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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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Abstract

The number concentration and properties of aerosol particles serving as cloud condensation nuclei (CCN) are important for understanding cloud formation, particularly in the tropical Atlantic marine boundary layer (MBL) where marine cumulus clouds reflect incoming solar radiation and obscure the low-albedo ocean surface. Studies linking aerosol source, composition, and water uptake properties in this region have been conducted primarily during summertime dust transport, despite the region receiving a variety of aerosol particle types throughout the year. In this study, we compare size-resolved aerosol chemical composition data to the hygroscopicity parameter κ derived from size-resolved CCN measurements made during the EUREC^{4}A and ATOMIC campaigns from January to February 2020. We observed unexpected periods of wintertime long-range transport of African smoke and dust to Barbados. During these periods, the accumulation mode aerosol particle and CCN number concentrations as well as the proportions of dust and smoke particles increased while the average κ slightly decreased ($\kappa = 0.45 \pm 0.1$) from marine background conditions ($\kappa = 0.52 \pm 0.08$) when the particles were mostly composed of marine organics and sulfate. Size-resolved chemical analysis shows that smoke particles were the major contributor to the accumulation mode aerosol during long-range transport events, indicating that smoke is mainly responsible for the observed increase in CCN number concentrations. Earlier studies conducted at Barbados have mostly focused on the role of dust on CCN, but our results show that aerosol hygroscopicity and CCN number concentrations during wintertime long-range transport events over the tropical North Atlantic are affected by African smoke more than dust. Our findings highlight the importance of African smoke for atmospheric processes and cloud formation over the Caribbean.

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Introduction

Aerosol particle number, size, hygroscopicity, and chemical mixing state determine cloud droplet formation and, thus, fundamentally affect the radiative properties and lifetime of clouds (Albrecht, 1989; McFiggans et al., 2006; Quinn et al., 2008; Twomey, 1977; Zuidema et al., 2008). Quantifying the effect of aerosols on cloud radiative forcing, however, is still the single largest source of uncertainty in predicting temperature increases associated with climate change (Forster et al., 2021). This uncertainty is especially important to resolve in marine regions where aerosol-cloud interactions are understudied, even though the majority of Earth's surface is covered by oceans (Carslaw et al., 2013). The existing literature that explores marine aerosol-cloud interactions does so primarily in the mid to high latitudes of the North Atlantic with few studies focusing in tropical latitudes where shallow cumulus clouds form (Allan et al., 2008; Behrenfeld et al., 2019; Klingebiel et al., 2019; Rauber et al., 2007; Sorooshian et al., 2020). Shallow cumulus clouds are important for Earth's climate as they are one of the most geographically pervasive cloud types and can influence Earth's radiative budget by reflecting incoming radiation over the low-albedo ocean surface.

Aerosol research conducted in the tropical Atlantic has focused mostly on the long-range transport of mineral dust from North Africa in the summertime. Long-range African dust transport occurs when emitted desert dust is lofted above the marine boundary layer (MBL) into the Saharan Air Layer (SAL) and is propagated westward (Carlson & Prospero, 1972). As dust is transported westward, it can mix into the underlying moist MBL and deposit into the Atlantic Ocean and Caribbean Sea as well as Western Atlantic land masses such as South America, the Caribbean islands, and North America (Barkley et al., 2019; Carlson & Prospero, 1972; Prospero et al., 1981, 2020). Some studies have attempted to understand the effects of long-range transported

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dust on cloud droplet formation and water uptake with varying results depending on the degree of aging that dust experiences during transport (Allan et al., 2008; Denjean et al., 2015; Rosenfeld et al., 2001). Ultimately, though, these studies provide conflicting results on whether dust particles are hygroscopic and numerous enough to impact CCN concentrations and, thus, do not thoroughly explain aerosol-cloud interactions in the tropical Atlantic. Due to the annual oscillation of the intertropical convergence zone (ITCZ), dust transport also exhibits a seasonality in terms of its geographic extent (Adams et al., 2012; Chin et al., 2014; Prospero & Lamb., 2003; Prospero, 1968; Prospero & Mayol-Bracero, 2013; Yu et al., 2019; Zuidema et al., 2019). However, marine shallow cumulus clouds form year-round in the tropical Atlantic regardless of dust transport, suggesting that other marine or other long-range transported sources are important for aerosol-cloud interactions in this region (McCoy et al., 2022).

Few studies have attempted to fully understand which aerosols are the most prominent CCN both during dust seasons and non-dust seasons in the tropical North Atlantic MBL. One particle type that may be important for CCN activation in the tropical north Atlantic yet has been understudied at dust receptor sites such as Barbados is African smoke (Wex et al., 2016). In contrast to dust, previous research has shown that smoke particles are an important source of CCN (Edwards et al., 2021; Lathem et al., 2013; Pierce et al., 2007; Spracklen et al., 2011) with more recent research showing that some smoke particles can activate at supersaturations as low as 0.05%.

There are a number of reasons that explain why smoke particles can be effective CCN. Smoke particles are often complex mixtures of both organic and inorganic components that change compositionally and morphologically during their residence time in the atmosphere (Reid et al., 2005). Smoke properties may also vary between fires depending on fuel type and moisture,

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94 combustion phase, wind conditions, etc. (Andreae, 2019; Miles et al., 1995; Reid et al., 2005). In
95 general, smoke particles are often found in the accumulation mode of the aerosol size
96 distribution and primarily contain particulate organic matter, black carbon, and inorganic
97 components including potassium chloride salts (Reid et al., 2005). Upon emission, smoke can
98 undergo chemical processing through photochemical and heterogeneous reactions, including the
99 loss of chloride and acquisition of sulfate and nitrate, creating potassium sulfate compounds in
100 smoke that are often used as tracers of aged smoke and can affect the hygroscopicity of smoke
101 particles (Capes et al., 2008; Hennigan et al., 2010, 2011; Reid et al., 2005; Zauscher et al.,
102 2013). The many variations and changes in the chemical and physical properties of smoke
103 particles during their residence time in the atmosphere makes it difficult to study these particles
104 thoroughly and can also affect their ability to act as CCN.

