



Mutual constraints on global aerosol composition from novel PACE data and ModelE

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Abstract. Aerosol climate impacts remain uncertain, in part because global aerosol composition observations have been unavailable to constrain Earth System Models (ESMs). We present the first comparison of aerosol composition from the Plankton, Aerosol, Cloud, ocean Ecosystem (PACE) satellite's Spectropolarimeter for Planetary Exploration one (SPEXone) against an ESM, co-locating Remote sensing of Trace gas and Aerosol Products (RemoTAP) retrievals with NASA GISS ModelE simulations to within ± 15 minutes over 2024–2025. In the fine mode, the two correlate well over land, capturing the same seasonal biomass burning and pollution cycles. The coarse mode, however, diverges: mineral dust agrees to within $\sim 10\%$ by volume over land, yet SPEXone exceeds ModelE eightfold over ocean, where it covaries with modeled sea salt and increases away from desert sources. Because SPEXone partitions the coarse mode on shape alone, we suggest this excess is nonspherical marine aerosol rather than mineral dust. Finally, ModelE carries a median 3.6 times the water per unit dry sea salt that SPEXone infers over ocean. Substituting reanalysis relative humidity (RH) and an observationally constrained hygroscopicity each move the model toward the retrieval, but neither alone nor together do they reproduce it. This factor amounts to only 0.027 in global median effective marine refractive index, amplified by the nonlinearity of the mixing rule used to infer water. Together, these results provide a first evaluation of modeled aerosol composition against PACE SPEXone retrievals.

1 Introduction

Atmospheric aerosols, hereon referred to as aerosols, are tiny liquid and solid particles suspended in Earth's atmosphere, ranging in size from a few nm to several μm in diameter. Despite being tiny, aerosols play a major role in determining Earth's energy balance. Aerosols are responsible for scattering and absorbing a significant portion of the Sun's light, and for facilitating the nucleation of nearly every cloud droplet and crystal, which themselves interact with light in complex ways (Seinfeld and Pandis, 2016). Aerosols are also important for air quality, with those smaller than $2.5 \mu\text{m}$ posing a risk to human health (Seposo et al., 2019). As such, studying aerosols, and accurately representing them in Earth System Models (ESMs), is essential for both the short-term and long-term well-being of humans.

As a result of their abundance, aerosols greatly impact Earth's energy balance. They do so both directly through their interactions with light, and indirectly through their ability to serve as cloud condensation nuclei (CCN) and ice nucleating



particles (INP). The sixth Intergovernmental Panel on Climate Change (IPCC) report found that aerosol-radiation interactions were responsible for -0.47 to 0.04 W m^{-2} of the change in effective radiative forcing from 1750 to 2019, compared to -1.45 to -0.25 W m^{-2} from aerosol-cloud interactions (ACI) (Forster et al., 2021). Collectively, this uncertainty is greater than that from all greenhouse gases combined (Forster et al., 2021). As a consequence, aerosols are believed to net cool Earth's surface, offsetting on the order of a third of the warming from greenhouse gases as of 2014 (Li et al., 2022). Importantly, a general decline in anthropogenic aerosol emissions in the United States, East Asia, and parts of Europe has been observed since 2015, which is why some believe global temperatures have been anomalously warm in the 2020s relative to ESM projections (Bauer et al., 2022; Quaas et al., 2022; Park et al., 2024). As such, accurately quantifying the aerosol direct and indirect effects is a major focus of research efforts today.

To constrain aerosol properties globally, a number of observational datasets have been developed. Ground-based networks such as the Aerosol RObotic NETwork (AERONET) provide column-integrated aerosol properties at fixed locations worldwide (Holben et al., 1998), while active satellite instruments such as the Cloud-Aerosol Lidar with Orthogonal Polarization (CALIOP) onboard the Cloud-Aerosol Lidar and Infrared Pathfinder Satellite Observations (CALIPSO) satellite infer detailed aerosol profiles with high vertical resolution but limited spatial coverage (Winker et al., 2006). Additionally, passive satellites have provided modelers with global aerosol datasets since the late 1990s, most commonly inferring Aerosol Optical Depth (AOD) (Torres et al., 2002; Remer et al., 2005). Despite these advances, passive satellites have historically been limited to cloud-free conditions globally (Christensen et al., 2017; Kaufman et al., 2005), face retrieval challenges in polar regions due to low sun angles, extended winter darkness, and persistent cloudiness (Wall et al., 2022), and have struggled to constrain aerosol microphysical properties (Li et al., 2019; Schutgens et al., 2021).

The last of these constraints has been partly addressed by polarimetry. From 2004 to 2012, the POLDER (Polarization and Directionality of the Earth's Reflectances) instrument onboard the PARASOL (Polarization and Anisotropy of Reflectances for Atmospheric Science coupled with Observations from a Lidar) satellite used polarimetric measurements to retrieve aerosol microphysical properties, though its sensitivity to aerosol composition and coarse-mode particles remained limited (Tanré et al., 2011). Since the mission ended, however, global observations of detailed aerosol composition and aerosol microphysical properties have remained a highly sought quantity for the modeling community.

A recent major advance in aerosol retrievals by satellites was the launch of the Plankton, Aerosol, Cloud, ocean Ecosystem (PACE) satellite on February 8, 2024 (Werdell et al., 2024). PACE carries three Earth-observing instruments, including the Spectropolarimeter for Planetary Exploration one (SPEXone), a multi-angle spectropolarimeter developed by the Space Research Organization Netherlands (SRON) and Airbus Netherlands (Hasekamp et al., 2019; van Amerongen et al., 2019). Due to SPEXone's ability to measure both radiance and linear polarization at unprecedented accuracy and spectral resolution, new inversion capabilities have become possible, allowing for the retrieval of aerosol microphysical properties globally for the first time. The Remote sensing of Trace gas and Aerosol Products (RemoTAP) algorithm, developed by SRON, is one such inversion applied to SPEXone observations, providing daily gridded column aerosol by species (Fu et al., 2025). Early validation efforts have shown strong agreement with AERONET and co-located flight campaigns (Fu et al., 2025; LeBlanc et al., 2024).



Despite the potential of these novel retrievals to reduce uncertainty of the direct and indirect effect in ESMs, PACE SPEXone aerosol chemical composition inversions have not yet been compared to ESM simulations. Here, we address this gap by presenting the first co-located comparison of SPEXone RemoTAP aerosol retrievals with an ESM, namely NASA Goddard Institute for Space Studies (GISS) ModelE, for 2024–2025. We compare column-integrated aerosol species including mineral dust, sea salt, black carbon (BC), absorbing organic aerosol (AOA), fine non-absorbing aerosol, and aerosol water, and identify regional and seasonal drivers of differences between the two products.

2 Data and methods

2.1 ModelE-MATRIX

In this work, we use the GISS ModelE3.1 ESM, a Coupled Model Intercomparison Project Phase 7 (CMIP7) evolution of ModelE2.1 (Kelley et al., 2020) with multiple updated structural and physical parameterizations (Cesana and Del Genio, 2021; Elsaesser et al., 2025). ModelE3.1's final resolution will be a C90 cubed-sphere grid; here we use an intermediate model version with $2^\circ \times 2.5^\circ$ horizontal resolution and 62 vertical layers. The model is given a one-year spin-up, and the years 2024–2025 are simulated with output saved every 30 minutes. Sea surface temperatures and sea ice concentrations are prescribed from the Hadley Centre Sea Ice and Sea Surface Temperature dataset version 1.1 (HadISST1.1) (Rayner et al., 2003), winds are nudged to National Centers for Environmental Prediction (NCEP) reanalysis data (Kalnay et al., 1996), and greenhouse gas concentrations follow the CMIP7 protocol (Nicholls et al., 2025). Land use and land cover are prescribed from Land-Use Harmonization 2 (LUH2) (Hurtt et al., 2020). Anthropogenic emissions are from the Community Emissions Data System (CEDS; Hoesly et al., 2018) prepared for CMIP7, and daily fire emissions from the Global Fire Emissions Database version 5.1 (GFED5.1) (van der Werf et al., 2025). Since CEDS emissions data for 2024 and 2025 were not yet available at the time of this analysis, unlike the biomass burning input files, the most recent available year (2023) was repeated under the assumption that anthropogenic emissions have not changed significantly over this period. This assumption is supported by the gradual nature of recent global emission trends, with year-to-year changes in key aerosol precursors such as SO_2 and BC typically on the order of a few percent (Hoesly et al., 2018; Bauer et al., 2022). Lastly, clear-sky conditions for ModelE are determined using a maximum overlap Monte Carlo approach based on the model layer-by-layer cloud fraction (Del Genio et al., 1996; Naud et al., 2008).

Within ModelE, we employ the Multiconfiguration Aerosol TRacker of mIXing state (MATRIX) aerosol microphysics module (Bauer et al., 2008). MATRIX uses the quadrature method of moments to track particle mass, number concentration, and size distributions of 16 unique aerosol populations mixed with combinations of eight chemical species: sea salt, mineral dust, organic aerosol (OA), BC, sulfate, nitrate, ammonium, and aerosol water (Bauer et al., 2008). MATRIX represents nucleation, coagulation, condensation, and internal mixing of aerosols, and is coupled to chemistry in ModelE including a modified Carbon Bond Mechanism 4 (CBM-4) chemical mechanism (Gery et al., 1989; Shindell et al., 2003, 2006; Bauer et al., 2020). For aerosol water, the EQUilibrium Simplified Aerosol Model (EQSAM) thermodynamics module is employed for sulfate-, nitrate-, and ammonium-containing aerosols (Metzger et al., 2002), while the Lewis parameterization is used for sea salt (Lewis and



Schwartz, 2004). Sea salt (Jaeglé et al., 2011), mineral dust (Miller et al., 2006), dimethyl sulfide (DMS), and natural carbonaceous emissions are calculated interactively, with 10% of terpene emissions assumed to form secondary organic aerosol (Bauer et al., 2008).

2.2 PACE SPEXone and RemoTAP

95 PACE is a low-orbit, polar-orbiting, sun-synchronous satellite, with a 1:00 pm equatorial crossing time and 98.3 minute orbital period that allows for complete global coverage from SPEXone observations approximately every 25 days (Werdell et al., 2024). SPEXone, one of three Earth-observing instruments onboard PACE, is a multi-angle spectropolarimeter that gathers light from five fixed viewing angles of 0° (nadir), $\pm 20^\circ$, and $\pm 50^\circ$, with a 100 km swath (Hasekamp et al., 2019). SPEXone collects data from 385–770 nm, with 2–5 nm resolution for radiance and 10–40 nm resolution for degree of linear polarization, 100 with polarization uncertainty of 0.003 in absolute sense and radiometric uncertainty of 2% in relative sense (Hoogeveen et al., 2023).

RemoTAP, developed by SRON, uses a full physics inversion to infer column-integrated aerosol properties from SPEXone observations by iteratively fitting a linearized radiative transfer model to the measurements (Fu et al., 2025; Hasekamp et al., 2024). The version of RemoTAP used here from Fu et al. (2025) assumes three log-normal aerosol modes per column: one 105 fine mode (effective radius between 0.02–0.65 μm) and two coarse modes consisting of a spherical mode assumed to represent hydrated sea salt and a nonspherical mode assumed to represent mineral dust (effective radius between 0.7–5.0 μm each) (Lu et al., 2022; Hasekamp et al., 2024). First, for the dry portion of the fine mode, chemical composition is inferred from the retrieved effective imaginary refractive index (IRI), partitioning into AOA, BC, and a non-absorbing remainder. Because the retrieved fine mode absorption is distributed between BC and AOA, this partitioning depends directly on the IRI assumed for 110 AOA, taken here from Kirchstetter et al. (2004) and tabulated in Table S2. Since a larger assumed value requires less AOA volume to reproduce the same absorption, partitioning is less certain than the fine mode total, a point returned to in Sect. 4.1. For the coarse mode, mineral dust is assumed to produce a nonspherical scattering signal with the IRI being retrieved assuming a spectral dependence from Torres et al. (2007). It is important to note that any nonspherical coarse particle that is not mineral dust is still necessarily reported as mineral dust given the assumptions above. The complete set of refractive index (RI) values 115 used in the RemoTAP retrieval algorithm is tabulated in Table S2, with the corresponding ModelE values in Table S1, and a spectral comparison of the two in Fig. S1. Lastly, aerosol water is determined assuming a volume-weighted mean real part of the complex RI of the dry aerosol and water endmembers (van Diedenhoven et al., 2022). The mixing rule constrains volume fractions, but both uptake schemes considered in Sect. 2.4 return water per unit dry aerosol, so we form the quotient of the two fractions throughout. Writing the retrieved effective real RI of the coarse spherical mode as n_{eff} , this assumption inverts to a 120 water-to-dry volume ratio

$$w \equiv \frac{V_w}{V_d} = \frac{n_{\text{dry}} - n_{\text{eff}}}{n_{\text{eff}} - n_{\text{water}}}, \quad (1)$$



with $n_{\text{dry}} = 1.50$ and $n_{\text{water}} = 1.33$. Equation (1) diverges as n_{eff} approaches n_{water} , so the inferred water content is a strongly nonlinear function of the retrieved index, and small differences in n_{eff} correspond to large differences in w . This sensitivity is quantified in Sect. 3.4. Early validation efforts have shown strong agreement to flight campaign nephelometer data and ECMWF Reanalysis v5 (ERA5) reanalyses relative humidity (RH) data (Mens et al., 2025). RemoTAP only retrieves data in clear-sky conditions, using a neural network cloud screening approach that filters pixels with cloud fraction exceeding 5%, and further filters by goodness-of-fit ($\chi^2 < 5$ over land, $\chi^2 < 10$ over ocean) (Fu et al., 2025).

The native SPEXone L2 pixel resolution is 4.6 km along-track by 5.4 km across-track. For the $1^\circ \times 1^\circ$ gridded product used here, native pixels are re-gridded by retaining only those with a quality flag of zero (highest quality) and requiring that at least five qualifying pixels fall within each $1^\circ \times 1^\circ$ bin (Fu et al., 2025). All SPEXone results presented here use the 12-Jun-2025 version of this gridded product, which is based on the full physics inversion described above. A hybrid approach combining collaborative neural networks with this inversion has since been developed for SPEXone and other multi-angle polarimeters (Fu et al., 2026), but is not used here as the new datasets had not yet been developed over the full two year analysis period this paper covers at the time of writing. The only use made of it here is the coarse mode partitioning test of Fig. S12, which draws on 348 daily files of the hybrid gridded product spanning February 23, 2024 through February 28, 2025, the end of the hybrid record at the time of analysis, aggregated without co-location to ModelE.

