Figure S1. Monthly ERA 5 winds (m s^{-1}) with monthly mean CAMS CO (ppb) at 950 hPa, 800 hPa, and 700 hPa illustrate aerosol transportation in a) June, b) July, c) August, and d) September. Red and orange stars denote the locations of Ascension Island and St. Helena Island. Grey contour lines denote sea-surface pressure (950 hPa) and geopotential height (800 hPa and 700 hPa).

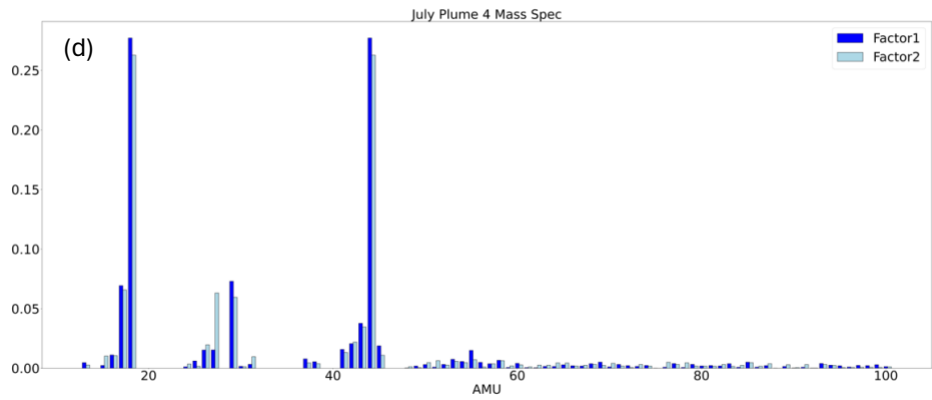
