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Saharan dust particles nucleate droplets in eastern Atlantic clouds

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[1] Many soil-derived particles dominated by insoluble material, including Saharan dusts, are known to act as ice nuclei. If, however, dust particles can compete with other atmospheric particle types to form liquid cloud droplets, they have a greater potential to change climate through indirect effects on cloud radiative properties and to affect the hydrological cycle through precipitation changes. By directly collecting and analyzing the residual nuclei of small cloud droplets, we demonstrate that Saharan dust particles do commonly act as cloud condensation nuclei (CCN) in the eastern North Atlantic. Droplet activation calculations support the measurements by showing that due to its slightly hygroscopic nature, even submicron dust can be important as CCN. Given the dual nature of Saharan dust particles as CCN and ice nuclei, this infusion of dust is expected to impact not only droplet size and albedo in small clouds, but ice formation in deep convective clouds.
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1. Introduction

[2] Vast quantities of crustal dust in the eastern North Atlantic free troposphere originate from north African sources. If dust acts as a cloud condensation nucleus, it may decrease mean droplet size and diminish precipitation [*Rosenfeld et al.*, 2001; *Mahowald and Kiehl*, 2003]. Additionally, dust particles with sources from the Saharan region have been identified as efficient ice nuclei in laboratory and field studies [*DeMott et al.*, 2003; *Twohy and Poellot*, 2005; *Field et al.*, 2006; *Prenni et al.*, 2007]. *Dunion and Velden* [2004] showed that the Saharan Air Layer (SAL) seemed to inhibit the development of hurricanes in the Atlantic, and *Evan et al.* [2006] demonstrated that dust is anticorrelated with tropical cyclone activity. They proposed that this effect is caused by dynamical and radiative effects related to the SAL, a dry, rapidly moving airmass. However, dust nucleation impacts on microphysics, latent heat release and vertical transport could also influence convection development and precipitation efficiency in complex ways [e.g., *Khain et al.*, 2008; *Seifert and Beheng*, 2006; *Van den Heever et al.*, 2006].

[3] In the NASA portion of the African Monsoon Multidisciplinary Activities experiment (NAMMA) [*Redelsperger et al.*, 2006], aerosol physiochemical characteristics and cloud size distributions were measured over the tropical eastern Atlantic Ocean from a DC-8 aircraft in late summer of 2006. Measurements of microphysical properties of the small cumulus clouds in this region revealed that in most cases, droplet number concentrations were higher than expected for clean marine clouds. Cumulus clouds in the Caribbean Ocean tend to have droplet concentrations of 50 to 150 cm^{-3} [*Rauber et al.*, 2007], while in the eastern Atlantic, mean droplet concentrations ranged from about 50 to 500 cm^{-3}, with seven of the eleven cases having mean concentrations higher than 150 cm^{-3}. This suggests that a substantial fraction of dust and other non-marine particles are acting as CCN in the eastern Atlantic.

2. Measurements

[4] On 5 Sept 2006, a mission was flown from the Cape Verde Islands (near the west coast of Africa) and over the Sahara Desert in Mauritania. This area is a major dust source region [*Goudie and Middleton*, 2001]. Figure 1 shows vertical profiles of aerosol scattering ratio (1a and 1b) and water vapor mixing ratio (1c) from a differential absorption lidar [*Browell et al.*, 2005], looking down through the dust layer which extended to about 5.5 to 6 km above sea level. The water vapor image shows that the absolute humidity in the SAL was higher over the ocean, where the leading edge of the SAL was likely mixing with the background moister environment. Small clouds formed at about 5 km and are indicated by the darkest backscatter regions toward the left of Figure 1a and in Figure 1b, where the lidar was fully attenuated below.

