Supplementary for

Transported African Dust in the Lower Marine Atmospheric Boundary Layer is Internally Mixed with Sea Salt Contributing to Increased Hygroscopicity and a Lower Lidar Depolarization Ratio

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S1. Overview of Airborne Aerosol Sampling

The Center for Interdisciplinary Research Projects in Airborne Science (CIRPAS), operated by the Naval Postgraduate School (NPS), provides a specialized Twin Otter research aircraft designed to support environmental and atmospheric science missions. The Twin Otter is a versatile aircraft well-suited for research flights enabling direct sampling of the lower troposphere. Equipped with multiple wing pylons, fuselage ports, and integrated power systems, the aircraft can accommodate a wide range of meteorological and aerosol instrumentation. CIRPAS adheres to rigorous calibration protocols and offers comprehensive logistical support to ensure high-quality data acquisition across multi-institutional field campaigns.

In situ measurements of air temperature (Rosemount total temperature probe, model E102AL) (Friehe and Khelif, 1992) and dew point temperature (Vigilant Chilled Mirror Hygrometer, Edgetech Instruments Inc., Hudson, MA) were used by flight scientists to determine the cloud base height (CBH). During the initial sounding at each sampling station, vertical profiles of air temperature and dew point temperature were monitored as they gradually converged with increasing altitude.

Aerosol measurements aboard the CIRPAS Twin Otter rely on the precise characterization of aerosol sampling dynamics. The aircraft's aerosol inlet has been evaluated under both flight and wind tunnel conditions to quantify its transmission efficiency across a range of particle sizes. Studies show that the inlet's transmission efficiency begins to decline near 3.5 μm particle diameter but stabilizes at 5.5 μm with a transmission efficiency just above 0.6 for particles up to 9 μm (Hegg et al., 2005). This behavior is likely attributed to sub-isokinetic aspiration flow, wherein the inlet flow velocity is lower than the freestream velocity, leading to inertial losses of larger particles.

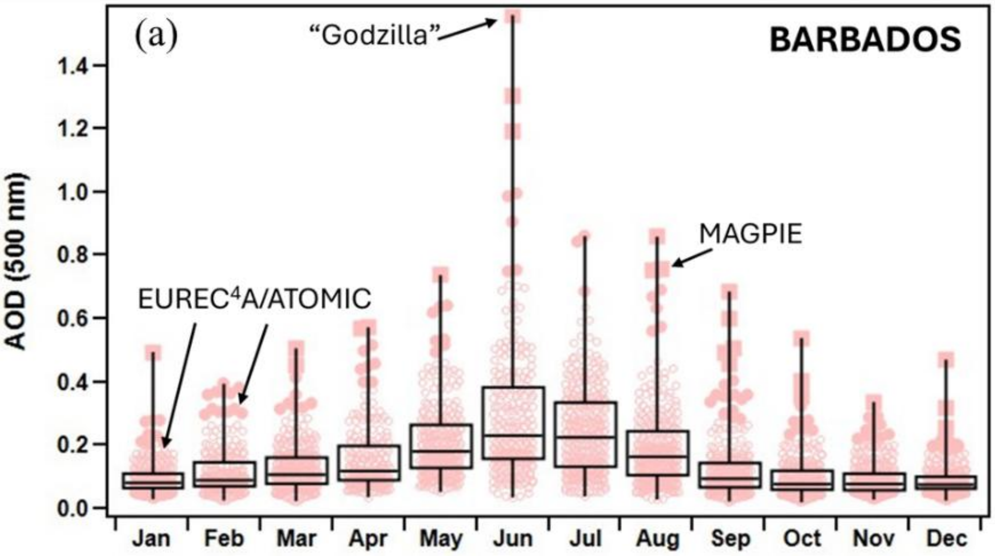

Figure S1. (a) Box plot showing monthly AOD at 550 nm measured using the AERONET (Holben et al., 1998) at Barbados for the last 17 years.

S2. Single Particle Aerosol Composition and Size Distribution

Single-particle chemical composition and morphology of aerosol samples collected during dust events were examined using computer-controlled scanning electron microscopy with energy-dispersive X-ray spectroscopy (CCSEM/EDX). The analysis revealed a diverse set of particle types with distinct chemistries and morphologies, including mineral dust, sea spray, aged sea spray, internally mixed mineral dust and sea spray, sulfates, and organics (Ault et al., 2014; Royer et al., 2023). Representative scanning electron microscopy (SEM) images and corresponding EDX spectra for each particle class are shown in Fig. 3c in main text.