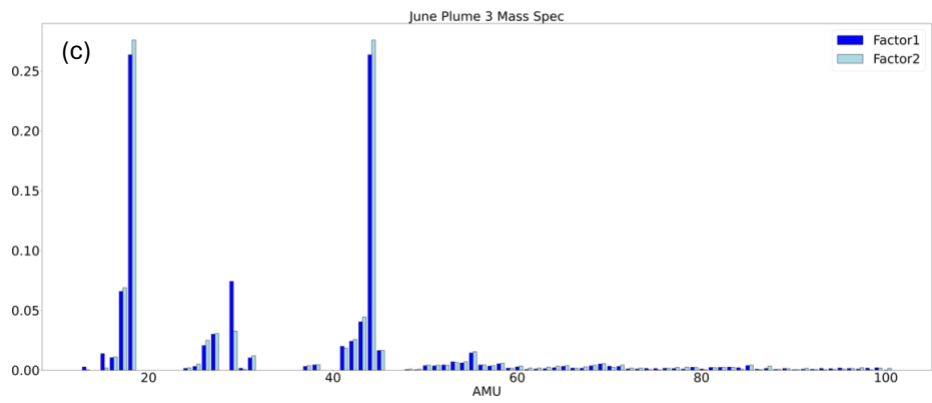
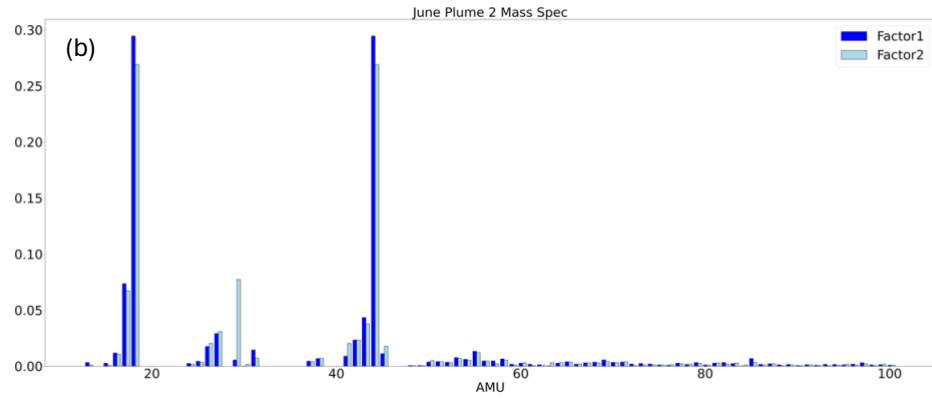
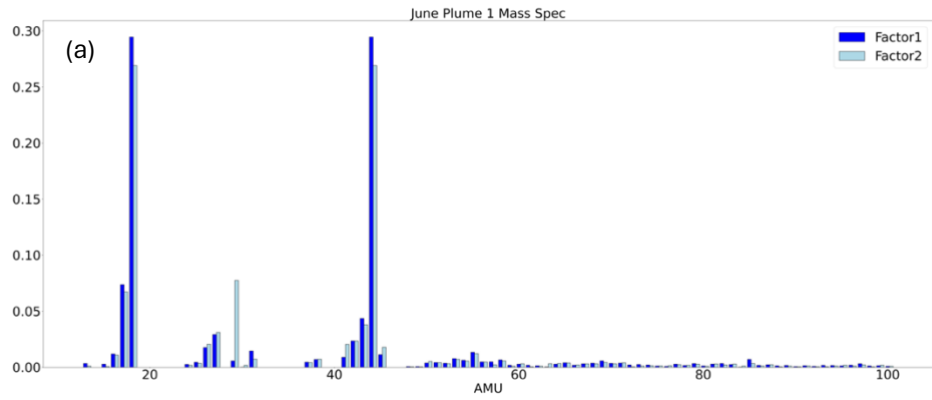
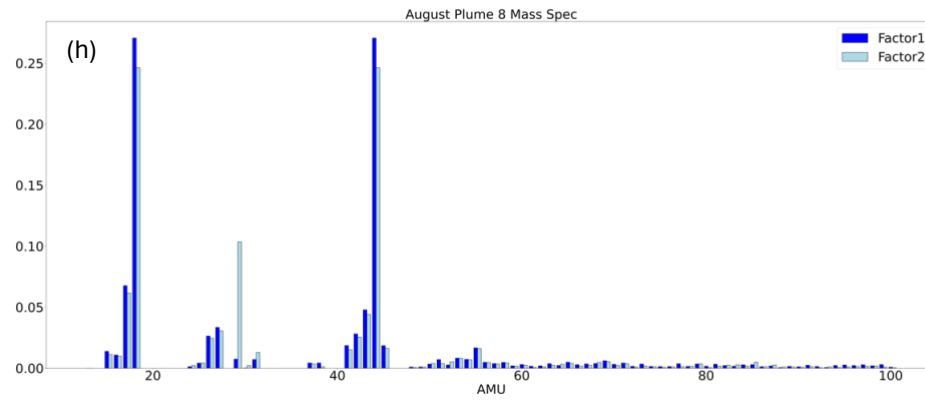
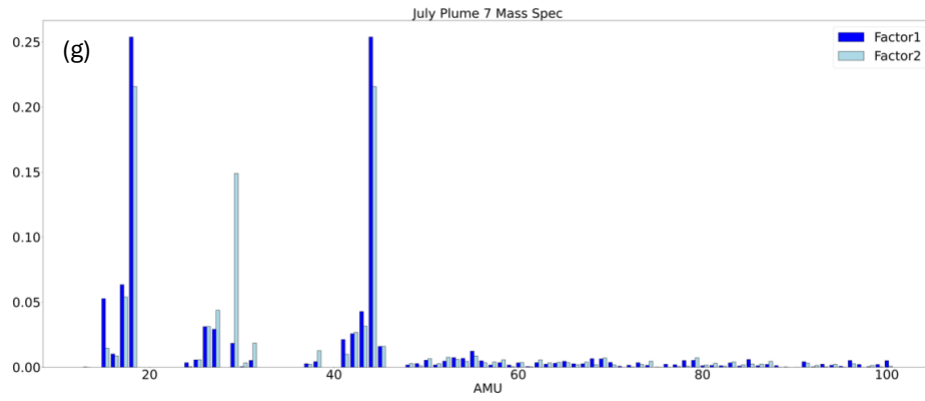
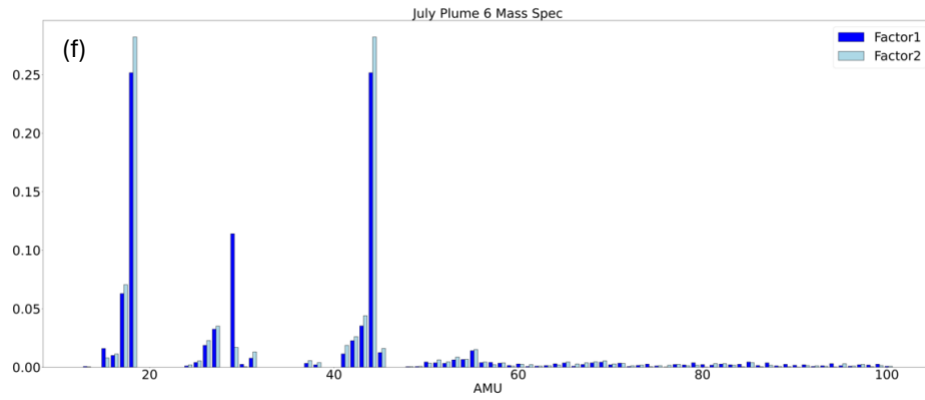
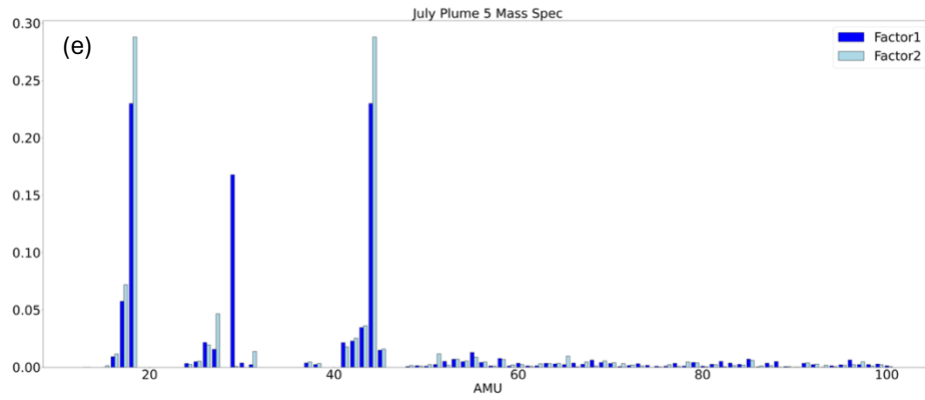
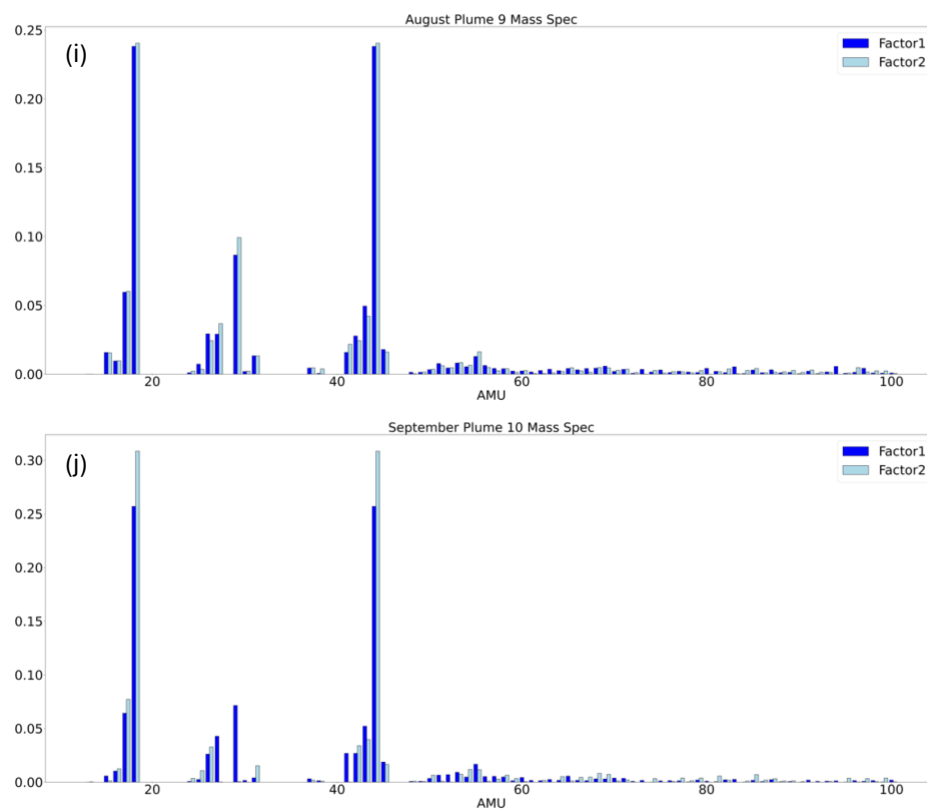



Figure S2. a-j. Average mass spectra from the ten selected plume events comparing factor 1(dark blue) and factor 2 (light blue).