[5] During NAMMA, aerosol particles and cloud residual particles were sampled with a counterflow virtual impactor (CVI) [*Noone et al.*, 1988] to assess the percentage and size of dust particles actually incorporated into clouds. In normal mode, the CVI rejects interstitial aerosol using a nitrogen counterflow while collecting and evaporating droplets or ice crystals. Individual non-volatile residual nuclei are retained. Ambient aerosol particles can also be collected by turning off the counterflow. Ambient aerosol or cloud residual particles were then collected by a two-stage jet impactor. The small particle stage captured particles approximately 0.1 to 0.5 μm in diameter, while the large particle stage captured particles from about 0.5 μm up to several microns in size. Samples were analyzed by transmission electron microscopy and energy dispersive X-ray spectrometry to detect chemical elements. Individual particles were identified as crustal dust, salts, metals, sulfate, carbonaceous, or mixtures of these types as was done by *Twohy and Poellot*

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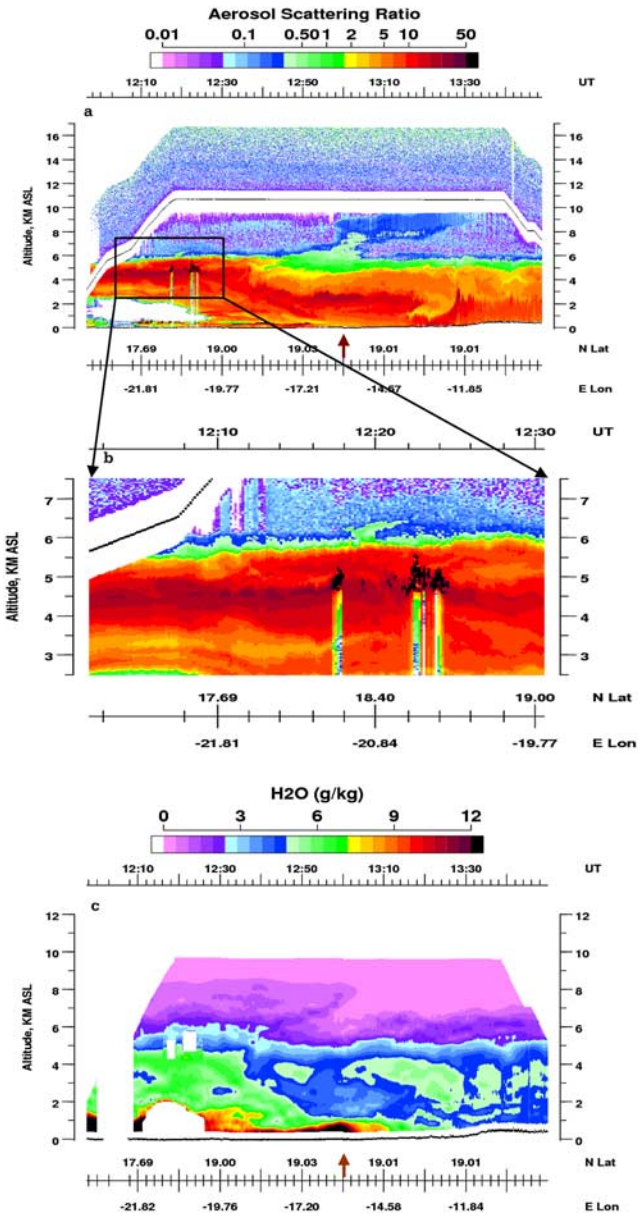

Figure 1. Onboard lidar images for 5 September 2006 from a west to east track, looking down through the Saharan Air Layer (SAL). (a) Aerosol scattering ratio (ASR) image. Western African coastline is marked with a brown arrow at the bottom. ASR are uncorrected for attenuation, but the uncorrected data better show the presence of clouds (black regions). Black curve surrounded by white region is DC-8 flight altitude profile surrounded by lidar blind (data void) region. The top of the SAL is located at about 6 km altitude where ASR values exceed 1.0. (b) Enlargement of area with embedded clouds (inset box in Figure 1a). (c) Corresponding water vapor mixing ratio (g kg^{-1}) image.