2.3 Co-location methodology

To enable a like-for-like comparison, clear-sky ModelE output is co-located in time and space to SPEXone inversions. First, the model is run outputting data at the finest temporal resolution possible, at every 30 minutes, and its output in native $2^\circ \times 2.5^\circ$ resolution re-gridded to match SPEXone's $1^\circ \times 1^\circ$ product using nearest neighbor interpolation. Only clear-sky ModelE data falling within ± 15 minutes of SPEXone overpasses in the corresponding $1^\circ \times 1^\circ$ grid cells are retained. The resulting sampling is sparse and spatially uneven, and is quantified in Sect. 3.1.

Next, the 62 vertical ModelE layers are reduced to column-integrated properties for comparison to SPEXone. For aerosol composition, mass mixing ratios ($\text{kg}_{\text{aerosol}}/\text{kg}_{\text{air}}$) per layer are converted to column volume totals per surface area ($\mu\text{m}^3_{\text{aerosol}}/\mu\text{m}^2_{\text{surface}}$) using the laboratory-based species densities of Ghan et al. (2001), tabulated in Table S3. Additionally, since RemoTAP infers AOA, all ModelE OA is assumed to be absorbing to make comparison possible, with the understanding that a significant non-absorbing fraction exists (Kirchstetter et al., 2004). We therefore write AOA for the retrieval category throughout and OA for the MATRIX tracer, which the comparison treats as equivalent only under this assumption. Comparisons span from February 23, 2024, the start of SPEXone data availability, through December 31, 2025, where the end date is constrained by the availability of prescribed input files required to drive ModelE.

To ensure retrieval quality while retaining low-AOD scenes characteristic of clean marine environments, we apply a hybrid uncertainty filter that retains retrievals with either relative uncertainty below 25% or absolute uncertainty below 0.03. This approach is motivated by the well-established structure of satellite AOD retrieval errors, which contain both absolute and relative components (Levy et al., 2013; Sayer et al., 2013). For the AERONET comparison of Fig. S2, AERONET AOD at 500 nm is converted to 550 nm using the 440–870 nm Ångström exponent reported with each record, $\tau(550) = \tau(500) (550/500)^{-\alpha}$,



applied per observation before averaging; the median conversion factor is 0.896. Agreement between the two products is summarized throughout by the coefficient of determination R^2 and the normalized mean bias (NMB), defined as the summed ModelE minus SPEXone difference over the summed SPEXone values, so that positive NMB indicates ModelE exceeding SPEXone.

160 2.4 Aerosol water diagnostics

Sect. 3.4 compares the two products in terms of the water carried per unit dry sea salt, $w \equiv V_w/V_d$. Two comparisons are made, each on a different subset of the co-locations. The thermodynamic and RH comparisons use every co-located ocean cell carrying both a non-zero dry salt burden and an RH profile. The RI comparison instead requires a retrieved effective index strictly between the two endmembers, since Eq. (1) is undefined at or below the water endmember and negative above the dry one, which excludes a further 11.8%. Those lie almost entirely at or below n_{water} , so their removal acts to lower the retrieved water volume. The magnitude of this effect is quantified in Sect. 3.4.

In ModelE, water associated with sea salt tracked by MATRIX is computed at each model grid box from the RH through a growth function $g(\text{RH})$, defined as the mass of water taken up per unit mass of dry salt. The water and dry salt burdens in grid box k are $W_k = m_{\text{ss},k} g(\text{RH}_k) A_k$ and $S_k = m_{\text{ss},k} A_k$, where m_{ss} is the dry sea salt mass mixing ratio summed over the accumulation (SSA), coarse (SSC), and mixed sea salt modes (MXX), and A_k is the grid box air mass per unit area. Summing over the 62 grid boxes of the column and dividing by the appropriate bulk densities ($\rho_{\text{SS}} = 2165$ and $\rho_w = 1000 \text{ kg m}^{-3}$; Table S3) converts these to volumes per unit surface area, and the model analog of the retrieved quantity is their ratio, $w_{\text{model}} = V_w/V_d$. Because the water in each grid box scales in direct proportion to the dry salt it carries at fixed RH, multiplying the salt in every grid box by a constant multiplies the water by the same constant and leaves w unchanged. A bias in ModelE's total sea salt burden therefore does not propagate into w , leaving it sensitive only to the relative vertical distribution of the salt.

The growth function of sea salt in ModelE is based on the parameterization of Lewis and Schwartz (2004), applied within the model at each timestep and subject to a phase-state branch that sets the water to zero for crystalline particles. The recomputations in this section apply the same uptake expression offline,

$$g_L(H) \equiv \frac{m_w}{m_d} = A + \frac{B}{1-H}, \quad H > 0.45, \quad (2)$$

where H is RH as a fraction, clamped to $[0.010, 0.995]$, and the constants $A = 0.23633$ and $B = 0.58185$ are those used within ModelE, following the growth parameterization of Lewis and Schwartz (2004). Equation (2) returns a mass ratio; conversion to the w of Eq. (1) requires the factor $\rho_{\text{SS}}/\rho_w = 2.165$ from Table S3. The model's phase-state branch cannot be reproduced offline, since instantaneous fields carry no RH history: between the efflorescence RH (ERH) of sodium chloride, 0.45, and its deliquescence RH (DRH) of roughly 0.75 (Armstrong Green et al., 2024; Cziczo et al., 1997), a particle may be crystalline or deliquesced depending on its trajectory. Equation (2) therefore hydrates all salt above 0.45. We bracket introduced bias by switching uptake on at the DRH instead, the maximally effloresced limit in which every grid box below $H = 0.75$ is crystalline and holds no water. Under ModelE's own RH this lowers the column ratio by under 1%, because 98% of the



column dry salt sits above the DRH, where no efflorescence treatment has any effect. The reanalyses are drier, so the bound is weaker: 2% under MERRA-2, which holds 94% of its dry salt above the DRH, and 22% under ERA5, which holds only 72%.
190 Hysteresis is therefore negligible for the model configuration compared in Sect. 3.4, and an upper bound rather than an estimate for the reanalysis recomputations. For comparison we also apply the single-parameter hygroscopicity framework of Petters and Kreidenweis (2007), which under the Kelvin-free approximation appropriate to coarse sea salt returns the equilibrium water-to-dry volume ratio directly,

$$g_K(H) \equiv \frac{V_w}{V_d} = \kappa \frac{H}{1-H}, \quad (3)$$

195 evaluated at $\kappa = 1.10$, constrained from laboratory measurements of real inorganic sea salt at RH = 90% (Zieger et al., 2017). Since κ is defined on volumes, Eq. (3) returns w directly, while Eq. (2) must first be converted. Equation (3) treats κ as independent of RH, an approximation Petters and Kreidenweis (2007) find adequate across the subsaturated range within measurement uncertainty, so we adopt the single reported value throughout. It is applied without the ERH cutoff of Eq. (2), since it describes equilibrium growth rather than a hysteresis branch; under 1.2% of the column dry salt sits below the ERH in
200 any of the three RH fields, so imposing that cutoff would change the column ratio by at most 0.2%. Both schemes diverge as $H \rightarrow 1$, so the column water is a strongly nonlinear function of the RH in the grid boxes that carry the dry salt.

Because the growth functions depend only on RH, the ModelE water burden can be recomputed under alternative RH fields while holding the modeled dry salt distribution fixed. We exploit this to evaluate ModelE's aerosol water against two independent reanalyses in addition to its online RH. So that the three are directly comparable, RH is computed identically
205 for all of them, as the ratio of specific humidity to its saturation value, with saturation vapor pressure over liquid water at all temperatures following Murphy and Koop (2005), from each product's archived temperature, specific humidity, and pressure. For ModelE this reproduces the diagnosis the model performs internally for its tracer modules. For MERRA-2, data are taken from the three-hourly pressure-level collection M2I3NPASM at $0.5^\circ \times 0.625^\circ$ resolution (Gelaro et al., 2017), and for ERA5 from the six-hourly pressure-level product at 1° resolution (Hersbach et al., 2020). The recomputed fields reproduce each
210 product's own reported RH to within 0.01 for MERRA-2 and 0.002 for ERA5 above 0°C . Below freezing the liquid-only values are lower, as expected from the lower saturation vapor pressure over ice, though this is of little consequence here since the median column carries only 0.03% of its dry sea salt below 0°C .

Both pressure-level products terminate at 1000 hPa, leaving the lowest few tens of meters of the marine column, where a substantial fraction of the sea salt resides, below the bottom of the reported profile. Each profile is therefore extended to the
215 surface using the corresponding 2 m fields: temperature, specific humidity, and surface pressure from the hourly single-level collection M2I1NXASM for MERRA-2, and 2 m temperature, 2 m dewpoint temperature, and surface pressure from the single-level product for ERA5, with dewpoint converted to specific humidity before entering the same expression. The resulting anchor at 2 m above the surface gives every ModelE grid box a corresponding reanalysis RH value by interpolation rather than extrapolation or rescaling. Without it, 30% of the dry sea salt volume for MERRA-2 and 16% for ERA5 lies below the lowest
220 reported level. ModelE, evaluated on its own native levels, requires no such treatment.



For each co-located SPEXone and ModelE grid cell, both reanalyses are sampled at the nearest grid point in space and at the time step nearest 13:00 local solar time, matching the PACE equatorial crossing. Since ModelE is also sampled at the SPEXone overpass itself (Sect. 2.3), all three RH fields describe the same time of day. The resulting profiles are linearly interpolated in height onto the ModelE levels of that column, so all three RH fields are evaluated on a common vertical grid, and the column integrals are computed on those native levels. Separately, and only for the composite profiles of Fig. 10a–c, each column is additionally interpolated onto a fixed 125 m grid before averaging across columns; because ModelE levels sit at varying geometric heights depending on the column, this allows a composite by-level comparison. ModelE RH bias is reported as a paired difference computed column by column before compositing, and column ratios w are reported as distributions on a logarithmic axis and summarized by medians throughout, since w is positive and strongly right-skewed. Where the two products are compared as a ratio, the statistic quoted is the median of the per-cell ratio $w_{\text{model}}/w_{\text{SPEXone}}$, given with its interquartile range.

3 Results

3.1 AOD comparisons and co-location validation

To begin, we quantify how much data survives co-location, and its impact on AOD. Figure 1a shows the spatial distribution of coincident SPEXone and ModelE clear-sky data, with the highest occurrence counts per grid cell across the tropics and mid-latitudes. Sampling is sparse and spatially uneven, owing to the narrow SPEXone swath, the differing clear-sky overlaps, and reduced retrieval coverage over bright surfaces and in low light. Across the 648 days for which SPEXone files are available in this period, SPEXone reports a value in 2,406,098 cell-days, 5.7% of all grid cell and day combinations, of which 758,926, or 1.8% of all combinations, survive cloud screening. Co-located ModelE is independently clear in a comparable 763,753 cell-days, and the two coincide in 365,140, or 48.1% of the SPEXone clear-sky subset. Applying the quality filter of Sect. 2.3 leaves the 357,302 pairs on which all following analysis rests. Figure 1c,d compare the mean clear-sky AOD at 550 nm for all ModelE data and co-located ModelE data, respectively, with Fig. 1b showing the corresponding zonal means alongside SPEXone. The co-located ModelE zonal means more closely reproduce the latitudinal variations observed by SPEXone, particularly between 50° N and 75° N, indicating that the co-location procedure effectively accounts for spatial sampling bias (Schutgens et al., 2016). This improvement likely reflects episodic boreal biomass burning at these latitudes: because ModelE is driven by daily fire emissions, the co-located output captures individual smoke events coincident with SPEXone overpasses that would otherwise be diluted under monthly or annual averaging.

