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African smoke particles act as cloud condensation nuclei in the wintertime tropical North Atlantic boundary layer over Barbados

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Details on the K-Means Clustering Algorithm and the Particle Identification Process

Data from CCSEM/EDX analysis were imported into MatLab 2019b (MathWorks, Inc.) as a matrix of relative elemental abundances for each single particle in a sample. The data were then analyzed using k-means clustering in which groups of similar particles (clusters) are generated based on the presence and intensity of elemental peaks in the spectra of individual particles. To utilize k-means clustering, the user sets the number of clusters that the algorithm will generate. The k-means clustering algorithm will then generate clusters based on similarities between the elemental composition of individual particles. Cluster numbers were chosen based on the optimal trade-off between error minimization and chemical composition representativeness of the dataset after running the algorithm with different cluster numbers for the same sample.

To determine the identity of particles in a cluster, 4 plots are utilized that provide detailed information on size, shape and chemistry. These plots include a particle size distribution plot, a circularity plot, a weight matrix detailing the relative % of each of the 16 elements of interest in the particles, and a digital color stack plot that provides information on the distribution of elements throughout the particles in the cluster. An absolute intensity of 1% for elemental abundance presented in the weight matrix was used as a threshold value to consider an element present in a cluster. Upon identification of particle types, number fractions for each particle type can be calculated by summing together similar particle types and dividing by the total number of particles analyzed. These number fractions were generated for Fig. 4, 5, and 7. For figures 4 and 7, specific particle type number fractions were determined by dividing the number of specific particles in a particle type by the total number of particles analyzed in one day of sampling. For Fig. 5, number fractions were generated by dividing the number of particles of one particle type by the total number of particles in each size range plotted in the figure.

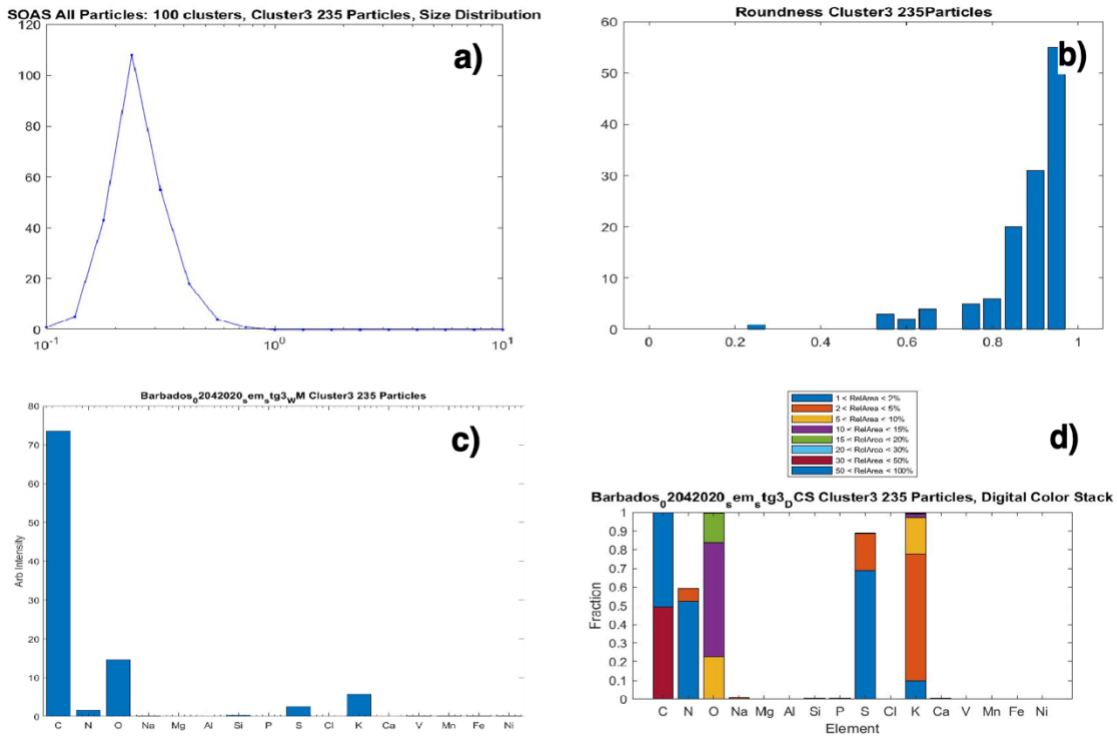

Figure S1 – Plots generated from the k-means clustering algorithm used for particle identification including a) a particle size distribution plot, b) a circularity plot, c) a weight matrix presenting relative abundance of elements in the cluster, and d) a digital color stack plot. The digital color stack plot describes the fraction of particles that contained each element of interest (height of bar) as well as the fraction of particles in the cluster in which an element made up a specific range of relative areas (size of colored bars and color of bar).