105 In this study, we investigated the relationship between submicron aerosol composition
106 and CCN in the tropical north Atlantic MBL during marine background conditions and
107 conditions affected by long-range continental aerosol transport (including dust and smoke). To
108 perform this work, we collected aerosol samples and size-resolved CCN data from January to
109 February 2020 at the Barbados Atmospheric Chemistry Observatory (BACO) during the
110 Elucidating the Role of Clouds–Circulation Coupling in Climate/Atlantic Tradewind Ocean-
111 Atmosphere Mesoscale Interaction Campaign (EUREC^{4}A/ATOMIC) campaigns (Quinn et al.,
112 2021; Stevens et al., 2021). Conducting this research during the boreal winter provided a unique
113 opportunity to explore aerosol-cloud interactions in different meteorological conditions than are
114 typically studied in the tropical North Atlantic. Dust primarily arrives to Barbados during the
115 summer months with peaks in June and July (Zuidema et al., 2019). As a result, dust receptor
116 sites in Barbados have historically been used to compare dust and marine background conditions

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during the boreal summer. During the winter, the southward shift of the ITCZ directs African dust to South America, resulting in a decrease in dust concentrations over Barbados during the winter months with days in December and January sometimes receiving no dust at all (Prospero, 1968; Prospero et al., 2014; Prospero & Lamb, 2003; Prospero & Mayol-Bracero, 2013). However, during the EUREC^{4}A/ATOMIC campaigns, we observed anomalous wintertime transport of African aerosols to Barbados, which provided novel sampling conditions to study the effects of various aerosol types on cloud droplet formation. Specifically, we were able to explore marine aerosols such as organics, sulfates, and sea salt and how the addition of continental aerosols like mineral dust and smoke particles affects CCN activity, thus comparing the impact of ocean-derived vs. long-range transported aerosol on water uptake properties and CCN concentrations. We conclude this manuscript by discussing the importance of our findings for cloud formation in the tropical North Atlantic.

Methods

Measurement Site and Sampling Period

Aerosol samples and size-resolved CCN data were collected at the Barbados Atmospheric Chemistry Observatory (BACO) on Ragged Point during the EUREC^{4}A and ATOMIC field campaigns from January 20, 2020 -February 20, 2020 (Quinn et al., 2021; Stevens et al., 2021). Ragged Point (13° 6' N, 59° 37' W), a prominence on Barbados' east coast, is an ideal location for studying the impact of long-range African aerosol transport on cloud-aerosol interactions as it is situated on the most easterly island in the Caribbean and is exposed to the steady easterly trade winds. Thus, the east coast of the island is subject to little anthropogenic aerosol influence from local islands to the west (Prospero et al., 2005; Savoie et al., 2002). Further, the island is at a latitude coinciding with the outflow of African aerosols such as mineral

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dust (Carlson & Prospero, 1972; Prospero, 1968) and biomass burning (Archibald et al., 2015) as well as tropical marine cumulus clouds (Stevens et al., 2016).

Air Mass Origins

During the sampling period, air masses of varying composition were observed at Ragged Point. To determine the origin of these air masses, 150 h back trajectories were generated every 6 hours (h) at heights of 500, 1000, and 1500 meters (m) throughout the campaign using the Hybrid Single Particle Lagrangian Integrated Trajectory (HYSPLIT) model calculated using model vertical velocity and meteorology from the National Center for Environmental Prediction (NCEP) 1-degree Global Data Assimilation System (GDAS) (Rolph et al., 2017; Stein et al., 2015).

Dust Concentration

To collect aerosols, BACO is equipped with a high-volume sampler and an isokinetic aerosol inlet on top of a 17 m tall tower situated on a 30 m bluff along the coast at Ragged Point. Daily dust mass concentrations were determined from filter-based measurements (Prospero et al., 2021; Zuidema et al., 2019) using a high-volume air sampler pumping at a rate of approximately 0.7 m^3/min across a 20 cm x 25 cm cellulose Whatman-41 (W-41) filter. W-41 filters were chosen for this analysis as they allow for high flow rates and yield a collection efficiency of 95% or better for dust (Kitto & Anderson, 1988). Upper particle diameter limits for W-41 filters with 20 µm pore size are approximately 80-100 µm or greater (Barkley 2021). After aerosol collection, the filters are placed in a furnace and combusted at 500°C for about 12 hrs. (i.e., overnight). Procedural blanks are also collected by placing a filter in the sampler for 15 minutes without turning on the pump. The resulting ash mass minus the mass of a filter blank is the approximate

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amount of mineral dust collected on the filter during the sample period. A correction factor of 1.3 is applied to the calculated dust concentrations to account for dust components such as bound water or soluble ions that are lost during the heating process (Prospero, 1999; Zuidema et al., 2019).

Aerosol Chemical Composition

Aerosol particles were sampled at ambient relative humidity (RH) through an isokinetic aerosol inlet and collected using a three-stage microanalysis particle sampler (MPS-3, California Measurements, Inc.), which samples particles from diameters of 5.0-2.5 µm (stage 1), 2.5 µm – 0.7 µm (stage 2), and <0.7 µm (stage 3). For each set of samples (1 set including 1 sample from each stage of the MPS), the MPS was run for 45 min at 2 L/min flow starting at approximately 9:30 local time or 13:30 coordinated universal time (UTC). Meteorological data from a local station was also used to manually check that wind direction fell between 335° and 130° and wind speeds were greater than 1 m/s during all sampling periods. Sampling during these wind conditions ensures that only air from the open ocean was sampled rather than local, anthropogenically-influenced air.

Particles were deposited onto carbon-coated copper grids (Ted Pella, Inc.) that were later analyzed at the Pacific Northwest National Laboratory using computer-controlled scanning electron microscopy coupled with energy dispersive x-ray spectroscopy (CCSEM/EDX; Quanta 3D) to determine the elemental composition of individual particles. Here we focus only on the submicron particle population which exerts a greater influence on CCN number concentrations and is more sensitive to chemical changes that affects its hygroscopicity. Thus, for this study we only present data from stage 3 of the MPS, representing <0.7 µm diameter particles. Data for the other stages will be the subject of a future manuscript.