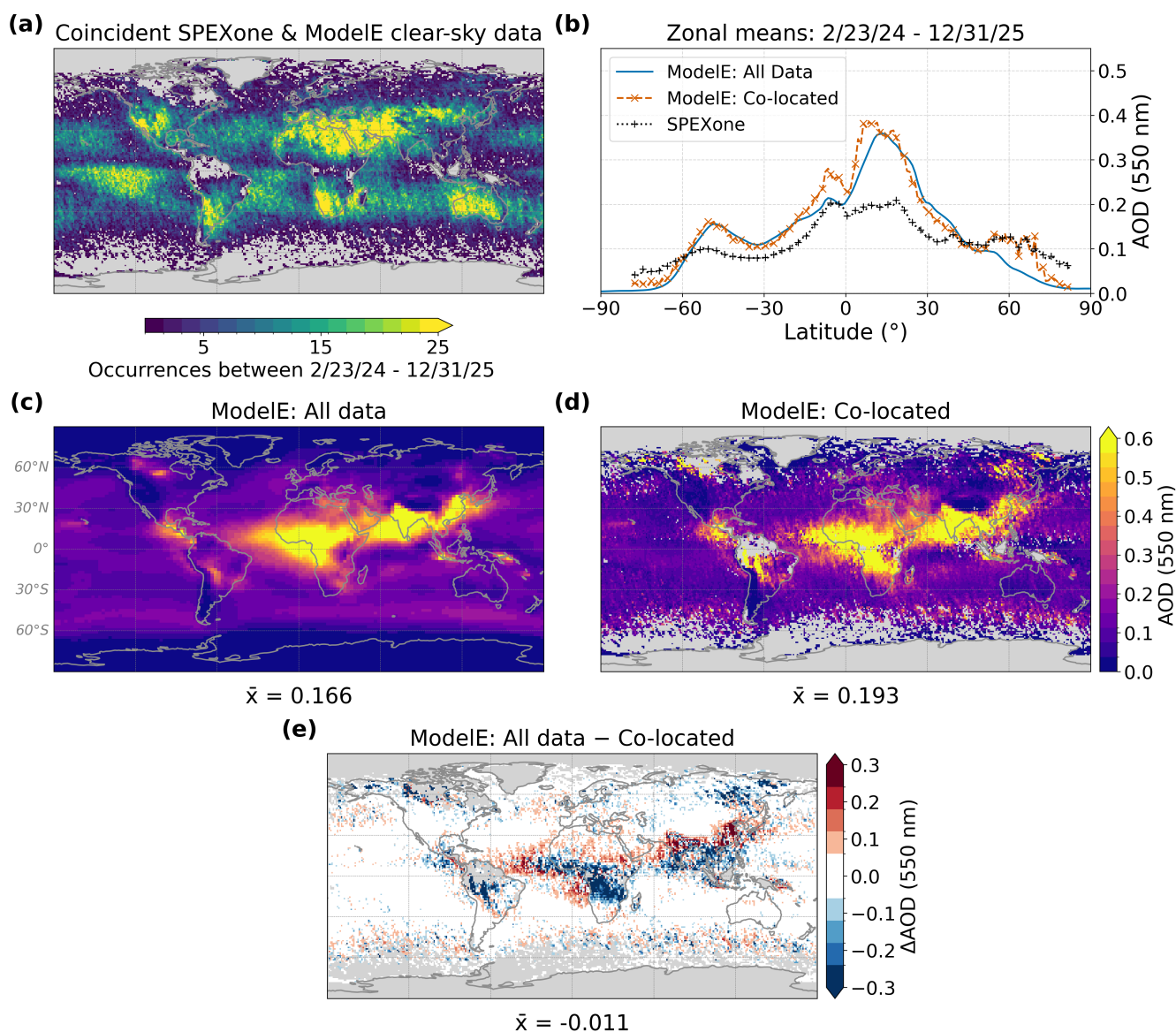


Figure 1. Spatial and temporal co-location of ModelE clear-sky data to SPEXone data from February 23, 2024 through December 31, 2025. (a) Number of coincident SPEXone and ModelE clear-sky occurrences per grid cell; the color scale saturates at 25. (b) Zonal mean aerosol optical depth (AOD) at 550 nm for all ModelE clear-sky data (solid blue), co-located ModelE (dashed orange), and SPEXone (dotted black). (c) Mean AOD at 550 nm for all ModelE clear-sky data ($\bar{x} = 0.166$). (d) Mean AOD at 550 nm for ModelE data co-located to within ± 15 minutes of a SPEXone overpass ($\bar{x} = 0.193$). (e) Difference between (c) and (d) ($\bar{x} = -0.011$), computed as the mean of per-cell differences over cells where both are defined, which is why it does not equal the difference of the means in (c) and (d). Gray regions in all map panels indicate missing data where either SPEXone did not retrieve or ModelE simulated cloudy conditions.



Co-locating ModelE to SPEXone overpasses also increases the global mean AOD by approximately 16%, from 0.166 to 0.193, consistent with SPEXone's limited coverage of poorly lit, low-aerosol polar regions, which remove low-AOD scenes from the co-located data (Chen et al., 2024). Figure 2b,e display the SPEXone AOD and its relative uncertainty (defined as the ratio of absolute uncertainty to the retrieved value) at 550 nm. The SPEXone global mean AOD is roughly 31% lower than that of co-located ModelE, with the difference in Fig. 2c concentrated over the biomass burning regions of central Africa and over South Asia. Split by surface, the deficit is larger over land (39%, 0.283 versus 0.171) than over ocean (25%, 0.161 versus 0.121). Figure 2d shows the ratio directly, and resolves structure the difference map does not: SPEXone exceeds ModelE across the high northern latitude ocean, while ModelE exceeds SPEXone over the Southern Ocean, where the absolute differences are too small to register in Fig. 2c. Part of the land difference may reflect the prescribed fire source, which remains poorly constrained, with emission factors often varying between studies by more than their own mean (van der Werf et al., 2025). As an independent check, both SPEXone and ModelE AOD are compared against co-located AERONET stations (Fig. S2). SPEXone shows less scatter and a smaller bias ($R^2 = 0.73$, RMSE = 0.108, bias = -0.028) than ModelE ($R^2 = 0.46$, RMSE = 0.268, bias = $+0.038$), but a log-log slope of 0.83 against ModelE's 0.96, underestimating the highest-AOD scenes. This remains poorer than the agreement reported for the same retrieval by Fu et al. (2025) (RMSE 0.053 over land, 0.043 over ocean) and Fu et al. (2026) (0.051 and 0.037), who average SPEXone retrievals within 20 km of a station over land and 50 km over ocean rather than the 1° cell used here. Since most AERONET sites sit near sources, the wider radius adopted here may contribute an apparent negative bias that need not indicate retrieval error. Both Fu et al. (2025) and Fu et al. (2026), however, report a small negative AOD bias over land (-0.017 and -0.015) alongside a small positive one over ocean ($+0.010$ and $+0.005$) even at their narrower radii, so the sign is likely not solely a sampling effect.

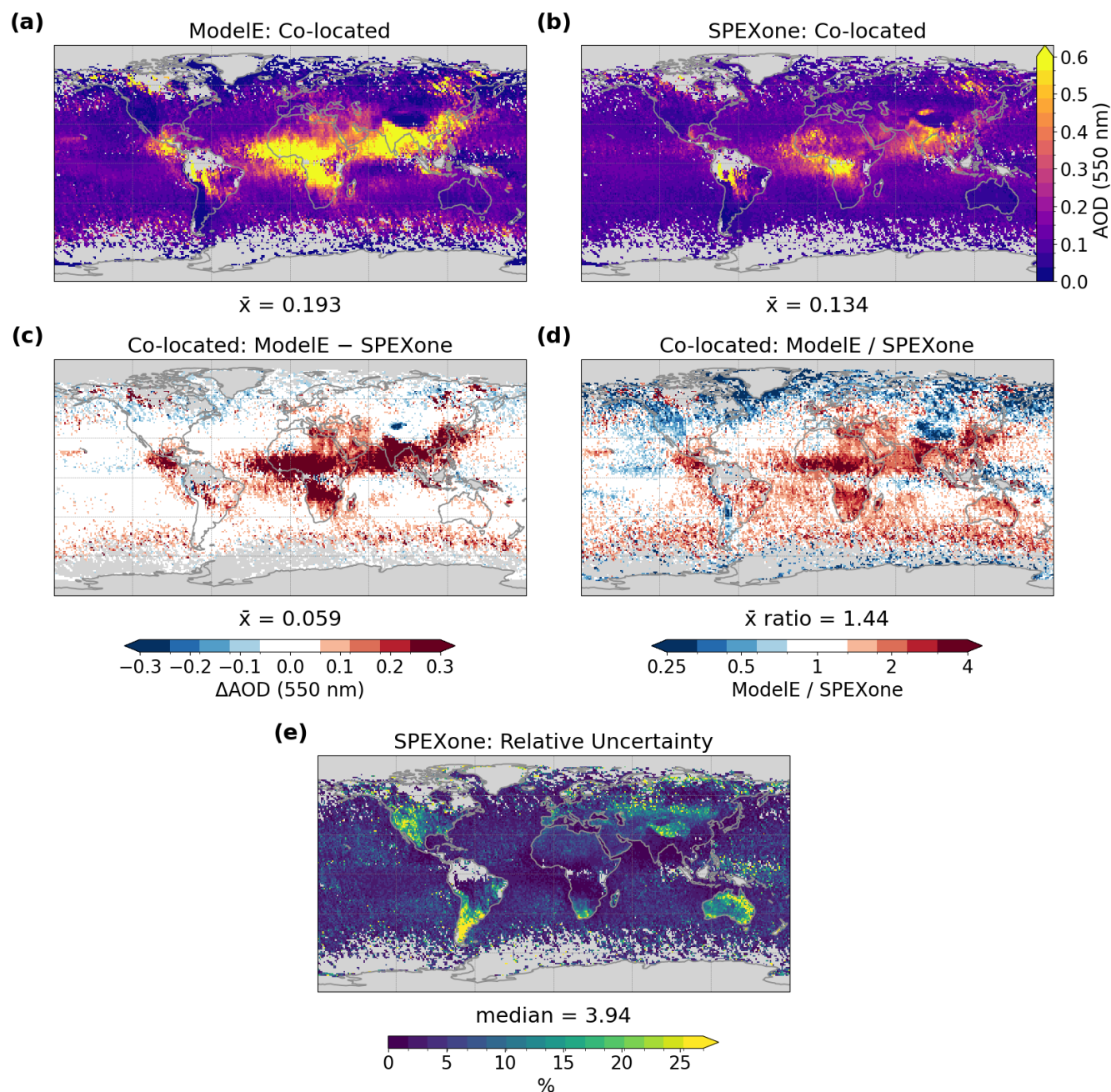


Figure 2. Co-located ModelE and SPEXone clear-sky aerosol optical depth (AOD) from February 23, 2024 through December 31, 2025. (a) Mean clear-sky AOD at 550 nm for ModelE data that co-located within ± 15 minutes of SPEXone overpass ($\bar{x} = 0.193$). (b) SPEXone AOD co-located to ModelE clear-sky data ($\bar{x} = 0.134$). (c) Their difference, ModelE – SPEXone ($\bar{x} = 0.059$). (d) Their ratio, ModelE / SPEXone ($\bar{x} = 1.44$), on a logarithmic scale. (e) SPEXone relative uncertainty (median = 3.94%) at 550 nm, highest over the western US, Australia, and southwest South America. Gray regions indicate missing data where either SPEXone did not retrieve or ModelE simulated cloudy conditions.

3.2 Global aerosol speciation comparisons

Having properly accounted for spatiotemporal co-location, we next evaluate aerosol chemical composition. Figure 3 shows the mean column burden of mineral dust, sea salt, and aerosol water for both ModelE and SPEXone alongside their differences from February 23, 2024 through December 31, 2025.

Of the three species shown, sea salt shows the closest agreement globally, with mean column burdens of 8.0×10^{-3} and $7.0 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ for ModelE and SPEXone, respectively, with both products showing peaks in the mid-latitude ocean bands consistent with strong surface wind-driven emissions (Jaeglé et al., 2011).

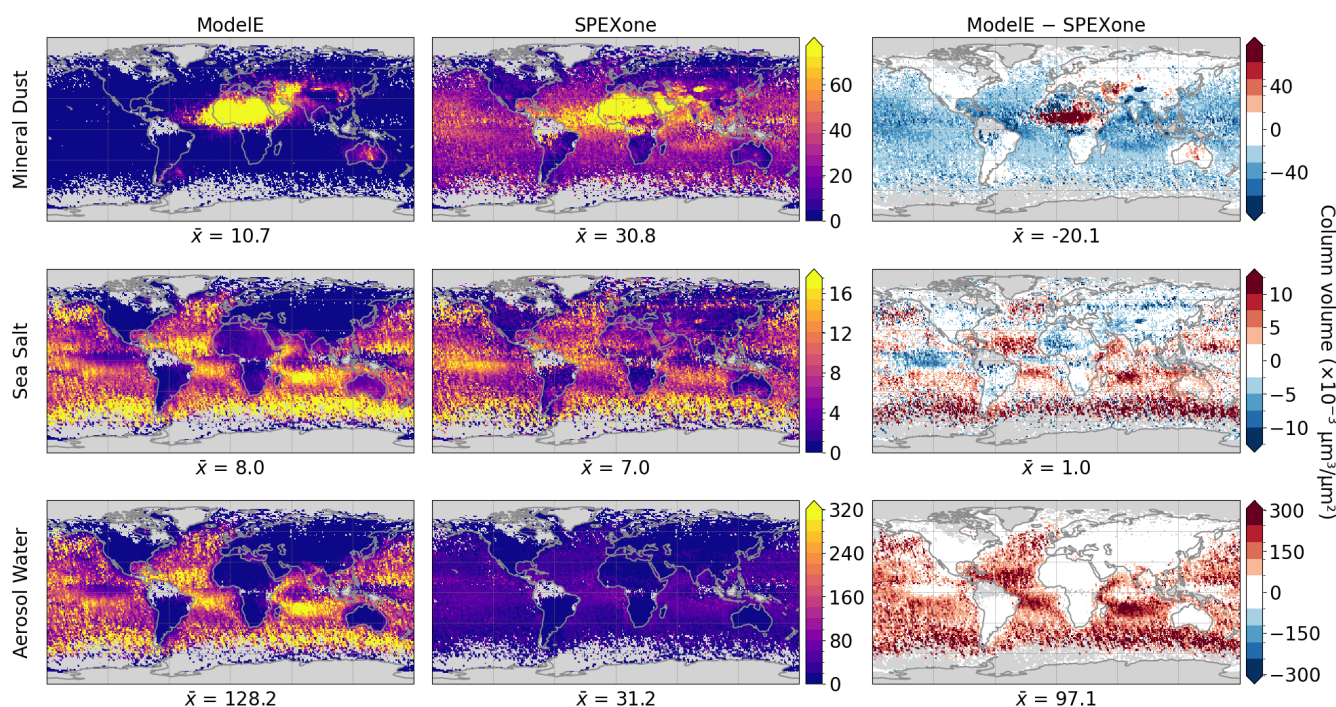


Figure 3. Mean co-located column aerosol burden ($10^{-3} \mu\text{m}^3 \text{ aerosol}/\mu\text{m}^2$) for mineral dust (top), sea salt (middle), and aerosol water (bottom) for ModelE (left), SPEXone (center), and their difference ModelE - SPEXone (right), from February 23, 2024 through December 31, 2025. Global means ($\bar{x}$) are shown below each panel. Gray regions indicate missing data where either SPEXone did not retrieve or ModelE simulated cloudy conditions.

Mineral dust, in contrast, shows an uneven disagreement. Globally SPEXone retrieves $30.8 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ against ModelE's $10.7 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$, but this conceals a sharp split between land and ocean. Over land the two products agree to within $\sim 10\%$ (SPEXone 34.0×10^{-3} versus ModelE $30.5 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$), and over the Sahara itself, where the coarse nonspherical signal is least likely to be contaminated by other species, agreement is comparably close (Fig. 9). Over the ocean, however, SPEXone exceeds ModelE by a factor of 8.4 (29.7×10^{-3} versus $3.5 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$). We defer interpretation of this split:



the regional analyses in Sect. 3.3 take up what the oceanic excess could represent, and whether long-range coarse mineral dust
280 transport is accurately represented in ModelE is returned to in Sect. 4.

