes		
	N	$r_{v,c}$ (μm)	N	Small $r_{v,c}$ (μm)	Large $r_{v,c}$ (μm)
Ragged Point	151	1.97 [1.83, 2.23]	105	0.95 [0.76, 1.45]	2.57 [1.99, 3.57]
Cape San Juan	269	2.19 [1.94, 2.69]	264	1.31 [0.88, 1.74]	3.44 [2.85, 4.20]
ARM Graciosa	305	2.30 [1.84, 2.88]	304	1.13 [0.78, 1.61]	3.51 [3.07, 4.02]
Amsterdam Is.	46	2.81 [2.64, 3.04]	70	1.12 [0.80, 1.43]	3.51 [3.13, 3.86]
MVD Total	1054	3.32 [2.85, 3.83]	1180	1.90 [1.29, 2.31]	4.44 [3.76, 4.96]
MVD SQ and EQ Summer	117	3.33 [2.83, 3.72]	147	1.78 [0.88, 2.31]	4.19 [3.23, 5.09]
MVD Low NO_2 (SQ and EQ)	68	3.06 [2.71, 3.54]	68	1.07 [0.82, 1.88]	3.48 [3.11, 4.17]

395 Tables 3 and 4 show that Montevideo's median coarse-mode r_v changes little relative to the MVD Total case when the mGMM is constrained to a single coarse mode. The $n_c = 2$ branch shows the clearer wind–season contrast. For the small-coarse mode component, the summer median for SQ and EQ decreases from $1.90 \mu\text{m}$ in MVD Total to $1.78 \mu\text{m}$, moving toward the marine-reference value of the comparison. The NQ and WQ winter median is $1.96 \mu\text{m}$, near the lower value of the



Table 4. Coarse-mode volume-weighted particle-radius means for Montevideo and the urban reference sites. Entries include [Q25, Q75] using Level 2.0 data. Here n denotes the number of inversions with exactly one coarse mode ($n_c = 1$) or exactly two coarse modes ($n_c = 2$); cases with $n_c \neq 1, 2$ are excluded. The Montevideo high-NO₂ row uses the Q75 definition stated above and is a descriptive sensitivity subset.

Site	Single Coarse Mode		Two Coarse Modes		
	N	$r_{v,c}$ (μm)	N	Small $r_{v,c}$ (μm)	Large $r_{v,c}$ (μm)
São Paulo	97	3.14 [2.87, 3.38]	62	2.33 [2.05, 2.49]	4.72 [4.45, 5.04]
Santiago	249	3.85 [3.62, 4.17]	670	2.27 [2.07, 2.52]	5.00 [4.73, 5.28]
Buenos Aires	790	3.45 [3.02, 3.89]	832	2.08 [1.57, 2.39]	4.64 [4.16, 5.08]
Beijing	476	2.83 [2.43, 3.25]	755	2.16 [1.85, 2.40]	4.87 [4.50, 5.29]
MVD Total	1054	3.32 [2.85, 3.83]	1180	1.90 [1.29, 2.31]	4.44 [3.76, 4.96]
MVD NQ and WQ Winter	174	3.43 [3.13, 3.86]	134	1.96 [1.50, 2.27]	4.50 [4.03, 4.72]
MVD High NO₂ (NQ and WQ)	182	3.45 [2.97, 3.97]	233	2.02 [1.59, 2.28]	4.62 [4.25, 4.96]

urban/continental comparison. The NO₂ selected data are retained here as descriptive sensitivity subsets and are interpreted in Section 4.4.

More generally, supermicron aerosol at a coastal–urban site can include sea salt as well as other particle types, such as dust or resuspended particles; therefore, we avoid attributing each fitted coarse mode to a unique aerosol species. Importantly, the appearance of $n_c = 2$ does not necessarily imply two resolved maxima in the summed coarse lobe: in the most onshore-influenced subset at Montevideo (SQ and EQ quadrants in DJF), only 8 of 145 analyzable $n_c = 2$ cases (about 6%) show two coarse peaks, with most cases presenting a single but broader and skewed supermicron lobe. Feng et al. (2017) derived bimodal supermicron volume distributions from optical particle sizing, reporting volume-weighted particle-radius means of 2.1 ± 0.1 and $5.1 \pm 0.2 \mu\text{m}$. Shipboard MOUDI (micro-orifice uniform deposit impactor) measurements along a West Pacific transect provide a second in-situ example: Na⁺ mass, used as a sea-salt tracer, shows a bimodal supermicron size distribution, with major contributions in the 1.8–5.6 and 10–18 μm stage intervals (Xu and Gao, 2015).

