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Retrieval of bimodal aerosol size distributions for stratospheric sulfate and smoke aerosols using multi–wavelength satellite measurements

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LOPC and WLPC aerosol size resolved concentration profiles are shown in fig. S1 for observations made on 13 September 2020 from Laramie, WY. Stratospheric aerosol concentrations are most concentrated with d_p of less than 560 nm. Limited coarse aerosol concentrations of d_p more than 560 nm and up to several micron are present up to ~20 km. SAGE III/ISS retrieved number densities show fairly strong agreement with the aerosol concentrations from the lowest measured size bins from the WLPC and LOPC, $N(d_p > 0.184\ \mu m)$ and $N(d_p > 0.300\ \mu m)$ respectively, ranging from 1–40 cm^{-3} (Fig. S1). SAGE III/ISS and OPC retrieved aerosol number densities below 18 km are reduced in the primary smoke perturbed layers.

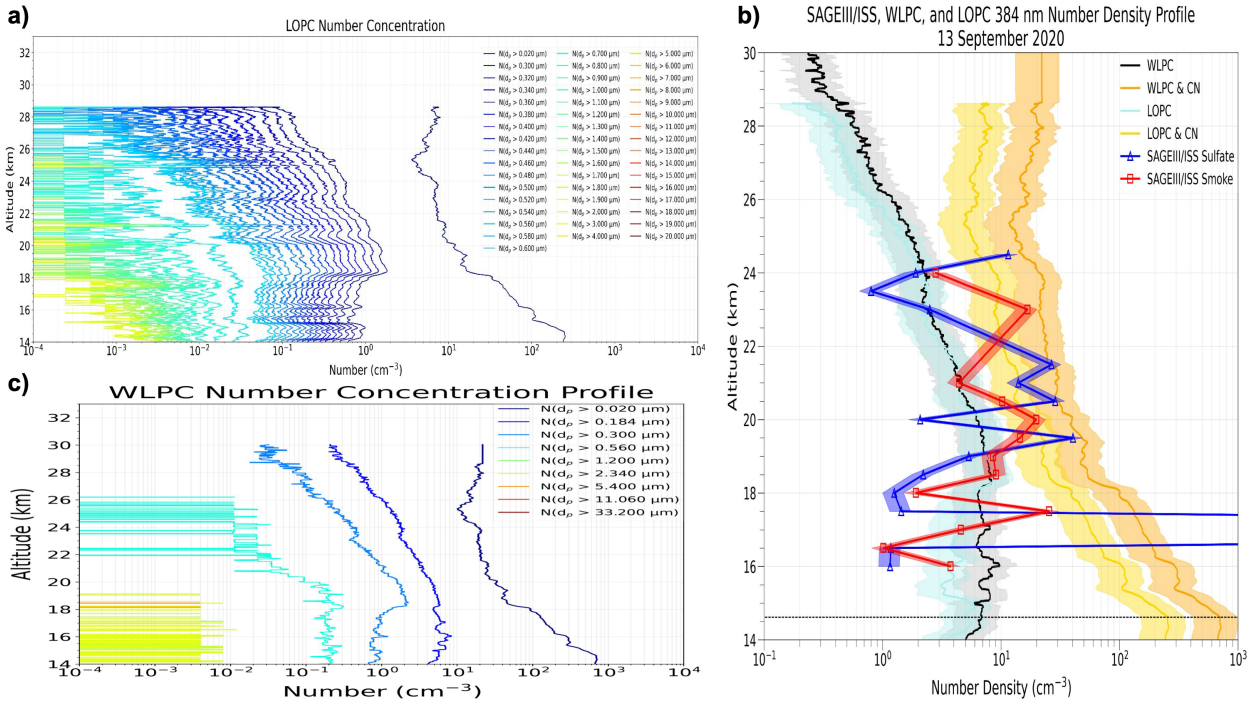


Figure S1: a) LOPC and c) WLPC measured aerosol size resolved concentrations on 13 September 2020 from Laramie, WY. b) SAGE III/ISS, WLPC, and LOPC aerosol number densities for the same observation.

The better part of the UTLS below 18.5 km is modulated by smoke aerosols with enhanced aerosol diameters and produce moderate aerosol optical and microphysical properties. From 15–18 km, SAGE III/ISS retrieved $d_{p,\,Fine}$ range from 350–500 nm for sulfate aerosols and 100–400 nm for smoke aerosols, with OPC measured values of 100–200 nm (Fig. 17b and S2a–b). Aerosol size distributions at 19 and 20.5 km indicate increased aerosol sizes in the in situ measurements which predominantly remain near several hundred nm (~300 nm) in broad polydisperse distributions (Fig. S2c–d). SAGE III/ISS retrieved aerosol distributions show the reduced aerosol concentrations which are a product of smaller aerosol concentrations and diameters (Fig. S2c–d).

