

S1 Supplementary tables

Tables S1 and S2 give the complete optical constants used by ModelE-MATRIX and by SPEXone-RemoTAP, and Table S3 the species densities used to convert ModelE-MATRIX mass mixing ratios to volumes.

Table S1: Complete set of real (n) and imaginary (k) refractive indices used by ModelE-MATRIX (Bauer et al., 2008), given for the six shortwave radiation bands of the GISS radiation scheme: sulfate (SU), black carbon (BC), organic aerosol (OA), mineral dust (DU), sea salt (SS), and aerosol water (H₂O). Nitrate and ammonium are assigned the same optical constants as sulfate. ModelE-MATRIX optical properties are band-averaged rather than monochromatic, so values are constant within each interval.

λ band (μm)	SU	BC	OA	DU	SS	H ₂ O
<i>Real index n</i>						
0.30–0.77	1.5281	1.8500	1.5277	1.5645	1.4500	1.3345
0.77–0.86	1.5200	1.8500	1.5200	1.5400	1.4500	1.3306
0.86–1.25	1.5079	1.8600	1.5080	1.5300	1.4500	1.3277
1.25–1.50	1.4972	1.9200	1.4972	1.5161	1.4500	1.3228
1.50–2.20	1.4831	1.9800	1.4831	1.5072	1.4500	1.3115
2.20–4.00	1.4610	2.1500	1.4610	1.4798	1.4639	1.2630
<i>Imaginary index k</i>						
0.30–0.77	1.00×10^{-7}	0.71	0.0145	2.21×10^{-3}	0	3.91×10^{-8}
0.77–0.86	1.00×10^{-7}	0.71	5.40×10^{-3}	8.00×10^{-4}	0	1.58×10^{-7}
0.86–1.25	1.64×10^{-6}	0.73	4.81×10^{-3}	1.79×10^{-3}	0	3.67×10^{-6}
1.25–1.50	1.98×10^{-5}	0.75	4.70×10^{-3}	3.78×10^{-3}	0	1.16×10^{-4}
1.50–2.20	5.17×10^{-4}	0.79	4.70×10^{-3}	5.84×10^{-3}	0	3.48×10^{-4}
2.20–4.00	0.0764	1.05	0.0762	0.0211	5.72×10^{-3}	0.0775

Table S2: Complete set of refractive indices used by SPEXone-RemoTAP in the forward model (Fu et al., 2025; Hasekamp et al., 2024), on the native wavelength grid. Three components are spectrally constant and are given once at the head of the table. The remaining wavelength-dependent quantities are the imaginary refractive index (IRI) of the organic component, which corresponds specifically to absorbing organic aerosol (AOA), and both indices of mineral dust. No sea salt or aerosol water refractive indices are carried as separate components in the RemoTAP state vector, and no real index is tabulated for black carbon.

<i>Spectrally constant components</i> (350–2500 nm, or 300–2500 nm for black carbon)							
Organic $n = 1.55$				Inorganic $n = 1.50, k = 0$		Black carbon $k = 0.700$	
λ (nm)	Organic	Mineral dust		λ (nm)	Organic	Mineral dust	
	k	n	k		k	n	k
300	–	1.53	0.0250	800	0.0005	1.53	0.0012
350	0.168	1.53	0.0170	900	0.0005	1.53	0.0012
400	0.112	1.53	0.0130	1000	0.0005	1.53	0.0010
450	0.063	1.53	0.0085	1100	0.0005	–	–
500	0.045	1.53	0.0078	1250	0.0005	1.53	0.0013
550	0.030	1.53	0.0055	1500	0.0005	1.42	0.0014
600	0.017	1.53	0.0045	1750	0.0005	1.37	0.0018
650	0.005	1.53	0.0045	2000	0.0005	1.37	0.0020
700	0.001	1.53	0.0040	2500	0.0005	1.37	0.0034
750	–	1.53	0.0040				

Table S3: Densities of aerosol species used to convert ModelE-MATRIX mass mixing ratios to column volumes per unit surface area (Ghan et al., 2001). Nitrate and ammonium are assigned the same density as sulfate. Aerosol water is taken as $1.000 \times 10^3 \text{ kg m}^{-3}$.

Aerosol species	Density (kg m^{-3})
Sulfate	1.770×10^3
Black carbon	1.700×10^3
Organic aerosol	1.000×10^3
Mineral dust	2.600×10^3
Sea salt	2.165×10^3

S2 Supplementary figures

Figures S1–S4 document the refractive indices, the AERONET validation, the total fine mode comparison, and the region boundaries used in analyses. Figures S5–S10 repeat the regional time series of Fig. 8 of the main text for the six regions not shown there. Figure S11 compares the two sea salt uptake schemes evaluated in Sect. 3.4 of the main text, and Fig. S12 the effect of partitioning the coarse nonspherical mode on absorption as well as shape.