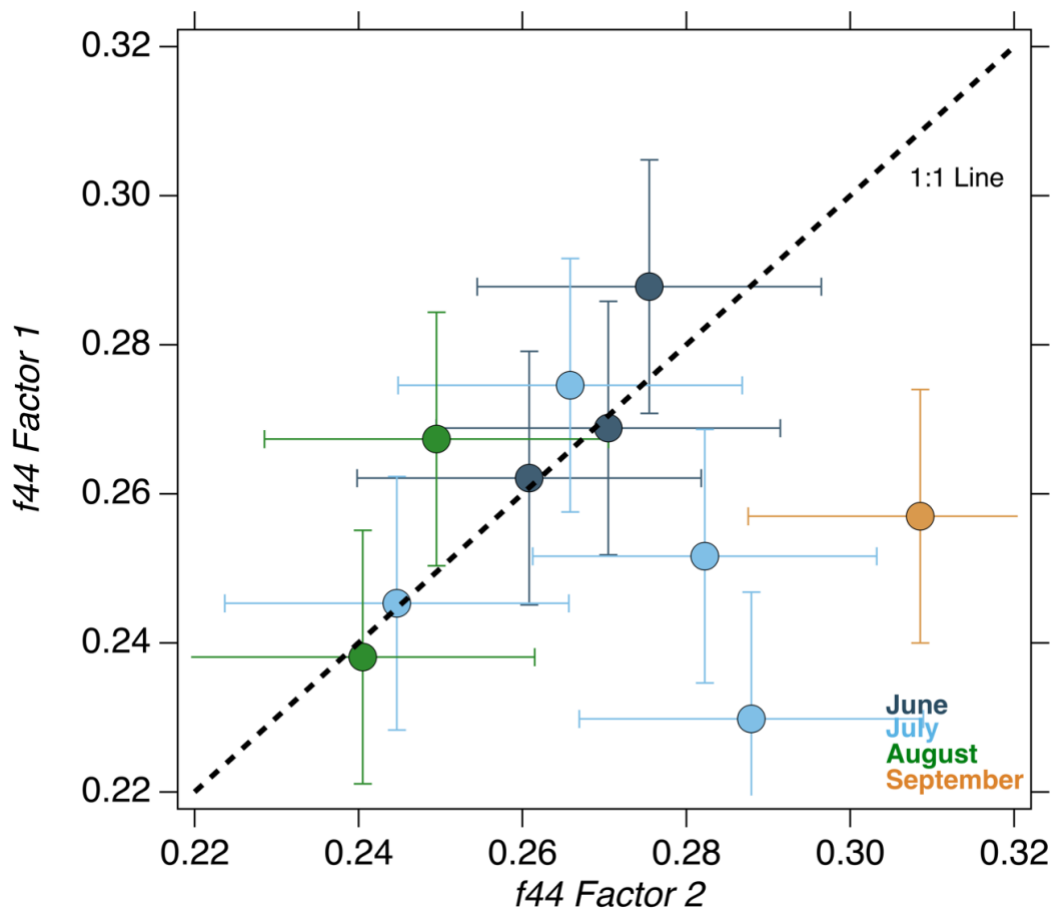
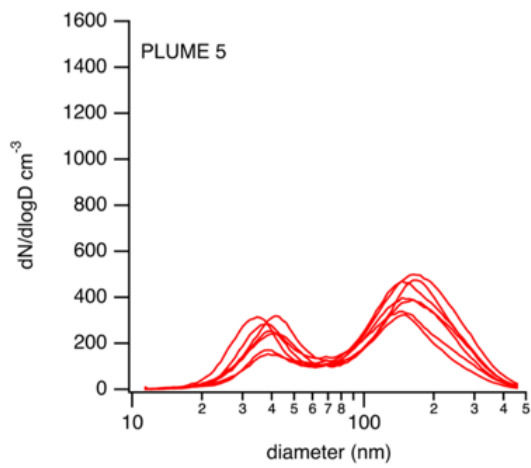
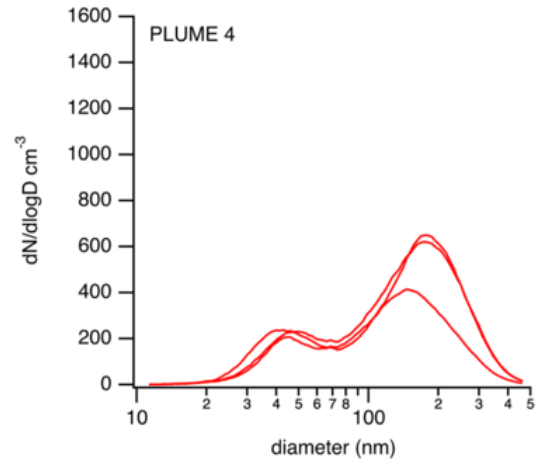
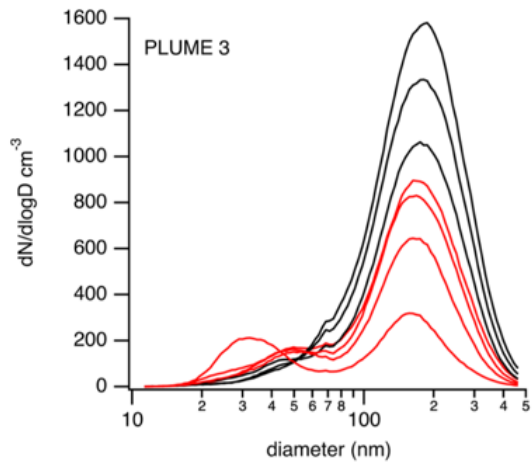
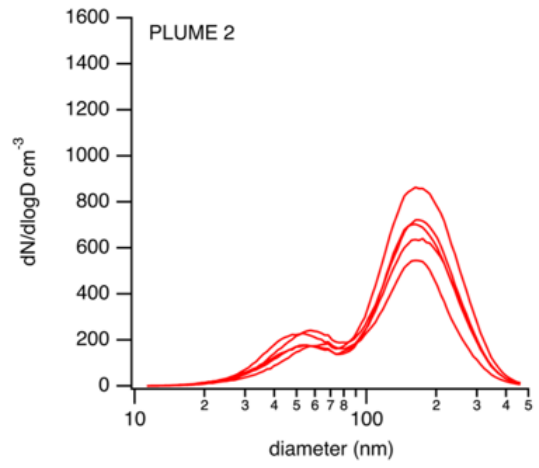
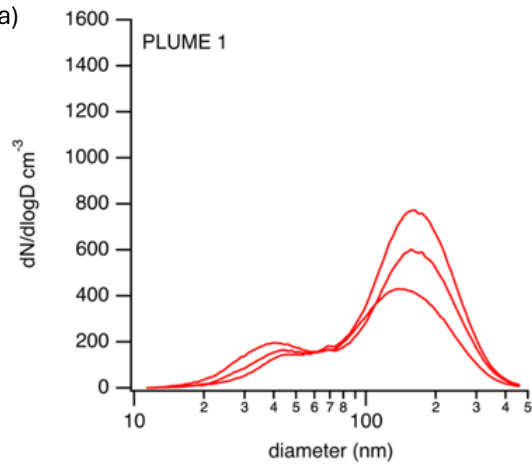