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EDX is considered a semiquantitative method providing the relative atomic fractions for elements of interest. Percent composition threshold values of 1% were used to ensure the presence of elements detected by the EDX. Elements of interest in CCSEM/EDX analysis were limited to 16 common elements indicative of organic material, sea spray, dust, and anthropogenic emissions: carbon (C), nitrogen (N), oxygen (O), sodium (Na), magnesium (Mg), aluminum (Al), silicon (Si), phosphorus (P), sulfur (S), chlorine (Cl), potassium (K), calcium (Ca), vanadium (V), manganese (Mn), iron (Fe), and nickel (Ni). The EDX peak for Cu is heavily influenced by a background signal from the Cu grid and is excluded from analysis. Samples collected on Si substrates confirmed the validity of the C signal in analyzed particles, as the carbon coating on the Cu substrates has the potential to generate a background signal as well. An excess of 1000 particles were analyzed in each sample. Due to size limitations of the CCSEM, only particles with diameters >0.1 μm were analyzed. Data products from CCSEM/EDX analysis were then analyzed in MATLAB (ver 9.6.0; The Mathworks, Inc.) using a K-means clustering algorithm (Ault et al., 2012; Shen et al., 2016). The algorithm operates by generating categories of similar particles (clusters) based on the presence and intensity of elemental peaks in individual single-particle EDX spectra. These clusters are then assigned to particle types based on their size, morphology, characteristic EDX spectra, and existing literature. Particle types typically observed in the supermicron aerosol loading, such as sea salt and dust, are not as abundant in our samples as we focus exclusively on the submicron aerosol loading.

Size-Resolved CCN Measurements and Data Analysis

To determine the size-resolved CCN activity of aerosol particles during the sampling period, we used a continuous-flow streamwise thermal gradient CCN counter (CCNC, model

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CCN-100, DMT, Longmont, Co, USA; (Roberts & Nenes, 2005; Rose et al., 2008)) combined with a differential mobility analyzer (DMA, model M, Grimm Aerosol Technik, Ainring, Germany) and condensation particle counter (CPC, model 5412, Grimm Aerosol Technik). Upon entering the system, sampled air was dried using a condensation drier to maintain a relative humidity (RH) between 20 and 30% and to ensure reliable hygroscopicity measurements. After drying, particles passed through a DMA which selected particles with a diameter (D) between 20 and 245 nm. The monodisperse aerosol-laden flow was then split between the CCNC and CPC. Inside the CCNC, particles were subjected to water vapor supersaturations (S) including 0.09, 0.16, 0.24, 0.43, and 0.74 %.

Calibrations of the CCNC supersaturations were performed according to the method described in Rose et al., 2008 by generating and size-selecting ammonium sulfate particles that were analyzed by the CCNC set to a designated temperature gradient as well as a CPC to measure total condensation nuclei (CN) values. Plots comparing CCN/CN and dry particle diameter were then used to determine the diameter at which 50% of the particles in an aerosol population activate as CCN at a particular S, also called the critical activation diameter (d_{50}). D_{50} values were then used to determine supersaturation. Supersaturations were plotted against the designated temperature at the calculated supersaturation. The resulting plot provided a linear curve that could be used to adjust the supersaturation shown by the instrument to the actual value of the column supersaturation. After calibrating, S values averaged 0.08, 0.15, 0.23, 0.41, and 0.71.

For ambient sampling, particles that activate as CCN at each S and D are counted in the CCNC as CCN, while all particles of a selected D are counted in the CPC to determine the total aerosol concentration of particles at each D. By scanning D at a given value of S, measurements

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231 from the CPC and CCNC are then used to calculate the d_{50}. These values, along with the particle
232 number size distribution determined by a scanning mobility particle sizer (SMPS, TSI model
233 3080 with CPC 3772) operating independently of the CCNC setup, are then used to calculate the
234 activation curve and the effective hygroscopicity parameter κ using equation (1) according to the
235 κ-Köhler model (Petters & Kreidenweis, 2007):

236 $$\kappa = \frac{4A^3}{27D_p^3 \ln^2 S_{crit}} \quad (1)$$

237 where D_p is the dry particle diameter, S_{crit} is the supersaturation set by the CCN counter, and A is
238 the Kelvin term calculated from equation (2):

239 $$A = \frac{4\sigma M_w}{RT\rho_w} \quad (2)$$

240 Where σ is the surface tension (σ=0.072 J/m^2), R is the universal gas constant, M_w is the
241 molecular weight of water, and ρ_w is the density of water. In the κ-Köhler model, higher values
242 of κ indicate a more hygroscopic particle that is more efficient at taking up water and can
243 activate as CCN at lower S. Calculations of activation curves, size-resolved CCN, CCN
244 efficiencies, and errors are described in detail in Pöhlker et al., 2016.

245 **Results and Discussion**

246 We find that, upon arrival of co-transported dust and smoke, smoke originating from fires
247 in the African Sahel dominate the accumulation mode particle population in the tropical North
248 Atlantic MBL, which results in an increase in CCN number concentration. Though dust and
249 smoke are both transported to the region, smoke dominates the accumulation mode number
250 concentration by an order of magnitude compared to dust. These findings are supported by data
251 products from dust mass concentrations, size-resolved hygroscopicity, single particle data (e.g.,

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CCSEM-EDX), and air mass history (e.g., NOAA'S HYSPLIT model), which all complement one another and provide unique insights into the aerosol sources, their single particle composition, and their effects on cloud droplet activation.

Sampling Conditions during the EUREC^{4}A and ATOMIC Campaigns

Barbados was influenced by two types of air masses during the sampling period: air masses that do not pass over land (referred to as clean marine conditions), and air masses influenced by continental regions (referred to as continental aerosol transport (CAT) events). To confirm the origins of the various air masses sampled, we performed back trajectory analysis throughout the campaign using NOAA's HYSPLIT model. Figure 1 shows that during periods with low dust mass concentrations and a bimodal size distribution, air masses originated from the remote Atlantic Ocean at higher latitudes with no land contact over 6 days. During time periods with high dust mass concentrations, air masses originated from continental Africa. Figure 2a shows that the total mass concentration of dust particles correlates very well with the arrival of air masses originating from Africa. During time periods when dust concentrations were low, the particle loading has a bimodal size distribution characteristic of clean marine air masses (Figure 2b) (Hoppel et al., 1986). Upon the increase in dust mass concentrations, the submicron particle size distribution correspondingly becomes unimodal and the smallest Aitken size mode is negligible, suggesting that transported particles are overwhelming the background marine particle loading or that smaller particles are coagulating onto larger transported continental aerosols (Tomlin et al., 2021).