These in-situ examples are reported as optical diameters or impactor-stage size ranges and are not directly comparable to the columnar AERONET-derived r_v values reported here. They provide contextual support for interpreting the supermicron lobe as broader or structurally more complex than a single lognormal coarse mode. This structure is consistent with distinct sea-spray production pathways, including jet drops and spume droplets (O’Dowd and De Leeuw, 2007). The fine-mode mGMM retrievals were constrained to a single fine mode (Table 5). Montevideo shows the greatest number of fine-mode retrievals (1678 of 2322 inversions, 72.3%). Multiple fine modes are retrieved in fewer cases (548 cases, 23.6%) and occur at smaller, closely spaced submicron radii. Treating those multiple fine modes as ordered small- and large-fine branches would make the comparison between fine-mode radii less stable.

Tables 5 and 6 provide the corresponding fine-radius comparison for the $n_f = 1$ subset. In these tables, the SQ and EQ summer subset gives the smallest Montevideo fine-mode r_v ($0.113 \mu\text{m}$) and lies at the low end of the marine-reference com-



Table 5. Comparison of fine-mode volume-weighted particle-radius means between Montevideo and marine reference AERONET sites, restricted to single-mode mGMM inversions ($n_f = 1$). Here N denotes the number of such inversions. Entries include [Q25, Q75] using Level 2.0 data. The Montevideo low-NO₂ subset uses the Q25 definition stated above and is retained for completeness as a descriptive sensitivity subset.

Site	N	$r_{v,f}$ (μm)
Ragged Point	137	0.115 [0.100, 0.138]
Cape San Juan	406	0.148 [0.119, 0.202]
ARM Graciosa	465	0.147 [0.122, 0.192]
Amsterdam Is.	71	0.150 [0.117, 0.261]
MVD Total	1678	0.143 [0.120, 0.196]
MVD SQ and EQ Summer	216	0.113 [0.102, 0.127]
MVD Low NO₂ (SQ and EQ)	106	0.127 [0.110, 0.177]

Table 6. Comparison of fine-mode volume-weighted particle-radius means between Montevideo and urban reference AERONET sites, restricted to inversions where the mGMM retrieves exactly one fine mode ($n_f = 1$). Here N denotes the number of such inversions. Entries show median [Q25, Q75] using Level 2.0 data. The Montevideo high-NO₂ row uses the Q75 definition stated above and is retained for completeness as a descriptive sensitivity subset.

Site	N	$r_{v,f}$ (μm)
São Paulo	108	0.170 [0.146, 0.196]
Santiago	554	0.137 [0.114, 0.184]
Buenos Aires	828	0.142 [0.112, 0.187]
Beijing	1020	0.162 [0.141, 0.192]
MVD Total	1678	0.143 [0.120, 0.196]
MVD NQ and WQ Winter	232	0.182 [0.142, 0.239]
MVD High NO₂ (NQ and WQ)	337	0.145 [0.128, 0.181]

parison. The NQ and WQ winter subset shifts to larger fine radii (0.182 μm), above the urban-reference medians listed here. The fine-radius tables add a size-scale counterpart to the wind–season ordering in $C_{v,f}/C_v$ (Section 4.2.2; Figure 14).

Overall, the multisite comparison places Montevideo between the selected marine and urban reference ranges. Under SQ and EQ summer conditions, $C_{v,f}/C_v$, the $n_c = 2$ small-coarse mode, and the $n_f = 1$ fine-mode radius move toward the margins of the marine reference ranges. Under NQ and WQ winter conditions, $C_{v,f}/C_v$ and the $n_f = 1$ fine-mode radius shift toward the margins of the urban reference ranges, while the $n_c = 2$ small-coarse mode lies near the lower urban margin. Taken together, these changes support a transport-regime interpretation of the modal shifts.



4.3 Model comparisons

Having characterized the modal structure at Montevideo and compared it with reference sites, we now evaluate the spectral performance of the mGMM approach. We first compare the spectral consistency of the mGMM and standard bimodal representations using the Ångström exponent (Sec. 4.3.1), and then illustrate the differences between our implementation and the original Taylor algorithm (Sec. 4.3.2).

