

# 1 Supplementary Figures

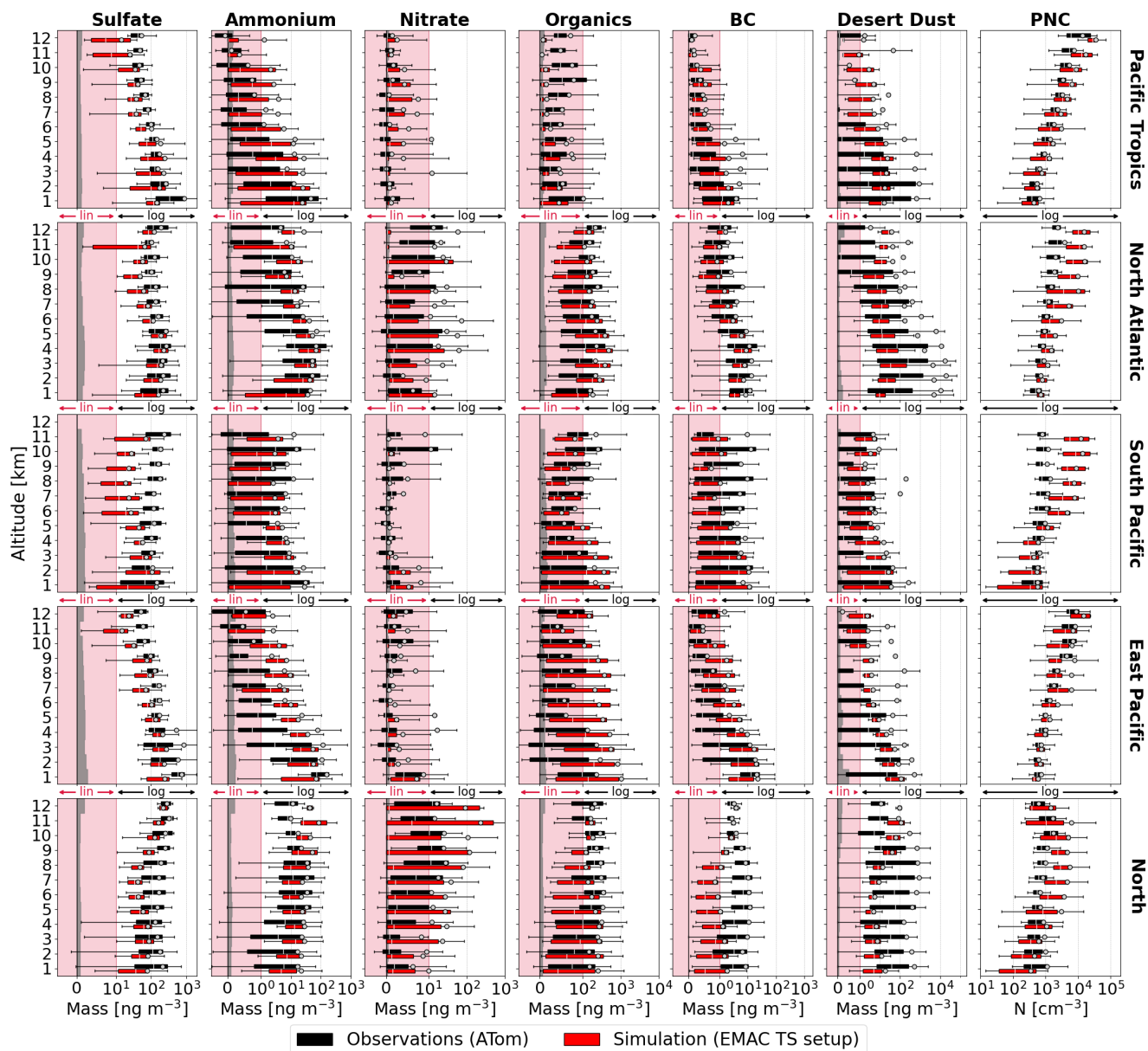
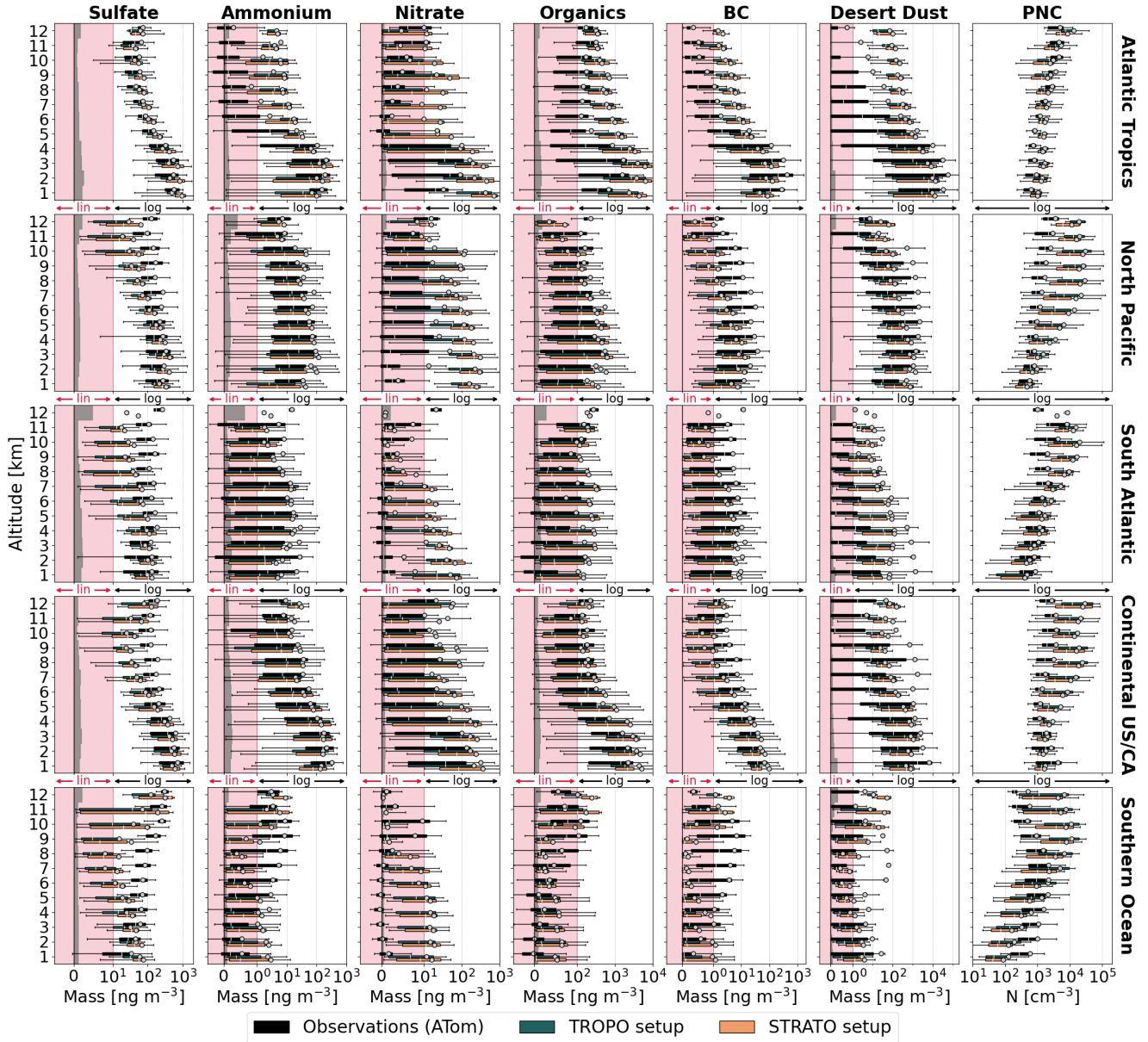


Figure S1. As Fig. 4 from the main text, but for the remaining regions.