Figure S3. *f44* factor 1 versus *f44* factor 2 for the 10 selected plume events. Markers are colored by month and error bars represent the standard deviation of the data set. The black dashed line represents the 1:1 relationship.

(a)

(b)

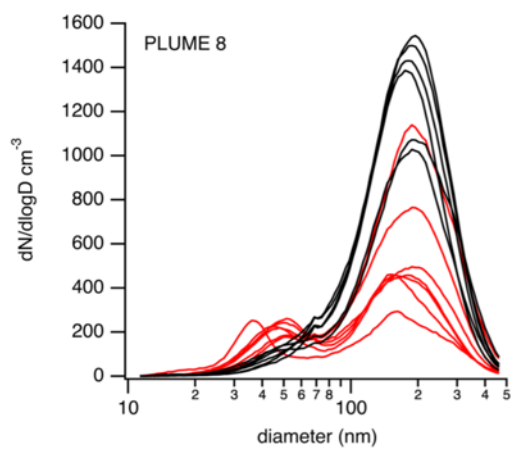
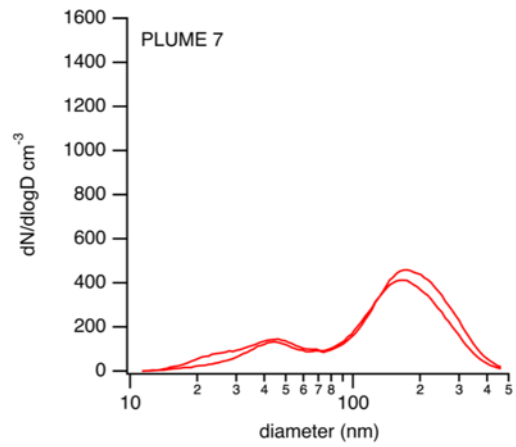
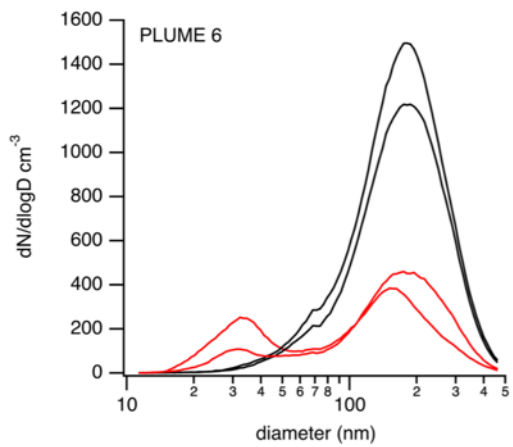
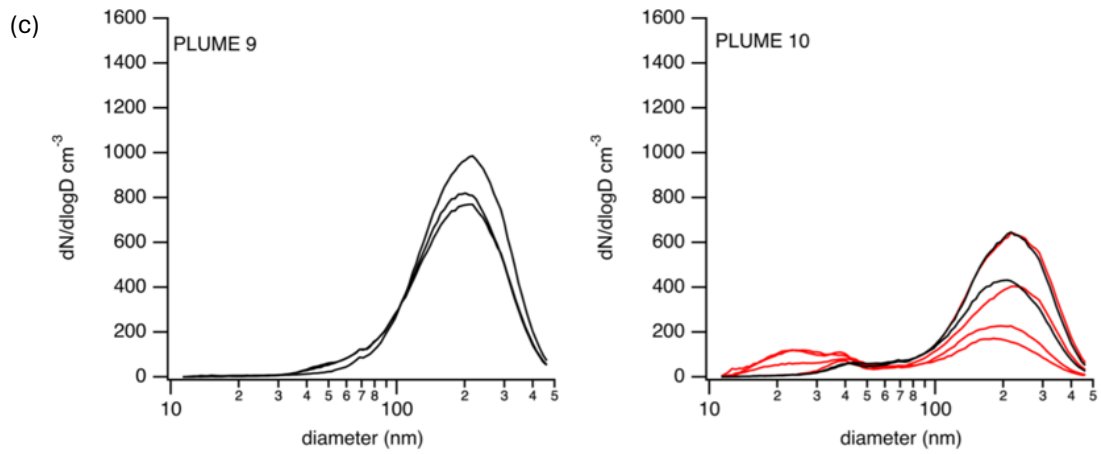



Figure S4. Daily average size for a) Regime 2, b) Regime 2, c) Regime 3. Red (black) size distributions represent bimodal (unimodal) distributions.