Temporal Chemistry Plots for the Supermicron Particle Loading

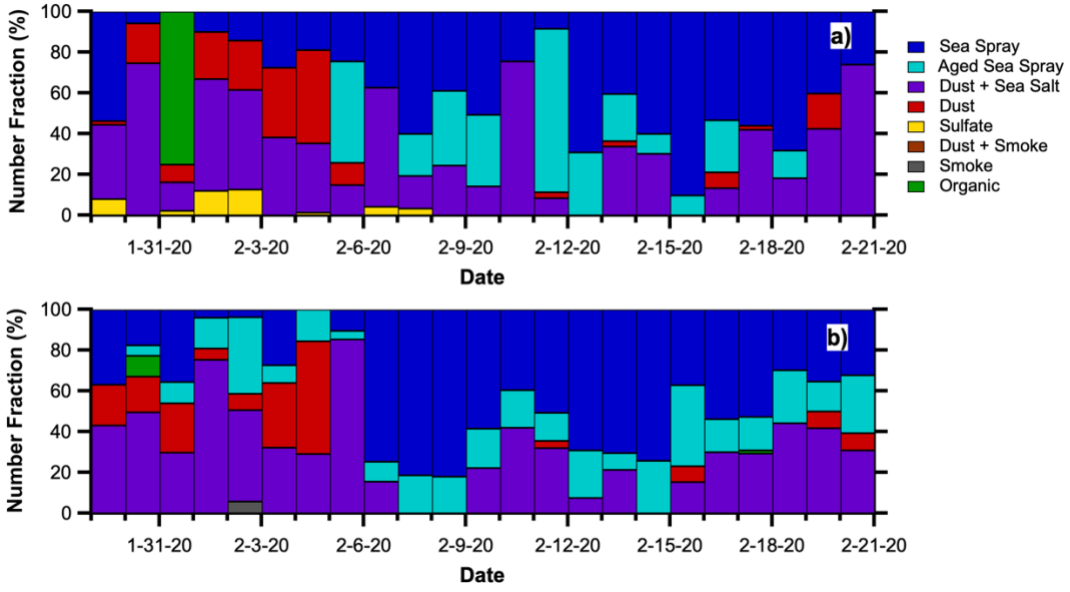

Figure S2 - Temporal evolution of particle type number fractions for a) stage 1 and b) stage 2 of the multistage particle sampler analyzed using CCSEM/EDX.

African Fire Distribution and Intensity During EUREC4A/ATOMIC

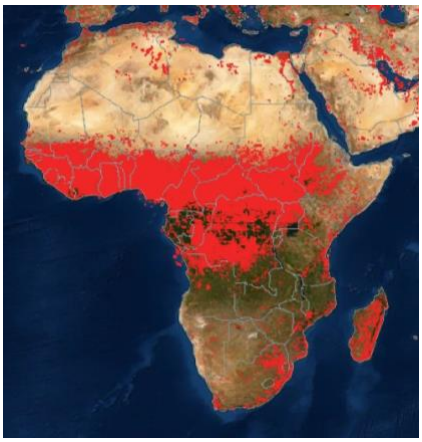

Figure S3 – Active fires (red shading) present between January 29th through February 20th of 2020 plotted using NASA's Fire Information for Resource Management System (FIRMS) model. Red shading is produced using NOAA's Visible Infrared Imaging Radiometer Suite (VIIRS NOAA-20), which shows active fire detections and thermal anomalies.