**Figure S2.** As Fig. 4 from the main text, but with simulated values from the TROPO and the STRATO setup.

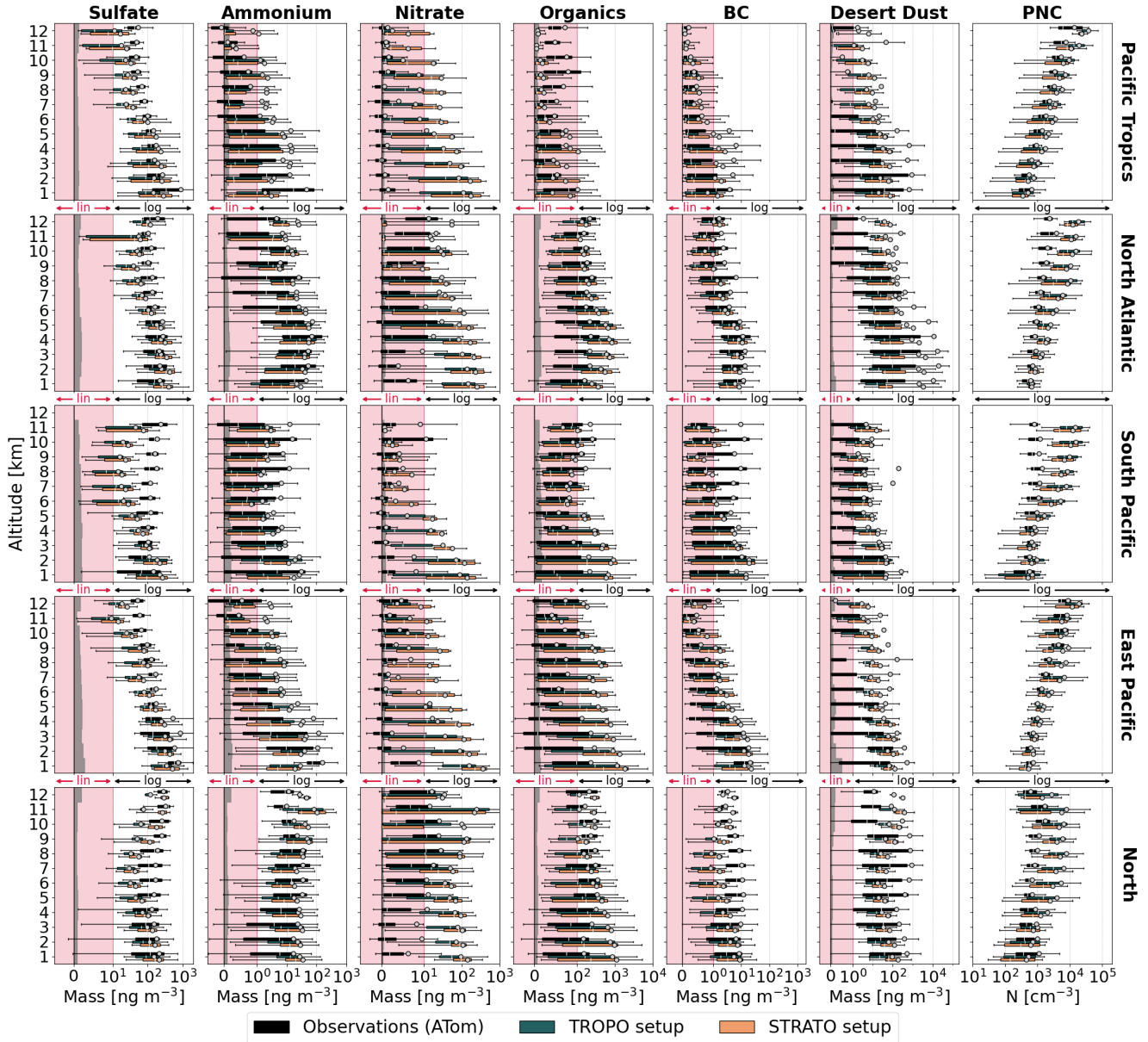
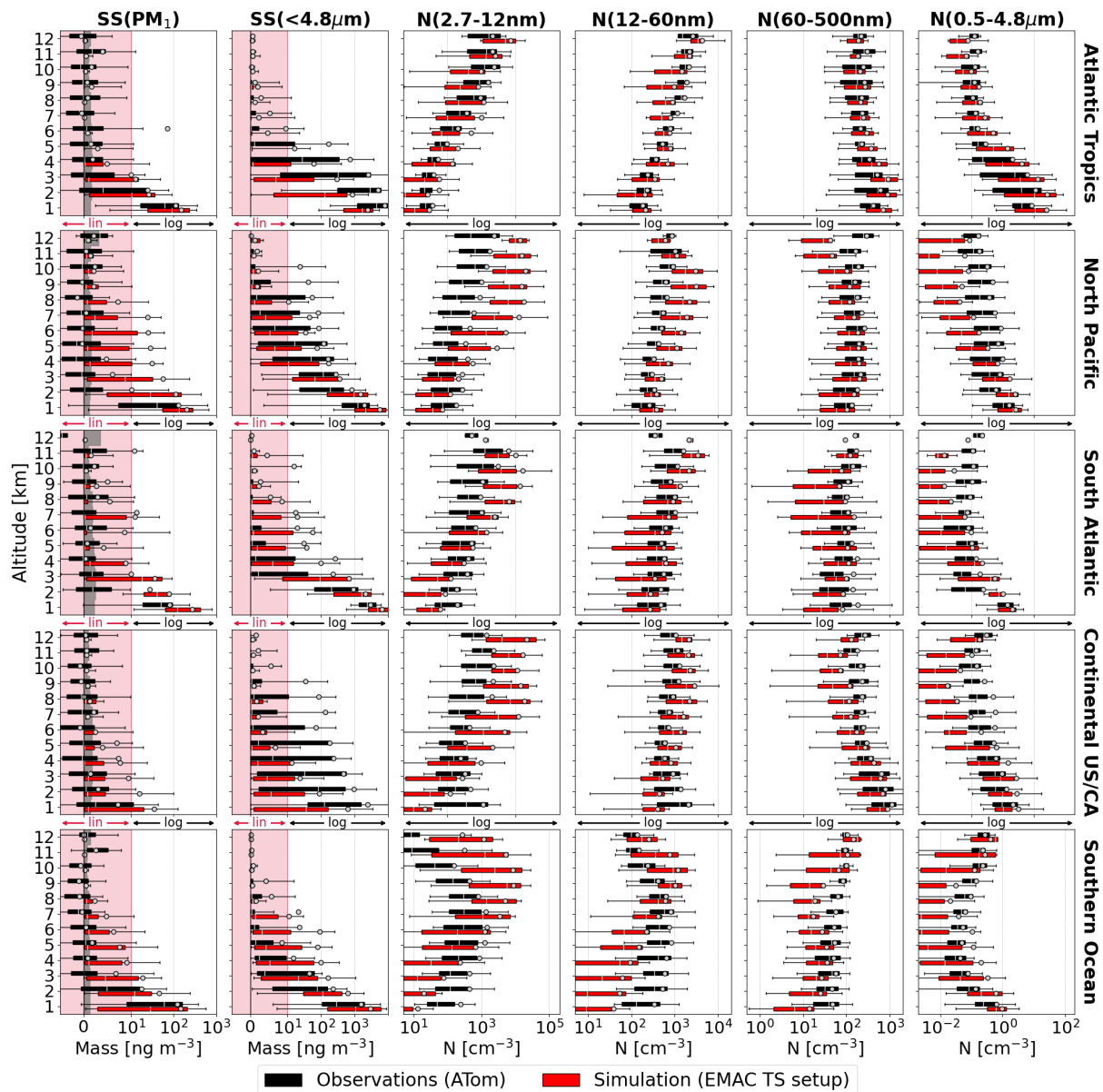
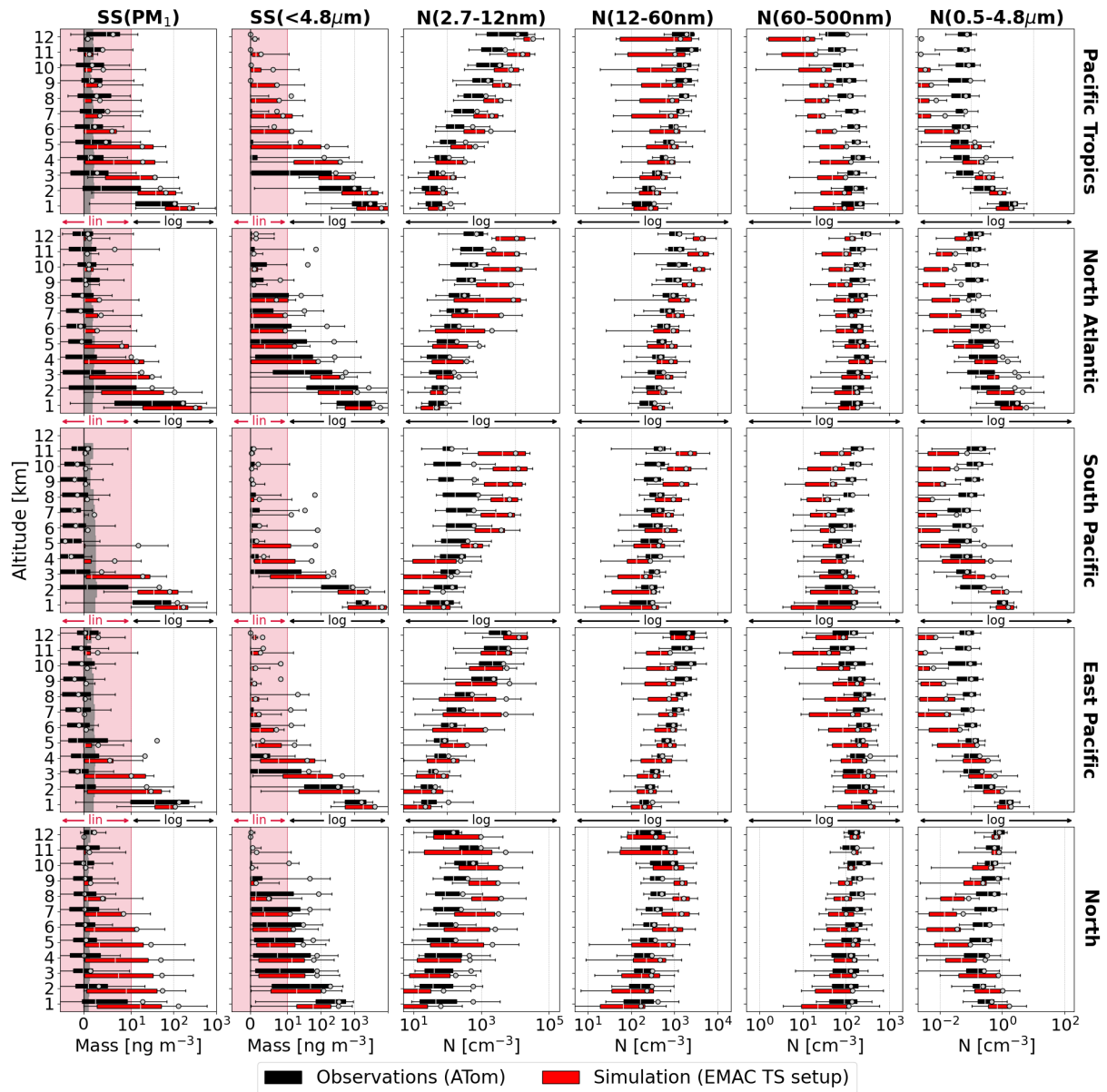