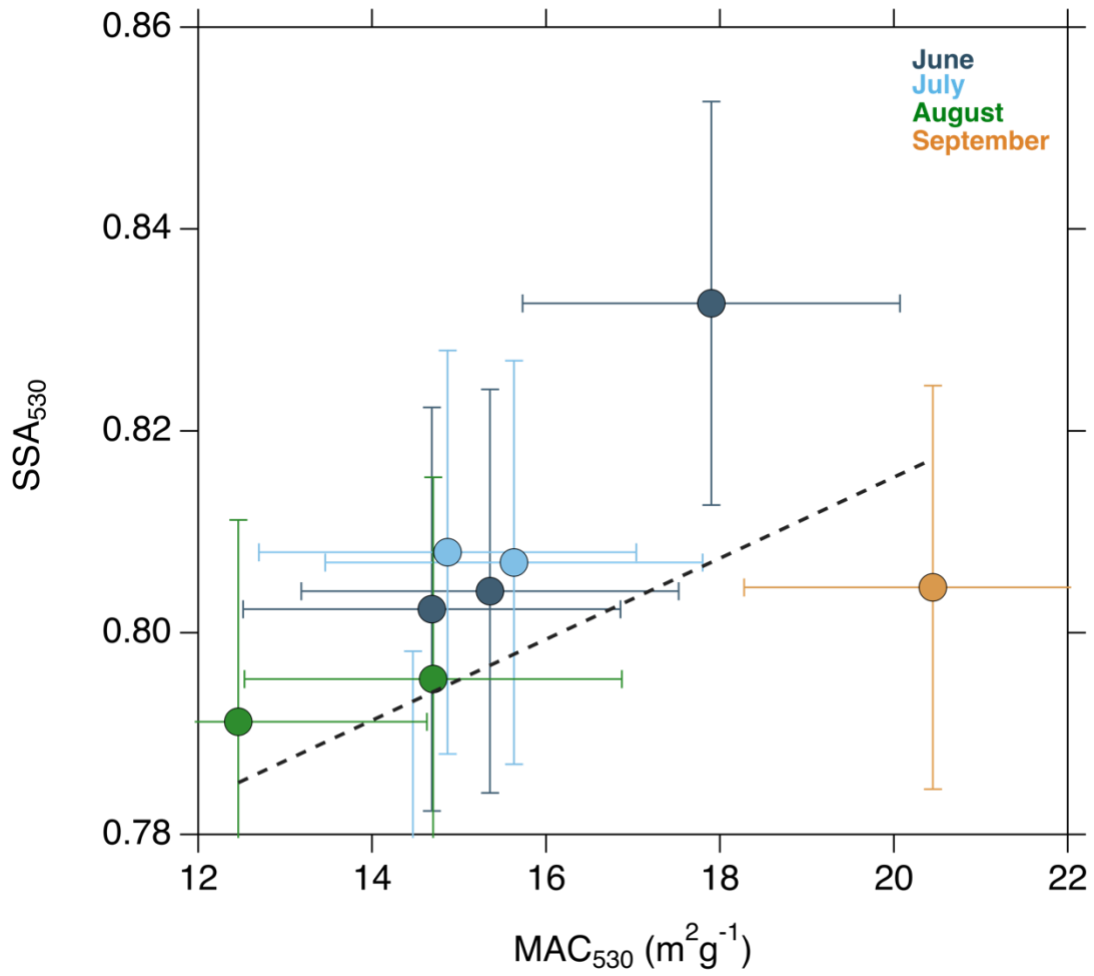



Figure S5. SSA$_{530}$ versus MAC$_{530}$ for the 10 selected plume events. Markers are colored by month and error bars represent the standard deviation of the data set. The black dashed line represents the best-fit line.

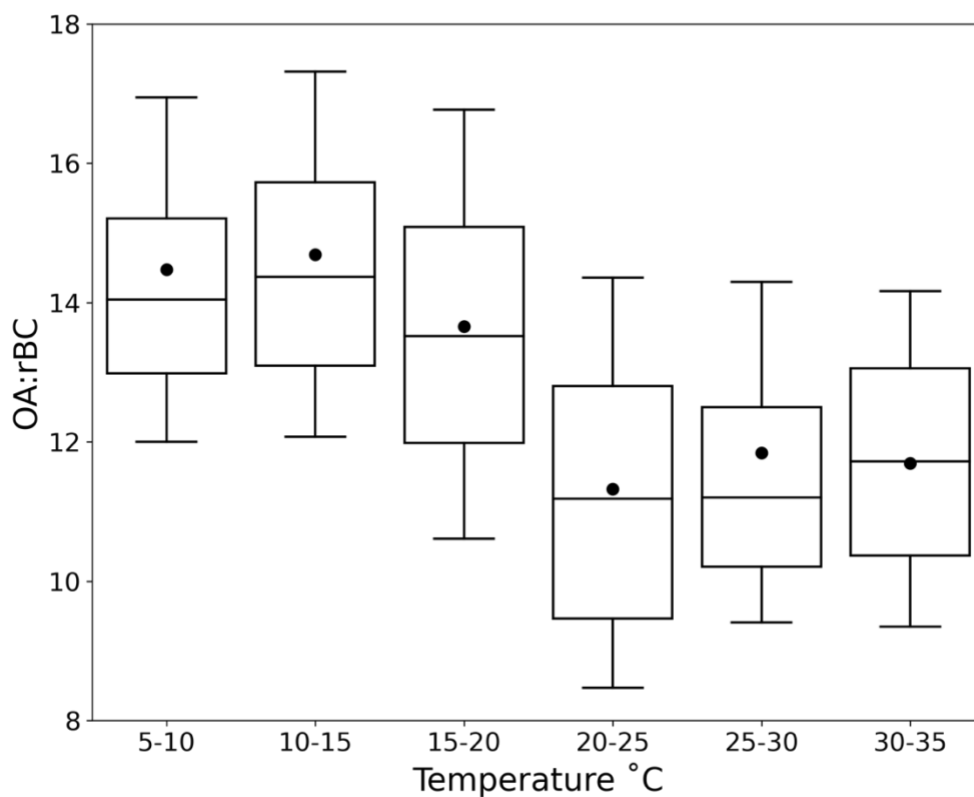



Figure S6. Box-whisker distributions using ORACLES aircraft data from 09/24/16 where OA>20µg m^{-1} to illustrate the dependence of OA:rBC on temperature (˚C). The FT data is used to demonstrate potential evaporation processes in the warm MBL. The lowest whisker is at the 10th percentile, with the lowest bar at 25th percentile, the middle bar at 50th percentile, the upper bar at 75th percentile, and the highest whisker at 90th percentile. Filled circles represent the mean.