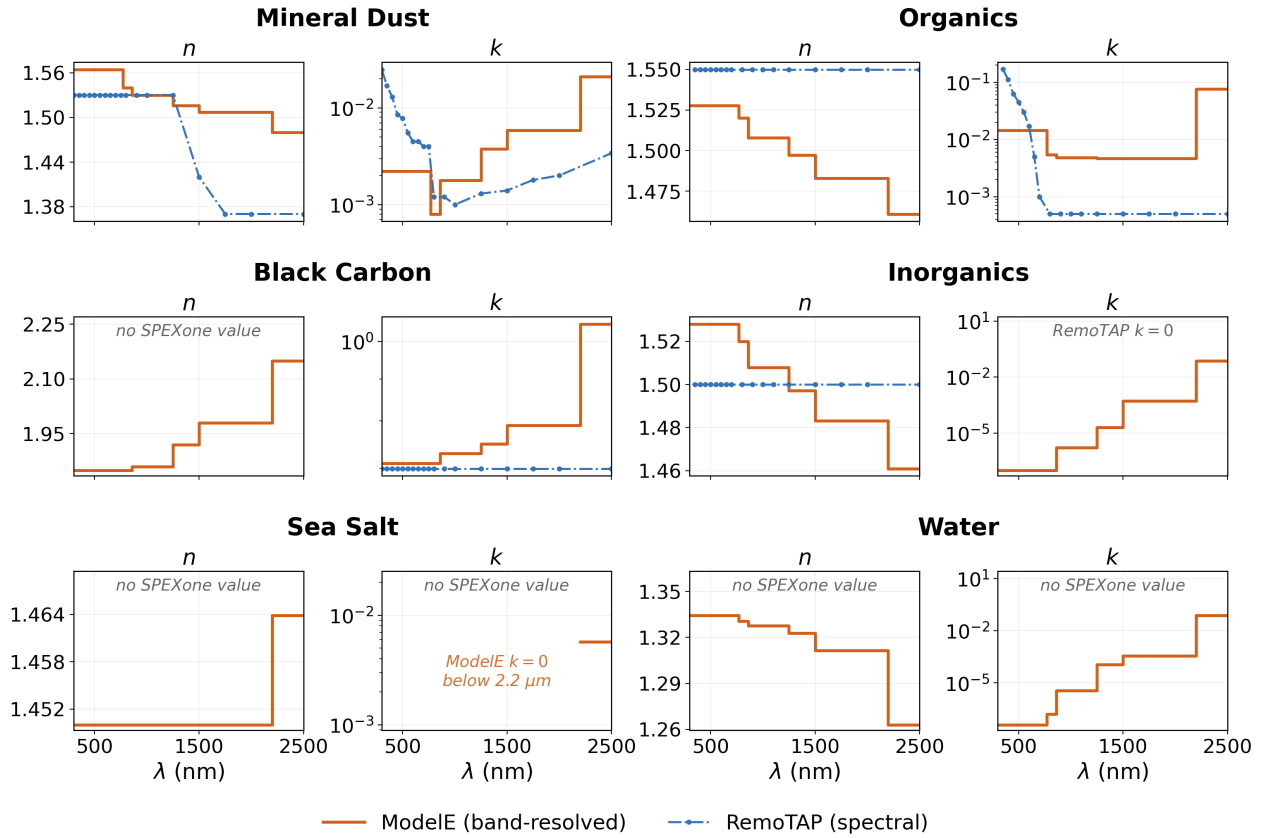


Figure S1: Spectral refractive indices of the aerosol components used to calculate optical properties in ModelE-MATRIX (Bauer et al., 2008), and for SPEXone-RemoTAP forward simulations (Hasekamp et al., 2024). ModelE-MATRIX values are band-averaged over the six shortwave intervals of the GISS radiation scheme and are therefore drawn as step functions; SPEXone-RemoTAP values are tabulated on a spectral grid. The RemoTAP organic component corresponds specifically to AOA, and the RemoTAP inorganic component is purely scattering ($k = 0$), which cannot be drawn on a logarithmic axis and is instead annotated. Panels labeled “no SPEXone value used” indicate optical constants absent from the RemoTAP a priori state vectors. ModelE-MATRIX sea salt is likewise non-absorbing below $2.2 \mu\text{m}$. The $2.2\text{--}4.0 \mu\text{m}$ ModelE-MATRIX band is shown only to 2500 nm , the long-wavelength limit of the RemoTAP tabulations.

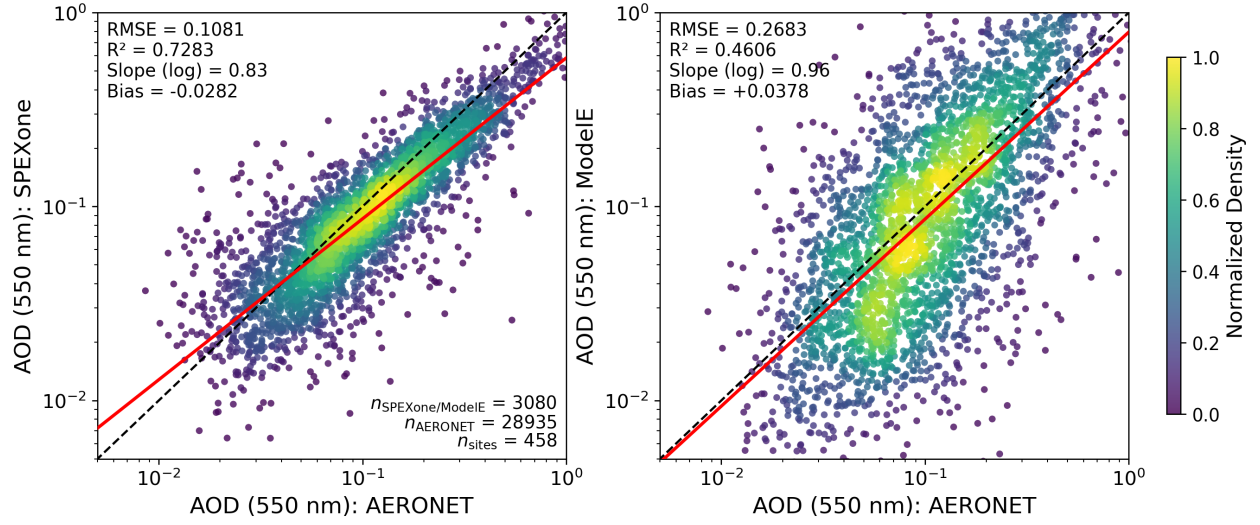


Figure S2: Density scatter plots comparing co-located AOD retrievals from SPEXone (left) and ModelE (right) against AERONET Level 1.5 AOD, all at 550 nm. AERONET reports AOD at 500 nm; each observation is converted to 550 nm using the 440–870 nm Ångström exponent reported with it, before averaging. Points are colored by normalized 2D histogram density. The dashed black line indicates the 1:1 relationship, and the solid red line shows the log-space linear regression. AERONET observations were re-gridded using nearest neighbor to the $1^\circ \times 1^\circ$ grid and temporally averaged within ± 30 minutes of SPEXone overpass time. A total of 28,935 individual AERONET measurements from 458 sites were averaged into 3,080 co-located grid-cell-day pairs spanning February 23, 2024 through December 31, 2025.