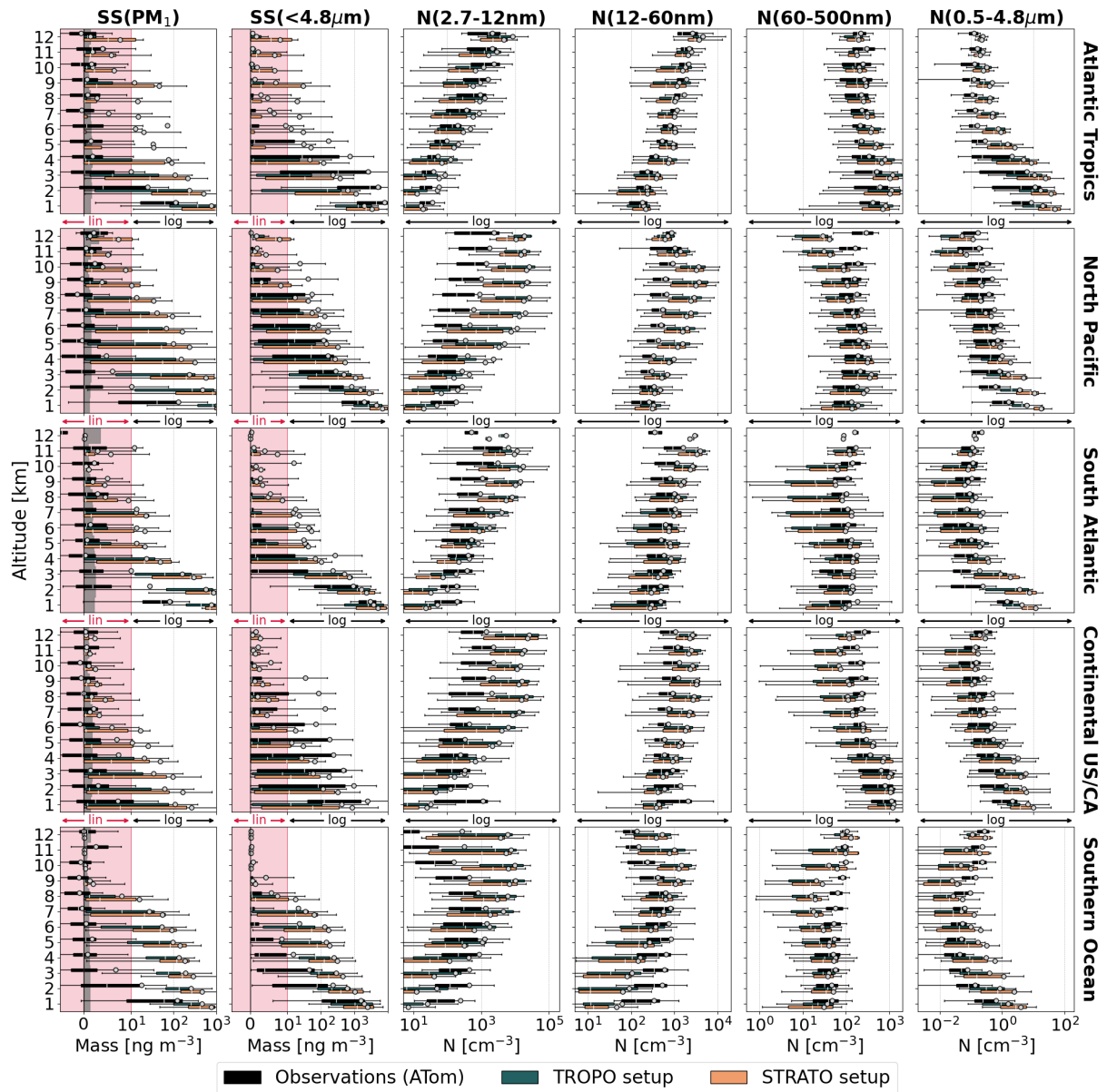
Figure S3. As Fig. S1, but with simulated values from the TROPO and the STRATO setup.