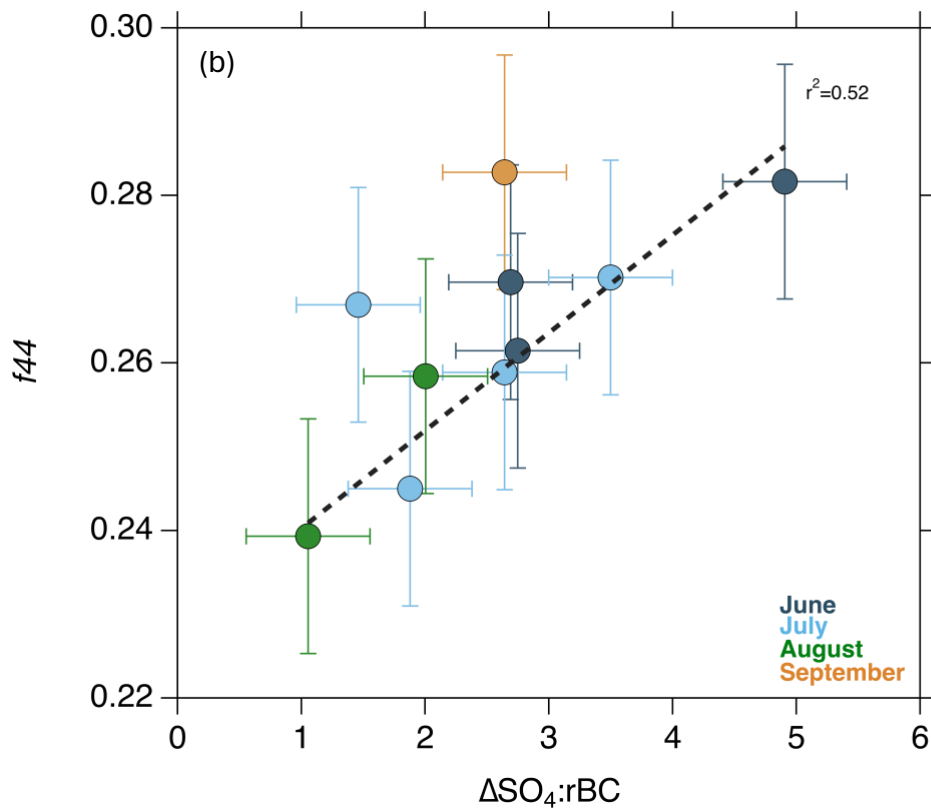
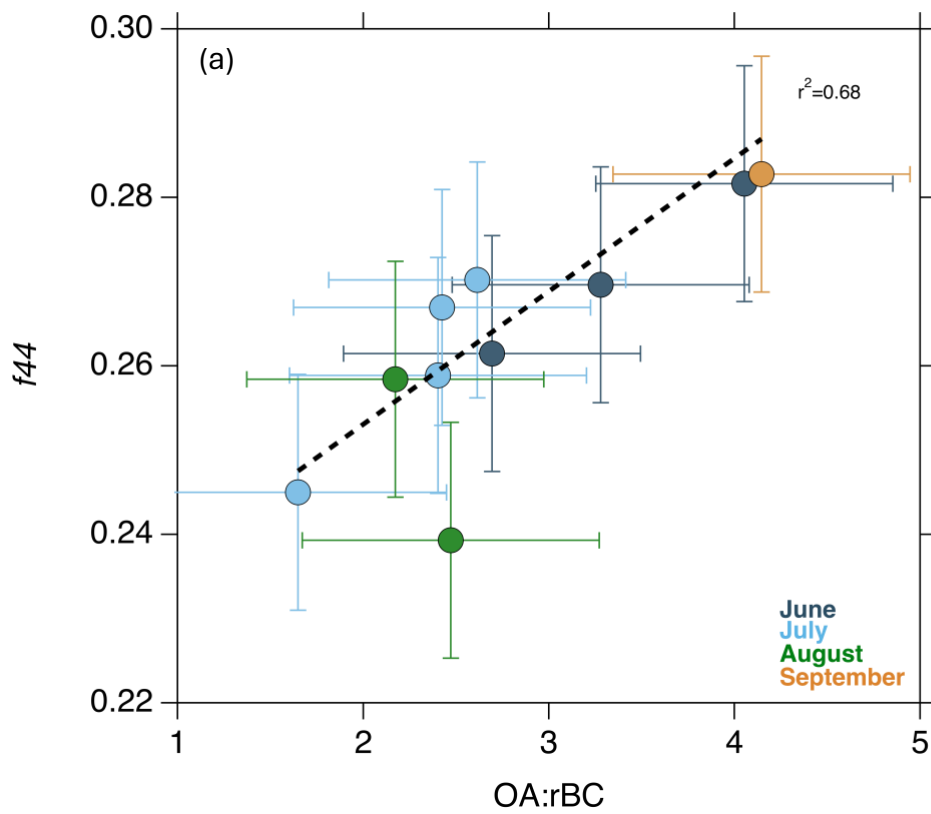



Figure S7. a) *f44* versus OA:rBC for the ten major plume events. b) *f44* versus SO₄:rBC for the ten major plume events. Markers are colored by month and error bars represent the standard deviation of the data set. The black dashed line represents the best-fit line.