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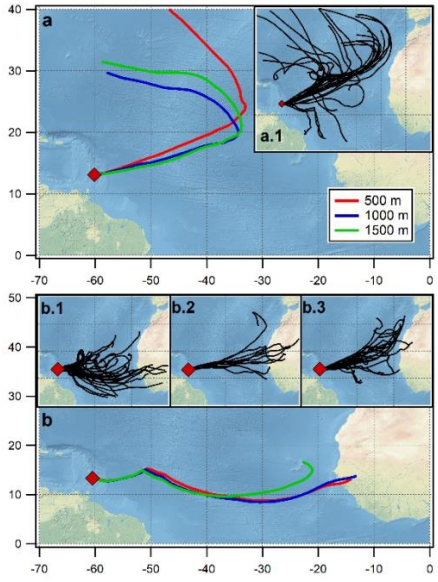

Figure 1: HYSPLIT back trajectories at Ragged Point, Barbados (red diamond) for the EUREC^{4}A/ATOMIC field campaign. (a) Back trajectories for 2020/2/8 18:00 UTC at heights of 500 m (red), 1000 m (blue), and 1500 m (green) exemplify air mass origins during clean marine sampling conditions. Subplot a.1 shows all back trajectories from clean marine sampling conditions collected at 6 h intervals with a release altitude of 1000 m from 2020/1/29 0:00 – 2020/1/29 12:00, 2020/2/6 12:00 – 2020/2/9 18:00 and 2020/2/12 12:00 – 2020/2/15 6:00 UTC. (b) Back trajectories for 2020/2/2 18:00 UTC at 500 m, 1000 m, and 1500 m exemplify air mass origins during continental sampling conditions. The subplots, b.1, b.2, and b.3 show all back trajectories for 3 time periods during which continental aerosols were sampled, including 2020/1/29 18:00 – 2020/2/6 6:00, 2020/2/10 0:00 – 2020/2/12 6:00, and 2020/2/15 12:00 – 2020/2/20 18:00 UTC. Trajectories for b subplots were also collected at 6 h intervals with a release altitude of 1000 m.

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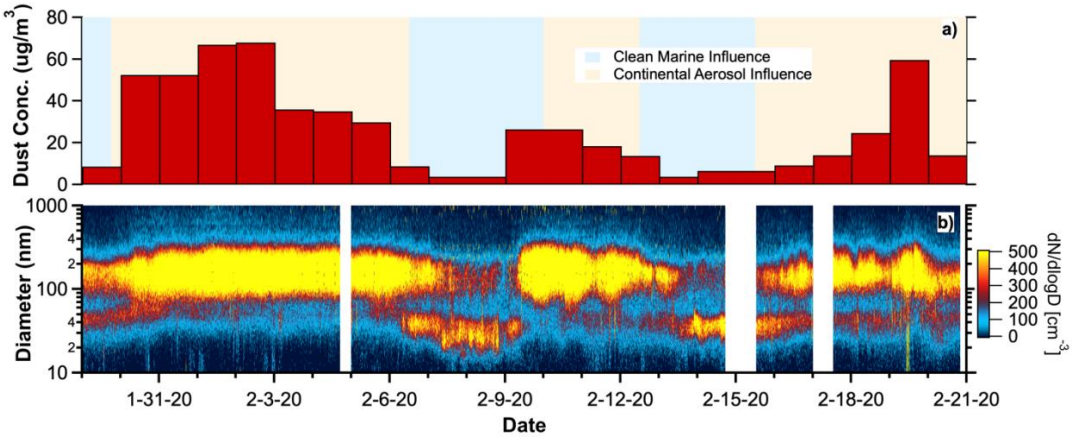

286

287 **Figure 2** – Temporal evolution of (a) dust mass concentrations determined from bulk aerosol
288 filter samples and (b) aerosol particle size distributions determined with an SMPS. Time for both
289 plots is given in UTC (-4 h local Atlantic Standard Time).

290

291 Single Particle Aerosol Composition

292 CCSEM/EDX analysis revealed the presence of several particle types in the submicron
293 aerosol loading with distinct morphologies and chemistries (Ault et al., 2014; Behnke et al.,
294 1997; Gaston et al., 2011a, 2013a) during the EUREC^{4}A and ATOMIC campaigns. Figure 3
295 presents SEM images (left) and EDX spectra (right) for each particle type detected on stage 3 of
296 the MPS (particle diameter <0.7 μm), including aged sea spray, mineral dust, sulfate, smoke,
297 internally mixed dust and smoke, and organics. Sea spray was a dominant component of the
298 supermicron aerosol loading but is only a minor component of submicron aerosol.

299 *Aged Sea Spray*

300 Aged sea spray was defined by the presence of sea salt components including Na, Mg, K, S,
301 and Cl. In contrast to freshly emitted sea spray particles, aged sea spray has a characteristically

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low or absent Cl signal with a strong presence of N or S. Sea spray can be aged through reactions with sulfuric acid (H_2SO_4), dinitrogen pentoxide (N_2O_5), and/or nitric acid (HNO_3) which results in Cl depletion and S or N enrichment (Ault et al., 2013, 2014; Behnke et al., 1997; Gaston et al., 2011, 2013; Sobanska et al., 2003).

Mineral Dust

Mineral dust is characterized by the presence of aluminosilicate elements such as Si, Al, Fe, K, Ca, and Mg in EDX spectra, which is consistent with previous studies of African dust (Denjean et al., 2015; Hand et al., 2010; Levin et al., 2005; Twohy et al., 2009). Elements such as S and N were not observed in this particle type (Kandler et al., 2018) suggesting that detected dust did not undergo chemical processing during transport.

Sulfate

Sulfate-rich particles are a prevalent component of marine submicron aerosol (O'Dowd & de Leeuw, 2007) and characterized here by a dominant S component - often with strong C, O, and N. These particles are likely sulfates bound to NH_4^+ such as ammonium sulfate ($(NH_4)_2SO_4$) or ammonium bisulfate (NH_4HSO_4) (Hand et al., 2010). The strong C component indicates a large organic fraction as well.