4.3.1 Comparison of mGMM with the standard bimodal model

Having evaluated the physical consistency of the mGMM retrievals under different wind-direction groupings, we now perform an internal consistency test comparing the optical representation of aerosols when moving from the standard bimodal model to the multimodal approach. By “standard bimodal model” we mean the sum of the fine- and coarse-mode lognormal distributions, with parameters provided by the AERONET inversion product (VolC-F, VMR-F, Std-F, and VolC-C, VMR-C, Std-C; VMR-F and VMR-C denote the product’s fine- and coarse-mode radius parameters), following the formulation given in Section 3 of Dubovik and King (2000). For both representations, spectral AOD was computed using Mie theory, adopting the same spectral refractive indices retrieved by the AERONET inversion algorithm (Dubovik and King, 2000; Dubovik et al., 2002) (Level 1.5) and the spectral extrapolation procedures described in subsection 3.3. Given the uncertainties in the retrieved refractive index at low AOD (Section 3.1.4), this comparison is not an absolute validation; rather, it tests whether the mGMM representation produces more consistent spectral behavior than the standard bimodal representation when the same refractive index is applied to both. We quantify this spectral consistency in terms of the Ångström exponent (AE, 440–870 nm). We present three illustrative cases selected from the upper tail of the $\Delta AE(\%)$ (bimodal-mGMM) distribution to highlight situations where the two representations diverge. These cases are not intended to be representative of the entire dataset; the quantitative evaluation of Table 7 below is based on the complete inversion subset.

Panels of Figure 16 show, for each date, comparison of three different size-distributions:

1. The PVSD retrieved from the 22 AERONET data values (red dots).
2. The standard AERONET bimodal distribution derived from the fine and coarse mode inversion parameters (blue line);
3. The distribution provided by the mGMM multimodal fit (green dashed line).

These results were obtained using Mie theory to the standard bimodal and mGMM size distributions using the same AERONET-derived refractive indices and the extrapolation procedure described in Sect. 3.3.

For the three selected cases shown in Figure 16, Table 7 compares the derived Ångström exponent (440–870 nm) with the Mie-calculated values from the standard bimodal and mGMM size distributions. These case studies illustrate that the multimodal scheme can reduce discrepancies in the inferred spectral slope. Extending the comparison to the complete 4-year inversion subset ($n = 2322$), we quantify the signed relative difference with respect to direct-sun observations, $\Delta(\%) = (AE_{\text{calc}} - AE_{\text{obs}})/AE_{\text{obs}} \times 100$. The median $\Delta(\%)$ decreases from +17 % (bimodal) to +3.4 % (mGMM), while the seasonal distributions (Figure 17) show that this improvement carries over into the seasonal statistics.

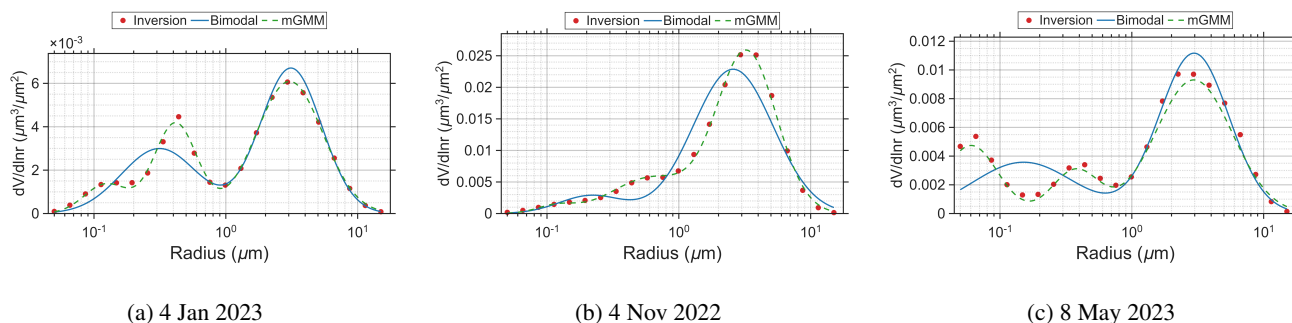


Figure 16. Comparison among the 22-bin AERONET inversion PVSD (red dots), the standard bimodal model derived from the AERONET inversion product (blue solid line), and the mGMM modal representation (green dashed line) for three selected days.

Table 7. Comparison of observed and Mie-calculated Ångström exponent values ($AE_{440-870}$) for the three dates shown in Figure 16. Calculated values are derived from the standard bimodal and mGMM size distributions using the same refractive-index inputs and Mie procedure. Signed relative difference: $\Delta(\%) = (AE_{calc} - AE_{obs})/AE_{obs} \times 100$.

Date	AE_{obs}	Bimodal		mGMM	
		AE_{calc}	$\Delta(\%)$	AE_{calc}	$\Delta(\%)$
4 Jan 2023	0.54	0.79	+45.9	0.59	+9.4
4 Nov 2022	0.35	0.46	+32.8	0.34	−1.5
8 May 2023	0.78	1.15	+47.4	0.78	−1.0

In summary, the mGMM approach shows greater spectral consistency than the standard bimodal model across the seasonal distributions examined here. This indicates that the additional flexibility in mode representation translates to measurable reductions in AE discrepancies.