CO Column Density Measurements during EUREC^{4}A/ATOMIC

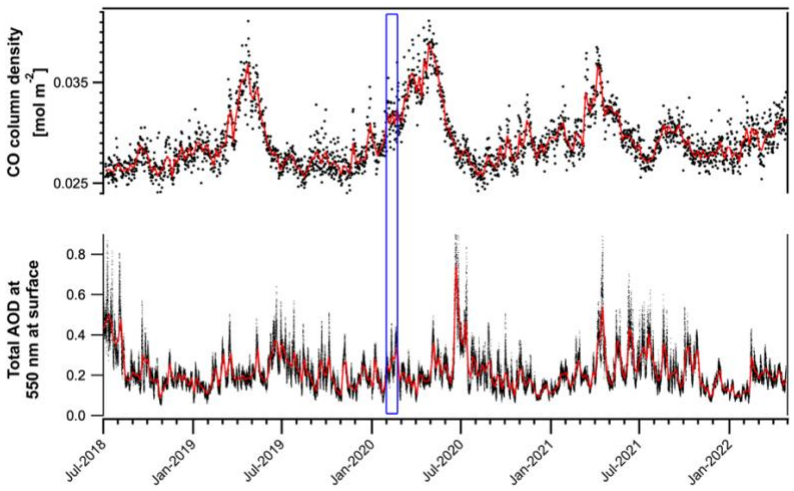

Figure S4 – Vertically integrated carbon monoxide (CO) column density measurements and total aerosol optical depth collected from July 2018 through January 2022 at BACO. CO measurements were obtained from the Sentinel-5P Near Real-Time Carbon Monoxide dataset. AOD data were obtained from the Copernicus Atmosphere Monitoring Service (CAMS). . Region outlined in blue box indicates time period for EUREC^{4}A/ATOMIC campaigns.

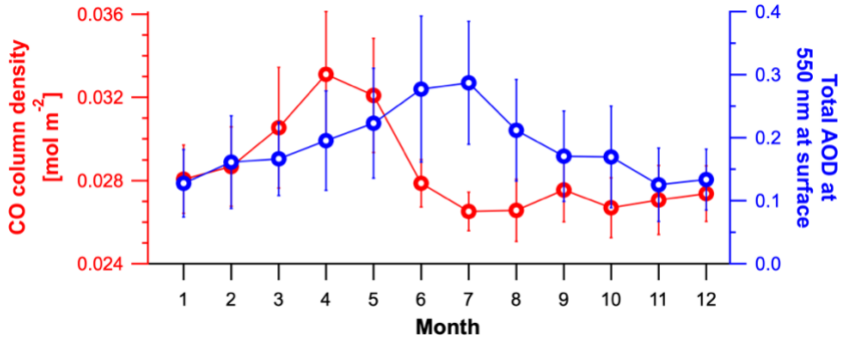

Figure S5 – Monthly averages for vertically integrated carbon monoxide (CO; a proxy for smoke) column density and aerosol optical depth collected at Barbados from 2018 – 2022, 2016 – 2022, and 1983 – 2022, respectively. CO measurements were obtained from the Sentinel-5P

Near Real-Time Carbon Monoxide dataset. AOD data were obtained from the Copernicus Atmosphere Monitoring Service (CAMS).