Perhaps the most striking disagreement is aerosol water, where ModelE shows a global mean of $128.2 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$, more than four times SPEXone's $31.2 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$. This disagreement is almost entirely an oceanic phenomenon: over land the two products are comparable (17.7×10^{-3} and $12.6 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ for ModelE and SPEXone, within a factor of 1.4), whereas over the ocean ModelE exceeds SPEXone by a factor of 4.4 (168.2×10^{-3} versus $37.9 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$).

285 Since sea salt is a major driver of aerosol water over the ocean, one might expect differences in sea salt burden to explain this ocean excess. The two products do report comparable coarse sea salt over the ocean (within 24%), though that agreement is provisional and subject to the retrieval reattribution proposed in Sect. 4.2. The analysis in Sect. 3.4 therefore works in w , the water carried per unit dry salt, a ratio left unchanged by scaling the salt in either product.

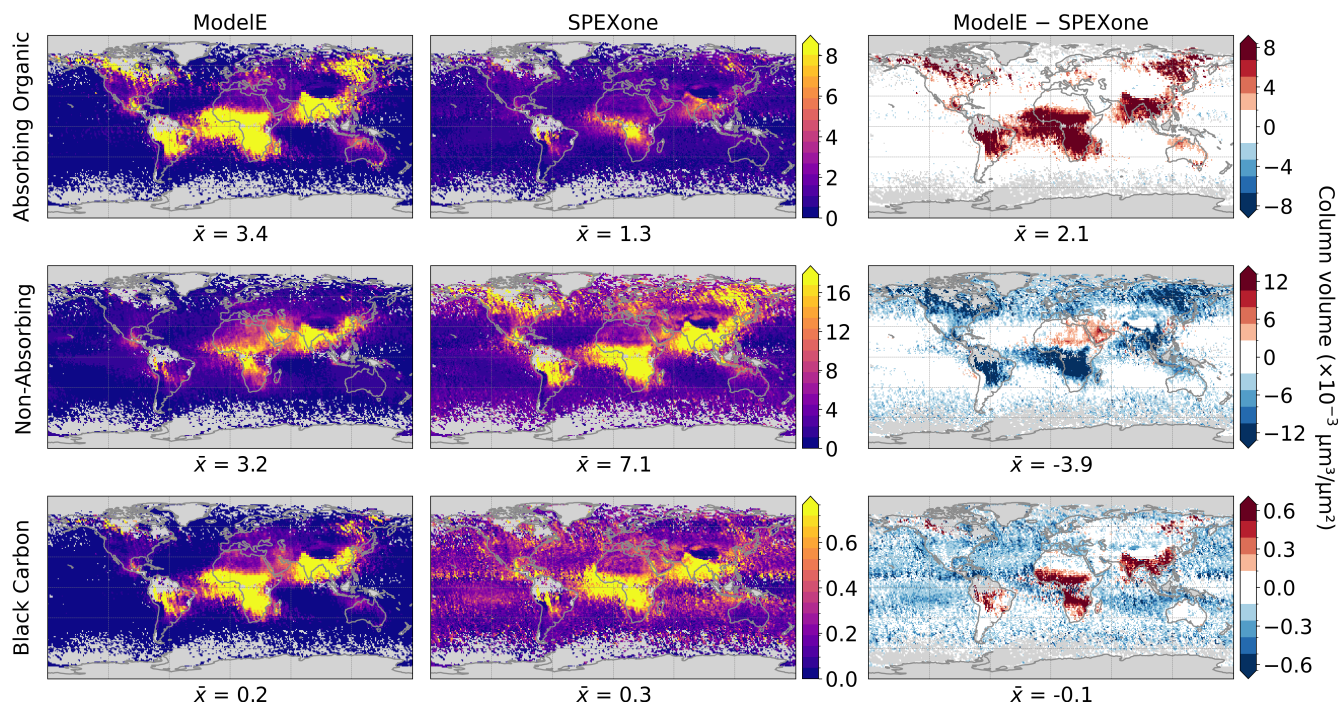


Figure 4. Same as Fig. 3 but for the fine mode: absorbing organic aerosol (AOA; top), fine non-absorbing aerosol (middle), and black carbon (BC; bottom). RemoTAP assigns only the absorbing fraction of organic aerosol to AOA and the remainder to the fine non-absorbing category, a split MATRIX does not make, so all ModelE OA is assumed absorbing here. The mirrored biases over biomass burning regions reflect the two products dividing the same organic material differently: ModelE exceeds SPEXone in AOA ($\bar{x} = 3.4$ versus $1.3 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$) but falls below it in fine non-absorbing aerosol (3.2 versus 7.1). Retrieved BC is highly sensitive to the assumed IRI and is expected to change in newer RemoTAP versions.

For the fine mode species shown in Fig. 4, AOA shows a large disagreement between PACE and ModelE, with ModelE's
 290 global mean of $3.4 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ being more than double SPEXone's $1.3 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$. The difference map shows ModelE exceeding SPEXone particularly over biomass burning and anthropogenic emissions hotspots. Fine non-absorbing aerosol shows a bias of the opposite sign in the same places, with SPEXone's global mean of $7.1 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ more than double ModelE's $3.2 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$. This mirroring likely follows from the assumption that all ModelE OA is absorbing, when in reality a significant non-absorbing fraction exists that SPEXone instead attributes to the fine non-absorbing category.
 295 Summing the three fine mode species removes this dependence, and the two products then agree to within 5% over land (Fig. S3). The assumption is necessary because ModelE cannot separate AOA within OA in the MATRIX configuration used here, although the capability exists in the One-Moment Aerosol (OMA) module (DeLessio et al., 2024).

Regardless of mirroring, SPEXone's fine non-absorbing excess is particularly pronounced over the North Pacific, North Atlantic, and Southern Ocean, where ModelE shows very little fine mode aerosol. The excess survives summation (Fig. S3), so
 300 it is not a partitioning artifact. Over the Northern Hemisphere oceans, transported biomass burning and anthropogenic aerosol



is a plausible contributor. Over the Southern Ocean, far from those sources, secondary marine aerosol formed from biogenic gas emissions including DMS, isoprene, monoterpenes, and amines is a candidate instead, having been shown to produce accumulation mode sulfate and organic particles in this environment (Russell et al., 2023). If real, this excess would have direct implications for the aerosol indirect effects in these regions, as candidate inorganic aerosols are likely efficient CCN (Fanourgakis et al., 2019).

Finally, BC shows relatively close global means of 0.2 and $0.3 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$ for ModelE and SPEXone, respectively, though SPEXone shows notably higher background values over the remote oceans. Retrieved BC volume is uncertain in these scenes for two reasons: it depends on the assumed IRI, and lensing from BC coatings can enhance apparent absorption (Liu et al., 2022). Both bear on an absorption signal that is already weak in the low-AOD remote marine scenes where the excess appears. Notably, the newer RemoTAP version retrieves less absorption over ocean (Fu et al., 2026), which would lower the retrieved BC burden there. Insufficient long-range BC transport in ModelE would produce the same excess from the model side, and the two cannot be separated here.

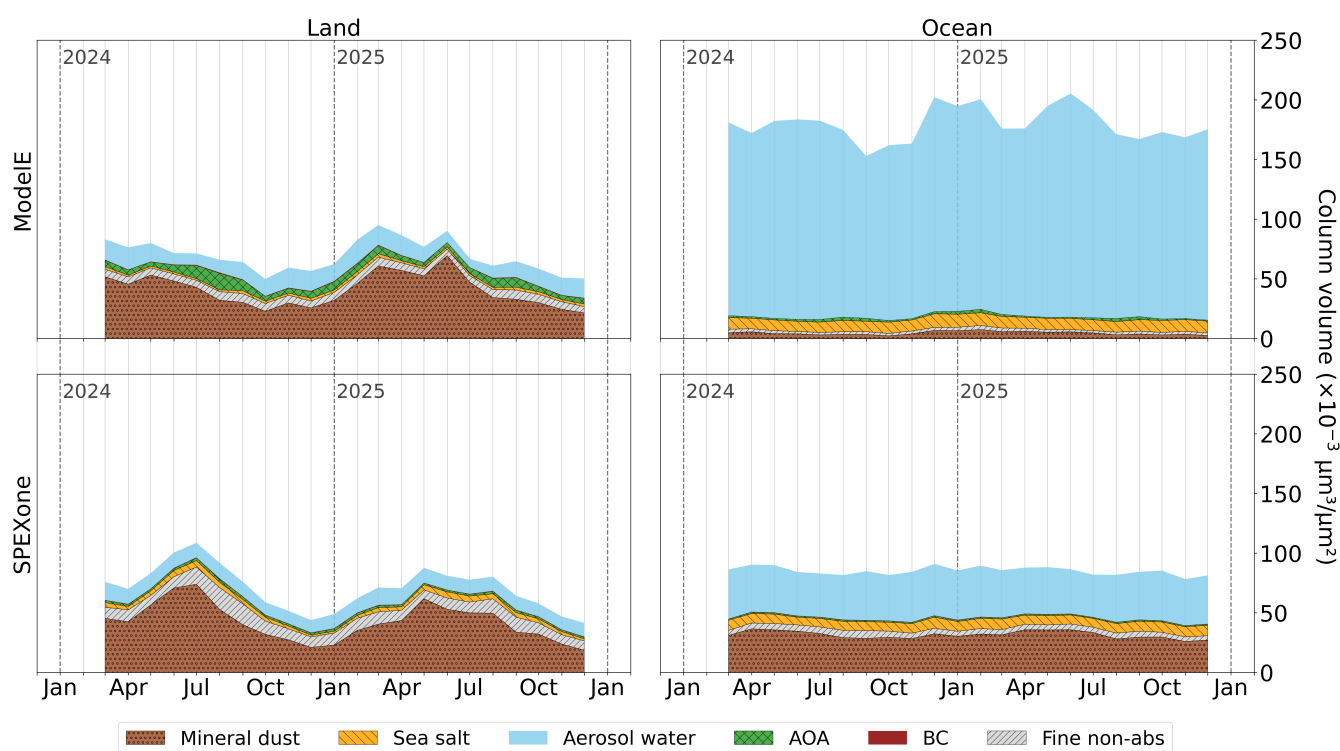


Figure 5. Monthly mean co-located column aerosol burden stacked by species for ModelE (top) and SPEXone (bottom), separated over land (left) and ocean (right), from March 2024 through December 2025. Over land the two products track one another in both magnitude and seasonality, with mineral dust dominating and peaking in boreal summer. Over ocean they diverge in composition regardless of seasonality: ModelE assigns 89.9% of its burden to aerosol water, while SPEXone splits its burden between aerosol water (46.5%) and mineral dust (37.0%). Both differences are addressed in Sects. 3.3 and 3.4.



The seasonality of these global differences is shown in Fig. 5, with monthly mean total column burden stacked by species over land and ocean for both products from March 2024 through December 2025. Over land, both products show comparable seasonal cycles, with boreal summer peaks consistent with the well-documented May through September maximum in Saharan dust storm activity (Engelstaedter et al., 2006). Over the ocean, however, the two products diverge substantially. ModeIE's total oceanic burden ($179.5 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$) is roughly 2.1 times that of SPEXone ($85.2 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$), driven almost entirely by aerosol water, which comprises 89.9% of ModeIE's oceanic amount compared to 46.5% for SPEXone. Conversely, SPEXone's oceanic mineral dust ($29.7 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$) is roughly eight times that of ModeIE ($3.5 \times 10^{-3} \mu\text{m}^3/\mu\text{m}^2$). In short, the two products disagree not only on total aerosol volume per surface area over the ocean, but also on composition: ModeIE attributes the bulk to aerosol water, while SPEXone splits it between mineral dust and aerosol water.

ModeIE also exceeds SPEXone in ocean AOD, by a factor of 1.33 against 2.1 in volume, as expected if the excess sits in coarse hydrated particles of low extinction efficiency. That ocean mean is not itself a water signal, however, since it mixes basins where water dominates AOD with those where it does not. Over the Northern Hemisphere ocean, mineral dust and transported fine mode aerosol both contribute substantially (Fig. 4), so the AOD difference there reflects other species. The Southern Ocean and southern subtropical basins come closer to permitting the reading, since hydrated sea salt is a major contributor to extinction, ModeIE exceeds SPEXone across them (Fig. 2d), and the water ratio of Sect. 3.4 is largest in the same regions (Fig. 11b,c). Even there, secondary marine fine mode aerosol contributes to extinction, and SPEXone retrieves more of it than ModeIE in these regions (Sect. 3.3), which works against the water difference rather than for it. AOD therefore points the same way without depending on the mixing rule, but it cannot isolate water from other aerosol types in each column.

As for global total volume overall, taken as area-weighted means of the co-located fields shown in Figs. 3 and 4, ModeIE is dominated by aerosol water (83.4%), followed by mineral dust (6.9%), sea salt (5.2%), AOA (2.2%), fine non-absorbing aerosol (2.1%), and BC (0.1%). SPEXone, on the other hand, divides its burden almost equally between aerosol water (40.1%) and mineral dust (39.6%), followed by fine non-absorbing aerosol (9.2%), sea salt (9.0%), AOA (1.7%), and BC (0.4%).



335 3.3 Regional aerosol speciation comparisons

To investigate regional drivers of the global differences described above, Figures 6 and 7 show mean column burden by species for SPEXone and ModelE across all longitudes at 15–35° N and 25–5° S, respectively, from February 23, 2024 through December 31, 2025. Together, these latitude bands capture many major aerosol source regions of both hemispheres, including North America, the Sahara, Central Africa, South America, and South Asia.

340 Fine mode aerosol shows good agreement in both latitude bands (NMB = –3% at 15–35° N, NMB = –14% at 25–5° S), with both products capturing peaks over well-documented source regions – over the eastern USA and South Asia in the Northern Hemisphere (Bauer et al., 2022), and over Central Africa and South America in the Southern Hemisphere (Feron et al., 2024). Sea salt also shows reasonable agreement in the Northern Hemisphere band (NMB = –1%), though ModelE shows more relative to SPEXone in the Southern Hemisphere (NMB = 30%), particularly over the South Atlantic and Indian Oceans
345 where stronger surface winds drive elevated emissions (Jaeglé et al., 2011; Erickson et al., 1986), though this comparison is provisional, since a coarse mode reattribution proposed in Sect. 4.2 would raise the retrieved sea salt burden.