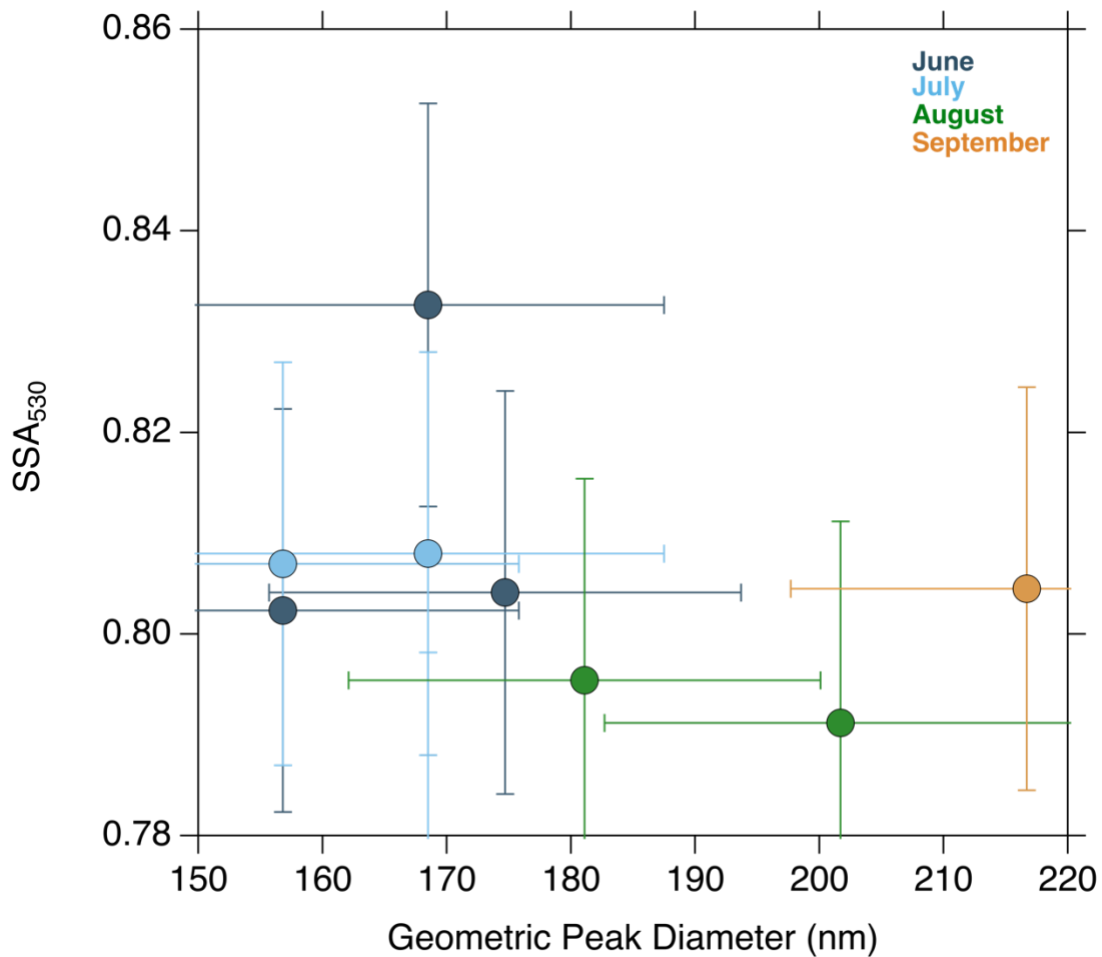



Figure S8. SSA$_{530}$ versus the geometric peak diameter of the accumulation mode (determined from the SMPS) for the ten plume events. Markers are colored by month and error bars represent the standard deviation of the data set.

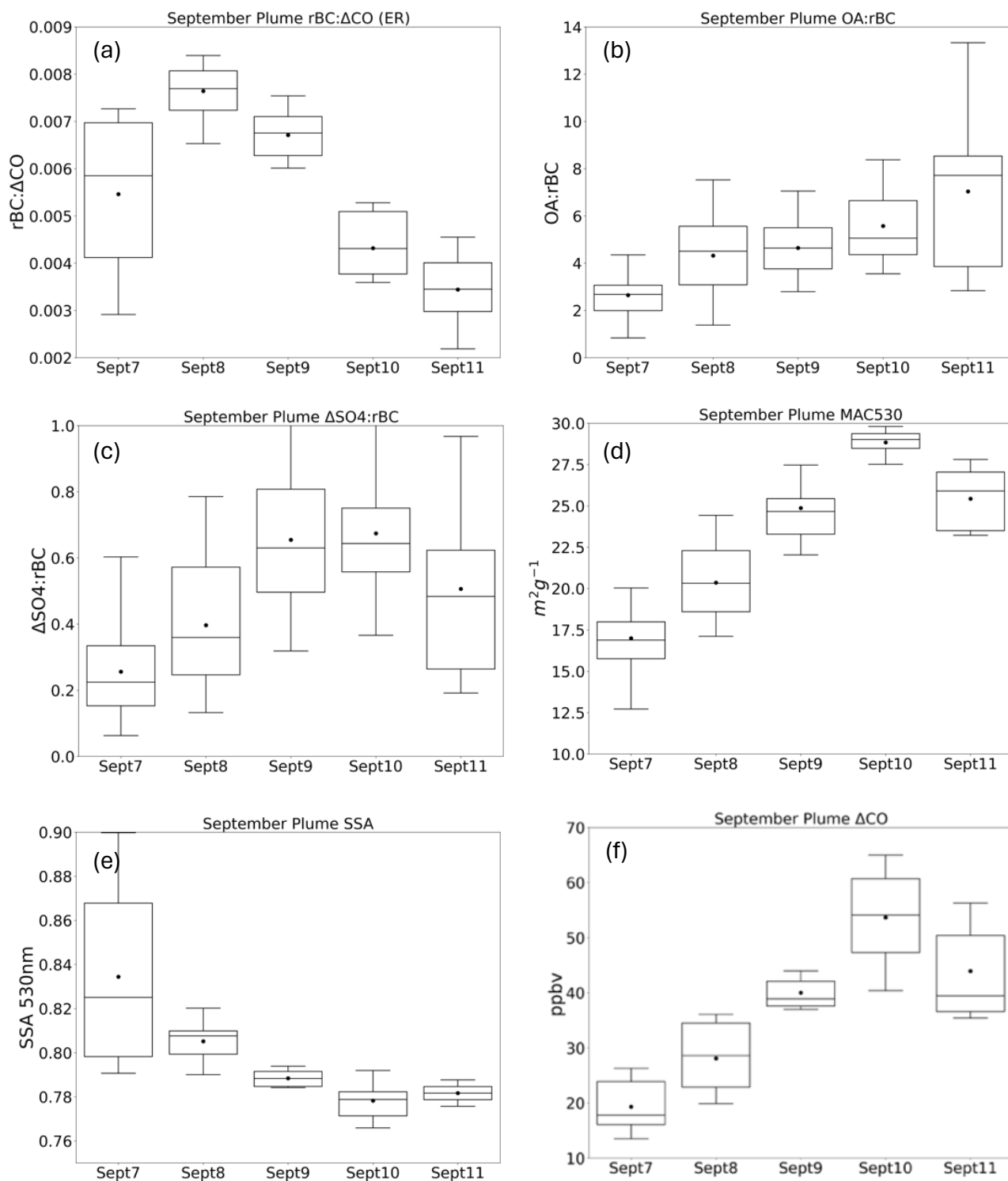



Figure S9. Daily averaged box-whisker distributions for a) rBC:ΔCO, b) OA:rBC, c) ΔSO_4:rBC, d) MAC_{530} (m^2 g^{-1}), e) SSA_{530}, f) ΔCO (ppb) for P10. Lowest whisker at 10th percentile, lowest bar at 25th percentile, middle bar at 50th percentile, upper bar at 75th percentile, and highest whisker at 90th percentile. Filled circles represent the mean. Red dashed lines distinguish the three regimes discussed in the text.