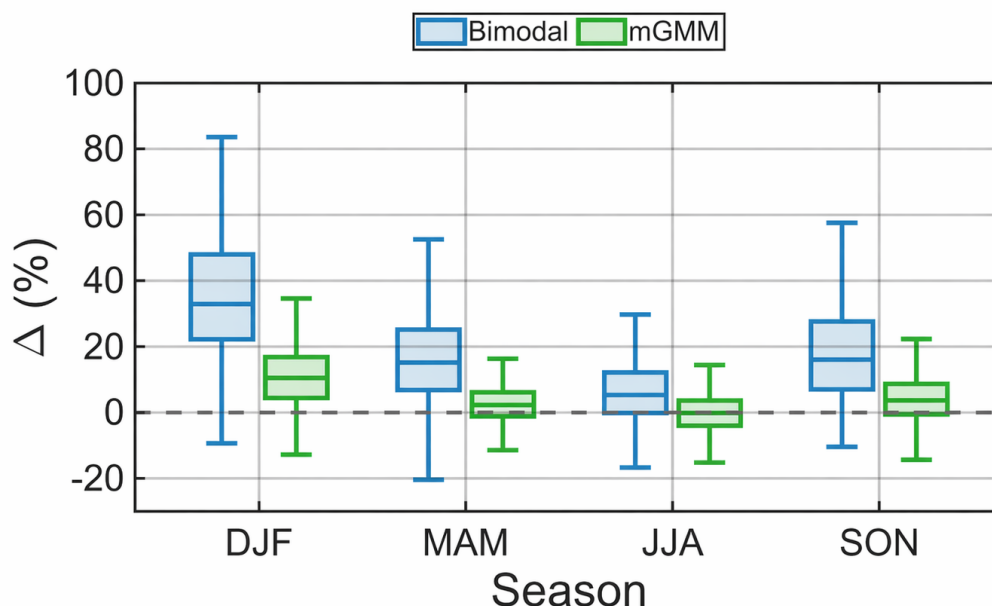


Figure 17. Seasonal distribution of signed relative Ångström-exponent differences, $\Delta(\%) = (AE_{calc} - AE_{obs})/AE_{obs} \times 100$, for the bi-modal and mGMM approaches. Values close to zero indicate better spectral consistency with direct-sun measurements.

4.3.2 Comparison of mGMM with the original Taylor et al. (2014) implementation

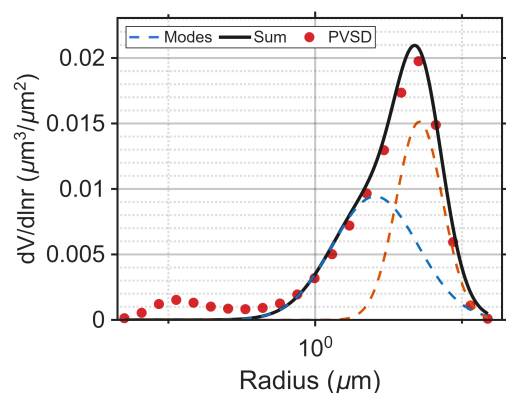
To isolate the impact of our implementation choices relative to the original Taylor framework, we compare the present mGMM with a Taylor-like implementation run on the same AERONET PVSDs. As described in Section 3.2, the present implementation uses a consistent L2-norm criterion for fitting and mode selection, preserves the native 22-bin resolution, computes modal volumes from analytical Gaussian integrals, and includes a dedicated fine-mode detection step. We illustrate these adjustments using three cases observed in Montevideo that show how the two fitting workflows respond to the same measured particle-volume size distributions:

- **Case 1 (23 Feb 2021):** A coarse-dominated distribution in which Taylor’s algorithm did not detect a fine mode, whereas the present mGMM identifies a small but discernible fine mode contribution.
- **Case 2 (14 Jun 2021):** A clearly bimodal distribution in which Taylor approach fitted 5 modes, whereas the present mGMM retained 2 modes while preserving the bimodal structure.
- **Case 3 (14 Sep 2022):** A trimodal distribution in which Taylor approach produced 6 modes, whereas the present mGMM reproduces the same overall structure with 3 modes.

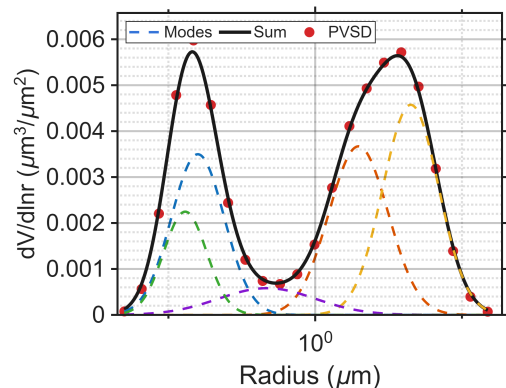
Taken together, these cases indicate that mGMM can recover fine structure and, in some cases, represent a given distribution with fewer modal terms.