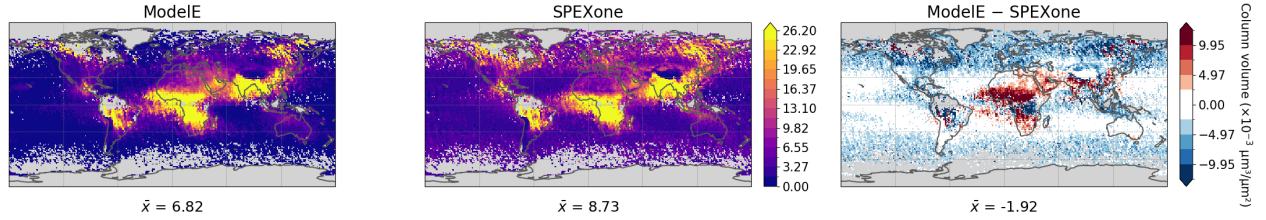


Figure S3: Total fine mode column burden ($10^{-3} \mu\text{m}^3 \text{ aerosol}/\mu\text{m}^2$), the sum of BC, AOA, and fine non-absorbing aerosol, for ModelE (left), SPEXone (center), and their difference (right), from February 23, 2024 through December 31, 2025. Summing removes the dependence on how each product splits organic aerosol between AOA and the non-absorbing remainder. The large opposing biases of Fig. 4 therefore largely offset over land (15.3 versus 16.2, NMB = -5%) but not over ocean (3.8 versus 6.1, NMB = -38%), where ModelE carries little fine mode aerosol.

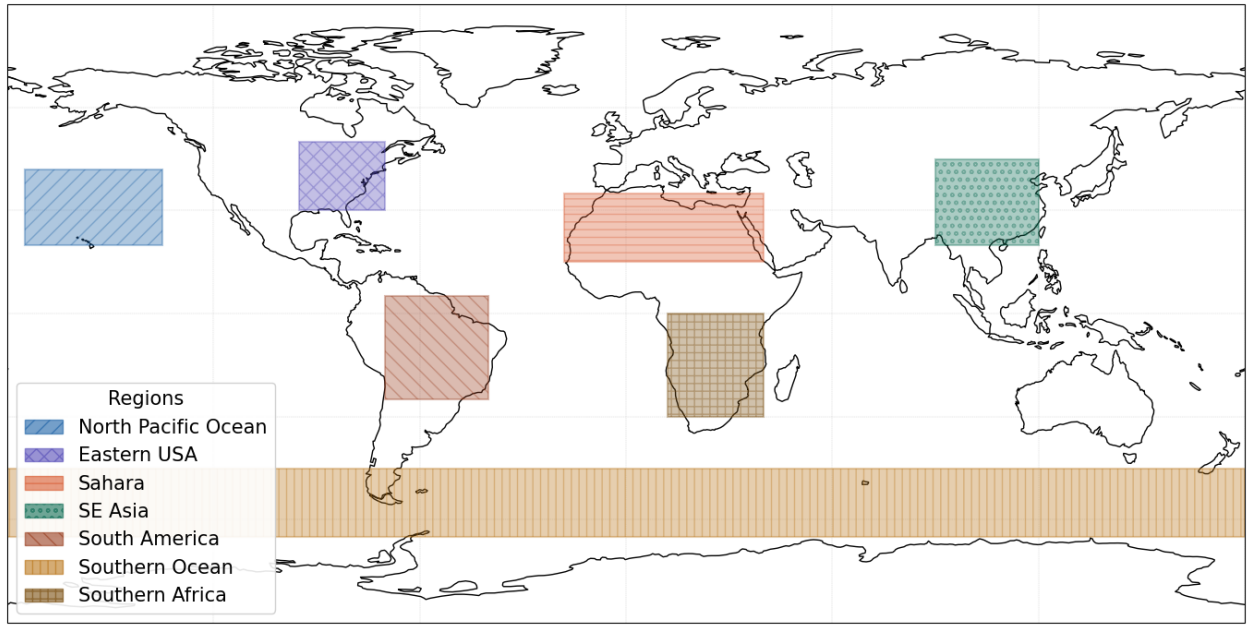


Figure S4: Region boundaries used in the analyses: North Pacific Ocean ($20\text{--}42^\circ\text{ N}$, $175\text{--}135^\circ\text{ W}$), Eastern USA ($30\text{--}50^\circ\text{ N}$, $95\text{--}70^\circ\text{ W}$), Sahara ($15\text{--}35^\circ\text{ N}$, $18^\circ\text{ W}\text{--}40^\circ\text{ E}$), SE Asia ($20\text{--}45^\circ\text{ N}$, $90\text{--}120^\circ\text{ E}$), South America ($25^\circ\text{ S}\text{--}5^\circ\text{ N}$, $70\text{--}40^\circ\text{ W}$), Southern Ocean ($65\text{--}45^\circ\text{ S}$, all longitudes), and Southern Africa ($30^\circ\text{ S}\text{--}0^\circ$, $12\text{--}40^\circ\text{ E}$).