Smoke

Smoke particles were identified by the presence of C with K and S likely representing internally mixed organic and black carbon with potassium-containing salts. K is a well-known indicator for biomass burning (Andreae, 1983; Hand et al., 2010; Hudson et al., 2004; Li et al.,

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2003; Murphy et al., 2006; Pósfai et al., 2003), especially in flaming conditions in Savannah fires as opposed to smoldering conditions (Echalar et al., 1995; Maenhaut et al., 1996). Morphologically, smoke particles can be spherical due to aging or coating but can also appear as aggregates or chains of spheroids (Dang et al., 2021; Hand et al., 2010; Miller et al., 2021; Pósfai et al., 2003).

Internally Mixed Dust and Smoke

Internally mixed dust and smoke particles are characterized by dust components such as Si, Al, Mg, Fe, Ca, and Mg with strong contributions of K, S, C, and O. Morphologically, internal mixtures of dust and smoke appear as aggregates of amorphous dust particles with clusters or spheres representing soot from smoke. Single particle chemical analysis of these particles shows distinctions between the dust and smoke portion of the particle, with the dust portion having typical dust components (Si, Al, Mg, Fe, Ca, and Mg) and the smoke portion having typical smoke components (K and S with C and O). Previous research has observed internal mixing of carbonaceous particles and dust particles in Africa when significant amounts of both biomass burning and dust were present (Hand et al., 2010); however, we show that these internal mixtures can be transported all the way to the Caribbean as well.

Organics

Organic particles are defined by strong signals of C and O with few other elements present, if any (Hand et al., 2010). This scarcity of additional elements includes S and N that, if present, would be indicative of sulfate and nitrate, respectively. Morphologically, organic particles are characterized as small individual spheres. The organics were likely marine in origin (Russell et al., 2010) as they were the smallest particle type observed both during clean marine conditions

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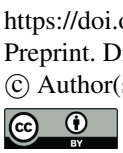

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and during long-range continental aerosol indicative of a "background" aerosol type (Russell et al., 2010).

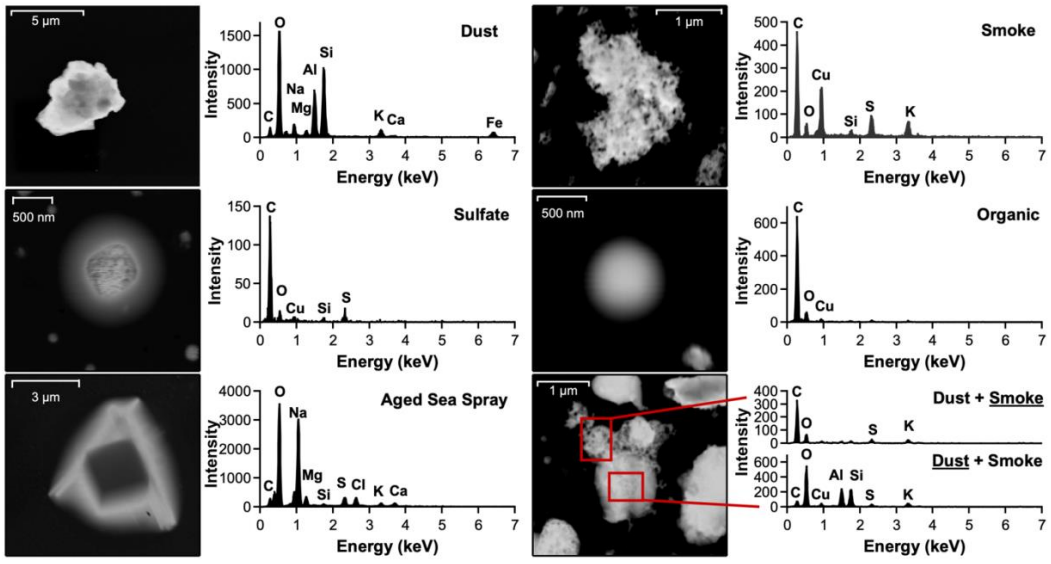

Figure 3: Characteristic aerosol particle types observed by means of SEM-EDX images (left) and spectra (right) in samples collected during the EUREC^{4}A/ATOMIC campaign. Plots for Dust + Smoke particle type represent different areas analyzed on the particle with EDX, denoted by the red boxes.

Arrival of Anomalous Wintertime Co-Transported Dust and Smoke

Figure 4 presents number fractions for particles detected in the submicron aerosol loading throughout the sampling period and reveals a similar trend in smoke particle number fractions to those of dust mass concentrations in Figure 2, suggesting that smoke and dust were co-transported to Barbados from Africa. During the boreal winter, the Sahel region in North Africa experiences its fire season in which large swathes of land are burned and large plumes of smoke

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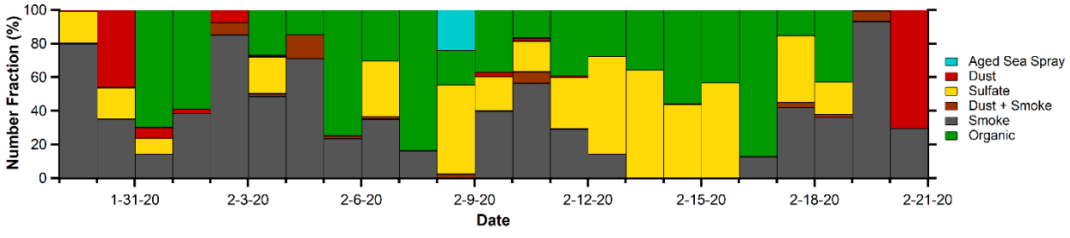
359 are emitted from the region (Figure S1;(Ansmann et al., 2009; Barkley et al., 2019; Roberts et
360 al., 2009). However, due to the southward shift in the ITCZ during the boreal winter, smoke is
361 expected to be transported primarily to South America (Moran-Zuloaga et al., 2018). In our
362 study, we observe the arrival of this smoke on Barbados. These findings are supported by
363 temporal carbon monoxide (CO) column density measurements that are often used as a tracer for
364 smoke (Figure S2 and S3). Periods that correspond to clean marine influence in the HYSPLIT
365 model from Figure 1 and low bulk dust mass concentrations in Figure 2 are dominated by sulfate
366 and organic particles in the submicron aerosol as exhibited in Figure 4. Upon arrival of
367 continental aerosols that correspond with continental aerosol influence from the HYSPLIT
368 model and bulk dust concentrations (2020/1/29 – 2020/2/7, 2020/2/9 – 2020/2/12 – 2020/2/16 –
369 2020/2/20 UTC), wildfire smoke appears to overwhelm the number fraction of the submicron
370 aerosol loading.