**Figure S4.** Comparison of simulated (red; TS setup) and observed (black; ATom mission) mass concentrations of sea salt ( $\text{PM}_{10}$  and  $0.1 \mu\text{m}$ – $4.8 \mu\text{m}$ ) and dust ( $0.1 \mu\text{m}$ – $4.8 \mu\text{m}$ ), and particle number concentrations in different size bins across selected regions. Box-whisker plots follow the format of Fig. 4 from the main text. All concentrations are provided at standard temperature and pressure (STP;  $T = 273.15 \text{ K}$  &  $p = 1013.25 \text{ hPa}$ ).



**Figure S5.** As Fig. S4, but for the remaining regions.



**Figure S6.** As Fig. S4, but with simulated values from the TROPO and the STRATO setup.

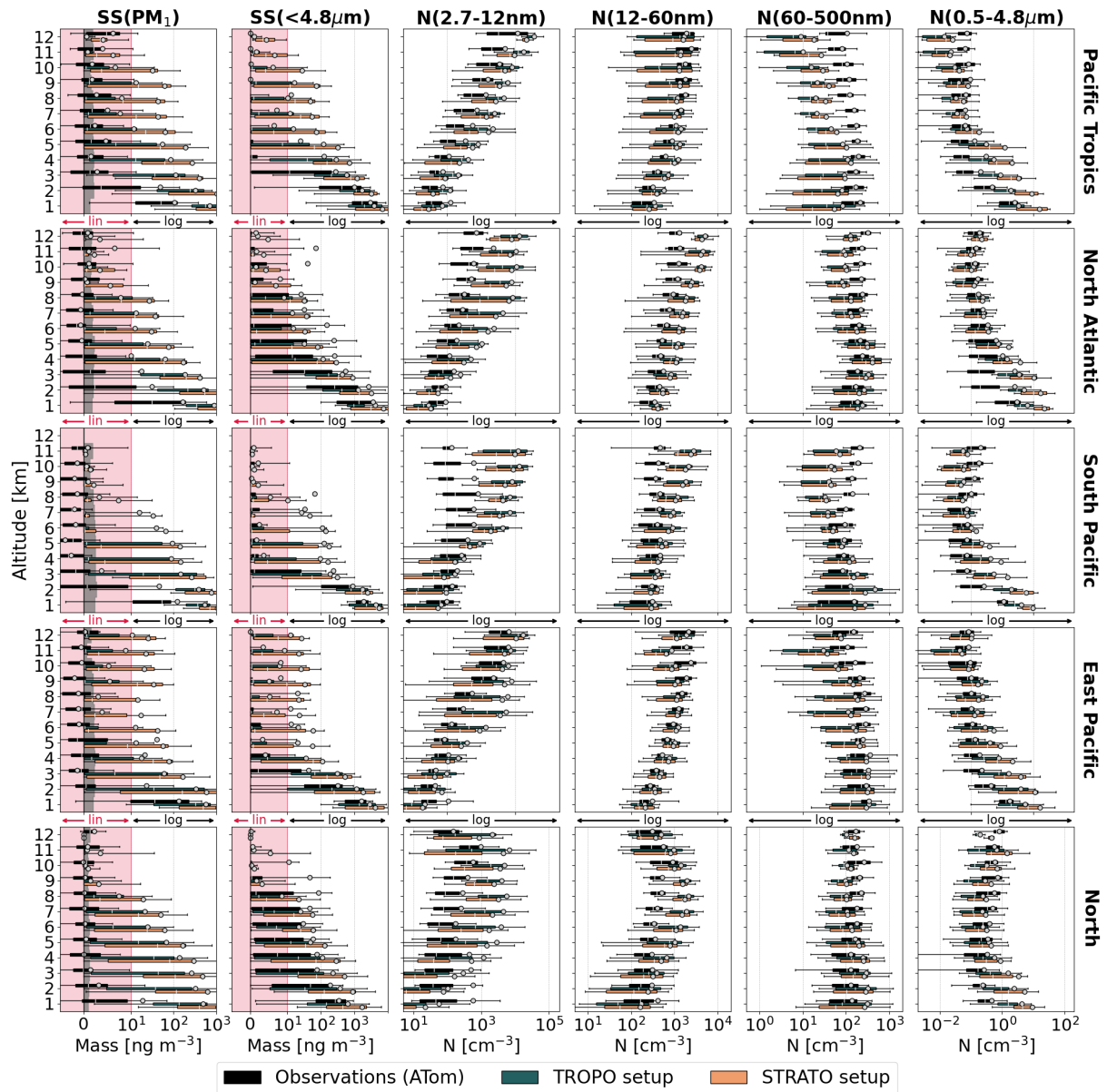