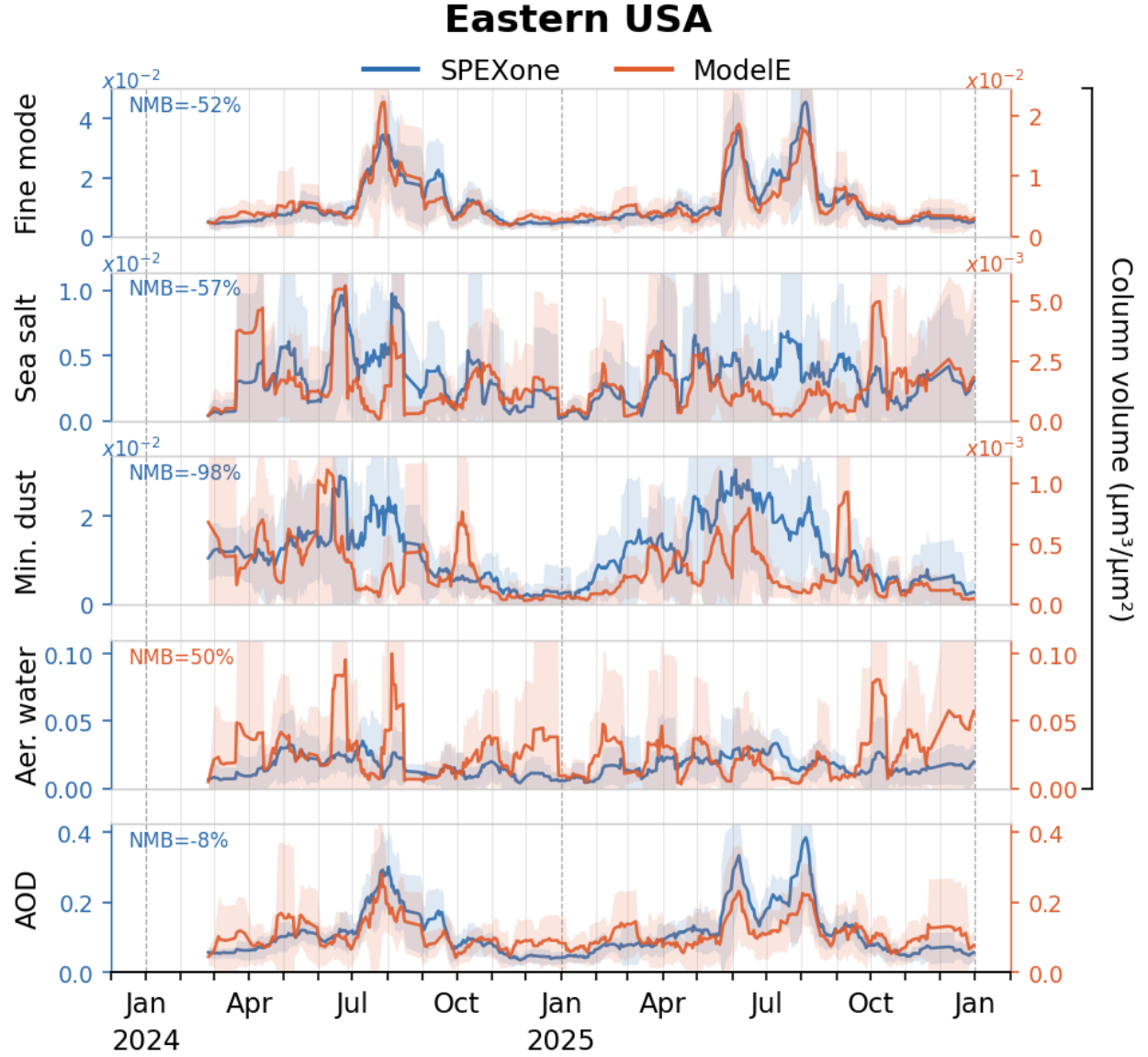


Figure S5: Same as Fig. 8 of the main text but for the Eastern United States ($30\text{--}50^\circ\text{ N}$, $95\text{--}70^\circ\text{ W}$). Fine mode aerosol covaries between the two products, though ModelE is relatively lower in magnitude ($\text{NMB} = -52\%$). Additionally, in August 2024, SPEXone mineral dust covaries with the fine mode, potentially an artifact of nonspherical wildfire smoke particles that arrived over the Eastern United States at that time, similar to the South American result.