371



372 **Figure 4**: Temporal evolution of submicron number fractions for different types of aerosol
373 particles determined by CCSEM/EDX analysis. The total number of particles analyzed for each
374 day ranges from 1000 to 20,000.

375 Figure 5 provides a detailed size-resolved plot of single particle chemistry from both
376 clean marine periods (clean marine), and periods that were influenced by air masses from
377 continental Africa (CAT event 1). Analysis of other CAT events (Figure S4) show similar

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chemical trends to those shown in the Cat Event 1 plot. Figure 5 shows that in both clean marine periods and CAT events, a small fraction of large particles have both a smoke and a dust signature. Both the clean marine and CAT event plot in Figure 5 demonstrate that dust and internally mixed dust and smoke particles tend to be the largest particle types in the submicron aerosol loading. This suggests that our clean marine conditions are "clean" compared to time periods influenced by dust and smoke, rather than pristine clean marine conditions without any continental aerosol influence. Smoke particles follow as the next largest particle type. The smallest particle types were found to be organics followed by sulfates suggesting a primary emission of marine organics and a secondary source for sulfate (Bates et al., 1992). Aged sea salt particles were on average smaller than most dust, internally mixed dust and smoke, and smoke particles. Figure 4 also shows that at a diameter of ~0.1µm (which is approximately the d_{50} of CCN at S 0.16% in clean marine and dusty conditions) the chemistry is dominated by sulfates and organics in the clean marine conditions, while smoke and organics dominate in the CAT event. A large decrease in sulfate number fraction suggests that marine biogenic sulfur precursors are condensing onto larger transported particles and might explain the loss of the Aitken mode observed in Figure 2.

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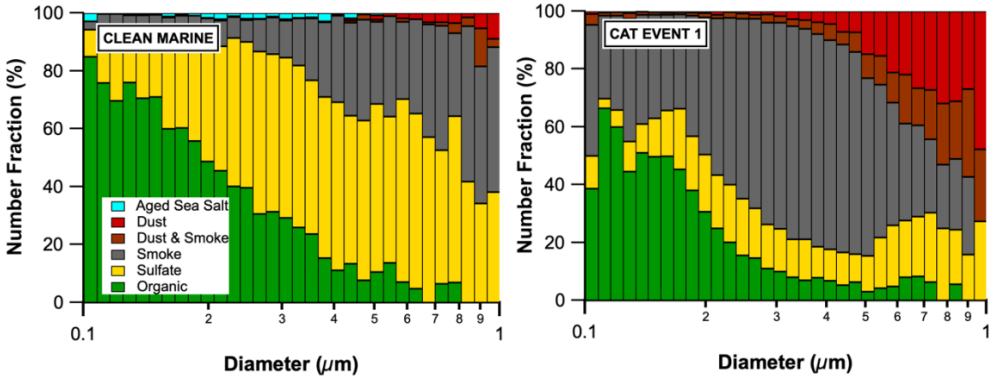

Figure 5: Number fractions of different types of submicron aerosol particles plotted against particle diameter. The "clean marine" plot (left) includes data from all clean marine sampling periods. The "CAT Event 1" plot (right) includes data from the first period in which dust and wildfire smoke were observed over Barbados (2020/1/29 18:00 – 2020/2/6 6:00 UTC). Bin sizes for each decade can range from 34 particles to up to 3041 with an average bin size of 493 particles for the Clean Marine plot and 973 for the CAT Event plot.

Changes in Aerosol Hygroscopicity during EUREC^{4}A/ATOMIC

Comparisons between size-resolved CCN measurements and submicron single particle elemental composition reveal that smoke particles lower submicron aerosol hygroscopicity compared to marine-derived submicron aerosol in the tropical North Atlantic. Figure 6 presents boxplots for κ values as well as average d_{50} measured at each S during both clean marine conditions and dusty conditions. Both plots show a similar trend in which average κ increases from 0.09% S to 0.24% S. Then, with each subsequent increase in S after 0.24% S, κ decreases likely due to smaller, less hygroscopic particles activating at higher supersaturations. Also of note is the κ of 0.6 observed for clean marine conditions at 0.24% S, which matches κ

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412 measurements for ammonium sulfate particles that can dominate along with sea spray organics
413 during clean marine conditions (Petters & Kreidenweis, 2007).

414 There is also a noticeable drop in average κ between the same supersaturations in clean
415 marine conditions compared to smokey conditions. For example, at 0.16% S, $\kappa=0.52\pm0.09$ for
416 clean marine conditions and $\kappa=0.46\pm0.10$ for continental aerosol transport. This is likely due to
417 the addition of less hygroscopic material such as dust and smoke particles that are acting as CCN
418 and are not present in clean marine conditions. As expected, trends in average d_{50} for both plots
419 show that activation diameters decrease with an increase in supersaturation, indicating that
420 smaller particles activate as CCN with larger supersaturations. Activation diameters during CAT
421 conditions are also larger than corresponding activation diameters in clean marine conditions for
422 the same supersaturation. This also suggests that the addition of less soluble material from
423 transported smoke particles lowers the hygroscopicity and increases the activation diameter.