[2005]. Percentages of types from both size stages were not substantially different for the cases presented here and have been averaged for simplicity of display. Additionally, only data from clouds without precipitation-sized particles, which can break up upon collection, are included.

[6] The first aerosol sample from 5 September was collected at 2.1 km directly over the desert. Analyses revealed that these particles were primarily unmixed crustal dust. The typical X-ray spectrum revealed particles that contained silicon and aluminum, as well as smaller amounts of iron, potassium, calcium and magnesium. As opposed to heavily weathered dust, which is more common closer to the equator [*Moreno et al.*, 2006], these younger, smectitic clays are expected to be more hygroscopic [*Hensen and Smit*, 2002]. Recently, the critical supersaturations required to activate submicron dust from the northern Sahara have been measured to be similar to those for slightly hygroscopic particles [*Koehler*, 2008]. Additionally, while silicon, aluminum, and iron in Saharan dust are associated with minerals with low solubilities [*Baker et al.*, 2006], potassium, magnesium and calcium compounds tend to have higher solubilities [e.g., *Kelly et al.*, 2007]. Significant amounts of water-soluble calcium and potassium have been attributed to Saharan dust [*Savoie and Prospero*, 1980].

[7] Figure 2a shows the percentage by number of different particle types by number in the sample taken over the Sahara. About three-quarters of the particles were unmixed crustal dust. Most of the smaller dust particles were on order 0.14–0.30 μm in diameter. A few particles were metals without significant silicon, which were probably different types of mineral dust. About 20% of the particles were in the "mixed" category, which is this case was primarily dust internally mixed with S and/or Cl. These elements usually indicate the presence of sulfates and chlorides which may be deposited on aerosol particles through heterogeneous reactions, and make particles more hygroscopic. This may be explained by the back trajectory analysis, shown below the composition plot, which predicts that this airmass had resided for about 70 hours over water and the Iberian Peninsula before passing over the Saharan source.

[8] The next sample (Figure 2b) is representative of Saharan dust that had traveled about 700 km offshore, but remained at mid altitudes with little interaction with the marine boundary layer. This sample from 3.7 km was almost entirely composed of dust aerosol without detectable S or Cl. This sample actually had a longer overall residence time over the Sahara than the previous sample. Total particle concentrations (diameter >0.01 μm) were about 450 cm^{-3}, with about 72% of them being non-volatile at 300°C, consistent with refractory soil dust. While most of the aerosol volume was present in particles larger than 1 μm, aerosol number was dominated by particles smaller than 1 μm in diameter.

[9] Seventy-nine percent of the residual nuclei from the liquid clouds embedded within the SAL layer (Figure 1b) were unmixed soil dust, with small amounts of salts and mixed particles present (Figure 2c). Most of the salts were not sea salt, but rather potassium or calcium-based salts. The dust particles collected ranged from 0.1 to 1.8 μm in diameter (median for small impactor stage = 0.26 μm and for large stage = 0.57 μm), well below the minimum size collected by the CVI. Therefore, these particles could only have entered the inlet if originally present inside a cloud droplet. The CVI collected about 65% of these droplets (remaining drops were smaller than the 7 μm cut size of the inlet.) The optical particle counter showed that 90% of the residual particles were larger than the 0.1 μm cut size of