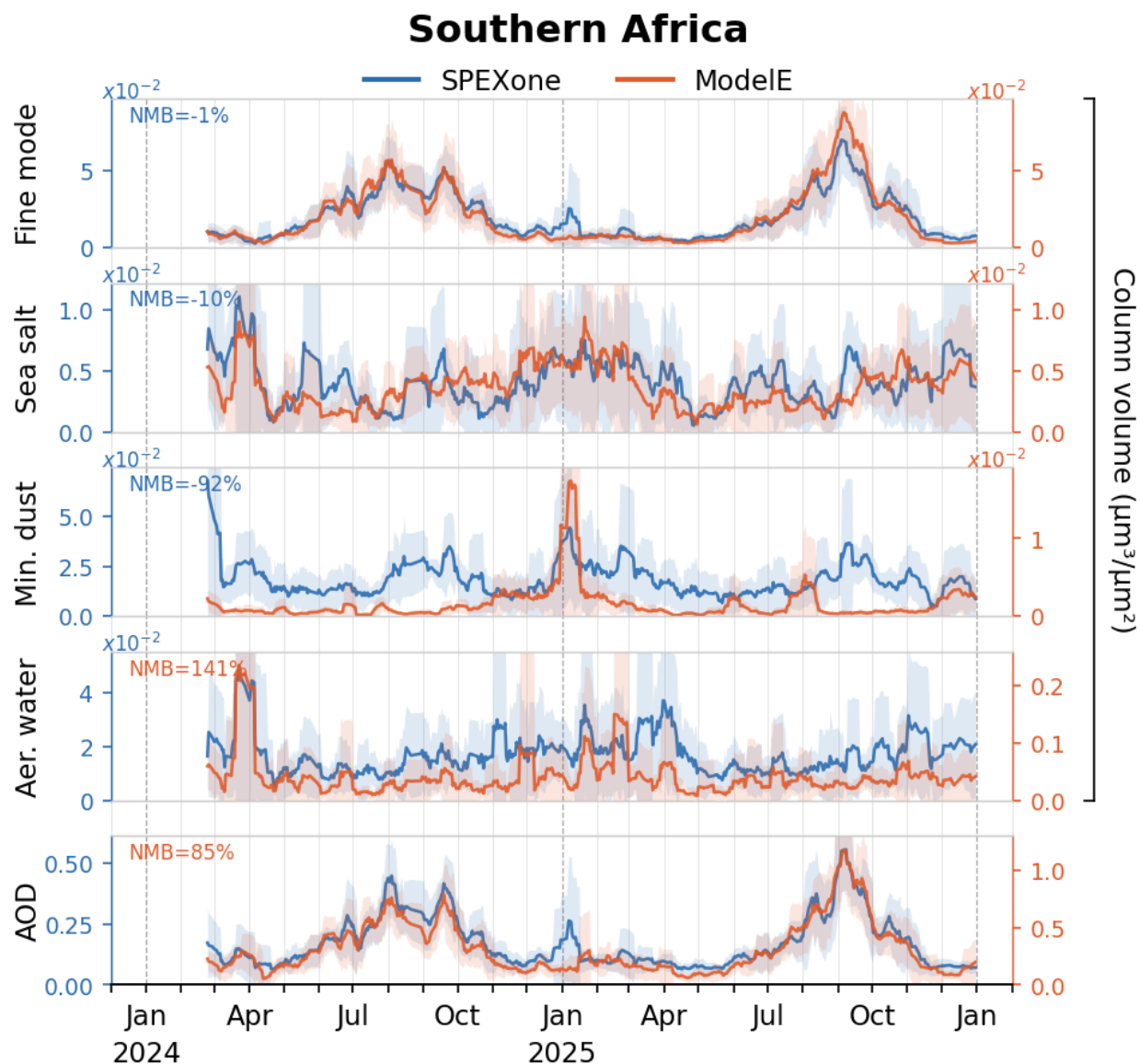


Figure S6: Same as Fig. 8 of the main text but for southern Africa (30° S–0°, 12–40° E). Both products capture the austral dry-season biomass burning maximum in fine-mode aerosol from July through October, consistent with widespread savanna and agricultural fires across sub-Saharan Africa. As in the South American case, SPEXone mineral dust covaries with fine-mode aerosol throughout the burning season in both years.

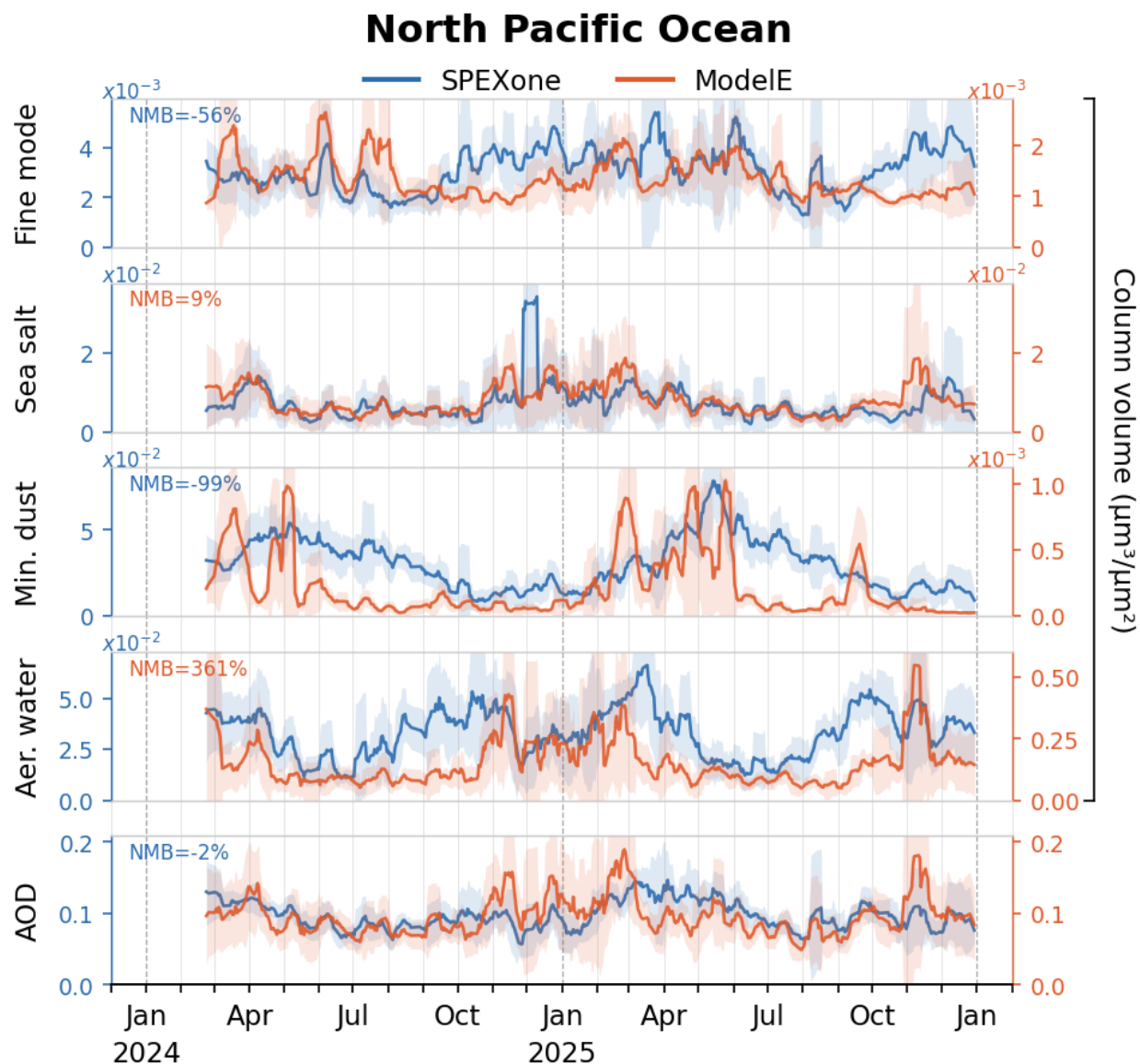


Figure S7: Same as Fig. 8 of the main text but for the North Pacific Ocean (20–42° N, 175–135° W). SPEXone mineral dust exceeds ModelE by nearly two orders of magnitude (NMB = –99%), maintaining a persistent baseline over the remote ocean where ModelE simulates negligible mineral dust. Aerosol water shows the second-largest disagreement (NMB = 361%), consistent with the over-ocean findings in the main text.