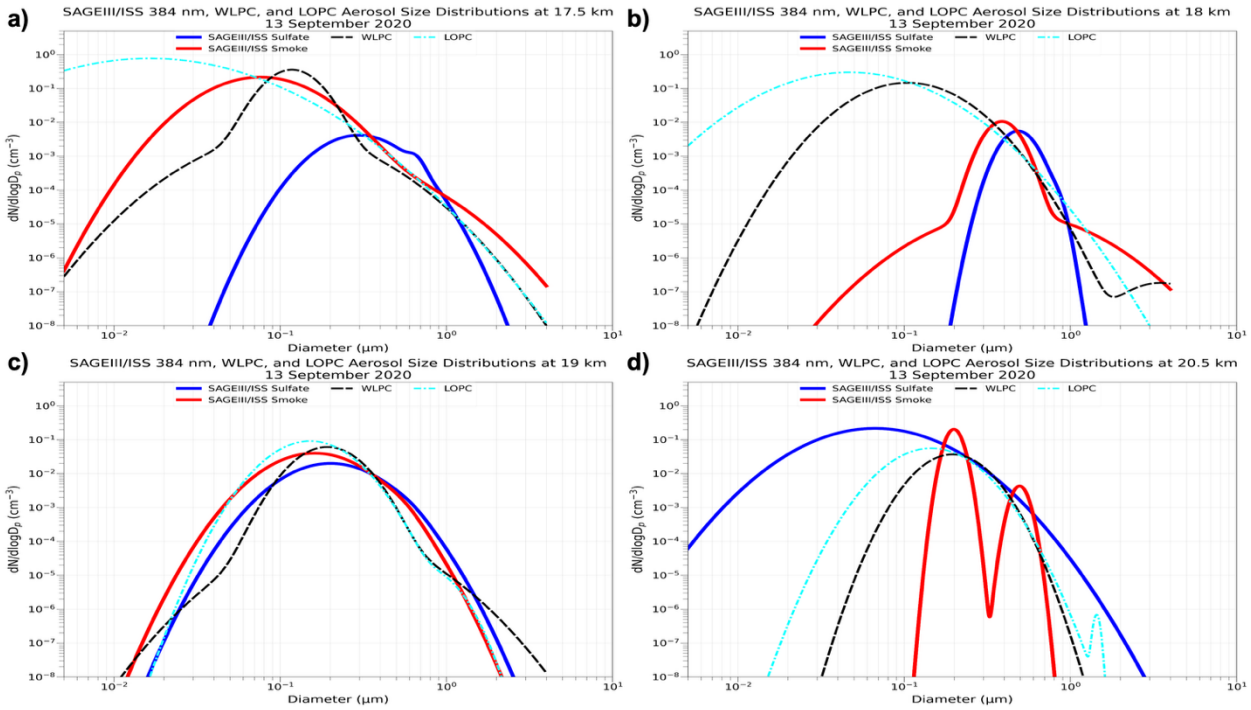


Figure S2: SAGE III/ISS, WLPC, and LOPC aerosol size distributions on 13 September 2020 at: a) 17.5 km, b) 18 km, c) 19 km, and d) 20.5 km.

Figure S3 show the retrievals described in this manuscript (SAGE III/ISS Sulfate or SAGE III/ISS Smoke), the operational SAGE III/ISS product (SAGE III/ISS v6), and in situ measurements from B^2SAP for 65 observations taken above Boulder, CO (40 °N, -105 °E) from 2019–2026 matching the coincidence criteria described in the manuscript but increasing the temporal window to within 48 hours. In situ measurement sampling dates which do not have SAGE III/ISS observations meeting these spatiotemporal criteria are not included, while averages are calculated for SAGE III/ISS retrieved values over multiple coincident flights. Retrieved aerosol optical and microphysical properties are shown as mean values for each 0.5 km altitude bin over the period from 2019–2026 for observations meeting the coincidence criteria.