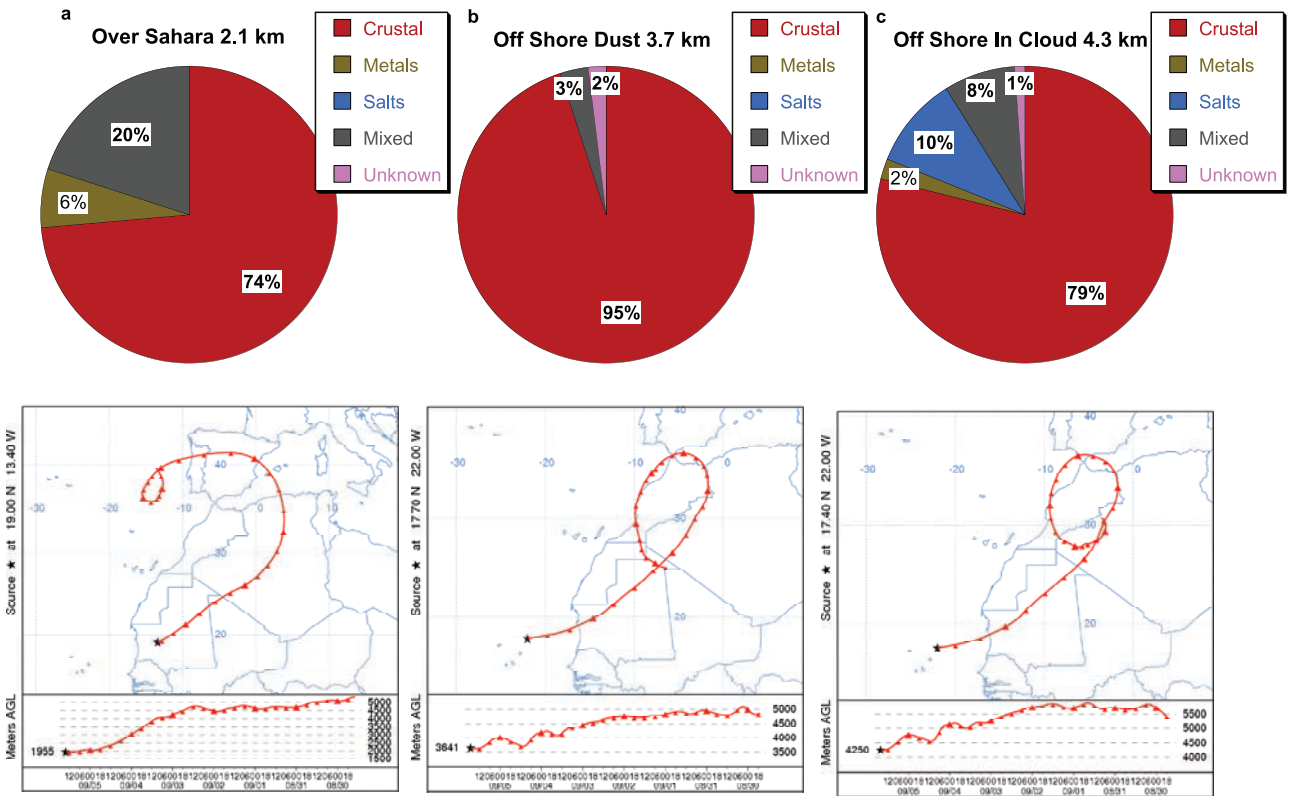

Figure 2. Percentage of different particle types by number from 5 Sept 2006 Saharan dust samples (100 particles for each sample, 50 from each of 2 size stages, were analyzed). The "unknown" category represents a few particles with no detectable-ray signature, most likely organic or nitrates. Below each pie chart is given the HYSPLIT 7-day back trajectory using NCEP/NCAR reanalysis data. Land area shown is the northwest coast of Africa. (a) The first ambient sample was taken directly over the Sahara (19.0N, 13.4W), at 2.1 km pressure altitude at 14:34 UTC. (b) The second ambient sample was over the Atlantic (17.7N, 22.0W) at 3.7 km pressure altitude at 15:11 UTC. (c) The third sample represents the composition of residual particles from a cloud embedded in the dust layer (17.6N, 21.9W) at 16:43 UTC. Cloud temperature was 273 K and particle size distributions indicated a liquid cloud with small droplets and no ice present. Maximum updraft velocities were about 0.5 m s^{-1}. Peak droplet concentrations (>3 μm) were about 150 cm^{-3}.

the impactor; therefore, these results represent the composition of about 58% of the droplets.