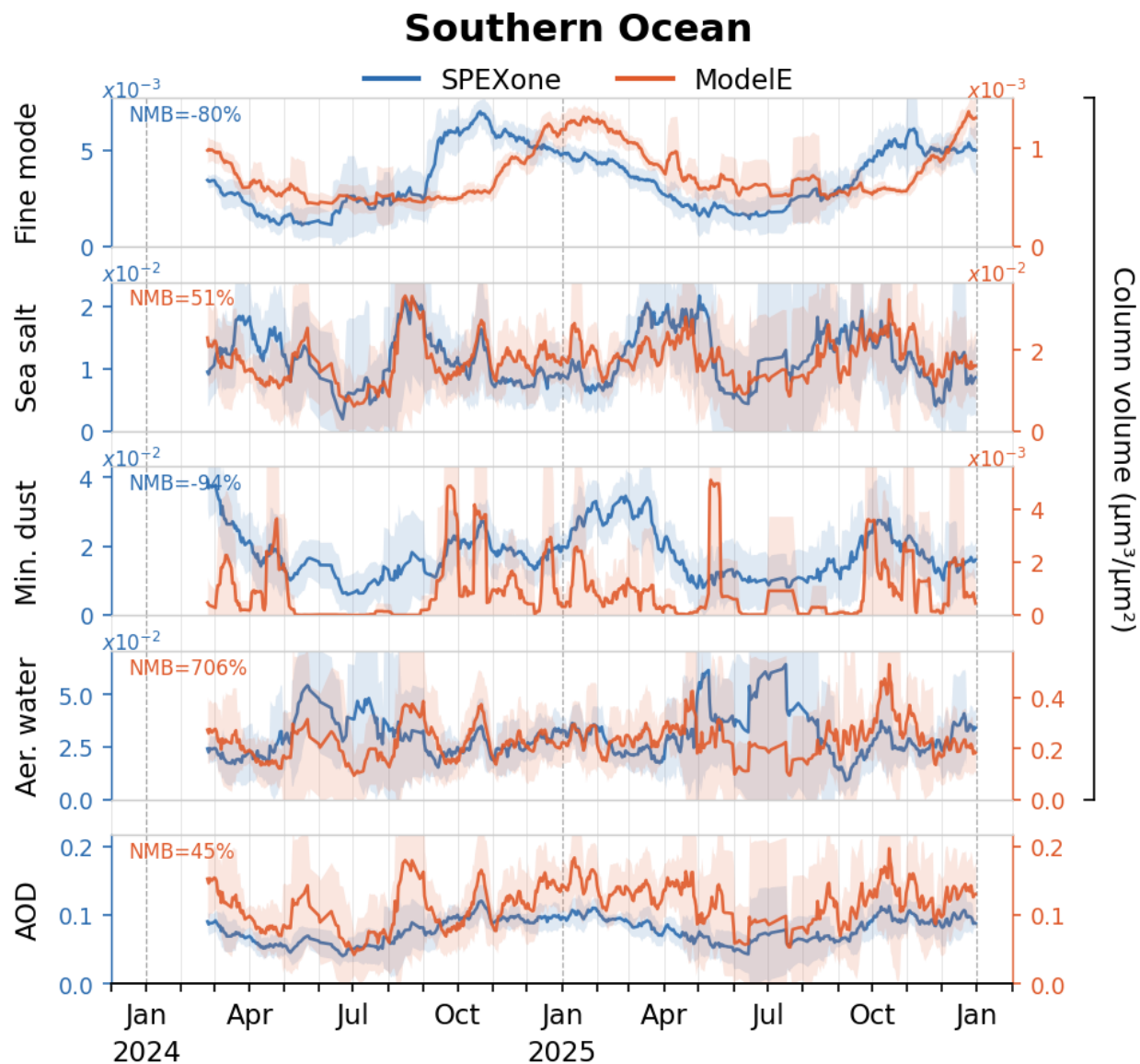


Figure S8: Same as Fig. 8 of the main text but for the Southern Ocean (65–45° S, all longitudes). Aerosol water shows the largest disagreement of any region (NMB = 706%). SPEXone mineral dust maintains a persistent baseline despite negligible expected mineral dust at these latitudes, consistent with the potential sea salt misattribution discussed in Sects. 3.3 and 4.2 of the main text.