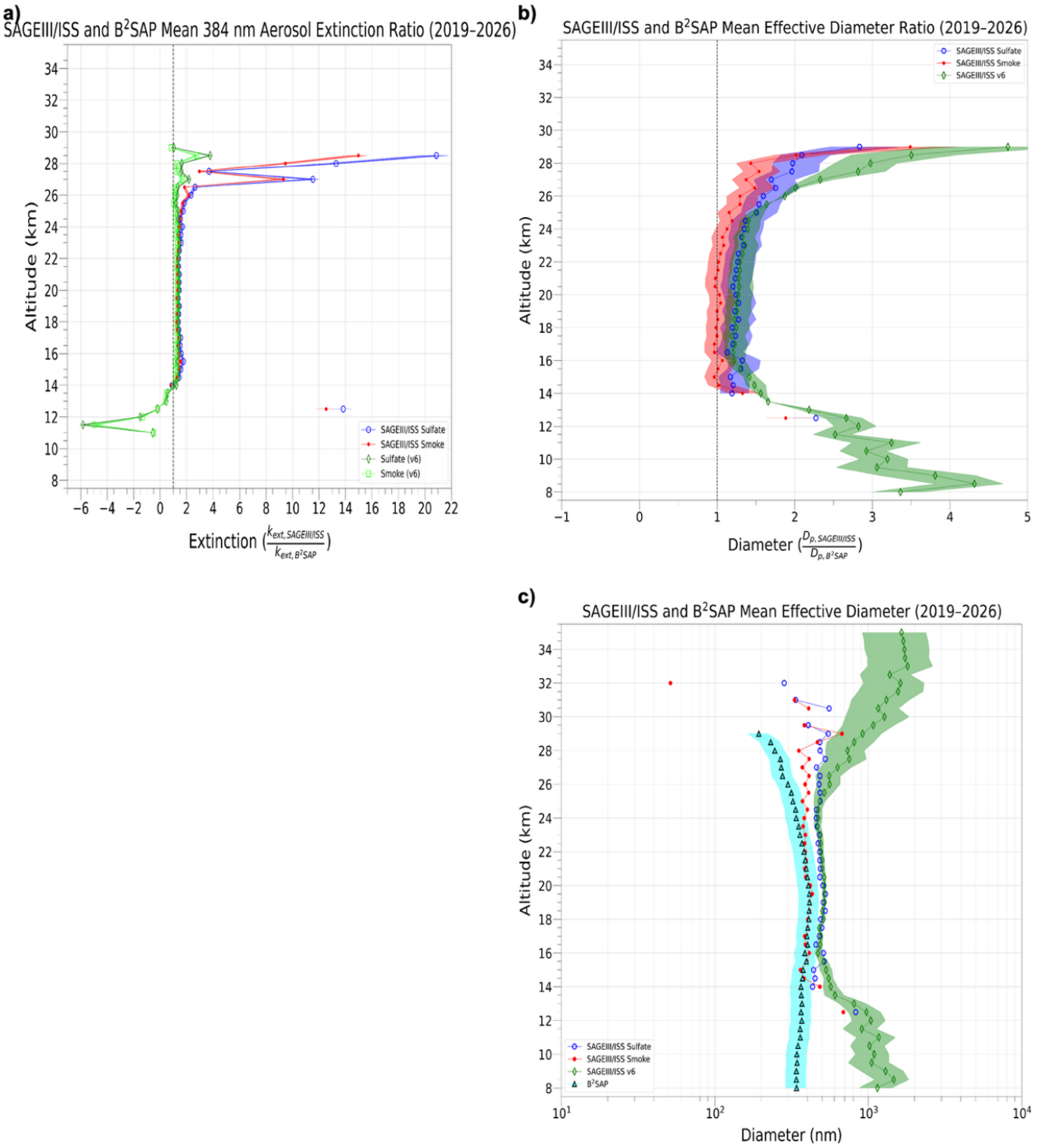


Figure S3: Ratios of SAGE III/ISS over B²SAP of a) 384 nm aerosol extinction coefficients and b) aerosol effective diameter. A vertical profile of mean aerosol effective diameters are presented in (c).

Figure S3 shows (a) 384 nm extinction ratio and (b) aerosol effective diameter ratio mean profiles over the same period. Higher altitudes (UTLS altitudes) display larger biases in the operational v6 product away from the in situ measurement and retrieval method described here as aerosol volumes decrease (increase). Aerosol effective diameters are clearly biased in the operational product due to the large particle influence on extinction and inversion products. Retrievals using the approach here for sulfate and smoke aerosols are in better agreement with the in situ measurement. The better agreement with in situ measurements seen in the retrieved effective diameters using the approach here reinforces that the new retrieval approach can reliably determine the aerosol size distribution and higher level moments for polydisperse aerosol populations. Both a primary and secondary mode can be retrieved, lessening the reliance of the inversion to only a single mode of larger particles, and instead weighing the concentrations of the small and large particle modes in the retrieved distribution.

Figure S3 (c) presents the aerosol effective diameter mean profiles used in (b) to form the ratios. In situ and satellite retrieved aerosol effective diameters range from 200–600 nm in most layers. Sulfate retrievals agree well with the SAGE III/ISS v6 product from 15–25 km where the aerosol effective diameter is ~400–500 nm. Smoke aerosols display a smaller effective diameter ~300–400 nm which has strong agreement with the in situ profile. Sulfate and smoke aerosol size sensitivity and retrieved diameters are described in sect. 3–5, with smoke aerosols tending to be smaller than sulfate aerosols for moderately attenuated layers, but with pyroCb retrieved samples containing concentrations with larger diameters overall. The v6 product assuming sulfate aerosols overestimates aerosol diameter at higher and lower altitudes in comparison to the retrieval approach here and in situ measurement. Mean values for the approach described here

are marginally increased at higher altitudes from the in situ measurement while remaining less than 600 nm, as the retrieved effective diameter increases as the measured extinction ratio increases.