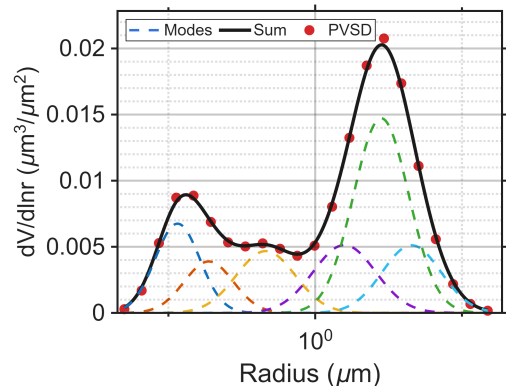
Taylor's approach



(a) 23 Feb 2021: no fine mode

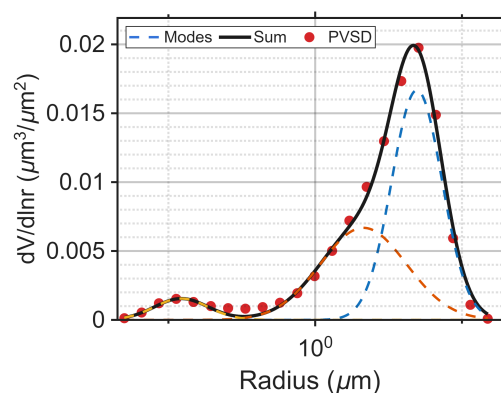


(c) 14 Jun 2021: 5 modes

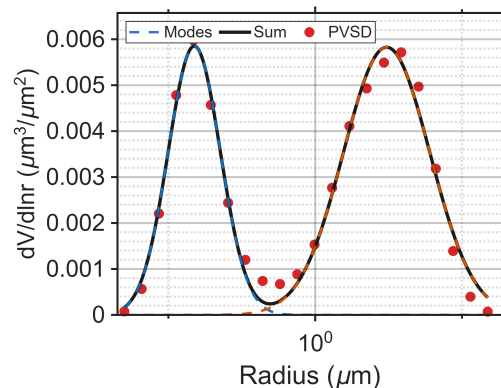


(e) 14 Sep 2022: 6 modes

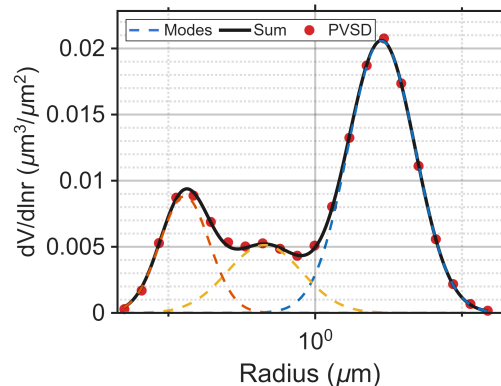
mGMM's approach



(b) 23 Feb 2021: fine mode detected



(d) 14 Jun 2021: 2 modes



(f) 14 Sep 2022: 3 modes

Figure 18. Comparison between the original Taylor et al. (2014) GMM implementation and the present mGMM for three representative AERONET PVSDs. Column headers identify the fitting workflow. Red circles are observed PVSD values on the native 22-bin radius grid, the black line is the fitted sum, and colored dashed curves are individual modal contributions.



4.4 Satellite-based context for the transport-regime interpretation

Tropospheric NO₂ columns values from TROPOMI (Sentinel-5 Precursor) provide independent satellite context for the transport-regime interpretation developed in Sections 4.2.2–4.2.3. During the period 2020–2023, NO₂ columns values are higher in NQ and WQ and lower in SQ and EQ, parallel to the aerosol ordering in which continental sectors show a larger fine-mode contribution and marine/estuarine-influenced sectors show lower fine fractions and relatively larger coarse-mode contribution. The Low/High NO₂ values reported in the multisite tables are therefore used as descriptive sensitivity subsets: the low-NO₂ SQ and EQ subset lies at the low fine-fraction end of the marine comparison and its $n_c = 2$ small-coarse mode moves close to the marine reference values, whereas the high-NO₂ NQ and WQ subset lies near the lower urban/continental reference side in fine-mode volume fraction and coarse-mode radii. Given their smaller sample sizes, these values are read as contextual support for the main wind–season pattern. The daily NO₂ diagnostics and figures are provided in Appendix B.

5 Conclusions

We analyzed four years (2020–2023) of observations from the Montevideo_FING station at Montevideo (AERONET Team (2026)) and established a systematic characterization of aerosol optical properties at a coastal-urban site in South America. The direct-sun products (AOD, FMF, and the SDA-derived decomposition into τ_f and τ_c), stratified by BLH-weighted winds, show low aerosol load (median AOD₅₀₀ = 0.067) and weak seasonality in total AOD. However, the balance between fine and coarse contributions exhibits a pronounced annual cycle: FMF is much higher in winter (median 0.72) than in summer (median 0.42). This is consistent with the seasonal behavior of the extensive AOD analogues (τ_f peaks in winter while τ_c peaks in summer).