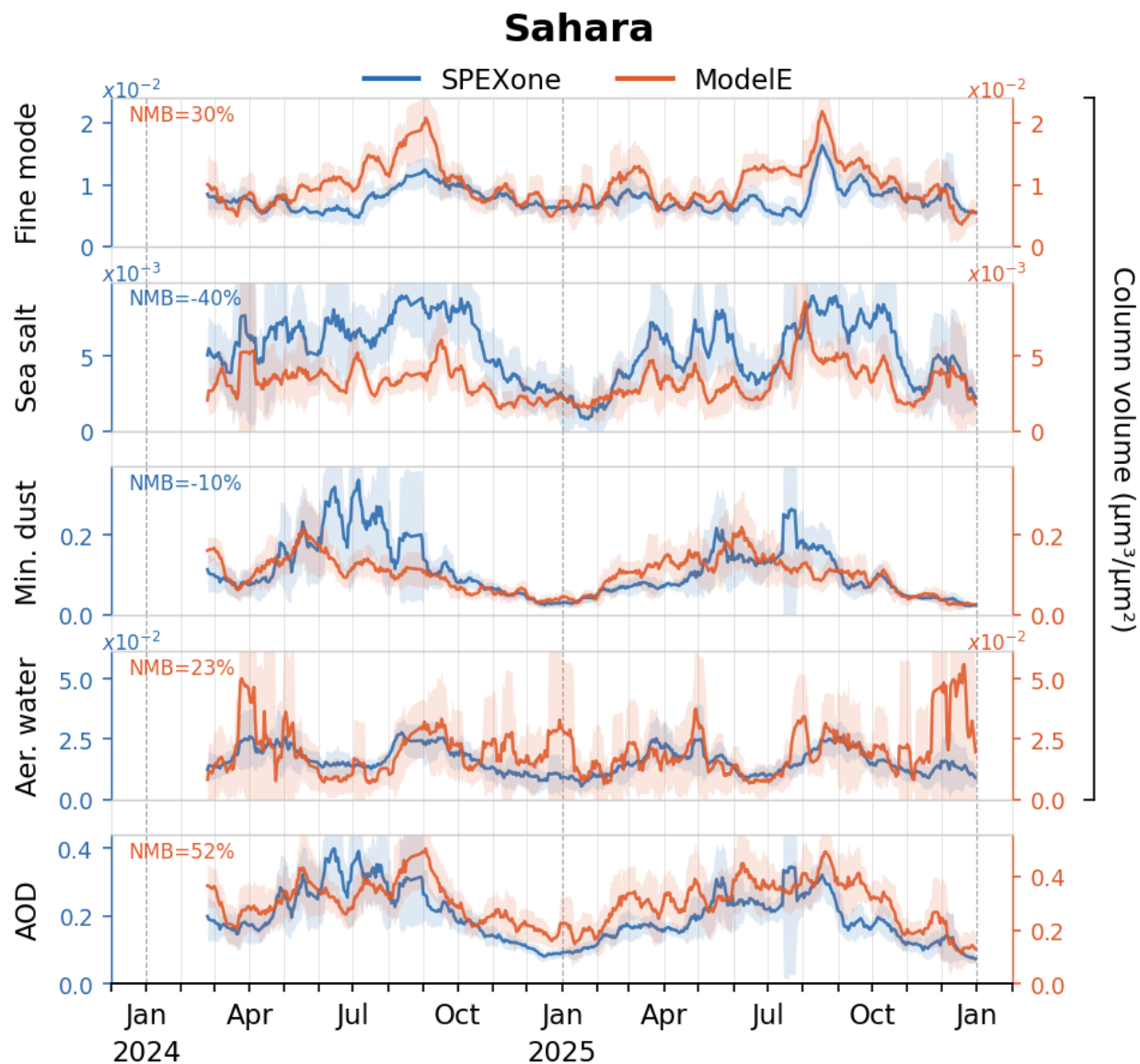


Figure S9: Same as Fig. 8 of the main text but for the Sahara ($15\text{--}35^\circ\text{ N}$, $18^\circ\text{ W--}40^\circ\text{ E}$). Mineral dust agreement is the closest of any region ($\text{NMB} = -10\%$), with both products capturing the boreal summer maximum in mineral dust storm activity. Fine mode agreement is moderate, with both products showing elevated fine mode aerosol during the boreal summer months.

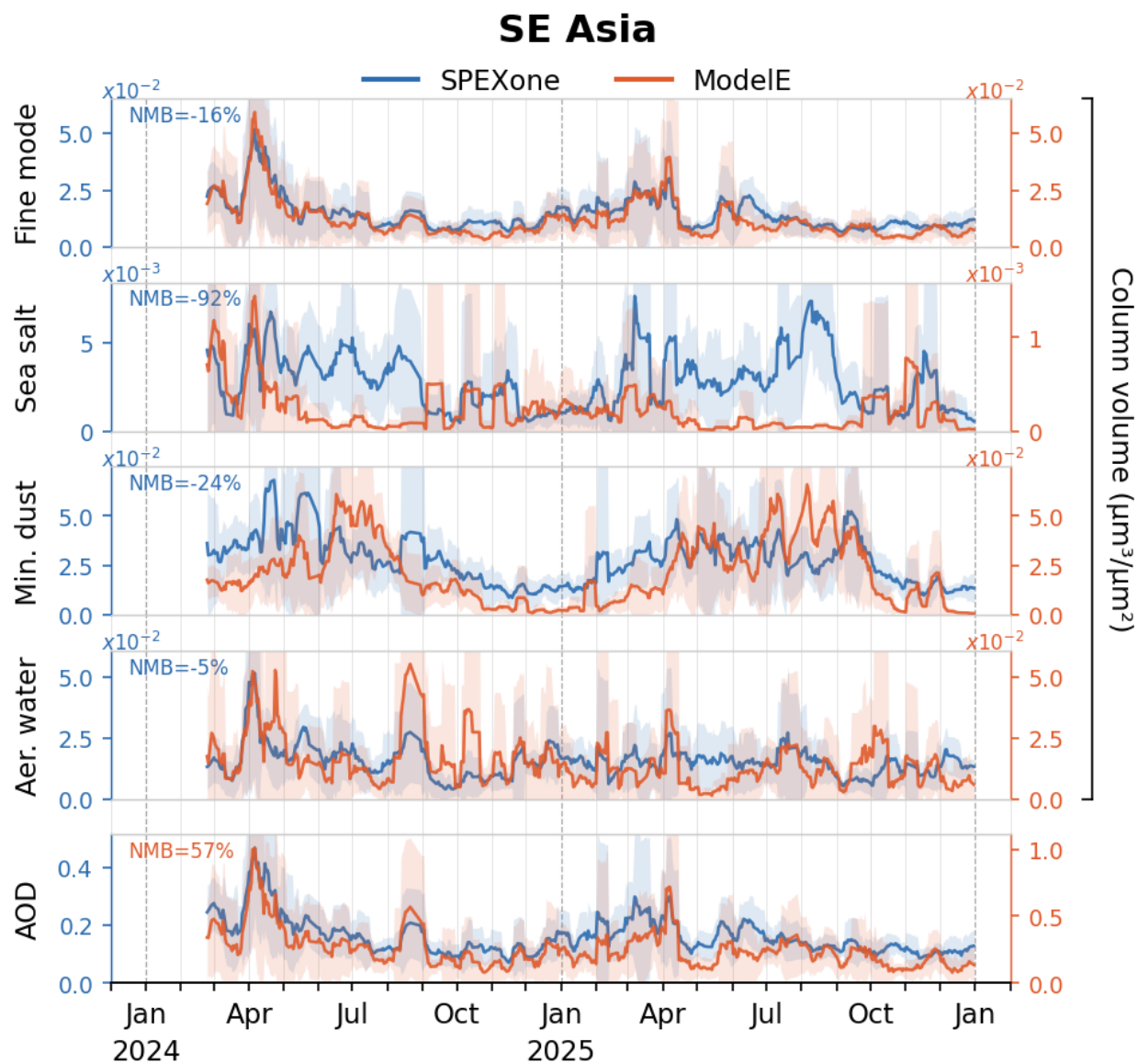


Figure S10: Same as Fig. 8 of the main text but for SE Asia (20–45° N, 90–120° E). Fine mode agreement is strong ($R^2 = 0.70$), with both products resolving seasonal cycles driven by anthropogenic emissions and spring agricultural burning.