[10] For the typical supersaturations expected in small cumuli (less than ~0.3%), an insoluble but wettable particle must be larger than ~0.7 μm to act as a CCN [*Koehler et al.*, 2007]. Smaller particles must have had nonzero hygroscopicity in order to nucleate cloud drops. Since most of the particles did not have detectable S or Cl, this suggests that dust particles do not have to be "aged", or coated with soluble material derived from pollution sources, to act as CCN. While thin coatings of volatile nitrate and organics could be missed by our detection technique, we show below that they are not necessary to explain the droplet nucleating ability of Saharan dust.

[11] The ability of dust to act as CCN is not confined to this one case of a cloud forming directly within the SAL layer. Shallow cumulus clouds located in the marine boundary layer below the dust layer were also sampled over the ocean on several other flights. For three samples, the percentage of cloud residuals that contained dust (either alone or mixed with S or Cl) ranged from 14% to 54% by number of the total particles collected. These are much larger percentages than have been found in droplet nuclei in other regions of the world [*Twohy and Anderson*, 2008], suggesting that substantial dust is entrained into the marine boundary layer.

3. Modeling

[12] A Lagrangian parcel model based on Feingold and Heymsfield [*Feingold and Heymsfield*, 1992] was used to simulate activation of dust to cloud droplets in the eastern North Atlantic under various conditions. The original model was modified to parameterize water activity and hygroscopic growth with a single parameter Kappa, κ [*Petters and Kreidenweis*, 2007]. Kappa parameterizes the relationship between dry diameter and critical supersaturation for activation, and is 0.00 for a completely insoluble but wettable particle. Although activation of dusts may proceed by complex mechanisms and the actual wetted size of the particle at activation is not yet established, CCN measurements for a subset of submicron Saharan dust particles indicate κ~0.05 represents the data well and this simple parameterization can be used to estimate the number of dust particles activated as a function of environmental supersaturation [*Koehler*, 2008].

[13] Four different model cases were run to assess competition between a bimodal aerosol distribution with a

Table 1. Aerosol Parameters Used in Dust Modeling Study

	Hygroscopic Mode				Dust Mode			
Case[a]	Kappa	N_d (cm^{-3})	d_g (μm)	σ_g	Kappa	N_d (cm^{-3})	d_g (μm)	σ_g
A	1.0	225	0.10	1.6	0.00	225	0.67	1.7
B	1.0	225	0.10	1.6	0.05[b]	225	0.67	1.7
D	1.0	225	0.10	1.6	0.00	225	0.10	1.6
E	1.0	225	0.10	1.6	0.05[b]	225	0.10	1.6

[a]Case letters correspond to same letters as panes in Figure 3.

[b]Kappa for a mixture can be computed by volume-weighting the individual constituents, and $\kappa = 0.05$ corresponds to a volume fraction of ~8% ammonium sulfate in an otherwise insoluble particle.

hygroscopic mode and an insoluble or slightly hygroscopic dust mode (Table 1). All cases were based on size distribution measurements in the SAL layer during NAMMA, with the simplification that there were only two submicron size modes. Cases A and B had a small hygroscopic mode and larger dust mode, while Cases D and E had both the hygroscopic and dust modes the same size, to assess the effects of composition alone. Two different κ values were modeled for the dust mode: $\kappa = 0.0$ and $\kappa = 0.05$. A range of updraft velocities simulating weak to strong convection in this region was modeled [*LeMone and Zipser*, 1980].

[14] Figure 3a shows the activation fraction for the two types of aerosol in Case A, where the dust is larger, but completely insoluble. For this case, primarily the smaller, more hygroscopic particles activate at low updrafts. However, the dust is large enough that greater than 40% of the dust particles activate at 0.5 m s^{-1}. Figure 3b shows that the small amount of hygroscopic material present naturally in the dust in Case B allows dust activation to occur with much higher efficiencies than in Case A; in fact, higher fractions of dust act as CCN than for the smaller, completely hygroscopic aerosol. Activation of the larger, slightly hygroscopic dust particles suppresses the maximum supersaturation achieved in cloud, as shown in Figure 3c. This reduces the number of smaller hygroscopic particles that can form drops, so a larger fraction of them are interstitial particles.