424 When comparing hygroscopicity data from this study to previous research, we find both
425 similarities and differences in κ trends. For example, Good et al., 2010 presents data collected in
426 the tropical eastern Atlantic that provides an ideal comparison to our findings. On average, their
427 values for κ in clean marine conditions and during observations of dust transport ($\kappa=1.15$-1.4 and
428 0.8-0.92, respectively) were much higher than our observed values. However, one finding of note
429 from Good et al., 2010 is the distinct drop in κ between clean marine conditions and conditions
430 influenced by continental aerosols from Africa owing to the addition of more hydrophobic
431 particles such as dust that activate as CCN. Our data shows a similar drop in κ from clean marine
432 conditions to conditions influenced by long-range transported African aerosols. Wex et al., 2016
433 present ground-based field sampling of CCN data in November and April at Ragged Point,
434 Barbados. They show a similar trend in κ in which values increase from 0.1% S, peak at 0.2% S,

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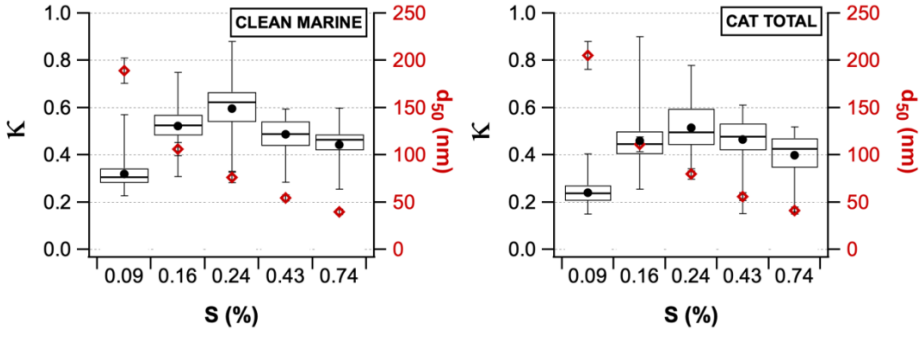
435 then decrease with each subsequent increase in S. Wex et al., 2016 also found a similar drop in κ
436 upon the arrival of long-range transported aerosols, likely due to less hygroscopic particles from
437 continental sources activating as CCN. A separate study from Kristensen et al., 2016 conducted
438 similar research at Ragged Point, Barbados during the boreal summer. The range in κ values of
439 0.2-0.5 match those observed in our work, especially during CAT events. However, Kristensen et
440 al., 2016 determined that concentrations of dust, sea salt, and soot were too small to influence
441 CCN, concluding that sulfates and organics were the primary CCN types. We find this is the case
442 for clean marine conditions but the change in κ between clean marine and CAT events indicates
443 the influence of an additional CCN particle type.



444

445 **Figure 6:** Hygroscopicity parameter κ (left axis, box plots) and corresponding mean critical
446 diameter "d_{50}" (right axis; red markers) for the investigated levels of water vapor supersaturation
447 (S). Black dots in the boxplot indicate κ mean values.

448

449

450 <u>African Smoke Particles Enhance CCN Concentrations</u>

451 Comparisons between smoke fractions and CCN counts suggest that smoke particles
452 enhance the number of CCN in the tropical N Atlantic MBL. Figure 7 presents two temporal

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plots of κ (Figure 7a) and smoke number fractions with CCN counts measured at 0.16% S (Figure 7b). Table 1 provides averages of CCN concentrations for each time period shown in Figure 7b. Fig 7a suggests that there is an inverse relationship between κ and smoke number fractions in which an increase in smoke particles results in a decrease in κ. This is likely due to the activation of smoke particles as CCN, which are on average less hygroscopic than the sulfate particles that act as CCN during clean marine conditions. In Fig 7b, there is a clear and direct relationship between smoke number fractions and CCN counts.

There are several possible explanations for why African smoke particles may act as CCN. As shown in Figure 5, smoke particles are larger than organics and sulfates, on average, and overwhelm sulfate and organic particle number concentrations upon arrival of long-range transported African aerosols. In this case, the relatively large size of the smoke particles makes for better CCN compared to organics or sulfates via the Kelvin effect (Dusek et al., 2006). The large number of smoke particles overwhelms the particle loading, providing more surface area for water condensation than is available on sulfate or organic particles. Another potential explanation for smoke particles acting as CCN could be the presence of water-soluble organic compounds (WSOC) such as dicarboxylic acids and humic-like substances that increase CCN numbers through the Raoult effect (Roberts et al., 2002). In addition to WSOC, aerosols can also contain organic surfactants that decrease surface tension and thus lower the vapor pressure necessary for CCN activation (Asa-Awuku et al., 2008; Facchini et al., 1999; Ovadnevaite et al., 2017). Aging of organic components can also potentially explain the ability of smoke particles to activate as CCN. Studies conducted on the aging of organic components show that a higher O:C ratio (a proxy for aerosol organic aging) increases aerosol hygroscopicity (Jimenez et al., 2009; Massoli et al., 2010). The long-range transport of smoke particles from Africa to Barbados

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would theoretically provide ample exposure time of smoke particles to oxidants such that aging of the particles could occur (Jimenez et al., 2009; Massoli et al., 2010). However, studies exploring the aging of biomass burning particles specifically show that aging results in a drop in κ, rather than an increase as observed in aging of secondary organic aerosols (Engelhart et al., 2012). Finally, the presence of salts in smoke particles has been shown to be an important component in smoke hygroscopicity and may explain why smoke is efficient as CCN. Previous studies have shown that smoke particles often contain hygroscopic salts such as potassium chloride, sulfate, and nitrate (e.g., KCl, KNO_3, and K_2SO_4) (Freney et al., 2009; Zauscher et al., 2013). Other research also shows that only small fractions of salts are needed to increase aerosol hygroscopicity (Roberts et al., 2002). It is likely that biomass burning hygroscopicity can be explained by a combination of factors. For example, previous research has found that the presence of salts can enhance the surfactant effect of hydrophobic organic compounds (Asa-Awuku et al., 2008).

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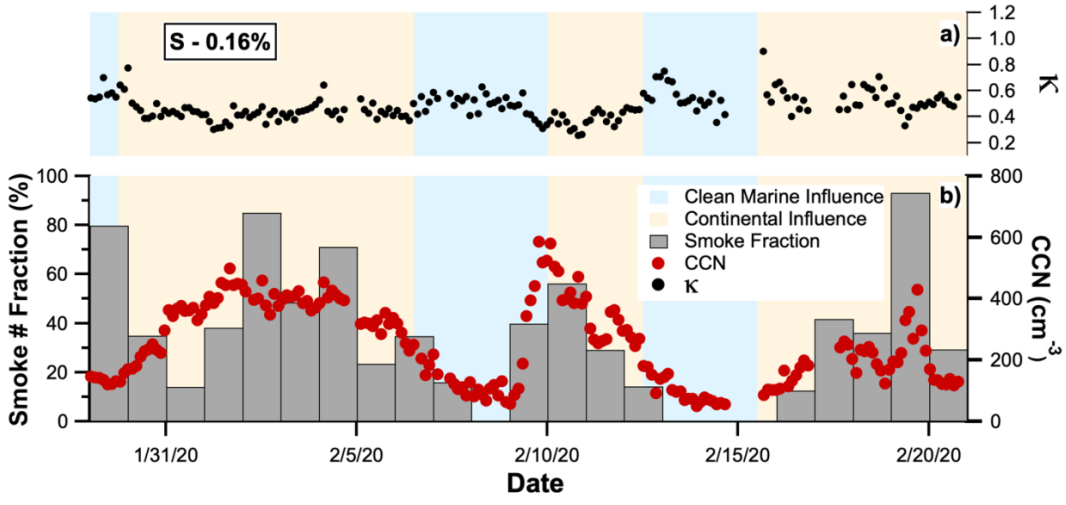

