Supplement to the Role of Particle Size Distribution in Simulating Aerosol Transport, Radiative Effects and Surface Area of the 2019/20 Australian pyroCb Plume

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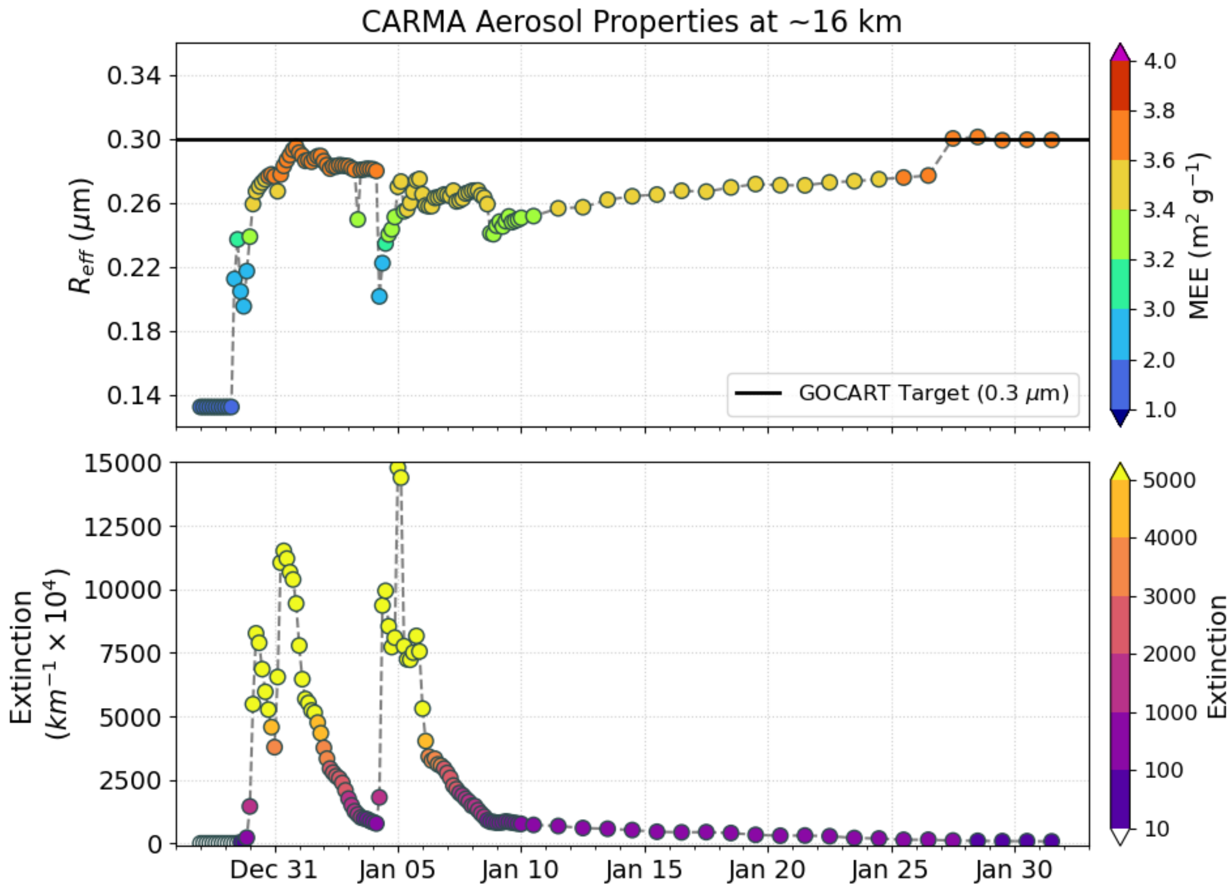