[15] The effect of dust size is also important. For cases D and E (Figures 3d and 3e), both the hygroscopic and dust modes were modeled with a d_g of 0.10, consistent with

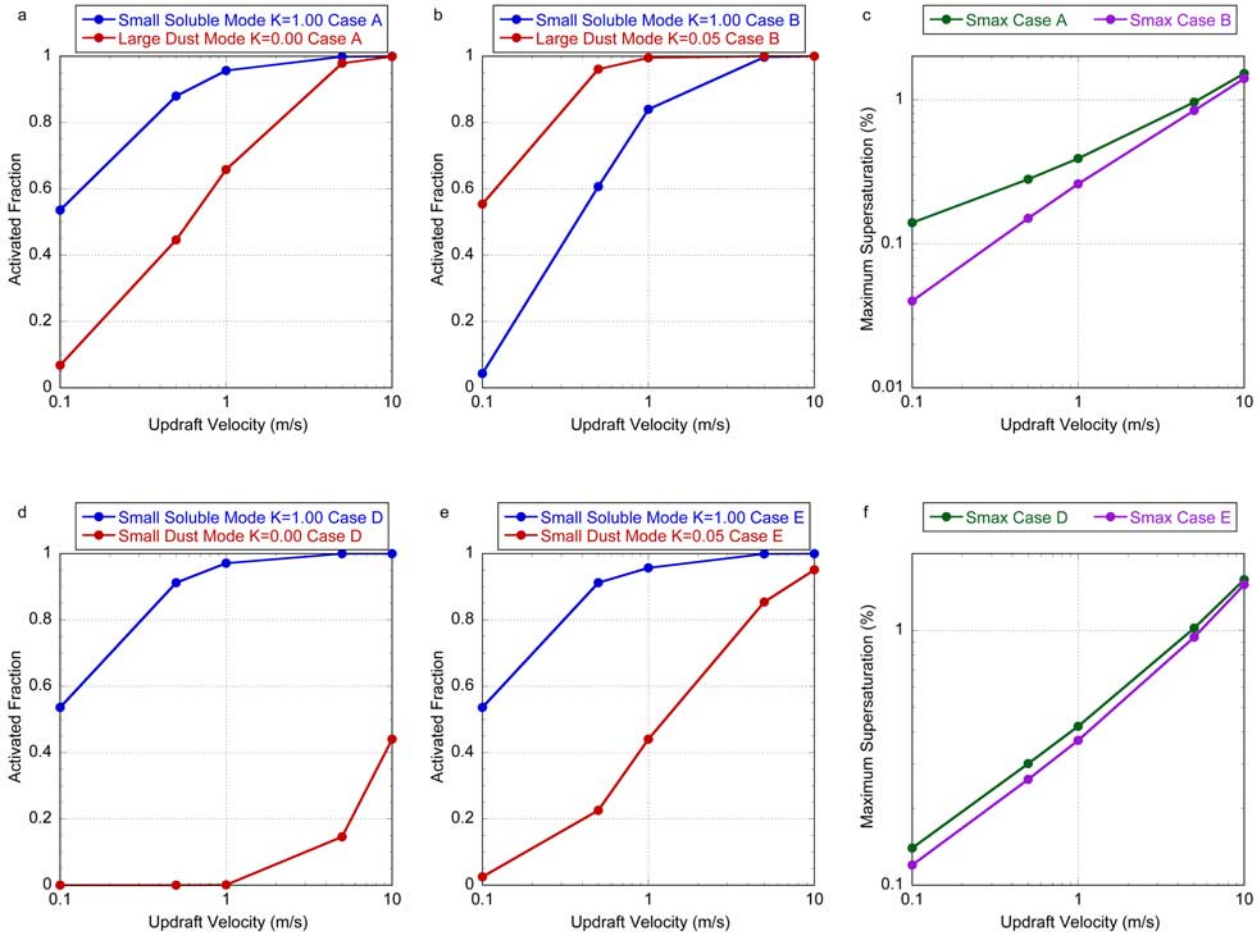

Figure 3. Fraction of aerosol particles activated or maximum supersaturation as a function of modeled updraft velocity for each individual particle mode as given in Table 1. Ambient conditions were 600 mb and 273 K. (a) Case A: Hygroscopic mode with $\kappa = 1.0$ and large dust mode with $\kappa = 0.00$. (b) Case B: Same as Case A, but with $\kappa = 0.05$ in the dust mode. (c) Maximum supersaturation predicted for Cases A and B. (d) Case D: Hygroscopic mode with $\kappa = 1.0$ and small dust mode with same size parameters but with $\kappa = 0.00$. (e) Case E: Same as Case D, but with $\kappa = 0.05$ in the dust mode. (f) Maximum supersaturation predicted for Cases D and E.

NAMMA volatility and single-particle measurements that indicated at least some of the dust was present in the small size range. The small-mode dust is activated with much lower efficiencies than the larger dust shown in Cases A and B. However, the simulation shows that even small dust with a $\kappa = 0.05$ can activate in substantial fractions of 0.2 to 0.7 at updraft velocities between 0.5 and 3 m s^{-1} (Figure 3e). The assumption of slightly hygroscopic dust can explain why substantial numbers of dust particles were observed in droplet residual nuclei for the 5 September case, where the particles were dominated by Saharan dust, and why some dust can activate in the marine boundary layer while competing with hygroscopic species like sea salt and sulfate.

4. Conclusions

[16] We conclude that the aerosol observations collected during the 2006 NAMMA field campaign are consistent with slightly hygroscopic, submicron Saharan dust particles that can act as cloud condensation nuclei in liquid clouds. Hygroscopic material can be present naturally in the dust at the source, as well as added afterward by interaction with atmospheric gases and particles. In high dust situations such as in the eastern Atlantic, dust can contribute substantially to the droplet numbers in small cumulus clouds. We estimate from measurements of this and other cases that approximately 30 cm^{-3} to 120 cm^{-3} of droplets in small clouds in this region are likely to contain Saharan dust.