Mineral dust tells a more complex story across the two latitude bands. In the Northern Hemisphere the NMB is –50%, but this bias is not distributed evenly with longitude: the two products track one another closely across the Saharan and Middle Eastern source longitudes, but their relative differences increase sharply over the Atlantic and Pacific basins on either
350 side, where SPEXone retains a substantial mineral dust burden that ModelE does not. The divergence is therefore confined to the ocean sectors of the band rather than being a uniform offset. In the Southern Hemisphere, however, the mineral dust disagreement is much larger (NMB = –92%), with SPEXone retrieving substantially more mineral dust than ModelE at nearly all longitudes, including over the remote South Atlantic and Indian Oceans far from any desert source regions. Notably, in Fig. 7, the SPEXone mineral dust signal closely tracks the sea salt signal in shape and magnitude across longitudes, and
355 increases away from desert source regions, decreasing over Australia rather than peaking there as would be physically expected for transported mineral dust. Quantitatively, at 25–5° S, SPEXone mineral dust correlates positively with ModelE sea salt ($r = +0.82$) but negatively with ModelE mineral dust ($r = -0.38$), based on 5° longitude-bin means across all surfaces. This suggests that a portion of the mineral dust inferred by RemoTAP in the Southern Hemisphere tropics stems from misattribution of nonspherical scattering signals from other nonspherical aerosols, most likely sea salt.

360 Sea salt aerosol that dries below its ERH adopts cubical or cuboidal shapes rather than remaining spherical (Kannigießer and Kahnert, 2021), and such particles depolarize backscattered light in a manner that optical models based on spherical sea salt do not reproduce (Bi et al., 2018). Critically, this is not solely a modeling expectation: airborne High Spectral Resolution Lidar (HSRL-2) measurements have observed elevated aerosol depolarization in marine layers attributable to nonspherical sea salt (Ferrare et al., 2023), establishing that a depolarizing, non-mineral-dust coarse marine population may be present
365 in the real atmosphere at the altitudes SPEXone integrates over. Because RemoTAP’s coarse mode partitioning assigns the nonspherical fraction entirely to mineral dust (Sect. 2.2), such particles are necessarily reported as mineral dust regardless of their composition. Whether the retrieved absorption can separate them is taken up in Sect. 4.2.

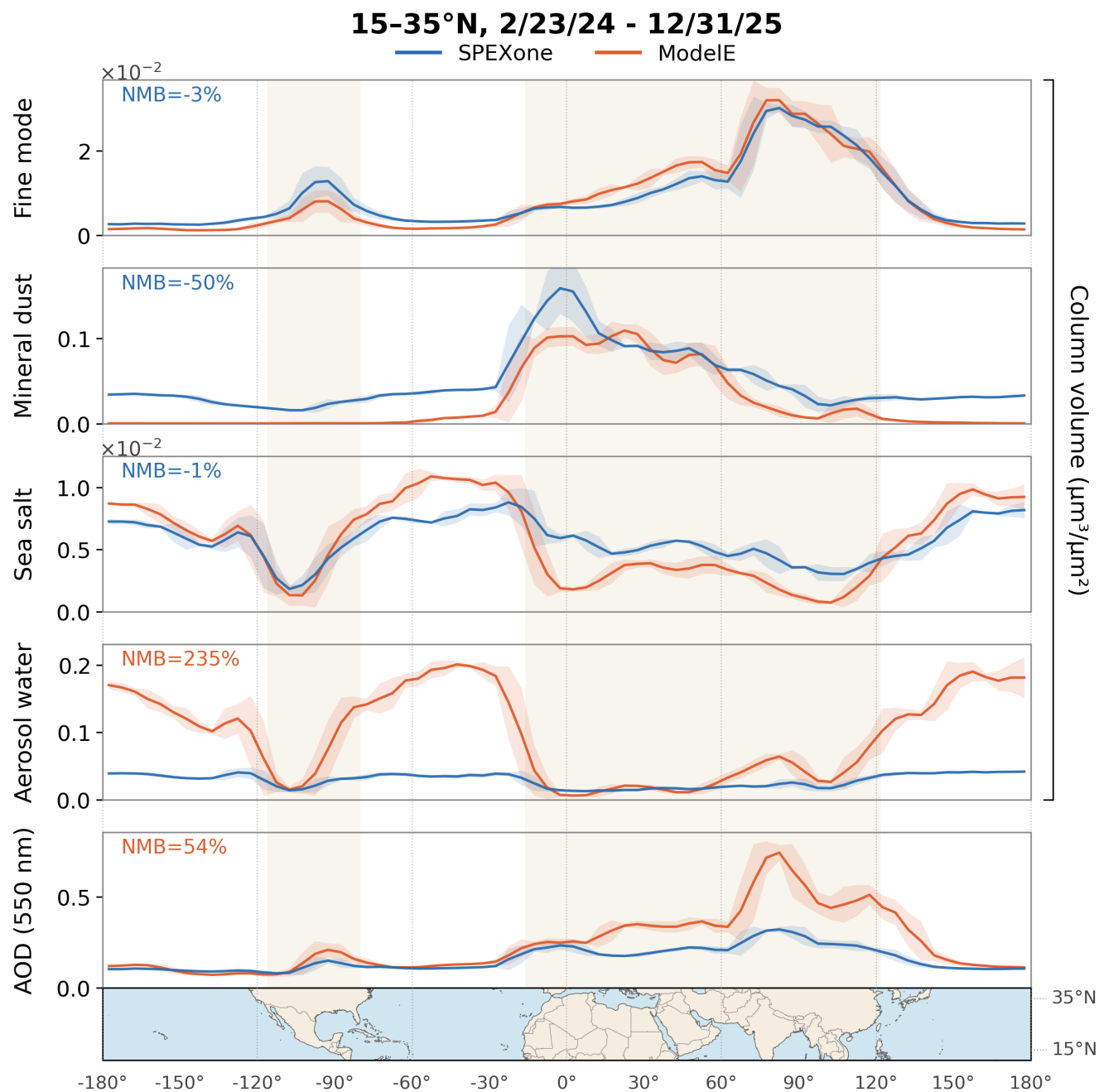


Figure 6. Longitudinal comparison of co-located column aerosol burden ($\mu\text{m}^3/\mu\text{m}^2$) for SPEXone (blue) and ModelE (orange) between 15–35° N, averaged over 5° longitude bins and smoothed with a 3-bin centered rolling mean, from February 23, 2024 through December 31, 2025. Shading indicates ± 1 rolling standard deviation. Normalized mean bias (NMB) values are shown for each row. Tan shading indicates longitudes where land area exceeds 25%.



Of all the species, aerosol water shows the largest and most consistent disagreement across both latitude bands (NMB = 235% at 15–35° N, NMB = 287% at 25–5° S), concentrated almost entirely over the ocean basins while the two products agree substantially better over land. Sea salt burdens remain comparable over the ocean (within 24%), though that comparison is subject to the nonspherical partitioning caveat of Sect. 4.2. The water difference does not rest on it: the retrieval infers water from the effective refractive index, so it is unchanged by any reattribution of coarse volume between the two species. Sect. 3.4 further analyzes this discrepancy over all ocean cells.

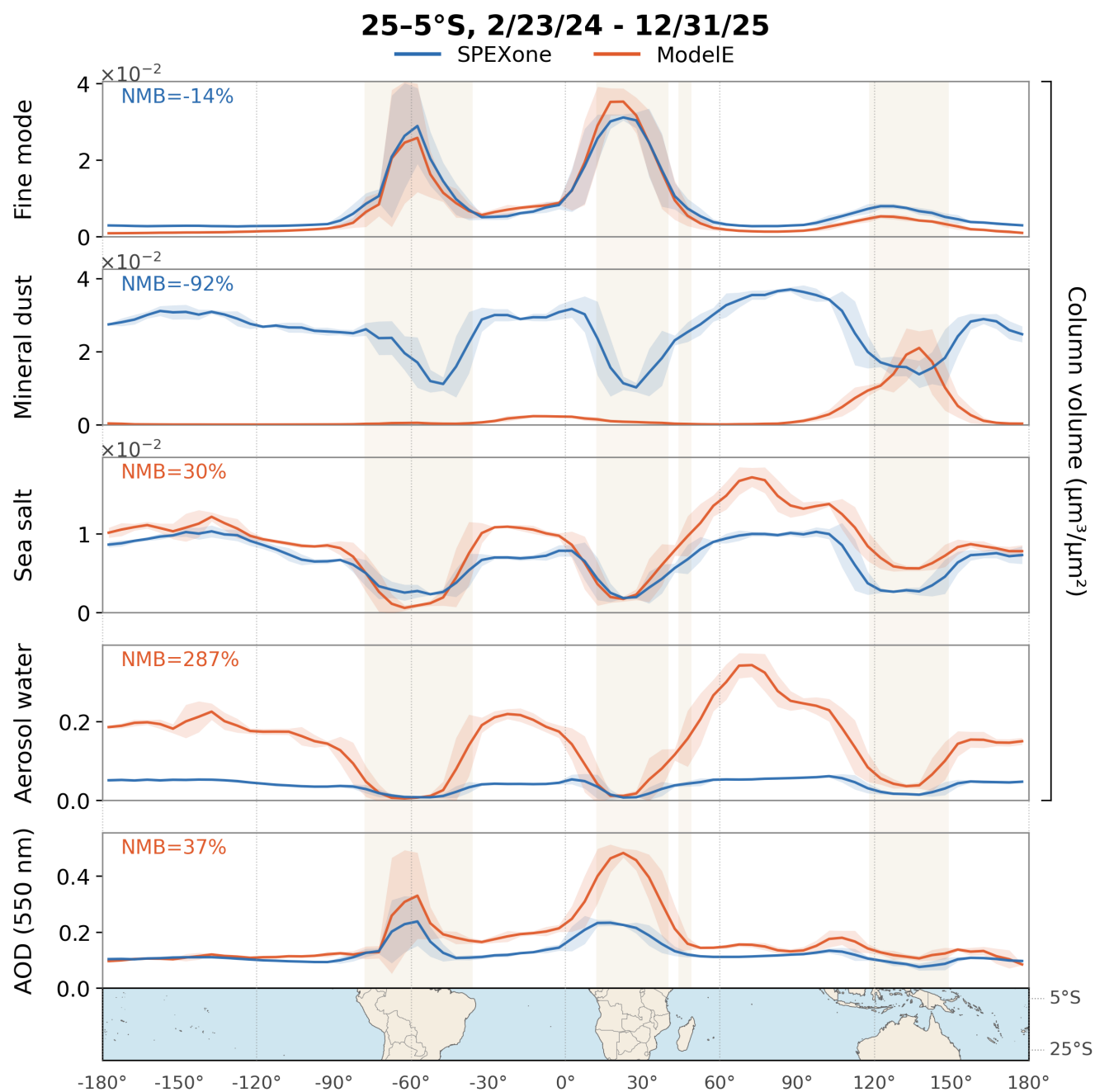


Figure 7. Same as Fig. 6 but between 25–5° S. Notably, SPEXone mineral dust covaries with sea salt strongly at this latitude band, and decreases over expected source regions, such as over Australia.



To further examine regional drivers of these differences, Fig. 8 shows time series of co-located column burden by species
375 over South America, alongside R^2 values and NMB. Both SPEXone and ModelE capture a pronounced austral spring peak in
fine mode aerosol (NMB = -16%), consistent with biomass burning in the Gran Chaco and southern Amazon (Feron et al.,
2024). Sea salt also shows reasonable agreement (NMB = -35%), with SPEXone slightly exceeding ModelE year-round, again
subject to the nonspherical partitioning caveat. Perhaps most notably, SPEXone retrieves a pronounced mineral dust signal over
South America (NMB = -95%) that peaks sharply during the austral spring burning season and closely tracks the fine mode
380 time series in both phase and shape. Previously, it has been shown that up to 30% of smoke layers over South America may
consist of nonspherical particles (Midzak et al., 2023; Liu and Mishchenko, 2020). Since central South America lacks major
proximal desert source regions, and transatlantic Saharan mineral dust transport does not peak in austral spring (Engelstaedter
et al., 2006), this covariance suggests that a portion of the mineral dust inferred by RemoTAP may stem from misattribution of
nonspherical scattering signals produced by coarse mode biomass burning smoke in this region.

385 These patterns recur across the regions in the Supplement, supporting both the aerosol water and nonspherical non-mineral-
dust findings. The biomass burning misattribution seen over South America reappears over the Eastern United States, where
SPEXone mineral dust covaries with the fine mode during an August 2024 smoke episode (Fig. S5), and over Southern Africa
during the austral dry season (Fig. S6). Over the remote oceans, where little mineral dust is expected, SPEXone retrieves
a persistent mineral dust baseline that ModelE lacks, most strikingly over the North Pacific (NMB = -99%, Fig. S7) and
390 Southern Ocean (Fig. S8), consistent with sea salt misattribution. Aerosol water disagreement peaks over these same regions
(NMB = 361% North Pacific, 706% Southern Ocean), while the Sahara and SE Asia are the regions of closest mineral dust
agreement (NMB = -10% and -24%, Figs. S9 and S10), reinforcing that the mineral dust discrepancy is smallest where
mineral dust is genuinely present and largest where it is not.