489

490 **Figure 7**: Temporal evolution of hygroscopicity parameter κ (black dots, upper panel) and CCN
491 number concentration (red dots, lower panel), both measured at S = 0.16%, and smoke particle
492 number fraction (grey bars, left axis, lower panel). Background color shadings indicate periods
493 of continental influence (orange) and clean marine influence (blue) determined by HYSPLIT
494 back trajectories and dust mass concentrations.

495 **Table 1** – Values for average CCN Concentrations and κ measured at 0.16% S during each clean
496 marine influence period and CAT event sampled during the EUREC^{4}A and ATOMIC
497 campaigns.

Sampling Period	**Day/Time**	**CCN Concentrations (pt/cm^3)**	**Average κ**
Clean Marine Period 1	2020/1/290:00 – 2020/1/29 12:00	140±10	0.58±0.07
CAT Event 1	2020/1/29 18:00 – 2020/2/6 6:00	340±90	0.44±0.08
Clean Marine Period 2	2020/2/6 12:00 – 2020/2/9 18:00	150±98	0.50±0.10
CAT Event 2	2020/2/10 0:00 – 2020/2/12 6:00	400±106	0.38±0.06
Clean Marine Period 3	2020/2/12 12:00 – 2020/2/15 6:00	100±42	0.55±0.10

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CAT Event 3	2020/2/15 12:00 – 2020/2/20 – 18:00	190±75	0.54±0.10

498

499 **Conclusions**

500 During clean marine conditions, the submicron aerosol loading consists primarily of
501 sulfate and organic particles. CCN measurements determine cloud activation by particles
502 approximately 80 nm in activation diameter with an average $\kappa = 0.52 \pm 0.08$. Comparisons
503 between particle size, hygroscopicity, and single particle elemental composition suggest that
504 sulfate particles (likely ammonium sulfate) are the primary CCN particles in clean marine
505 conditions. During the EUREC^{4}A/ATOMIC campaign, Barbados received three African aerosol
506 transport events during which we detected mineral dust and smoke particles from northern
507 Africa. Upon the arrival of African aerosols to BACO, CCN average activation diameter
508 increased to approximately 200 nm while the average hygroscopicity of activated particles for all
509 CAT events decreased to $\kappa = 0.45 \pm 0.1$. Upon arrival of high concentrations of smoke particles to
510 Barbados, smoke particles overwhelm the accumulation mode particle loading, decrease aerosol
511 hygroscopicity, and also increase CCN number concentrations, which could also increase the
512 cloud droplet number concentration and alter cloud radiative properties (Twomey, 1974).
513 Overall, we find that smoke has a larger effect on CCN number concentrations than dust.

514 The observation of smoke transported to Barbados during the boreal winter also indicates
515 the large geographic extent of African smoke that can impact the MBL. Building upon recent
516 work from Ragged Point and other parts of the tropical and subtropical Atlantic (Holanda et al.,
517 2020; Kacarab et al., 2020; Schill et al., 2020; Zuidema et al., 2018) this work also indicates a
518 need for greater consideration of the impacts of smoke in the MBL. Previous research conducted
519 at Ragged Point has primarily focused on African dust, which reaches its maximum during the

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boreal summer when smoke transport is low (Zuidema et al., 2019). To better contextualize our findings, we analysed carbon monoxide column density (a tracer for smoke) as well as aerosol optical depth (AOD) and aerosol optical thickness (AOT) (tracers for dust) from 2018-2022 (Figure S2 and S3). Figure S2 shows the temporal trends while Figure S3 show seasonal averages. As expected, AOD and AOT peak in July when dust transport reaches a maximum. However, Figures S2 and S3 indicate that smoke is decoupled from dust, reaching a maximum in the spring around April and a minimum in the summer when dust transport is highest. This finding suggests that while the dust transport during the EUREC4A/ATOMIC campaigns is higher than average dust loadings during this month (Zuidema et al., 2019), the amount of smoke observed is not unique, but rather characteristic of the region. Thus, smoke may be playing an important role on CCN formation throughout a large portion of the year. This is especially true considering the large size of long-range transported smoke plumes that have a wide geographic extent in which they can affect cloud formation. To conclude, this work highlights the need to characterize African smoke transport to Ragged Point and better understand the role of smoke in cloud formation, radiative forcing, and climate (Pechony & Shindell, 2010; Shindell et al., 2009).

Data Availability

The data will be made publically available in the University of Miami data repository and will be linked with a doi.

Author Contributions

Conceptualization of this work was done by HMR, MLP, OK, and CJG. Collection of samples was conducted by HMR, OK, EB, and PS, while analysis was done by HMR, MLP, OK, NNL, and ZC. The development of the methods used in this work was done by HMR, MLP, OK, ZC, SC, APA, and CJG. Instrumentation used to conduct this work were provided by MLP, SC, APA, and CJG. Formal analysis of data was performed by HMR, MLP, CP, and OK. Validation of data products was performed by HMR, ZC, SC, APA, and CJG. Computer code used for data analysis was provided by MLP, OK, and APA. Data visualization was performed by HMR, MLP, and OK. PKQ, PZ, CP, and UP helped interpret results. Supervision and project

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547 administration duties were done by MLP and CJG. HMR wrote the original draft for publication,
548 and all co-authors reviewed and edited this work.

549 **Competing Interests**

550 Some authors are members of the editorial board of Atmospheric Chemistry and Physics. The
551 peer-review process was guided by an independent editor, and the authors have no other
552 competing interests to declare

553

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