Mineral Dust: Mineral dust particles were primarily characterized by the presence of aluminosilicate elements such as Si, Al, Fe, K, Ca, and Mg, consistent with long-range transported Saharan dust (Royer et al., 2023, 2025; Twohy et al., 2009; Levin et al., 2005;

Krueger et al., 2004; Hand et al., 2010; Denjean et al., 2015). Notably, approximately only 25% of the dust particles analyzed contained sulfur and nitrogen, suggesting atmospheric chemical aging either during transport or during entrainment into the marine boundary layer (MBL). This is consistent with earlier findings at the site that reported minimal evidence of aging in mineral dust (Kandler et al., 2018; Royer et al., 2025).

Sea Spray and Aged Sea Spray: Fresh sea spray particles were identified by a high relative abundance of Na and Cl and exhibited crystalline morphologies indicative of halite (NaCl). Small peaks of Mg further confirmed their marine origin. Aged sea spray particles were distinguished by depleted chloride content and enriched sulfur and nitrogen signatures, likely resulting from heterogeneous reactions with atmospheric acidic gases such as sulfuric acid (H_2SO_4), nitric acid (HNO_3), and dinitrogen pentoxide (N_2O_5). These interactions are known to produce Cl-depleted sea spray and form secondary aerosol coatings (Ault et al., 2013a; Royer et al., 2023, 2025; Gaston et al., 2011). The aspect ratio of sea spray particles in the smaller size bins (Fig. 5d) is higher than in the larger bins, potentially due to the presence of gypsum rods commonly observed attached to these particles and partial drying of the particles on the substrate. However, it should be noted that the number of particles observed in these smaller size bins is significantly lower than in larger bins.

Internally Mixed Mineral Dust and Sea Spray: These particles exhibited a heterogeneous composition, containing both dust-derived (Si, Al, Fe, K, Ca, Mg) and marine-derived (Na, Cl) components (Royer et al., 2023, 2025; Kandler et al., 2018). Elemental distributions varied within individual particles, indicating spatially localized mixing of the two sources. The generally low percentage of nitrogen and sulfur suggests limited atmospheric aging of this particle type. This is contrary to wintertime observations at Barbados by Royer et al. (2025), where SEM/EDX elemental mapping showed that signs of aging, such as the presence of sulfur or nitrogen, were confined to the sea spray portions of these mixed particles. Previous work at the same site proposed that this internal mixing likely occurs locally, potentially driven by turbulent interactions between airborne dust and sea spray (Kandler et al., 2018).

Organics: Organic particles were predominantly composed of carbon and oxygen (>95%), with minor contributions from inorganic constituents (<5%). Several larger particles were observed, potentially corresponding to marine gels, characterized by Mg-rich shells and sea spray-dominated cores (Ault et al., 2013b; Gaston et al., 2011). These marine gels are typically formed from bubble-bursting processes at the ocean surface, where hydrophobic organic compounds concentrated in the sea surface microlayer become aerosolized and associate with divalent cations during gel formation (Chin et al., 1998).

Sulfates: Sulfate-rich particles were identified based on dominant sulfur signals accompanied by carbon, oxygen, and nitrogen. These particles are consistent with marine secondary aerosol components such as ammonium sulfate ($(NH_4)_2SO_4$) and ammonium bisulfate (NH_4HSO_4) (Hand et al., 2010; Royer et al., 2023). The elevated carbon content suggests these particles also contain a substantial organic fraction, a common feature in marine submicron aerosols (O’Dowd and de Leeuw, 2007).