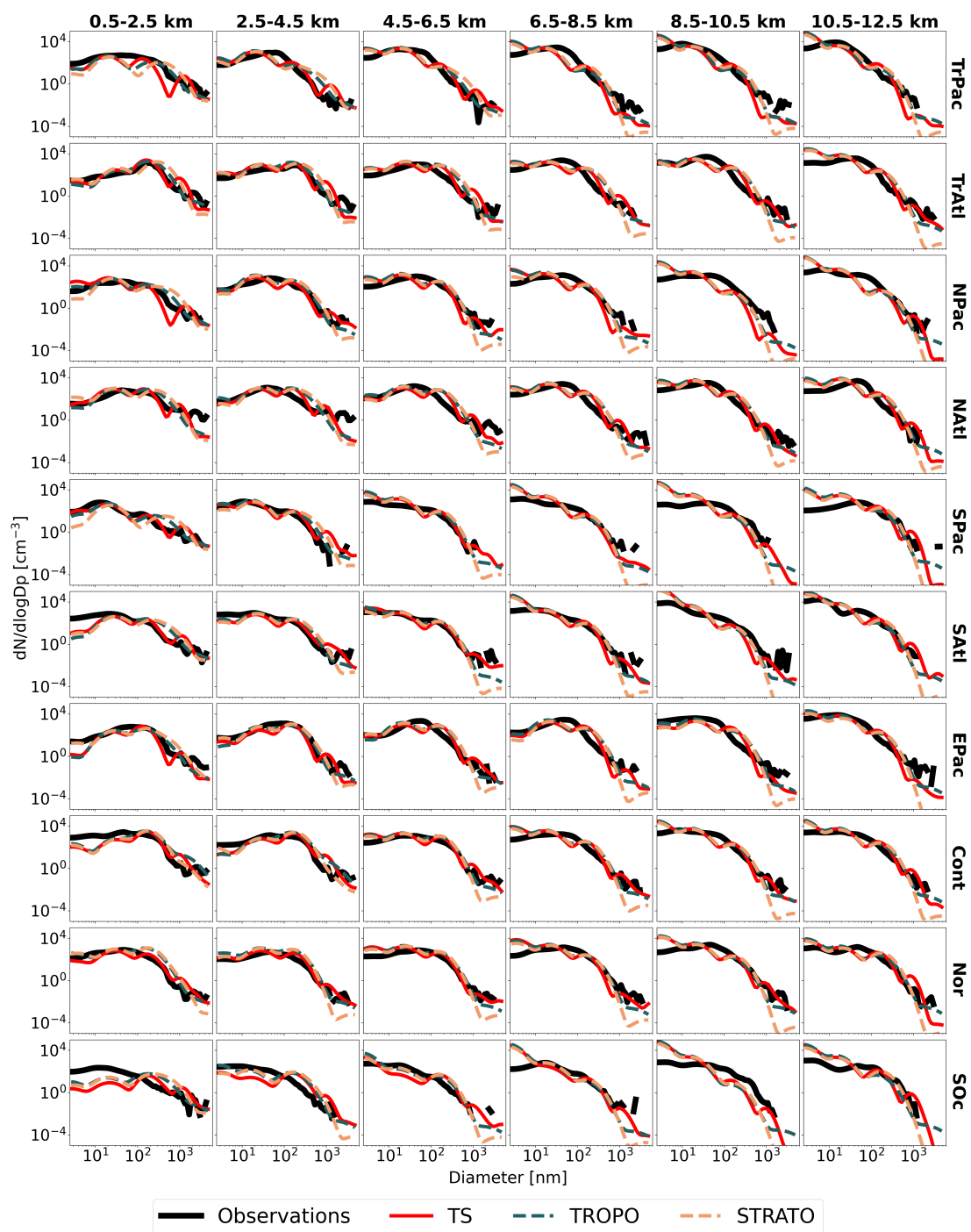
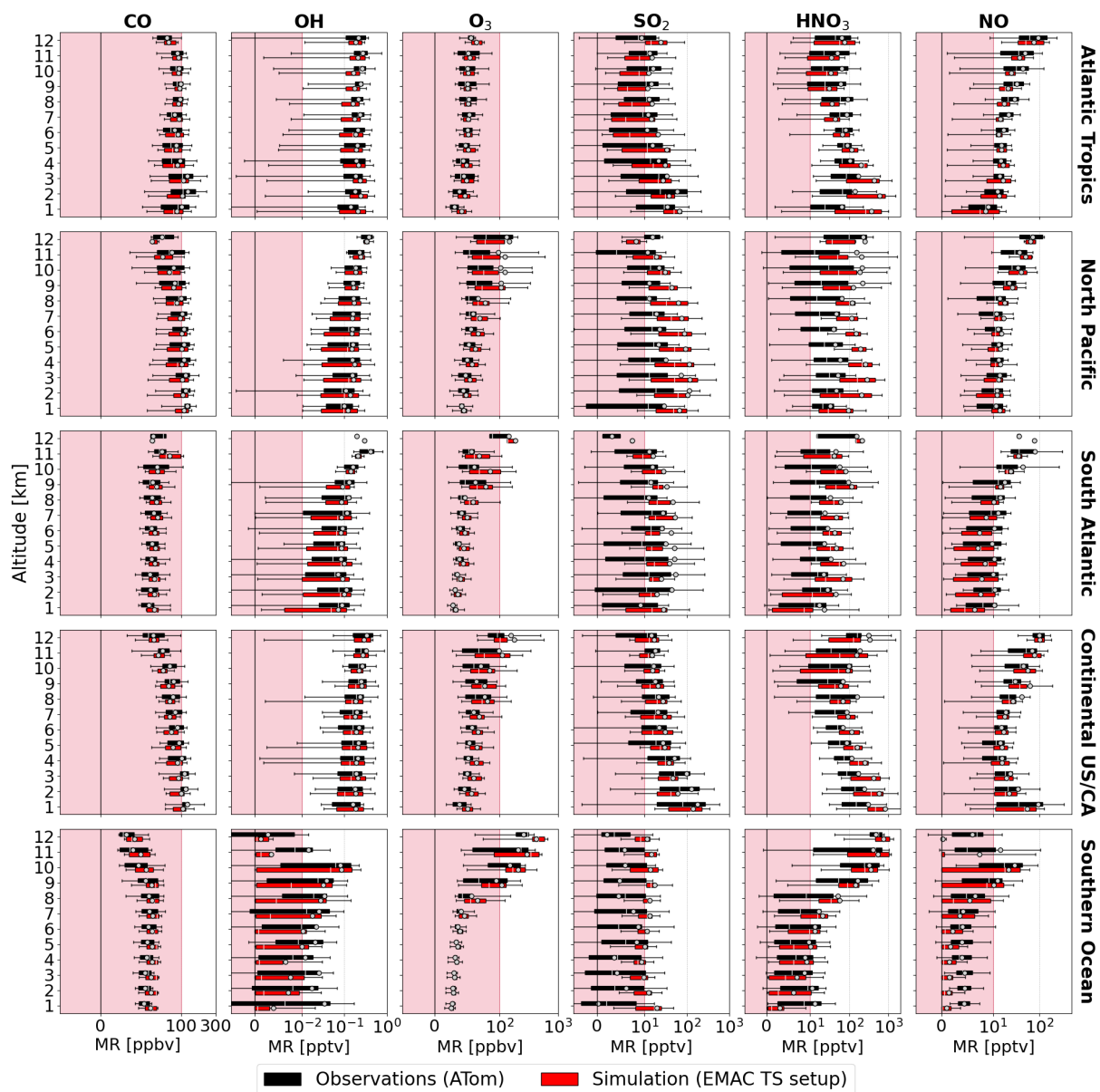


Figure S7. As Fig. S5, but with simulated values from the TROPO and the STRATO setup.



**Figure S8.** Comparison of simulated particle size distributions from the three model setups with ATom observations (black) across selected regions.



**Figure S9.** Comparison of simulated (red) and observed (black) mixing ratios (MR) of carbon monoxide (CO), the hydroxyl radical (OH), ozone (O<sub>3</sub>), sulfur dioxide (SO<sub>2</sub>), nitric acid (HNO<sub>3</sub>), and nitric oxide (NO) from the ATom mission across selected regions, as defined in Fig. 1 from the main text. Box-whisker plots follow the format of Fig. 4 from the main text.