[17] These relatively high concentrations of droplets containing dust are important not only for their impact on microphysical properties and precipitation efficiency of warm clouds, but also because dust can serve as an ice nucleus in mixed-phase clouds. Their additional action as CCN demonstrated here is important, since both Saharan and Asian dusts activate more efficiently as condensation freezing nuclei than as deposition nuclei at temperatures warmer than about 240 K [*Field et al.*, 2006]. Since ice crystal number concentrations in most clouds are typically about three orders of magnitude smaller than droplet concentrations, substantial changes in ice number concentration and size are possible as a result of dust entering droplets as CCN.

[18] Considerable uncertainty exists regarding current and future dust source emissions. Assuming, however, that dust emissions increase due to land use and climate changes, the propensity of dust to act as CCN would be expected to result in smaller droplets, enhanced albedo, and decreased precipitation from small cumulus clouds without ice. This could exacerbate dust emissions from the Sahel and other dust source regions due to heightened drought conditions [*Rosenfeld et al.*, 2001]. Additionally, changes in latent heating profiles in deep convection due to dust acting as IN are likely to change not only cloud microstructure and macrostructure but also cloud dynamics, with additional impacts on precipitation efficiency and storm lifetime. Dust influence on precipitation patterns over the Atlantic could impact ocean primary productivity and CO_2 flux, since aquatic biota are fertilized by iron and other trace nutrients from Saharan dust. Interestingly, modeling studies have shown that increased CCN and ice nuclei can increase or decrease precipitation and convective intensity, depending on environmental conditions and the stage in the storm's lifecycle [*Khain et al.*, 2008]. Future, more sophisticated modeling should examine this effect for the eastern Atlantic convective environment.

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