This seasonal contrast persists in the almucantar inversion subset, where the microphysical retrievals provide an inversion-based metric of the same fine–coarse balance. In particular, the fine mode volume fraction ($C_{v,f}/C_v$) shifts systematically from more coarse-dominated conditions in summer toward more fine-dominated conditions in winter.

Stratification by wind direction quadrants further supports a compact and internally consistent interpretation the optical and microphysical properties of aerosols: NQ and WQ quadrants are, on average, associated with enhanced fine mode contribution, whereas SQ and EQ quadrants are associated with enhanced coarse mode contributions. Within the selected wind–season subsets, the coarse-mode r_v values of the inversion subset also tend to be smaller under SQ and EQ summer conditions than under NQ and WQ winter conditions. Taken together, this wind-direction framework provides a practical context for interpreting both optical variability (direct-sun) and microphysics derived from inversions at the Montevideo coastal urban interface (without implying unique source attribution for individual events).

The inversion results indicate that strict bimodality is not the dominant state at Montevideo: 68% of the cases were classified as multimodal ($K \geq 3$), suggesting that a bimodal (fixed) representation may not accurately describe the retrieved modal structure in a mixed coastal–urban environment. To address this, we implemented a modified Gaussian Mixture Model (mGMM) that preserves compatibility with the native AERONET sampling (22 bins) while improving statistical consistency (least squares fitting, and improved sensitivity to the fine mode).



510 An internal consistency test of the spectral slope of the AOD against direct-sun observations shows that the multimodal representation better characterizes that spectral slope than the standard bimodal model: the median signed relative difference in the Ångström exponent decreases from 17.0% to 3.4%. This suggests that, in mixed cases, increased adaptability leads to a measurably more consistent Ångström exponent comparison when Mie calculations are applied to the retrieved size distributions.

515 Multisite comparisons place Montevideo between remote marine and urban-continental environments. The fine mode volume fraction shifts from a rather coarse-fraction-dominated value for SQ and EQ winds in summer (median $C_{v,f}/C_v = 0.123$) to a rather fine-fraction-dominated value for NQ and WQ winds in winter (0.353), while the mGMM-derived r_v values shift coherently towards the margins of the marine or urban reference ranges. These patterns are consistent with mixed coastal and continental influences, whereas directly attributing each mode to a single source type is not representative of this site.

520 Tropospheric NO₂ columns values from TROPOMI are higher in the NQ and WQ quadrants and decrease with increasing wind speed. This pattern is consistent with the separation observed in the aerosol metrics and provides an independent check on lower-tropospheric aerosol tendencies at this location. The SQ and EQ subset retained for low-NO₂ days ($\text{NO}_2 \leq \text{Q25}$) shows an even lower fine-mode volume fraction ($C_{v,f}/C_v = 0.098$) and a small-coarse mode close to the marine reference values. The resulting statistics provide contextual support for the wind–season pattern, with the wind-regime classification defined by ERA5.



525 Appendix A: BLH-weighted wind treatment: implementation details

This appendix provides the technical implementation details for the BLH-weighted wind treatment summarized in Section 3.4.

A1 Vertical reference and AGL-to-a.s.l. conversion

We use ERA5 level profiles (horizontal components and geopotential z) and boundary layer height (blh) from the CAMS global composition reanalysis (EAC4), both based on IFS (Hersbach et al., 2020; Inness et al., 2019; Copernicus Atmosphere
530 Monitoring Service, 2025). Geopotential z is converted to geometric height above sea level (a.s.l.) as $Z = z/g_0$. To anchor BLH (defined "above ground level," AGL) to a.s.l. elevations, we adopt terrain elevation at the site from the ETOPO1 global model: $Z_{\text{sfc}} = 17$ m a.s.l. (Amante and Eakins, 2009), and add this elevation to BLH: $\text{BLH}_{\text{asl}}(t) = 17 \text{ m} + \text{BLH}_{\text{AGL}}(t)$. Additionally, we ignore levels below the instrument.