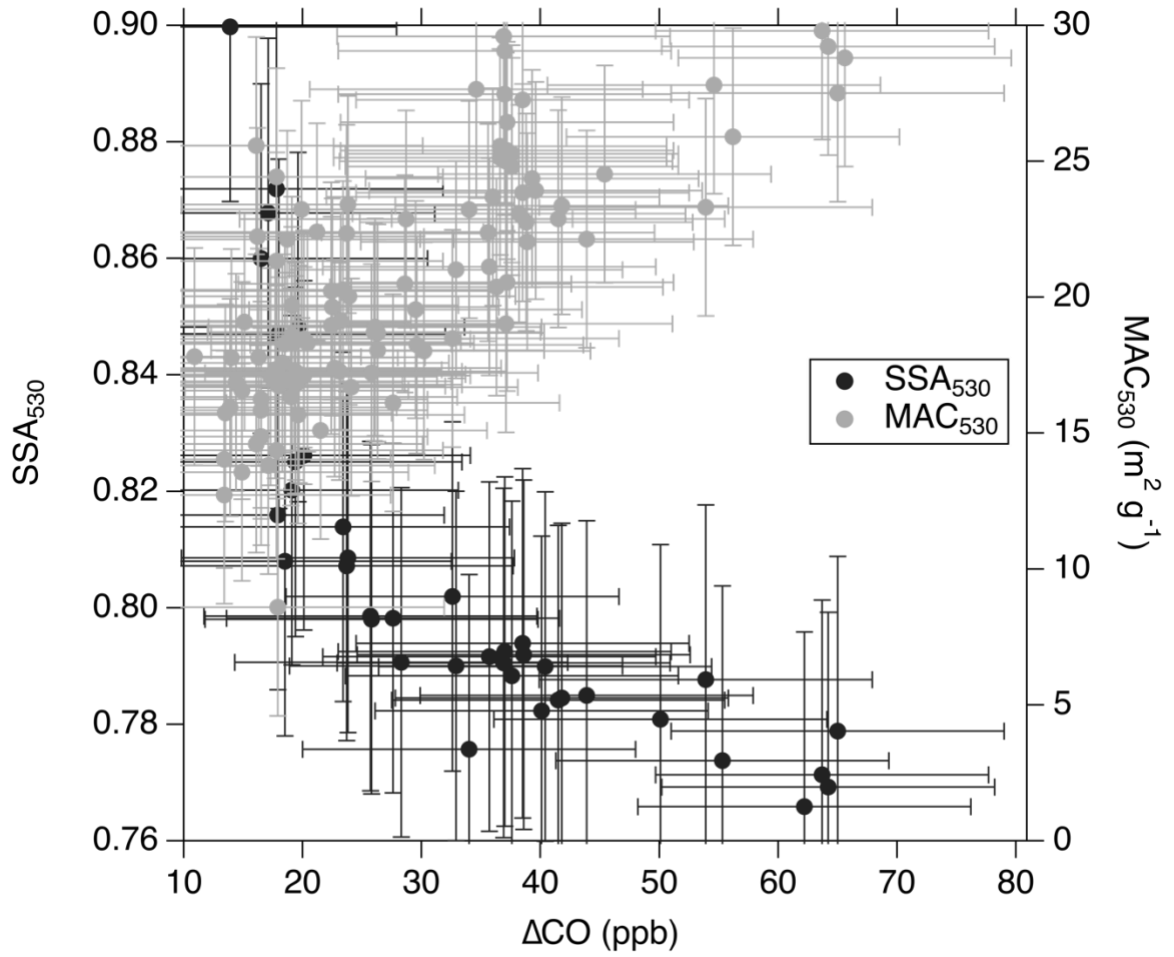



Figure S10. SSA$_{530}$ (black) and MAC$_{530}$ (grey) versus ΔCO for P10. Error bars represent the standard deviation of the data set.

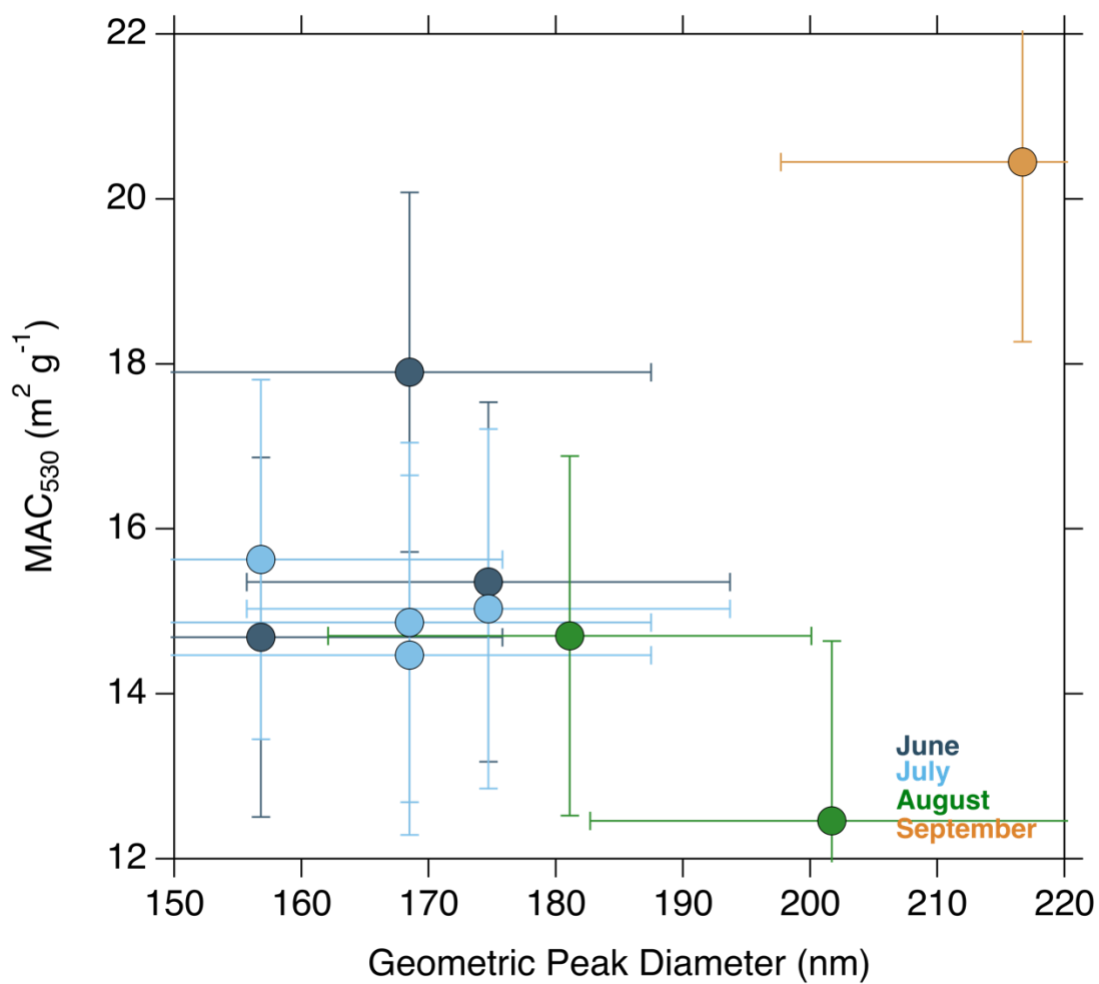



Figure S11. MAC$_{530}$ versus the geometric peak diameter of the accumulation mode (determined from the SMPS) for the ten plume events. Markers are colored by month and error bars represent the standard deviation of the data set.