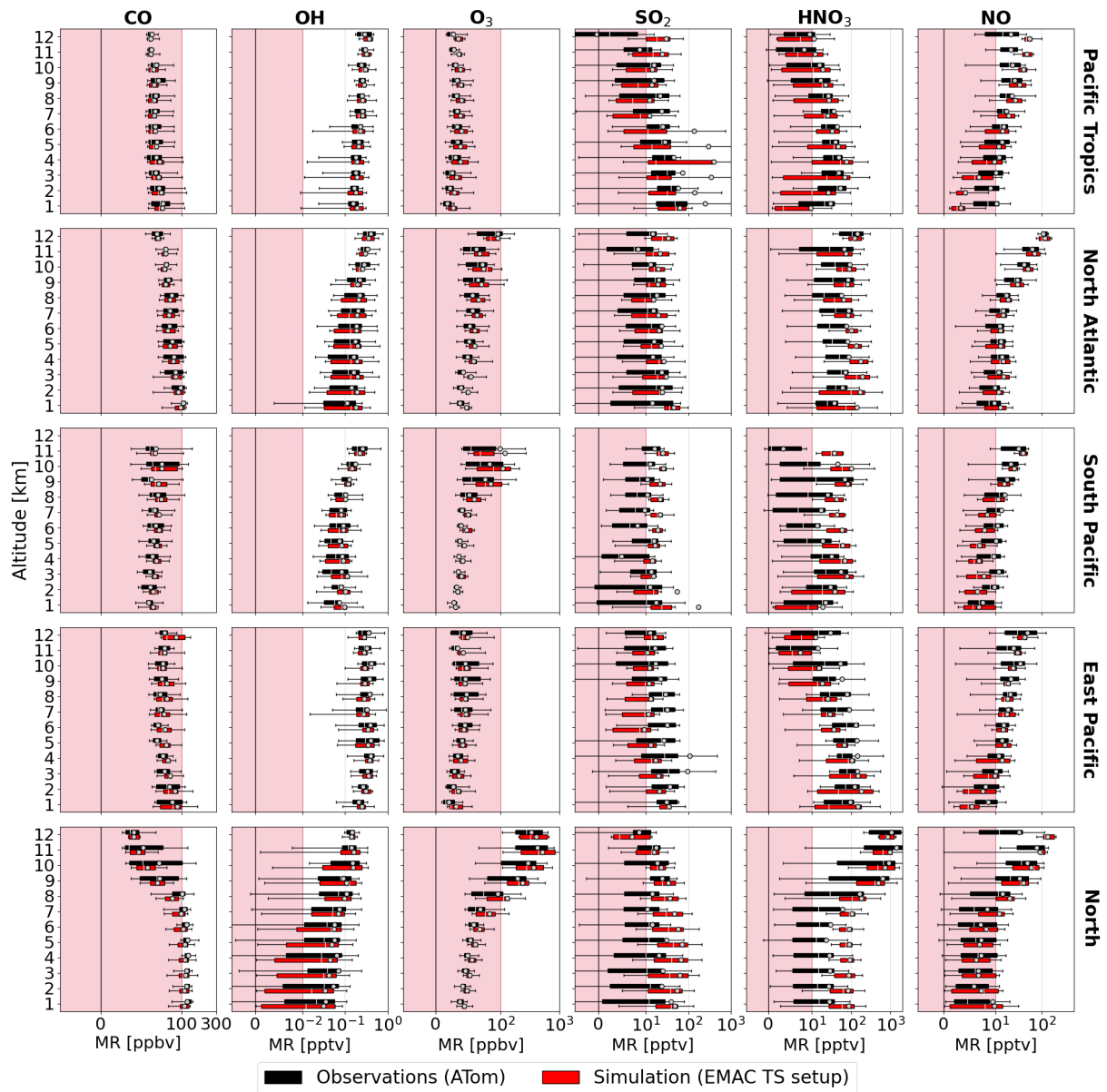
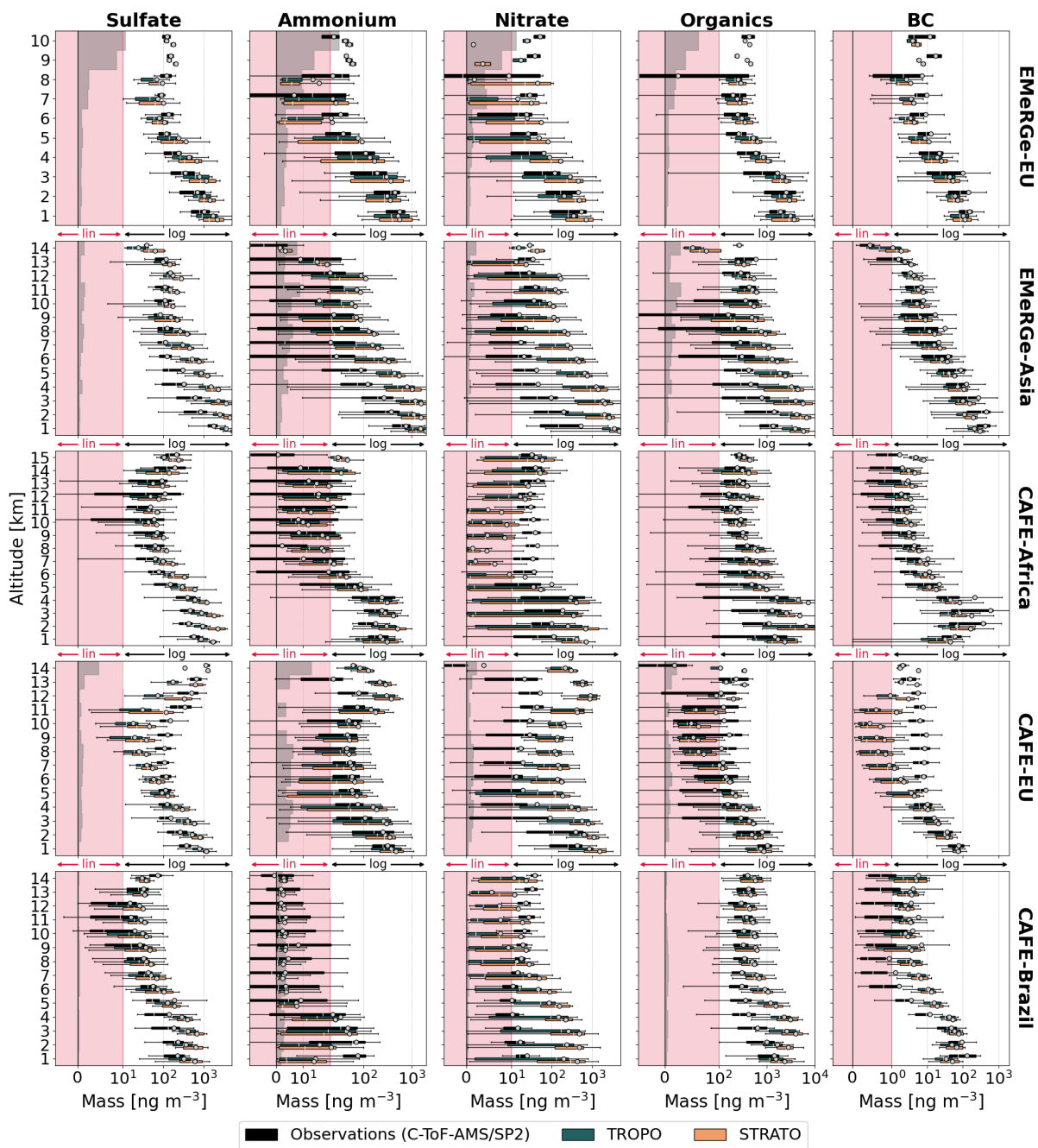
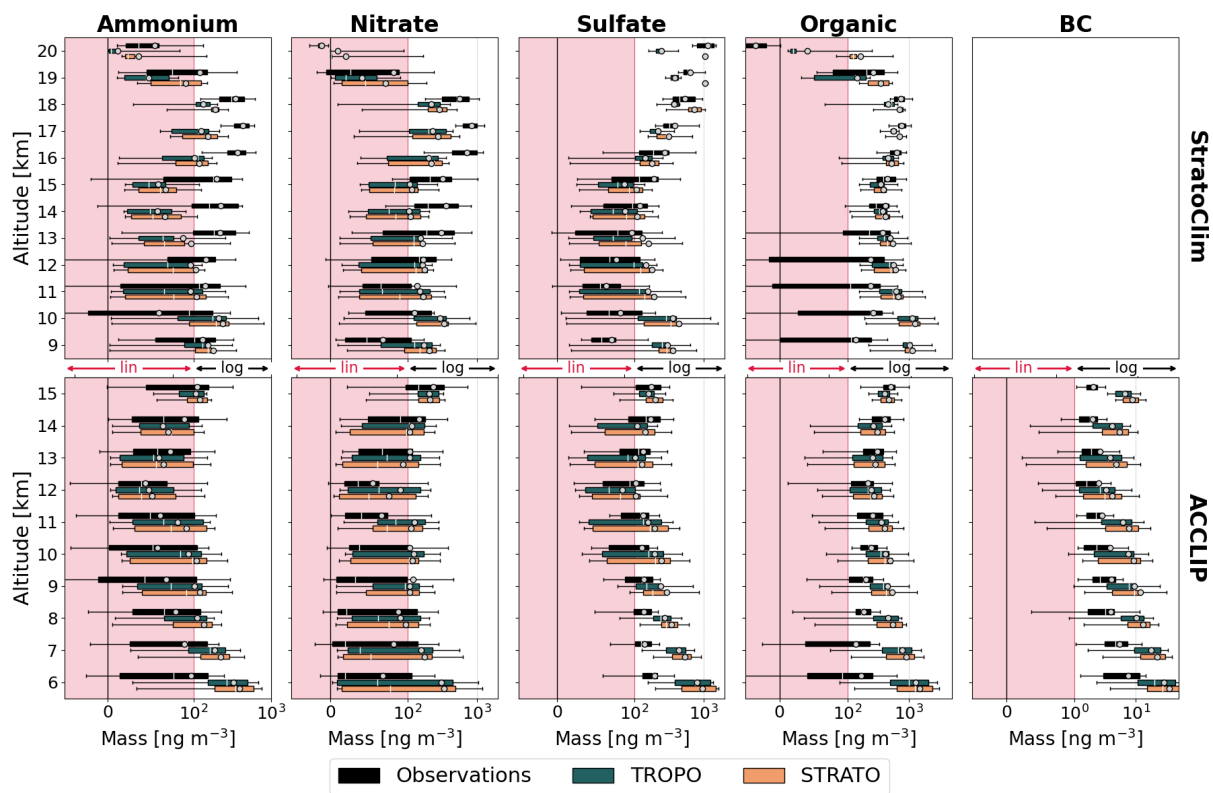