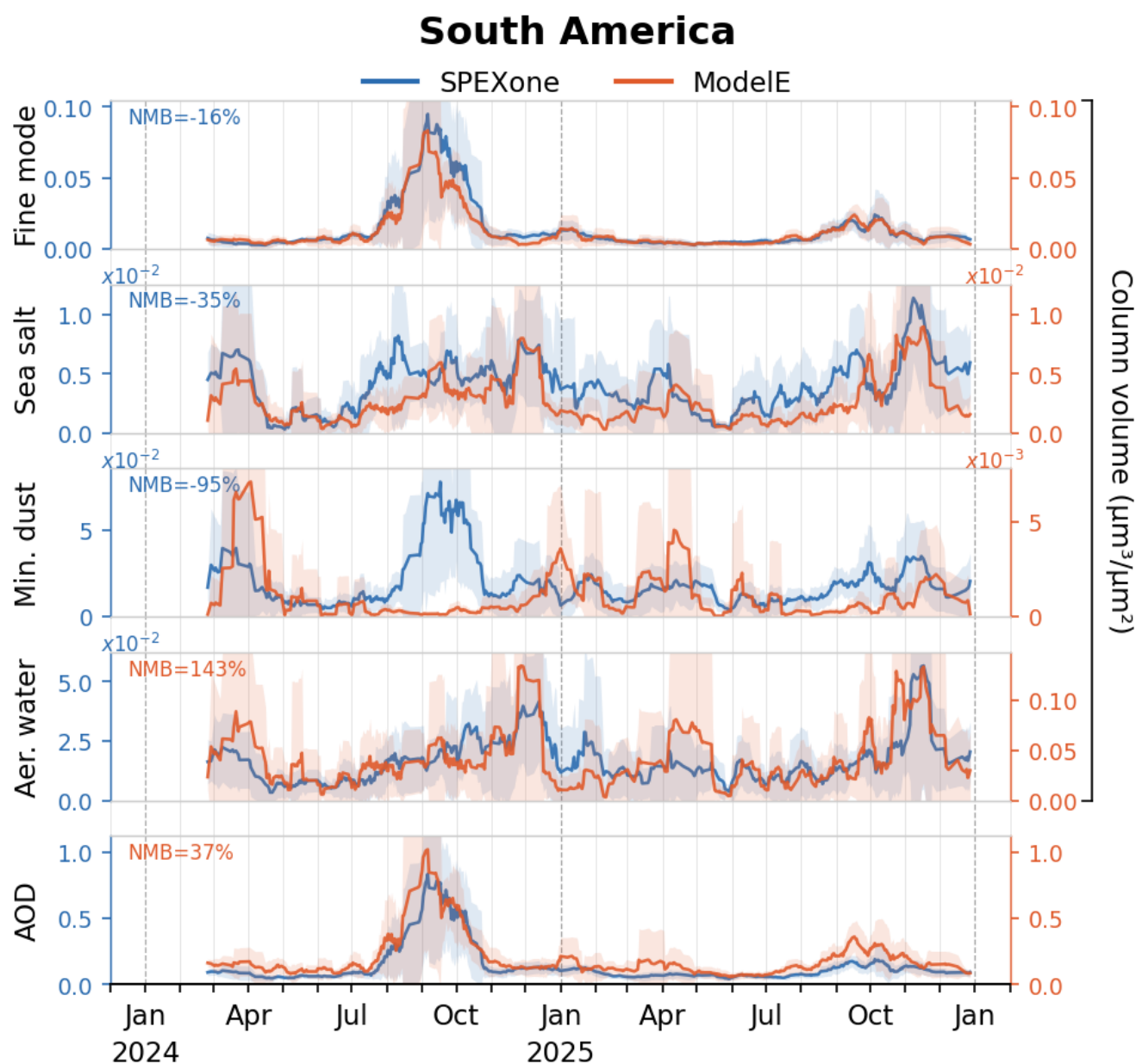


Figure 8. Mean aerosol column densities ($\mu\text{m}^3/\mu\text{m}^2$) and aerosol optical depth (AOD) at 550 nm over South America (25° S–5° N, 70° W–40° W) from SPEXone (blue) and ModelE (orange), from February 23, 2024 through December 31, 2025. Panels show, from top to bottom, fine-mode aerosol (BC + AOA + fine non-absorbing), sea salt, mineral dust, aerosol water, and total column AOD. Curves are 10-day centered rolling means with ± 1 standard deviation shading. Axes are scaled independently when the normalized mean bias (NMB) exceeds $\pm 50\%$ and shared otherwise.



Finally, Fig. 9 synthesizes several key findings from repeating the temporal analyses from Fig. 8 for each of Sahara, Eastern
395 USA, SE Asia, Southern Africa, as well as the North Pacific & Southern Oceans. Region boundaries are given in Fig. S4.
Firstly, total fine mode agreement is strongest over the biomass burning regions ($R^2 = 0.70, 0.81, 0.74$ for SE Asia, South
America, and Southern Africa), weaker over the Sahara and the Eastern USA (0.38 and 0.50), and collapses over the oceans
($R^2 = 0.08$ and 0.02 for the Southern and North Pacific Oceans, respectively). Additionally, within the fine mode, AOA and fine
non-absorbing biases mirror each other across all land regions, consistent with the assumption that all ModeIE OA is absorbing
400 when a significant non-absorbing fraction exists (Kirchstetter et al., 2004). This is confirmed by the much better statistical
scores of the total fine mode shown in the second column that eliminates this assumption, and by the total fine mode maps of
Fig. S3.

For the coarse mode, the mineral dust bias is strikingly region-dependent. Agreement is closest over the Sahara (NMB =
–10%) and SE Asia (NMB = –24%), the two regions where coarse nonspherical aerosol is most likely to genuinely be mineral
405 dust, and degrades monotonically away from them, reaching –95% over South America and –99% over the North Pacific.
Correlation follows the same ordering, with R^2 falling from 0.20 over the Sahara to near zero over the oceans: not only does
the model fail to reproduce the magnitude of the retrieved ocean mineral dust, it fails to reproduce its day-to-day variability,
which would not be expected if the two were tracking the same geophysical quantity at different amplitudes. Both features
are consistent with SPEXone overestimating mineral dust away from source regions through misattribution of nonspherical
410 scattering from sea salt. Finally, the aerosol water bias is modest over land (NMB = +23% over the Sahara, –5% over SE Asia)
but grows dramatically over the oceans (+361% over the North Pacific, +706% over the Southern Ocean). The disagreement is
therefore squarely a marine one, and is the subject of the following Sect. 3.4.

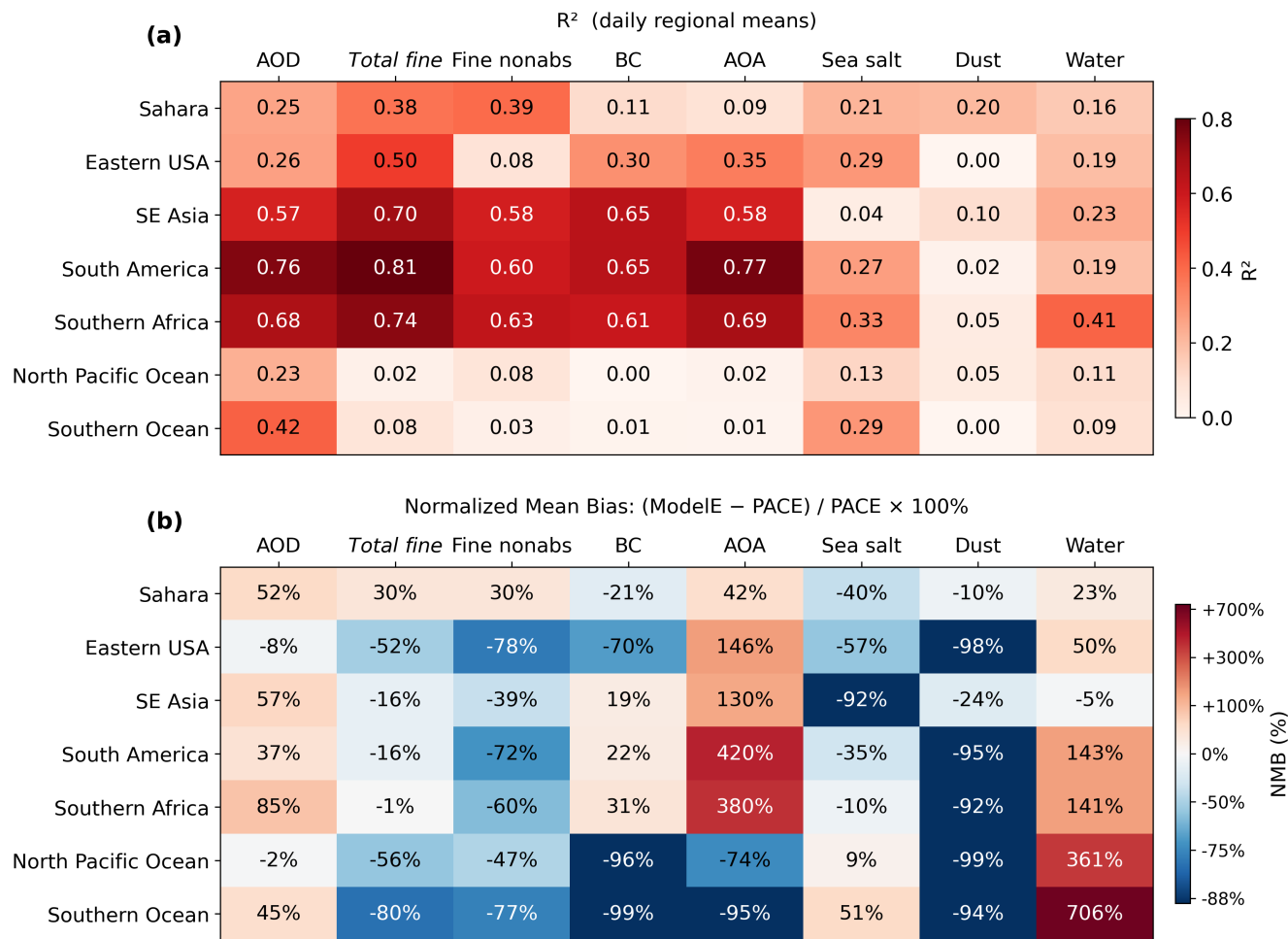


Figure 9. Heatmaps of (a) coefficient of determination (R^2) and (b) normalized mean bias (NMB, %) between ModelE and SPEXone daily regional mean aerosol properties across seven regions. Species shown are total column aerosol optical depth (AOD, 550 nm), total fine-mode (BC + AOA + fine non-absorbing), individual fine-mode components, sea salt, mineral dust, and aerosol water.

3.4 Attribution of the marine aerosol water discrepancy

The regional analyses above localize the aerosol water disagreement to marine aerosol but do not identify its cause. Here we examine it directly over ocean, using the water associated with sea salt, which dominates marine aerosol water uptake and corresponds to the coarse spherical mode from which RemoTAP infers its water product. Sea salt can account for over 90% of sea spray aerosol mass, with most of that mass in particles greater than 1 μm dry diameter (Russell et al., 2023), so treating it as the only contributor to marine aerosol water is a reasonable approximation. It holds best in the remote marine boundary layer



and degrades toward coastlines, where sulfate, nitrate, and transported pollution contribute hygroscopic volume that ModelE carries, so the model side of the comparison is a lower bound there.

Following Sect. 2.4, we work with the water-to-dry-salt volume ratio w of Eq. (1) rather than the water burden or the water volume fraction f_w , since both uptake schemes reduce to it and referencing to a dry volume makes any difference in w a difference in water alone. The cost is that w diverges as $n_{\text{eff}} \rightarrow n_{\text{water}}$, so both a small index difference and any endmember error are amplified. We therefore report the comparison in all three quantities: w for the thermodynamics, and f_w and n_{eff} for its magnitude. Two comparisons follow. The first tests whether ModelE can be brought to the retrieval thermodynamically, recomputing its water under two reanalysis RH fields and two uptake schemes while holding the modeled vertical dry salt distribution fixed. The second, since w is inferred from an RI rather than observed, places both products on Eq. (1) and compares them in index space.

The first set of experiments asks whether bias in the RH ModelE carries where the salt sits can account for the difference from SPEXone. The relevant property is not the column mean RH but rather the salt-weighted column RH,

$$\overline{\text{RH}} = \frac{\sum_k \text{RH}_k m_{\text{ss},k} A_k}{\sum_k m_{\text{ss},k} A_k}, \quad (4)$$

where RH_k is the RH of model grid box k , $m_{\text{ss},k}$ the dry sea salt mass mixing ratio of that grid box, and A_k its air mass per unit area, so that each grid box counts in proportion to the dry salt it carries and salt-free grid boxes do not contribute. Figure 10a,b confirm the offset: ModelE shows $\overline{\text{RH}} = 0.858$ against 0.792 for MERRA-2 and 0.762 for ERA5. Differenced column by column, the median excess is 5.7 and 8.6 percentage points respectively, persisting through the depth of the boundary layer and peaking near 1.7 km. It is not, however, sufficient to bring the modeled w to the retrieved value. Recomputing ModelE sea salt water under each reanalysis RH while holding the modeled dry salt distribution fixed lowers the median column ratio from 12.2 to 9.3 and 6.9 (Fig. 10d), against a SPEXone median of 3.2.

We next test the sensitivity of the modeled w to the uptake scheme itself, substituting the single-parameter framework of Petters and Kreidenweis (2007) for Eq. (2). Applying κ -Köhler at $\kappa = 1.10$, the value Zieger et al. (2017) constrain from laboratory measurements of real inorganic sea salt, lowers the median to 9.1 under model RH, 6.6 under MERRA-2, and 4.5 under ERA5. Lewis is therefore the more hygroscopic of the two. Because both schemes carry the same $H/(1-H)$ divergence, dividing it out leaves κ itself for Köhler and, for Lewis, an effective hygroscopicity

$$\kappa_{\text{eff}}(H) = \frac{\rho_{\text{SS}}}{\rho_w} \left[A + \frac{B}{1-H} \right] \frac{1-H}{H}, \quad (5)$$

the value of κ that would reproduce the Lewis uptake at that RH. Evaluated at the three $\overline{\text{RH}}$ above, $\kappa_{\text{eff}} = 1.55, 1.72$ and 1.81 (Fig. S11), against 1.10 from measurement, and the $\kappa \approx 1.4$ Petters and Kreidenweis (2007) give as an upper bound for the most hygroscopic atmospheric species. Lewis therefore takes up more water than laboratory sea salt across the relevant RH range, and more than that upper bound allows for any species, which suggests it is too hygroscopic as applied here. Still, no combination of RH source and uptake scheme examined here reproduces the retrieved w : the closest, ERA5 RH with the



450 observationally constrained κ , still exceeds it by a factor of 1.4, while the model's own configuration exceeds it by the median per-cell factor of 3.6.