A2 Level selection and exponential weighting

535 For each hour t , with wind profiles $\{u_k(t), v_k(t)\}$ and heights Z_k , we select levels in

$$\mathcal{B}(t) = \{k : Z_{\text{ref,low}} \leq Z_k \leq 17 + \text{BLH}(t)\}.$$

The vector average is calculated with exponential weights (w_k) depending on depth within the mixing layer, measured from $Z_{\text{ref,low}}$:

$$w_k(t) = \frac{\exp\left(-\frac{Z_k - Z_{\text{ref,low}}}{0.20 \text{BLH}(t)}\right)}{\sum_{j \in \mathcal{B}(t)} \exp\left(-\frac{Z_j - Z_{\text{ref,low}}}{0.20 \text{BLH}(t)}\right)}, \quad U = \sum_{k \in \mathcal{B}(t)} w_k u_k, \quad V = \sum_{k \in \mathcal{B}(t)} w_k v_k. \quad (\text{A1})$$

540 At this decay scale (20% BLH), approximately 64% of the weight falls into the first 0.2 BLH and 92% into the first 0.5 BLH, making the lower layer more influential while preserving representativeness for the remaining mixed layer.

A3 Temporal smoothing and wind-vector diagnostics

To reduce high-frequency variability in the hourly wind components, we apply a backward running mean with parameter $N = 5$, averaging over 6 samples (hours $i - 5$ through i , inclusive):

$$545 \quad \bar{U}_i = \frac{1}{6} \sum_{j=i-5}^i U_j, \quad \bar{V}_i = \frac{1}{6} \sum_{j=i-5}^i V_j. \quad (\text{A2})$$

Meteorological wind direction and speed are then computed from the smoothed components: $\text{WD} = \text{mod}(450^\circ - \arg(\bar{U} + i\bar{V}), 360^\circ)$ and $\text{WS} = \sqrt{\bar{U}^2 + \bar{V}^2}$, where the sign convention ensures direction indicates where the wind blows from.

A4 Equivalent height and local summary values

We define equivalent wind height as $Z_{\text{eq}}(t) = \sum_{k \in \mathcal{B}(t)} w_k(t) Z_k$, summarizing the representative altitude of the weighted flow
550 in coordinates above sea level (a.s.l.). In the period (2020–2023), BLH from CAMS EAC4 presents (p10, median, p90) = (202,



511, 1033) m; the mixing domain top is then BLH+17m with percentiles (219, 528, 1050) m. The Z_{eq} calculated with the above scheme yields percentiles (107, 174, 293) m.

Appendix B: NO₂ data from TROPOMI instrument

As satellite context for the transport-regime interpretation, we examine tropospheric NO₂ columns values from TROPOMI. Figure B1 summarizes tropospheric NO₂ columns values over the site surroundings for 2020–2023. The period median is $1.77 \times 10^{-5} \text{ mol m}^{-2}$ ($p_{90} = 3.73 \times 10^{-5} \text{ mol m}^{-2}$). Seasonally, winter (JJA) columns values exceed summer (DJF) values by 28.3%. It is also observed that the median values for working days are higher than those for weekends, especially in winter, which coincides with the weekly cycles recorded by TROPOMI in other urban contexts (Goldberg et al., 2021; Fioletov et al., 2025). However, our main focus here is the wind-direction stratification discussed above.

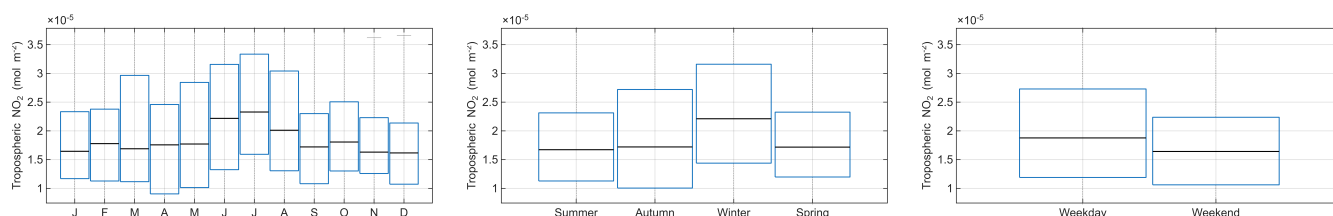


Figure B1. Tropospheric NO₂ (TROPOMI, Sentinel 5 Precursor) observations during 2020–2023: (a) monthly and (b) seasonal distributions. Panel (c) compares working days and weekends.

Using the BLH-weighted wind treatment (Section 3.4), we stratify daily NO₂ columns values by wind quadrant (NQ, SQ, EQ, WQ) and wind-speed tercile (Low/Medium/High). Median NO₂ columns values are higher under NQ and WQ winds ($2.02 \times 10^{-5} \text{ mol m}^{-2}$ and $2.13 \times 10^{-5} \text{ mol m}^{-2}$ for northerly and westerly winds, respectively) and lowest under S winds (1.34×10^{-5}), with E intermediate (1.67×10^{-5}). Within each quadrant, the values in the NO₂ columns decrease with increasing wind speed (Figure B2) which is consistent with aerosol results in Montevideo. NO₂ column values are higher under overland winds from NQ and WQ, where FMF also increases (Figure 10, panels (a) and (b)), whereas the lower NO₂ columns under SQ and EQ are compatible with cleaner marine or estuarine inflow.