Figure S1. Evolution of stratospheric smoke aerosol properties at ~16 km derived from CARMA sectional microphysics. *(a) Time series of the effective radius (R_{eff}, μm) colored by the Mass Extinction Efficiency (MEE, $m^2\ g^{-1}$). The solid black line indicates the 0.3 μm target R_{eff} adopted for the bulk GOCART PyroCb optics simulation. (b) Corresponding aerosol extinction at 870 nm. The rapid growth of R_{eff} during the high-extinction injection pulses (late December and early January) demonstrates that coagulation quickly shifts the size distribution, justifying the use of an aged, 0.3 μm particle size for simulating the long-term transport of the plume.*

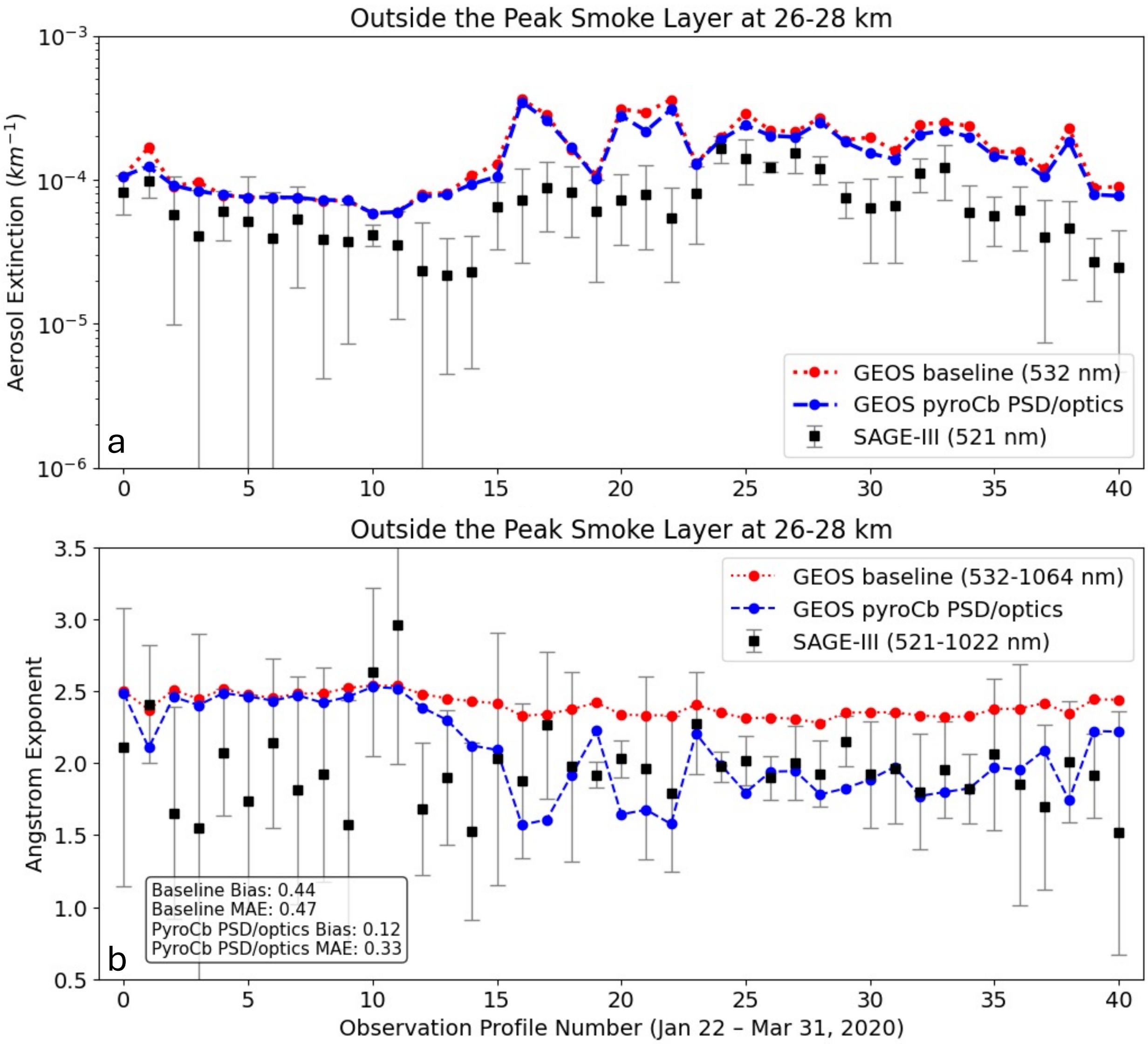

Figure S2. Comparison of modeled aerosol optical properties with SAGE-III/ISS observations outside the peak smoke layer (26–28 km) from January 22 to March 31, 2020. *(a) Aerosol extinction at ~532 nm from GEOS baseline (red) and GEOS pyroCb PSD/optics (blue) simulations relative to SAGE-III (black squares). (b) Angstrom Exponent (532–1064 nm). The updated GEOS pyroCb PSD/optics (blue) shows significantly better agreement with the observed