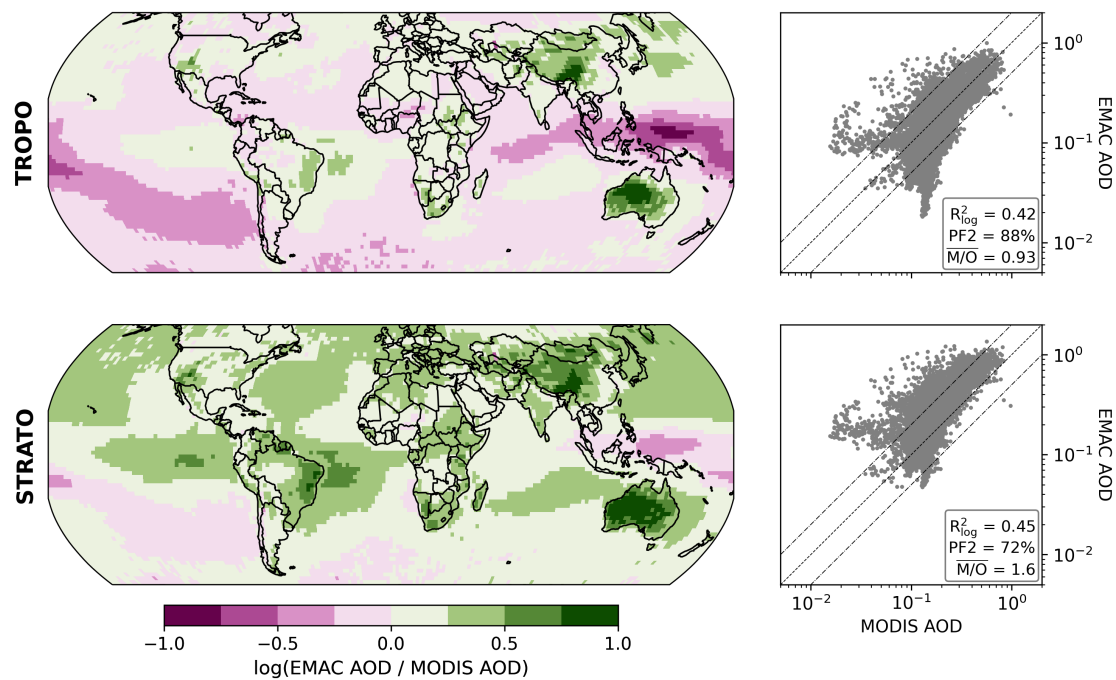
Figure S10. As Fig. S9, but for the remaining regions.



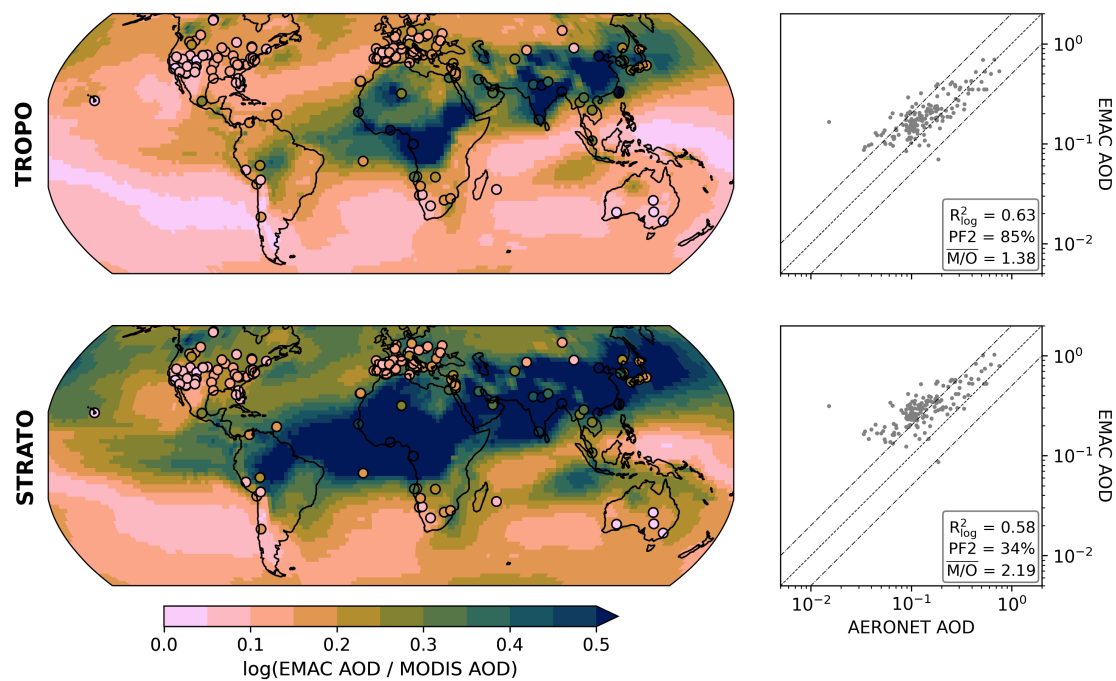
**Figure S11.** As Fig. 5 from the main text, but with simulated values from the TROPO and the STRATO setup.