Separately of note, 1.8% of ocean retrievals sit at the first-guess index $n_{\text{eff}} = 1.35$, which implies $w = 7.5$. In other words, the retrieval's own starting point is closer to ModelE than to where the retrieval converges, which drifts toward markedly drier aerosol.

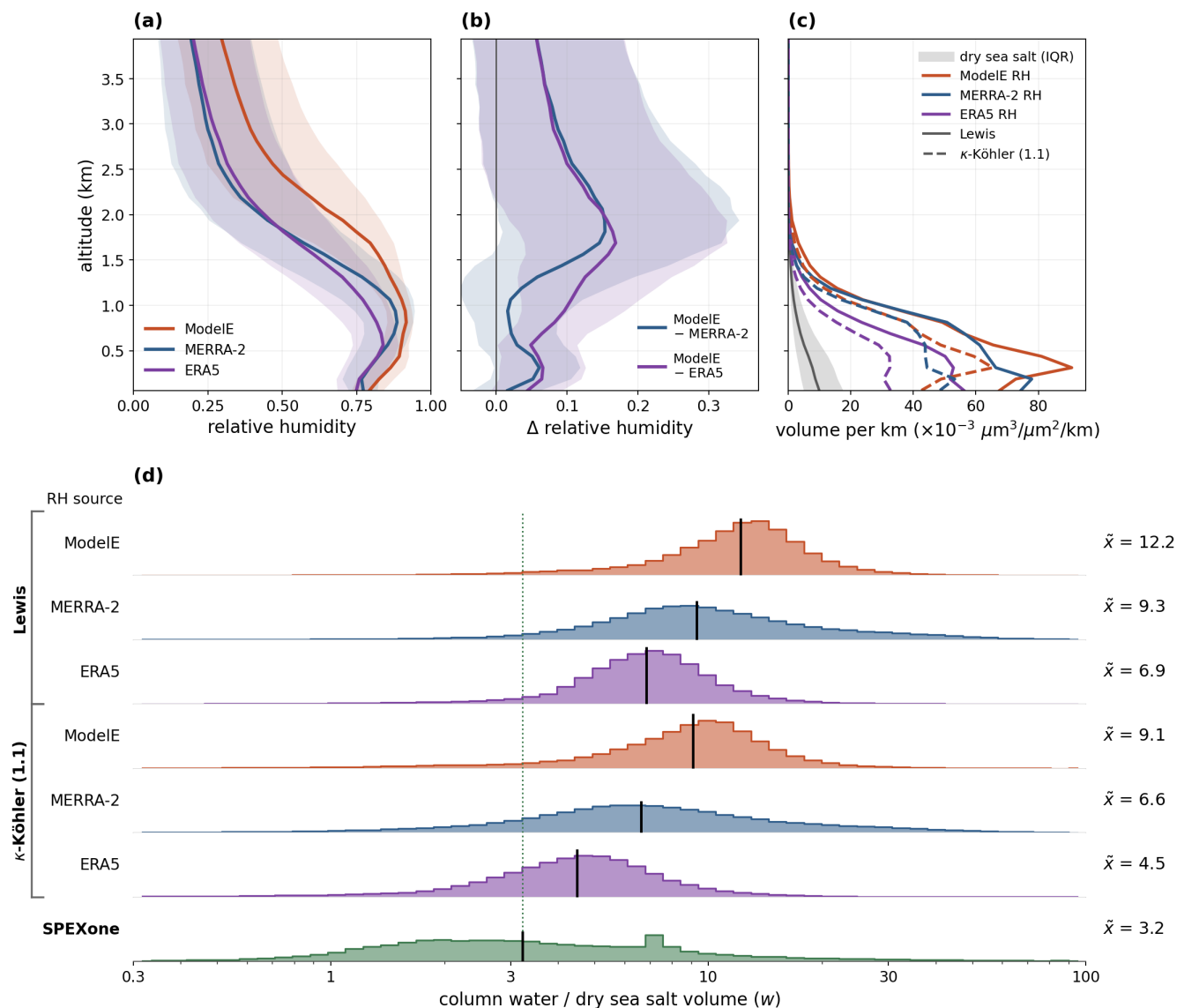


Figure 10. Co-located marine sea salt water, February 23, 2024 through December 31, 2025. Profiles in (a)–(c) are composited by computing quartiles across columns within each day and averaging across days. (a) Relative humidity (RH) for ModelE, MERRA-2, and ERA5: median (line) and interquartile range (shading). (b) ModelE minus reanalysis RH, differenced column by column before compositing; median and interquartile range. (c) Dry sea salt volume per unit height, median (gray line) and interquartile range (gray shading), and the associated water, computed under each RH field (color) and each uptake scheme (solid, Lewis; dashed, κ -Köhler with $\kappa = 1.10$); water curves show medians only. (d) Distributions of the column ratio w over all co-located ocean cells for the same six scheme–RH combinations, with the SPEXone retrieved distribution at bottom. Densities are normalized on the log axis and share a common vertical scale, so heights are comparable across rows; black bars show medians, with values also printed at right, and the dotted line marks the SPEXone median.



455 Since no thermodynamic adjustment closes the gap, the second analysis asks how large the water discrepancy is in the quantity the retrieval actually constrains. Recast in RI space, the SPEXone median over the full record is $n_{\text{eff}} = 1.370$ against ModelE's 1.343 (Fig. 11a). This difference of 0.027 spans 16% of the interval between the two endmembers, $n_{\text{dry}} - n_{\text{water}} = 0.17$, and therefore corresponds to a coarse spherical mode that is 76% water by volume in the retrieval against 93% in the model. The two also differ in spread as much as in central value: the interquartile range of n_{eff} , and hence of f_w , is 5.9 times
460 broader in SPEXone than in ModelE (Fig. 11a). The model therefore occupies a narrow band near the water endmember while the retrieval samples much of the interval between the endmembers, a difference in variability independent of the offset between the medians. Expressed as the ratio $w = f_w/f_d$ in which both uptake schemes are formulated, however, the same difference becomes a factor of 3.6 globally, taken as the median of the paired per-cell ratio, with an interquartile range of 1.5 to 7.1 (Fig. 11a). This stems from the amplification not being symmetric: the derivative of Eq. (1) evaluates to -106 at the
465 retrieved median but -1046 at the model median, so an identical index error carries roughly ten times the consequence for the model-derived water.

Also of note, this discrepancy is not a constant offset. Binned by latitude, the ratio ranges from approximately 5.5 near 40° S to 1.1 at 70° N (Fig. 11b,c). The retrieved w reaches its minimum in the Southern Ocean storm belt, where sea spray production is greatest and a well mixed marine column would be expected to be wettest, while the model peaks in the tropics.
470 Finally, the retrievals excluded at the water endmember (Sect. 2.4) bear on the size of this separation. Because f_w is linear in n_{eff} and remains defined beyond the endmembers, their effect can be evaluated exactly rather than bounded: including them all raises the retrieved median water fraction from 0.764 to 0.794, against 0.93 in the model. The exclusion therefore accounts for a small part of the separation and cannot explain it.

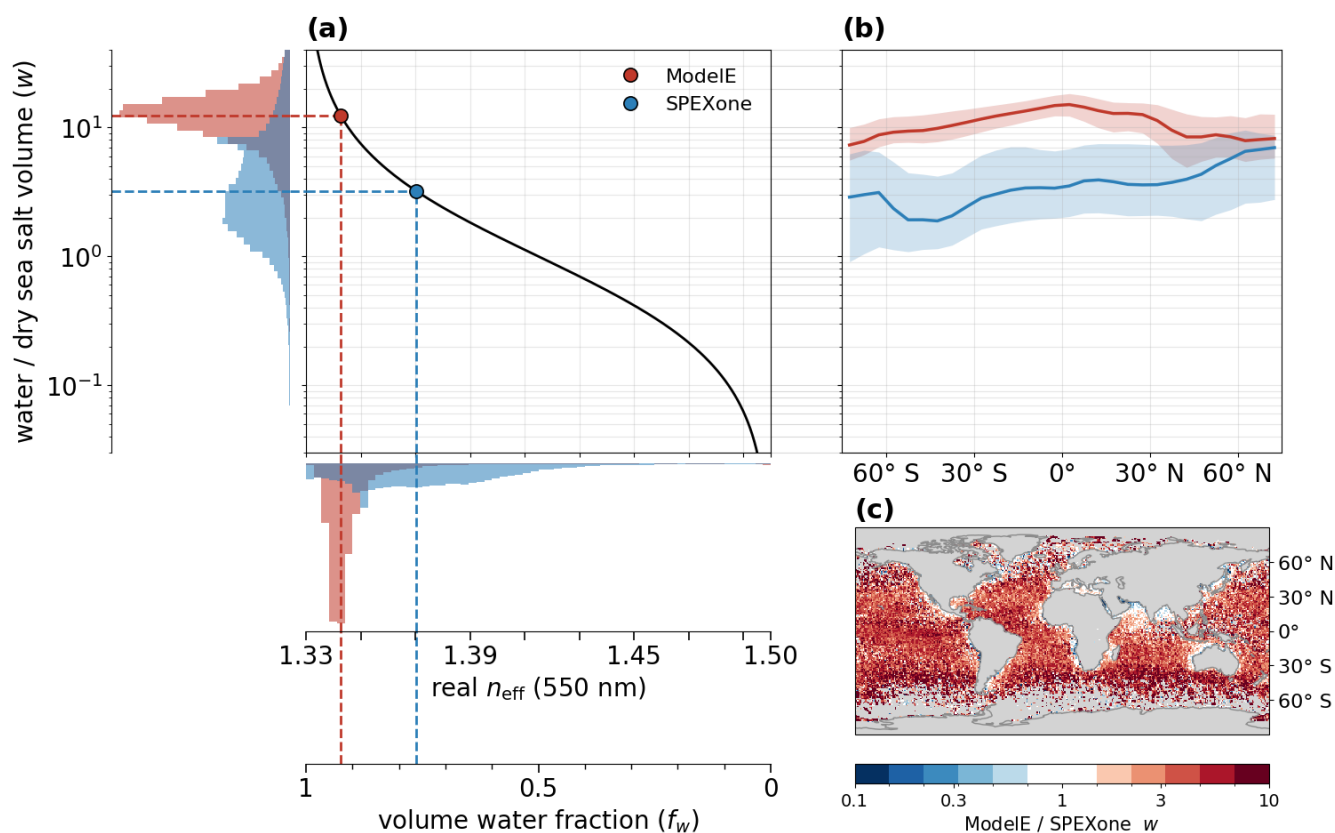


Figure 11. Marine sea salt water over co-located ocean columns recast in refractive index space, with both products placed on the single relation of Eq. (1). (a) Position of each product on Eq. (1), which relates the effective real refractive index of the coarse spherical mode at 550 nm to the water-to-dry-salt volume ratio w . The retrieved w follows directly from Eq. (1); the modeled w is computed by the Lewis parameterization, and its corresponding index is obtained by inverting the same volume-weighted mixing rule with the same endmembers ($n_{\text{dry}} = 1.50$, $n_{\text{water}} = 1.33$). The two axes are related analytically rather than independently measured, so both products lie on the curve by construction and differ only in their position along it. Median column values are marked with dotted lines. (b) Zonal mean w for both products with interquartile shading. (c) Geometric mean of the per-cell ratio of modeled to retrieved w on the 1° grid.



4 Discussion

4.1 Fine mode agreement

Fine mode agreement over land is the most robust result of this comparison, with both products resolving the same biomass burning and anthropogenic seasonal cycles. The mirror-image AOA and fine non-absorbing biases reflect assumptions on both sides: MATRIX does not separate absorbing from non-absorbing OA, so treating all ModelE OA as absorbing inflates AOA and depletes fine non-absorbing, while on the retrieval side the IRI assumed for AOA (Kirchstetter et al., 2004) is large enough so that little AOA volume is required to reproduce the retrieved absorption, leaving a larger non-absorbing remainder (Sect. 2.2, Table S2). Because the bias cancels when the two are summed (Fig. 9), the disagreement is likely one of partitioning, not of total fine mode amount. Notably, a newer RemoTAP product iteration (Fu et al., 2026) adopts a substantially smaller IRI for AOA; preliminary comparisons leave the coarse mode and water results largely unchanged while altering the AOA–non-absorbing split. The fine mode partitioning reported here is therefore specific to the 12-Jun-2025 product version, with a note that constraining that index empirically, from scenes confidently dominated by AOA, would remove the largest current ambiguity.

One fine mode difference not explained by the partitioning assumption is that SPEXone retrieves substantially more total fine mode aerosol than ModelE over the North Pacific, North Atlantic, and Southern Ocean. Over the Northern Hemisphere basins, insufficient long-range transport of biomass burning and anthropogenic aerosol in ModelE is plausible, since continental sources lie upwind. Over the Southern Ocean, far from those sources, secondary marine aerosol from biogenic gas emissions is the likelier candidate (Russell et al., 2023). The excess survives summation (Fig. S3), so neither is a partitioning artifact, and either would have implications for ACI in these regions.

4.2 Shape alone cannot identify mineral dust

The two mineral dust products agree within 10% by volume over both the Sahara and land overall, while the SPEXone excess is much larger over remote ocean basins than downwind of sources. The mechanism follows from the SPEXone assumption of Sect. 2.2: the coarse mode is partitioned based on shape alone, so every nonspherical coarse particle is reported as mineral dust regardless of composition. At least two populations plausibly violate that assumption in an influential way.

The first is sea salt. At 25–5° S retrieved mineral dust covaries with modeled sea salt ($r = +0.82$), anticovaries with modeled mineral dust ($r = -0.38$), and increases away from Australia (Fig. 7). There is a plausible physical basis for this assignment, since sea salt drying below its ERH adopts cubical habits rather than remaining spherical (Kannigießer and Kahnert, 2021; Bi et al., 2018), and instruments such as HSRL-2 have observed the resulting depolarization in marine layers directly (Ferrare et al., 2023).