SAGE-III Angstrom Exponent compared to the baseline simulation (red), indicating that the updated size distribution better captures the physical state of the stratospheric aerosol.*

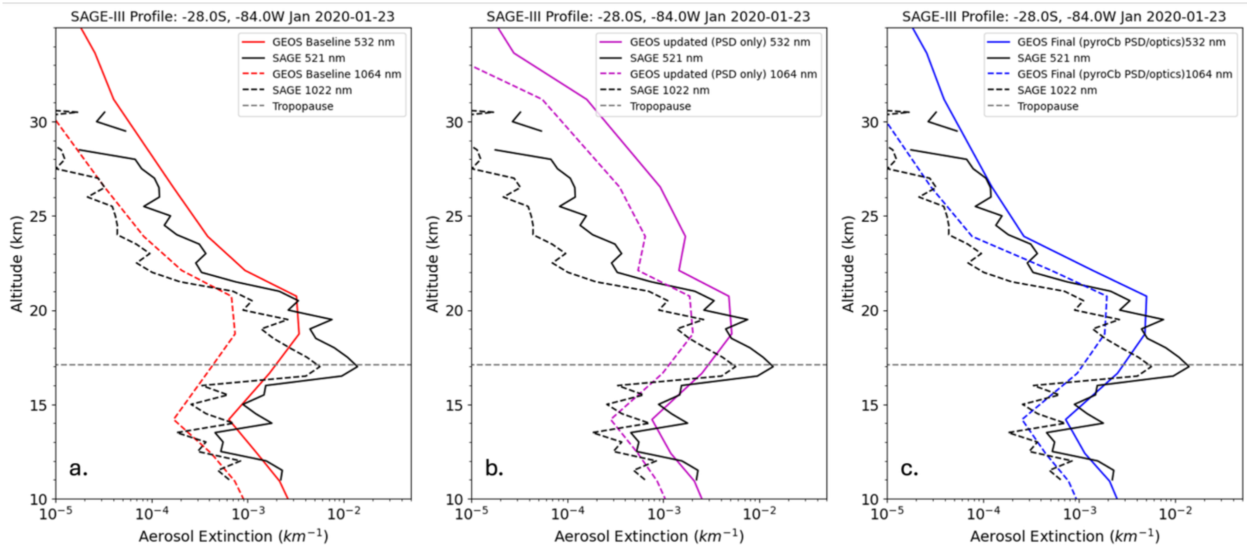

Figure S3. *Comparison of simulated vertical aerosol extinction profiles against a SAGE-III/ISS solar occultation measurement for a single unsaturated profile captured on January 23, 2020 (28.0°S, 84.0°W). The panels illustrate the progressive impact of the updated microphysical and optical assumptions on simulated aerosol extinction (km^{-1}) profile at visible (521/532 nm; solid lines) and near-infrared (1022/1064 nm; dashed lines) wavelengths. Black lines indicate SAGE-III/ISS observations. Colored lines indicate GEOS model simulations for: (a) the baseline GEOS configuration (red), (b) an intermediate GEOS configuration with only the updated Particle Size Distribution ("PSD only"; purple), and (c) the final updated GEOS pyroCb PSD/optics configuration (blue) which incorporates both the updated PSD and the observationally constrained organic aerosol absorption. The horizontal dashed line denotes the local tropopause height.*