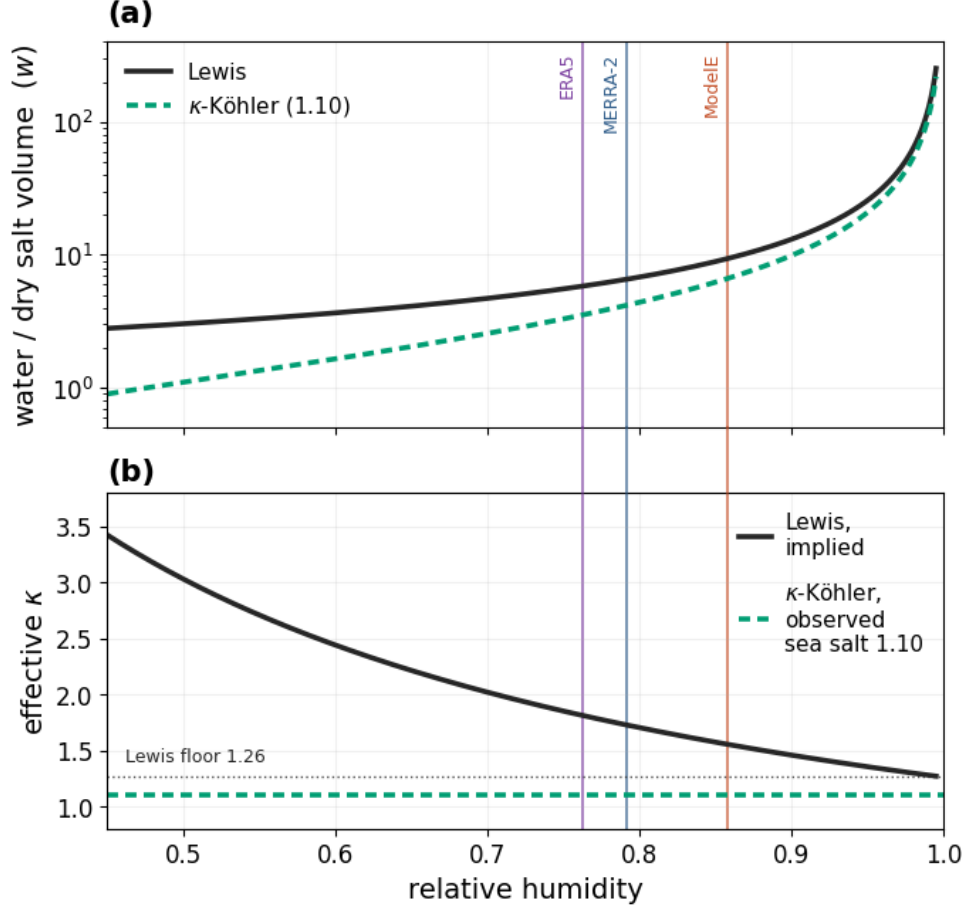


Figure S11: The two sea salt uptake schemes of Sect. 3.4 as functions of RH. (a) Equilibrium water-to-dry-salt volume ratio w : Eq. (2) of the main text, converted from a mass ratio by $\rho_{SS}/\rho_w = 2.165$, against Eq. (3) at $\kappa = 1.10$ (Zieger et al., 2017), which is already a volume ratio and needs no conversion. (b) The same with $H/(1 - H)$ divided out, leaving κ itself for Köhler and, for Lewis, the effective hygroscopicity of Eq. (5), which drifts downward with humidity, approaches 1.26 as $H \rightarrow 1$, and exceeds 1.10 throughout. Vertical lines mark the median over all co-located ocean cells of the salt-weighted column relative humidity, Eq. (4). Both curves are analytic; only the vertical lines use the co-located dataset. These are single RH values, whereas Fig. 10 integrates water and salt over the whole column and so corresponds to no single RH.

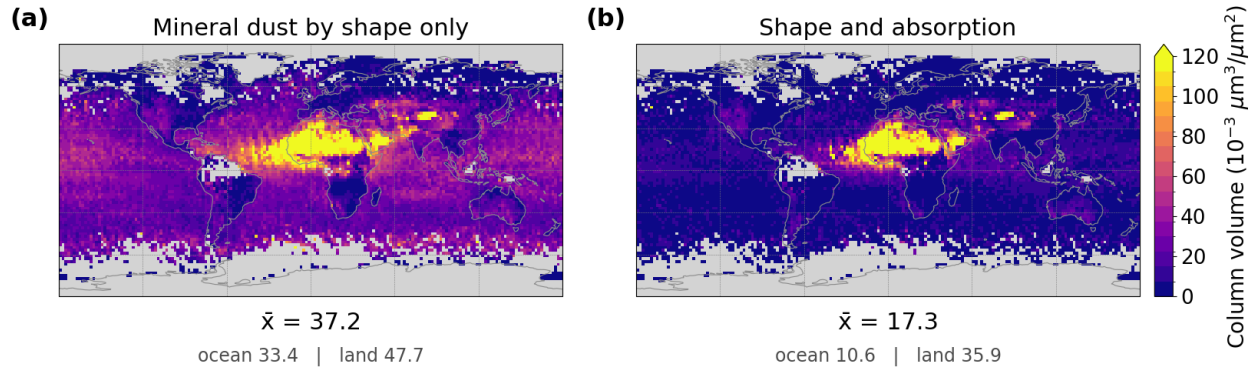


Figure S12: Retrieved mineral dust column volume ($10^{-3} \mu\text{m}^3/\mu\text{m}^2$) from the SPEXone-RemoTAP hybrid product (Fu et al., 2026), February 23, 2024 through February 28, 2025 (348 daily files), the end of the hybrid record at the time of analysis. (a) The partition as shipped, assigning all nonspherical coarse particles to mineral dust. (b) The refinement of Sect. 4.2, reassigning to sea salt the mineral dust volume in cells where the retrieved imaginary refractive index (IRI) of the coarse nonspherical mode falls below 10^{-7} , effectively at the 10^{-9} lower bound of the inversion. This removes 68% of the ocean mineral dust but only 25% over land. Means are area-weighted; the product is not co-located with ModelE.

References

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