The second is biomass burning smoke, which produces a smaller version of the same effect over land. SPEXone mineral dust over South America tracks the biomass burning fine mode in phase and shape, peaking when Saharan transport is at its seasonal minimum (Fig. 8). Smoke is likewise often nonspherical, with up to 30% of smoke layers over South America found to potentially contain nonspherical particles (Midzak et al., 2023; Liu and Mishchenko, 2020). Other nonspherical populations,



including pollen (Bohlmann et al., 2021) and airborne microplastic fragments and fibers (Revell et al., 2021), have been documented but are unlikely to contribute comparably at the global scale given their far smaller atmospheric burdens. We note that in the new version of the SPEXone product from Fu et al. (2026), a fine non-spherical mode has been introduced to account for fine-mode non-spherical smoke and mineral dust. This is expected to improve the representation of fine non-spherical aerosol.

The comparison also opens an unintended route to constraining coarse sea spray aerosol morphology. Shape alone cannot separate absorbing mineral dust from non-absorbing sea salt (Table S2), but the retrieved IRI can. The 12-Jun-2025 product analyzed here does not report an IRI for the coarse nonspherical mode, so we test this on the newest hybrid RemoTAP product of Fu et al. (2026), which does. Over remote ocean, far from desert sources, that index falls to its lower bound of 10^{-9} , indicating an essentially non-absorbing population. Reassigning to sea salt the volume in cells where that index falls below 10^{-7} , effectively at the bound, removes 68% of the retrieved mineral dust over ocean but only 25% over land, leaving the desert source regions and their plumes intact (Fig. S12), and would bring the eightfold oceanic mineral dust excess of Sect. 3.3 to roughly a factor of three. Being a different product version and not co-located with ModelE, this is indicative rather than a correction to our results.

Read this way, the retrieved ocean mineral dust field may be a first global-scale indication of how much coarse marine aerosol is nonspherical, and the implied magnitude is large. Nonspherical particles scatter differently from the spheres assumed for sea salt in ModelE and in most radiative transfer treatments, so the marine direct effect may be computed on the wrong phase function. Nonsphericity also implies partial or complete crystallization, and a crystallized particle carries little water. It is therefore reasonable to ask whether the two principal findings of this work share a cause, though that is not resolved here: implicating crystallization would likely require vertical depolarization measurements paired with co-located RH profiles, which SPEXone does not supply. The Atmospheric Lidar (ATLID) aboard the Earth Cloud, Aerosol and Radiation Explorer (EarthCARE), launched in May 2024, measures depolarization as a vertical profile and therefore responds to particle shape directly, and is complementary to SPEXone in this respect: pairing the two with reanalysis RH could plausibly constrain the height and RH at which the retrieved nonsphericity occurs (Donovan et al., 2024).

4.3 Marine aerosol water and the volume mixing rule

The aerosol water disagreement is the largest systematic bias identified in raw magnitude, and Sect. 3.4 turns it into a test of the model's uptake scheme. Working in the water-to-dry-salt ratio w removes any dependence on the sea salt burden, leaving a disagreement about the water held per unit dry sea salt, which is what an uptake scheme predicts. Equation (1) then translates between w and the retrieval's native units: the median per-cell factor of 3.6 in w corresponds to just 0.027 in effective RI. Most of that apparent magnitude follows from the divergence of Eq. (1) near the water endmember, since the model sits close enough to n_{water} that an identical index error propagates ten times more strongly into its water ratio than into the retrieval's. Ocean AOD is consistent with this reading and does not depend on the mixing rule, though other species in the column keep it from isolating water (Sect. 3.1).



540 Regardless of Eq. (1) sensitivity, neither RH nor the uptake scheme closes the difference on its own, though both act in the same direction. ModelE is humid by 5.7 and 8.6 percentage points in salt-weighted column RH against MERRA-2 and ERA5 respectively, yet substituting either reanalysis still leaves the model ratio two to three times the SPEXone w . Its Lewis parameterization (Lewis and Schwartz, 2004) also takes up more water than κ -Köhler does at the measured hygroscopicity, corresponding to κ_{eff} of 1.55 to 1.81 across the three RH fields against the 1.10 measured for real inorganic sea salt (Zieger et al., 2017). The closest any combination comes to matching the SPEXone w is ERA5 RH with κ -Köhler at $\kappa = 1.10$, which brings the model ratio to 4.5 against a retrieved 3.2 (Fig. 10d), accounting for much of the excess but leaving a residual factor of roughly 1.4. Two lines therefore point the same way: the model carries higher RH than either reanalysis where the salt sits, and its uptake scheme is more hygroscopic than measurement supports. Ocean AOD is consistent with both, since ModelE exceeds the retrieval in the regions where the water ratio is largest, though it cannot isolate water from other aerosol types in the column. ModelE therefore appears biased high in marine aerosol water, with its RH and uptake scheme each contributing but neither, nor the two combined, accounting for the full difference.

The retrieval side carries its own limitations. First, no uncertainty is reported on the RI, which is the quantity Eq. (1) is most sensitive to. Second, the assumed dry endmember of 1.50 sits at the low end of plausible values, and therefore could bias the inferred water fraction low. Finally, Eq. (1) presumes a homogeneous particle, whereas marine aerosol carrying an organic fraction can phase-separate and thereby alter its optical properties (Freedman, 2020). Therefore, neither product's marine aerosol water should be treated as a reference for the other, and independent constraints from in-situ nephelometry (Mens et al., 2025) and lidar remain possible routes to resolving the residual difference.

4.4 Limitations of the comparison

Neither of the coarse mode findings above establishes that the model's coarse aerosol is itself well represented. A genuine deficit in simulated long-range mineral dust transport, if present, would act in the same direction as the misattribution described here, and the evidence available cannot separate the two. Separating them will likely require the retrieved IRI to distinguish marine from mineral dust particles, which we suggest shape alone cannot do in current RemoTAP versions (Sect. 4.2). The remote marine scenes carrying the largest mineral dust excess also tend to carry weaker optical signals, which limits the polarimetric constraint on particle shape. How sensitive the retrieved nonsphericity is to diminishing AOD is the subject of ongoing evaluation.

This study has also not addressed aerosol size distributions or mixing state, both of which influence optical properties nonlinearly. On size, SPEXone reports fine mode effective radii validated against AERONET, but constructing an analogous MATRIX quantity requires decisions about which of the 16 populations to include and how to weight them vertically and across modes, a comparison we are actively developing. Mixing state raises a related difficulty. Internally mixed BC can absorb up to twice as strongly as an external mixture (Bauer et al., 2013; Liu et al., 2022; Bond and Bergstrom, 2006), so whether SPEXone's volume-weighted mixing assumptions hold in regions where BC and inorganic aerosol are internally mixed must also be quantified (Mallet et al., 2005).



A further limitation is that SPEXone retrieves column-integrated properties while ModelE-MATRIX resolves 62 layers, raising the question of whether a vertical weighting kernel is needed to build a genuine satellite analog. Forward simulating SPEXone-analog radiance and polarization from ModelE output and re-retrieving with RemoTAP would address this. A PACE forward simulator already exists (Zhai et al., 2022), and reducing the 62 model layers to a form suitable for it is ongoing work, analogous to the Cloud Feedback Model Intercomparison Project Observation Simulator Package (COSP) developed for cloud comparisons (Bodas-Salcedo et al., 2011). Beyond the vertical dimension, SPEXone's horizontal sampling is also limited, since a 100 km swath yields sparse coverage and polar regions go largely unobserved. HARP2 (Gao et al., 2023) and OCI (Werdell et al., 2024) onboard PACE offer near-daily global coverage but either lack the information content or accuracy (in current product versions) that enables species-level partitioning.

4.5 Implications and outlook

SPEXone data are relevant for ESM development because mineral dust and sea salt emissions are tuned rather than prescribed from inventories, and that tuning rests on few field measurements. Across the 15 CMIP5 models evaluated by Wu et al. (2020), global mineral dust emission varies by a factor of 4–5 at fixed size range, and current estimates of oceanic fluxes to aerosol and aerosol precursors span over two orders of magnitude (Russell et al., 2023). A retrieval trusted at the species level over ocean would therefore supply an observational target that does not currently exist, and the coarse mode refinement identified in Sect. 4.2 may be a step towards one.

Lastly, though this study treats clear-sky aerosol, the differences identified carry direct implications for ACI, since droplet number and cloud albedo depend on CCN number, size, and composition (Fanourgakis et al., 2019; Virtanen et al., 2025), and hygroscopic growth sets the wet radius at activation (Petters and Kreidenweis, 2007). Future work analyzing aerosol data from SPEXone alongside cloud data from OCI and HARP2 could help to better constrain ACI in ESMs, the dominant uncertainty in anthropogenic forcing (Forster et al., 2021).

5 Conclusions

We present the first global comparison of aerosol chemical composition between a satellite retrieval and an ESM. PACE SPEXone retrievals from the RemoTAP algorithm are co-located in time and space with NASA GISS ModelE-MATRIX simulations from February 2024 through December 2025, and column-integrated volumes of mineral dust, sea salt, BC, AOA, fine non-absorbing aerosol, and aerosol water are compared directly.

Fine mode aerosol shows the strongest agreement, with both products resolving the same seasonal cycles over land tied to biomass burning and anthropogenic sources. Its internal partitioning, however, is less certain than its total, since the mirrored AOA and fine non-absorbing biases cancel when summed and reflect assumptions on both sides rather than a deficiency in either. The coarse mode is where the products diverge the most, and where the disagreement proves most informative. Mineral dust agrees closely over land and the Sahara, yet SPEXone exceeds ModelE eightfold over ocean, with the excess largest over remote basins rather than downwind of sources. That oceanic excess covaries with modeled sea salt in the Southern



605 Hemisphere tropics, indicating that nonspherical marine aerosol is being reported as mineral dust. A smaller version of the same effect appears over land, where retrieved mineral dust tracks the biomass burning fine mode over South America, pointing to nonspherical smoke. Coarse sea salt agreement is stronger, though only provisionally: the refinement we propose, partitioning the coarse nonspherical mode on the retrieved IRI rather than shape alone, would reassign part of that population to sea salt and raise the retrieved burden. Lastly, aerosol water is the largest bias in raw magnitude, with ModelE exceeding SPEXone
610 by a median factor of 3.6 per cell, but this amounts to a difference of only 0.027 in the effective RI of the coarse spherical mode, amplified by the divergence of the mixing rule through which the retrieval infers water. A high RH bias in ModelE and an uptake scheme more hygroscopic than observations support each contribute, but together they leave a residual, motivating future work.

Read together, the two products constrain one another. SPEXone separates absorbing from non-absorbing fine aerosol and is
615 sensitive to particle shape, neither of which ModelE can observe; ModelE resolves species by origin across 62 vertical layers, which a column-integrated retrieval cannot recover. Crucially, the constraint runs in both directions. Because ModelE places mineral dust and sea salt in their established regions and seasons, it allows us to identify which SPEXone retrievals are likely misassigned, and points to the retrieved IRI as the quantity that could separate them. Applied that way, the retrieval would yield a global measure of what fraction of coarse marine aerosol is nonspherical, a quantity no model currently constrains. An ESM
620 can therefore serve as more than a target for satellite evaluation; it can be a tool for improving the retrieval itself, and a first step toward using PACE-era aerosol composition to reduce uncertainty in the aerosol direct and indirect effects.

Code and data availability. GISS ModelE3.1 simulation code is publicly available via the NASA GISS ModelE repository at <https://simplex.giss.nasa.gov/snapshots/>. The model was driven by prescribed sea surface temperatures and sea ice from HadISST1.1 (Rayner et al., 2003), NCEP reanalysis winds for nudging (Kalnay et al., 1996), anthropogenic emissions from the Community Emissions Data Sys-
625 tem (Hoesly et al., 2018), and daily fire emissions from the Global Fire Emissions Database version 5.1 (van der Werf et al., 2025). AERONET Level 1.5 data used for validation in Fig. S2 are available at <https://aeronet.gsfc.nasa.gov/>. MERRA-2 reanalysis fields are available from the NASA Goddard Earth Sciences Data and Information Services Center, using the three-hourly pressure-level collection M2I3NPASM (inst3_3d_asm_Np) and the hourly single-level collection M2I1NXASM (inst1_2d_asm_Nx) (Gelaro et al., 2017). ERA5 reanalysis fields are available from the Copernicus Climate Change Service Climate Data Store, using the ERA5 pressure-level and
630 single-level reanalysis products (Hersbach et al., 2020). SPEXone RemoTAP Level 2 retrievals, re-gridded to $1^\circ \times 1^\circ$, are publicly available at <https://public.spider.surfsara.nl/project/spexone/RemoTAP-SPEXone/> (Fu et al., 2025). The hybrid gridded product used for Fig. S12 is available at <https://public.spider.surfsara.nl/project/spexone/RemoTAP-Hybrid-SPEXone-GRIDDED/> (Fu et al., 2026). The co-located dataset and analysis scripts used to produce all figures in this study are archived at <https://doi.org/10.5281/zenodo.22037356>.

Author contributions. SF performed all analyses and prepared the manuscript with contributions from all co-authors. SEB and KT supervised
635 the work, led relevant ModelE development efforts, and contributed to the study design. OH and GF developed the RemoTAP retrieval



algorithm used to develop the SPEXone data products, provided guidance on the retrieval assumptions described in Sect. 2.2, and proposed the absorption-based coarse mode criterion tested in Sect. 4.2.

Competing interests. At least one of the co-authors is a member of the editorial board of Atmospheric Chemistry and Physics.

640 *Acknowledgements.* Climate modeling at GISS is supported by the NASA Integrated Modeling Virtual Institute (IMVI) program, and re-sources supporting this work were provided by the NASA High-End Computing (HEC) Program through the NASA Center for Climate Simulation (NCCS) at Goddard Space Flight Center. SPEXone is a public–private initiative funded by the Netherlands Space Office (NSO), the Netherlands Organisation for Scientific Research (NWO), SRON, and Airbus-NL. We thank the principal investigators (PIs) of the AERONET network and their staff for establishing and maintaining the different sites used in this investigation. The authors thank Brian Cairns for helpful discussions on satellite–model comparison, Seyed Mohammad Hassan Erfani for helpful discussions on coding infrastruc-
645 ture, Maegan DeLessio for helpful comments on AOA, and Ron Miller for helpful discussions on ModelE mineral dust.

Financial support. SF has been supported by the Future Investigators in NASA Earth and Space Science and Technology (FINESST) program (grant no. 80NSSC22K1442). OH has been supported by the European Space Agency (ESA) Airsense project (grant no. 4000142902/23/INS).



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