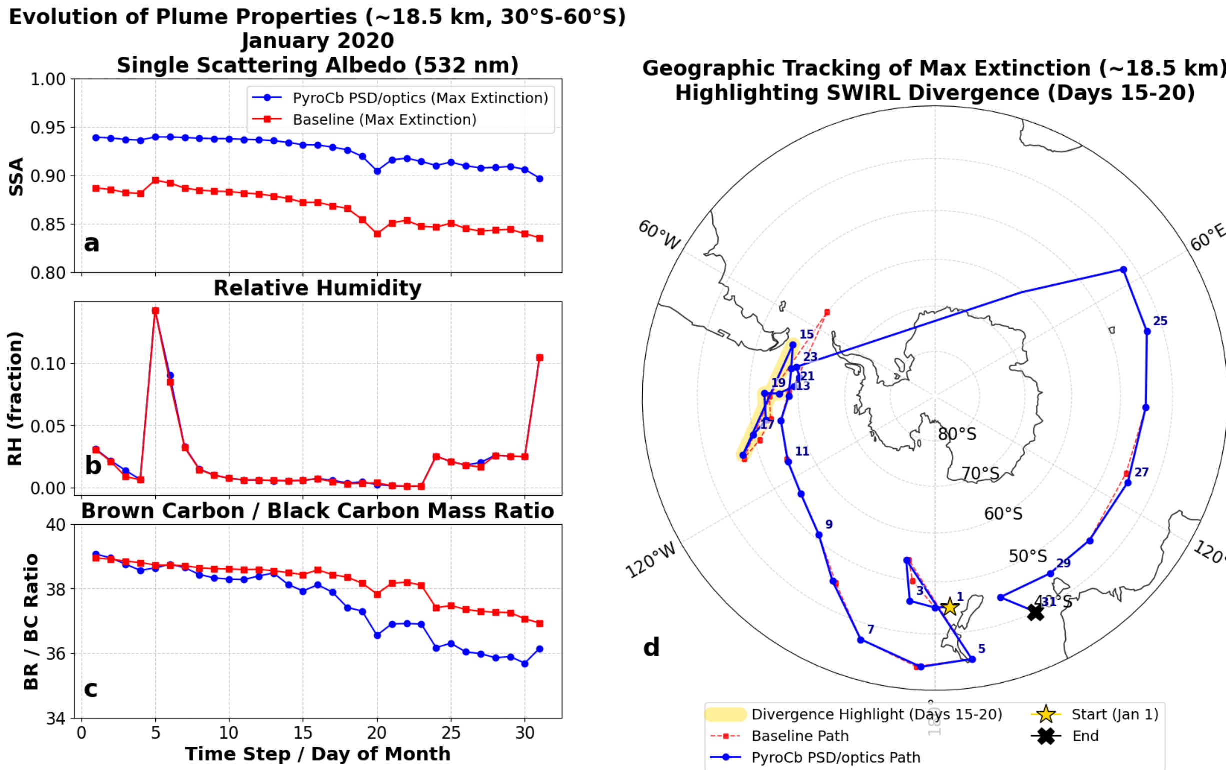

Figure S4. *Evolution and trajectory of the simulated ANY pyroCb plume core during January 2020.* ***(a–c)*** *Time series of physical and optical properties extracted along the tracked plume core trajectories for the updated pyroCb PSD/optics (blue) and baseline (red) configurations, showing:* ***(a)*** *Single Scattering Albedo (SSA) at 532 nm,* ***(b)*** *fractional Relative Humidity, and* ***(c)*** *Brown Carbon (or OA) to Black Carbon mass ratio.* ***(d)*** *Geographic tracking of the plume core, defined by the grid cell with maximum aerosol extinction at the ~18.5 km altitude level. The map highlights the dynamic transport divergence between the two configurations, most notably the distinct smoke vortex trajectory south of Chile between days 15 and 20.*