Size-Resolved Chemistry of CAT Event 2 and 3 During EUREC^{4}A/ATOMIC

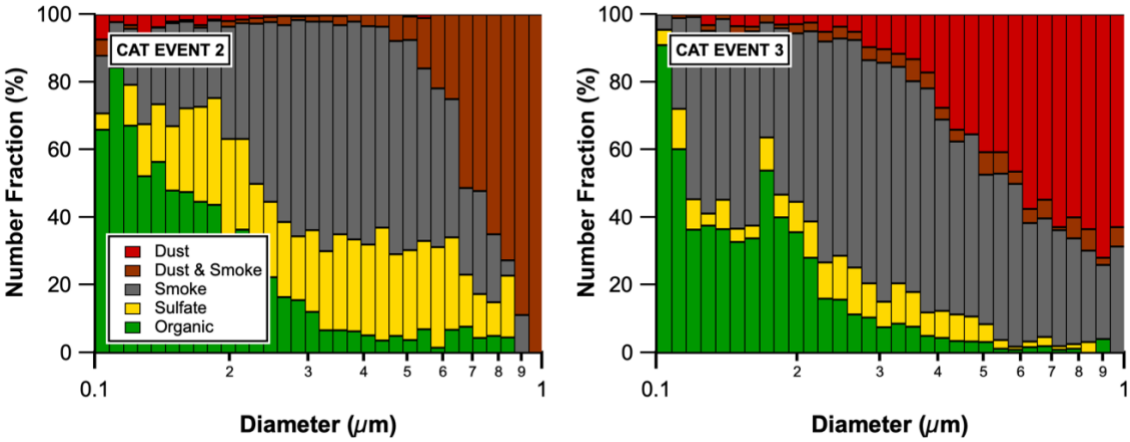

Figure S6 – Number fractions of 6 main submicron particle types plotted as a fraction of aerodynamic diameter (Da). CAT Event 2 and 3 includes size-resolved chemical data from the 2nd (2/10/2020 0:00 – 2/12/2020 6:00 GMT) and 3rd (2/15/2020 12:00 – 2/20/2020 18:00 GMT) period in which dust and wildfire smoke were observed over Barbados, respectively. Particle counts in bins for CAT Event 2 range from 9 to 476 particles, with an average bin size of 253 particles. Particle counts in bins for CAT Event 3 range from 22 to 792 particles, with an average bin size of 266 particles.

Average Particle Diameters for Submicron Aerosols

Sampling Period	Day/Time	Average Diameter (μm)					
		Organic	Sulfate	Smoke	Aged Sea Spray	Dust	Dust + Smoke
Clean Marine	2020/1/29 0:00 – 2020/1/29 12:00	---	0.40±0.15	0.26±0.11	---	0.70±0.29	---
CAT Event 1	2020/1/29 18:00 – 2020/2/6 6:00	0.18±0.08	0.26±0.15	0.27±0.13	---	0.56±0.29	0.48±0.34
Clean Marine	2020/2/6 12:00 – 2020/2/9 18:00	0.20±0.10	0.28±0.12	0.39±0.23	0.26±0.08	0.69±0.24	0.74±0.61
CAT Event 2	2020/2/10 0:00 – 2020/2/12 6:00	0.19±0.08	0.28±0.12	0.30±0.12	---	0.18±0.07	---
Clean Marine	2020/2/12 12:00 – 2020/2/15 6:00	0.23±0.08	0.30±0.15	0.22±0.08	---	---	0.62±0.35
CAT Event 3	2020/2/15 12:00 – 2020/2/20 – 18:00	0.19±0.08	0.27±0.11	0.29±0.16	---	0.56±0.30	0.39±0.29

Table S1 - Average submicron particle diameters for each particle type determined for each sampling condition observed during the field campaign.

Mean and Median values of Total Smoke Particle Count Analyzed with CCSEM/EDX

Sampling Period	Day/Time	Mean Smoke Particles Analyzed (#)	Median Smoke Particles Analyzed (#)
CAT Event 1	2020/1/29 18:00 – 2020/2/6 6:00	1985	1011
CAT Event 2	2020/2/10 0:00 – 2020/2/12 6:00	1094	848
CAT Event 3	2020/2/15 12:00 – 2020/2/20 – 18:00	948	500

Table S2 - Mean and median values of total smoke particles analyzed using CCSEM/EDX for each CAT Event observed throughout the field campaign.

Comparison of Smoke # Fraction and CCN counts for each day of EUREC4A/ATOMIC

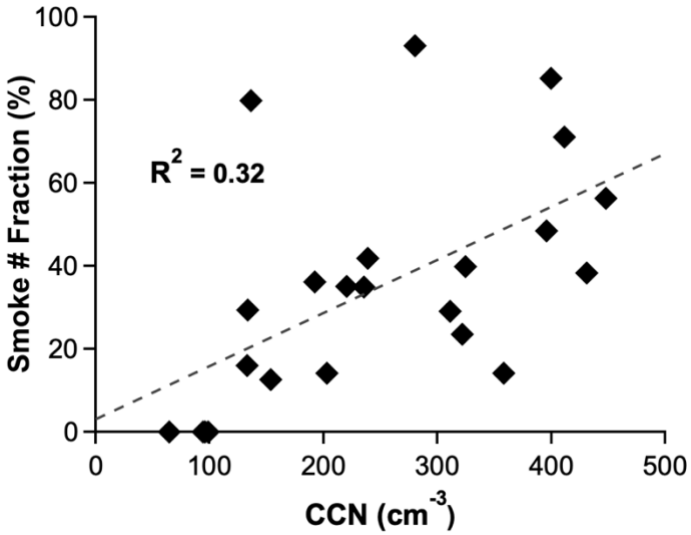

Figure S7 - Correlation plot comparing smoke particle number fraction from samples collected on stage 3 of the particle impactor and analyzed with CCSEM/EDX to CCN counts averaged for each day of the sampling period.