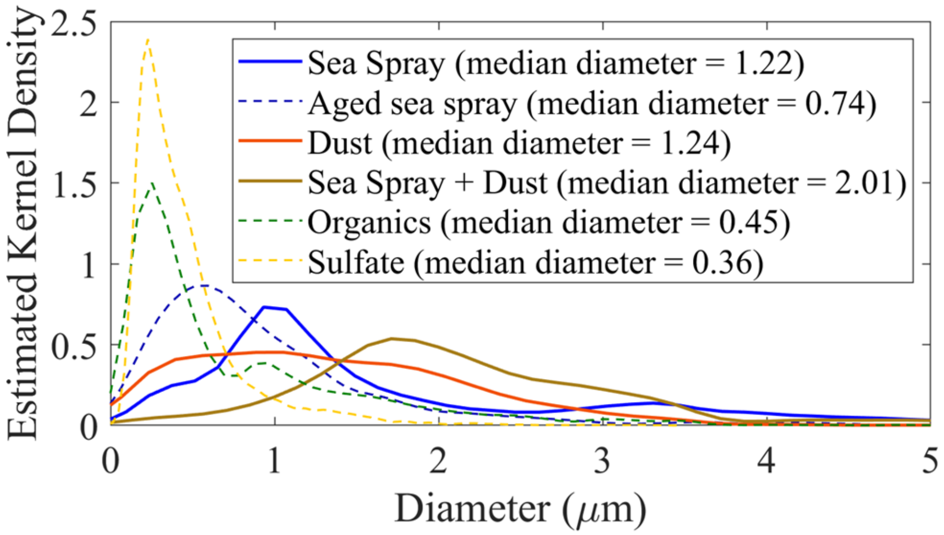

Fig. S2. Kernel density of different particle types vs particle diameter during the dust event.

S3. Estimation of Expected Lidar Depolarization Ratio

S3.1 Modeling expected depolarization ratio mixture.

To estimate the expected aerosol depolarization ratio (δ_{expected}), we begin by modeling the total aerosol backscatter as a mixture of contributions from dust and marine aerosols. The total backscatter coefficient (ν) is expressed as:

$$\nu = \nu^{(d)} + \nu^{(m)} \quad (1)$$

where, $\nu^{(d)}$ and $\nu^{(m)}$ are dust and marine air mass backscatter, respectively.

The total parallel ($\nu_{\parallel}$) and perpendicular ($\nu_{\perp}$) components of the backscatter can be similarly expressed as:

$$\nu_{\parallel} = \nu_{\parallel}^{(d)} + \nu_{\parallel}^{(m)} \qquad (2)$$

$$\nu_{\perp} = \nu_{\perp}^{(d)} + \nu_{\perp}^{(m)} \qquad (3)$$

The depolarization ratio mixture is then defined as:

$$\delta = \frac{v_{\perp}}{v_{\parallel}} \qquad (4)$$

Using this formulation, the expected depolarization ratio can be alternatively written as:

$$\delta expected = \frac{v_{\perp}^{(d)}}{v_{\parallel}^{(d)} + v_{\parallel}^{(m)}} + \frac{v_{\perp}^{(m)}}{v_{\parallel}^{(d)} + v_{\parallel}^{(m)}} \qquad (5)$$

This formulation requires estimation of the aerosol-specific backscatter and their polarized components which are computed as described in the following sections.

S3.2 Estimating the aerosol specific polarized backscatter

To calculate the expected depolarization, we first estimate extinction β [m^{-1}] using the observed particulate backscatter (v) and lidar ratio μ [sr]:

$$\beta = v\mu \qquad (6)$$

Next, the aerosol specific backscatter components for marine and dust contributions are derived using their respective mass concentrations $M^{(d)}$ and $M^{(m)}$ [μg m^{-3}], and lidar ratio cut offs for dust (μ^{d}) and marine aerosols (μ^{m}) were taken as 40 and 20, respectively:

$$v^{(m)} = \frac{M^{(m)}}{M^{(m)} + M^{(d)}} \frac{\beta}{\mu^{m}} \qquad (7)$$

$$v^{(d)} = \frac{M^{(d)}}{M^{(m)} + M^{(d)}} \frac{\beta}{\mu^{d}} \qquad (8)$$

S3.3 Compute the marine and dust parallel and perpendicular backscatters

Using the campaign-derived in-situ linear depolarization ratios for dust and marine aerosols in-situ marine $\delta^{(m)}$ and dust $\delta^{(d)}$ (i.e., 0.3 for dust and 0.02 for marine), we calculate the aerosol specific backscatter into polarized components.

a. the parallel polarized marine and dust backscatters is calculated as:

$$v_{\parallel}^{(m)} = \frac{v^{(m)}}{(1 + \delta^{(\mathrm{m})})} \qquad (9)$$

$$v_{\parallel}^{(d)} = \frac{v^{(d)}}{(1 + \delta^{(d)})} \qquad (10)$$

b. the perpendicular polarized marine and dust backscatters is calculated as:

$$v_{\perp}^{(m)} = \delta^{(m)} \, v_{\parallel}^{(m)} \qquad (11)$$

$$v_{\perp}^{(d)} = \delta^{(d)} \, v_{\parallel}^{(d)} \qquad (12)$$

Finally, the expected linear depolarization ratio is computed by substituting Eqs. (7) – (12) into Eq. (5).

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