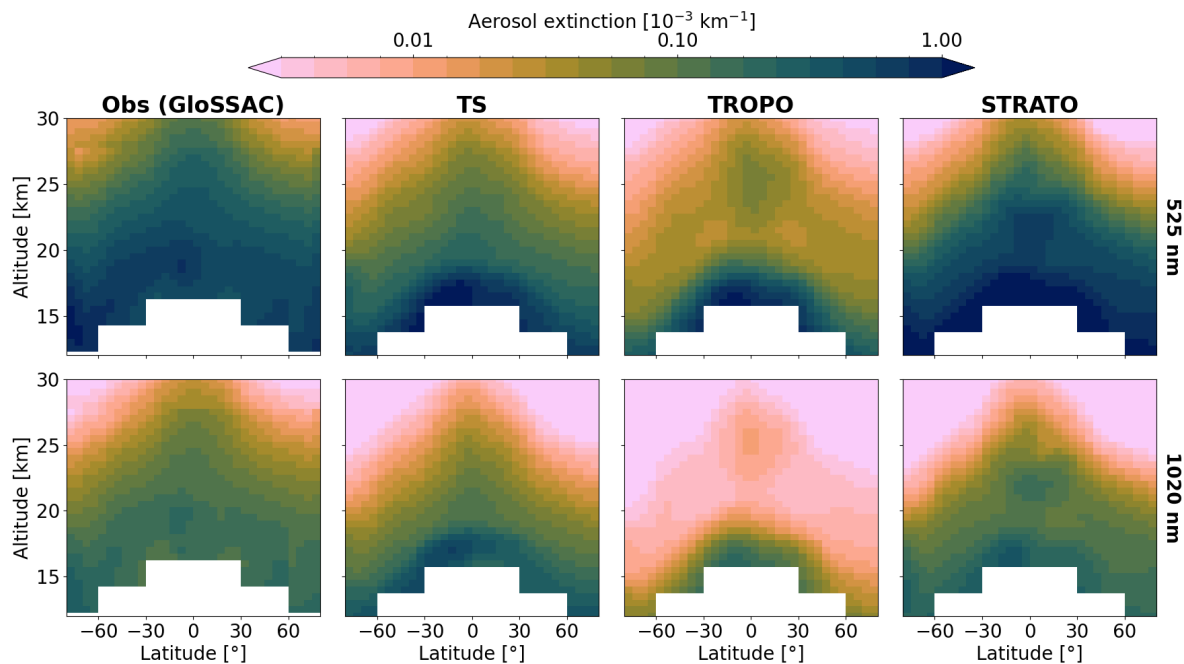
**Figure S12.** As Fig. 6 from the main text, but with simulated values from the TROPO and the STRATO setup.



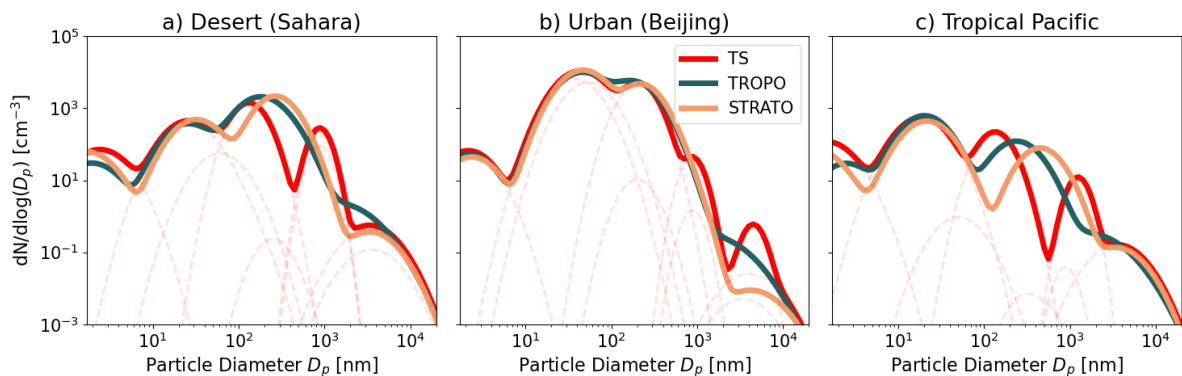
**Figure S13.** As panels a and b from Fig. 3 in the main text, but with simulated values from the STRATO and the TROPO setup.



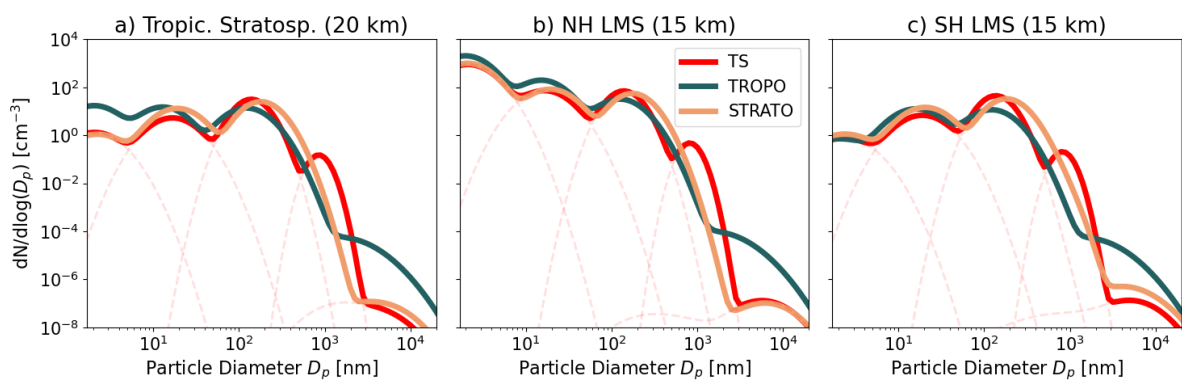
**Figure S14.** As panels c and d from Fig. 3 in the main text, but with simulated values from the STRATO and the TROPO setup.



**Figure S15.** Zonally and temporally (2016–2018) averaged profiles of aerosol extinction, derived from EMAC model simulations compared to GLoSSAC observations at 525 (upper part) and 1020 nm (lower part).



**Figure S16.** Simulated Particle Size Distributions (PSD) at the Earth's surface (averaged over June 2017) for the different model setups in: a) the Sahara Desert, b) an urban region (central Beijing), and c) the Tropical Pacific. The red dashed lines indicate the different modes in the TS setup.



**Figure S17.** Simulated stratospheric Particle Size Distributions (PSD; averaged over June 2017) for the different model setups in: a) the tropical stratosphere ( $20^{\circ}\text{S}$ – $20^{\circ}\text{N}$ ) at 20 km, b) the Northern Hemisphere (NH) lowermost stratosphere (LMS;  $45^{\circ}$ – $80^{\circ}\text{N}$ ) at 15 km altitude, and c) the Southern Hemisphere (SH) lowermost stratosphere (LMS;  $45^{\circ}$ – $80^{\circ}\text{S}$ ) at 15 km altitude. The red dashed lines indicate the different modes in the TS setup. Note the differing y axis from Fig. S16.