The NO₂ column patterns, with maxima in the NQ and WQ quadrants and in winter, are consistent with the wind-regime separation identified in FMF and the mGMM analysis. The coincidence of higher FMF and higher NO₂ columns values under NQ and WQ winds is consistent with stronger urban/overland influence in those sectors. Together, these diagnostics provide satellite context for the transport-regime interpretation used in the main text.

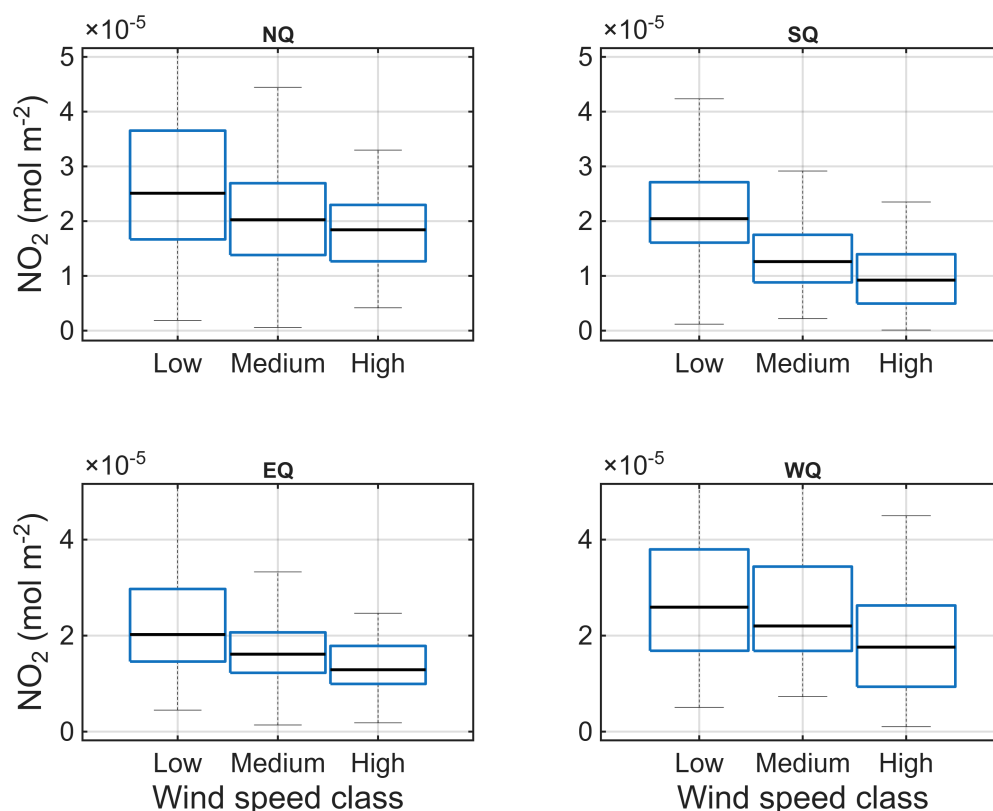


Figure B2. Tropospheric NO₂ columns values (TROPOMI, Sentinel 5 Precursor) by quadrant (NQ, SQ, EQ, WQ) and BLH-weighted wind speed class (Low/Medium/High; terciles). Higher NO₂ columns values are observed with overland winds (N and W) and decrease within each quadrant with increasing speed.

Code availability. The analysis and figure generation were performed using free trial version of MATLAB and public Python libraries. The Mie calculations used the custom *MiePy_CUDA* package (Python module *MiePy*). The analysis and figure-generation scripts are available from the corresponding author upon reasonable request.

Data availability. AERONET Version 3 Level 2.0 products are available from the AERONET data archive (<https://aeronet.gsfc.nasa.gov/>). ERA5 reanalysis data are available from the Copernicus Climate Data Store (CDS; <https://cds.climate.copernicus.eu/>). CAMS EAC4 composition reanalysis data are available from the Copernicus Atmosphere Monitoring Service (CAMS; <https://atmosphere.copernicus.eu/>). Tropospheric NO₂ columns from TROPOMI (Sentinel-5P OFFL L3) were accessed via Google Earth Engine.

575



Author contributions. AA conceptualized the study, processed and analyzed the AERONET, ERA5, CAMS, and TROPOMI data, developed the mGMM workflow, prepared the figures. EF led the conceptualization, contributed to the interpretation of the results, supervised the work. NTO'N and BH provided fundamental comments on the interpretation of AERONET inversion and SDA products, contributed to the methodological framework for modal aerosol analysis. All co-authors contributed to the revision of the manuscript.

Competing interests. The authors declare that they have no conflict of interest.

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