Measurements of Saharan Dust in Convective Clouds over the Tropical Eastern Atlantic Ocean*

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ABSTRACT

Mineral dust particles have been shown to act as cloud condensation nuclei, and they are known to interact with developing tropical storms over the Atlantic downwind of the Sahara. Once present within liquid droplets, they have the potential to act as freezing ice nuclei and further affect the microphysics, dynamics, and evolution of tropical storms. However, few measurements of mineral dust particles in tropical convective clouds exist. This study indicates that about one-third of droplets sampled in small convective clouds in the tropical eastern Atlantic contained dust particles, and dust was the dominant residual particle type sampled in ice crystals from anvil outflow. However, estimated number and mass concentrations of dust in anvil ice were small compared to the amount of dust available within the Saharan air layer itself.

1. Introduction

Mineral dust is one of the largest contributors to aerosol mass in the atmosphere, and blankets vast areas over Earth's continents and oceans (Heintzenberg et al. 2000; Darwin 1839). Dust is also an important aerosol type for cloud formation, since it may act as both a cloud condensation nucleus (Twohy et al. 2009; Karydis et al. 2011) and an ice nucleus (DeMott et al. 2003; Twohy and Poellot 2005; Sassen et al. 2003; Connolly et al. 2009). Microphysical changes in cloudy areas impacted by dust may therefore affect cloud albedo and precipitation efficiency (Albrecht 1989; Rosenfeld et al. 2001; Mahowald and Kiehl 2003; Koehler et al. 2007; Twomey 1974).

In deep convective clouds, dust particles acting as cloud condensation nuclei (CCN) can result in more, smaller droplets, which when lifted above the freezing

level can subsequently release latent heat through ice formation and growth (Khain et al. 2005; Jenkins and Pratt 2008). This effect can strengthen updrafts and prolong the storm's life cycle (Andreae et al. 2004) and impact anvil characteristics (Fan et al. 2013), although effects vary with life cycle stage (van den Heever and Cotton 2007) and environmental conditions (Khain et al. 2008). In the case of dust particles, their activity as ice nuclei may change the distribution of latent heat and storm characteristics further. The removal of dust by precipitating clouds can also influence ocean productivity (Fung et al. 2000; Mahowald et al. 2011; Deboudt et al. 2012) through transfer of limiting nutrients such as iron from the atmosphere to the oceans.

Over the Atlantic, large quantities of dust particles travel westward with the Saharan air layer (SAL)—a warm, dry, rapidly moving air mass originating from the African continent that is Earth's largest dust source (Carlson and Prospero 1972). Aircraft measurements indicated that SAL particles larger than 0.1- μm diameter are primarily unprocessed mineral dust without detectable soluble sulfates or salts (Chen et al. 2011). These aluminosilicates can still act as CCN owing to partially soluble components (Kelly et al. 2007; Koehler et al. 2009) or through surface adsorption alone (Kumar et al. 2011). Individual droplets from a liquid cloud embedded directly within the SAL contained unprocessed dust particles, and studies have shown that even submicron dust particles can be activated under moderate updraft

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conditions (Twohy et al. 2009; Heymsfield et al. 2009). In two low-altitude samples from the eastern Atlantic marine boundary layer, dust was common in clear-air particles larger than $0.1\text{ }\mu\text{m}$ (Chen et al. 2011).

Observations suggest that interaction of the SAL with Atlantic storms can initially invigorate them (Jenkins et al. 2008; Zipser et al. 2009; Storer et al. 2014) but over time may inhibit their development (Dunion and Velden 2004; Evan et al. 2006). These effects may be due to the characteristically dry, high-shear air mass or to microphysical–thermodynamic actions of the dust particles themselves, or both. Additionally, the extent to which the SAL is able to penetrate to protected inner areas of developing tropical storms is unknown (Montgomery et al. 2012). This work summarizes the composition of residual particles extracted from droplets in small warm clouds and from anvil ice in deep convective clouds, near the Saharan dust source. In addition, dust concentrations within small cumuli and anvil ice are estimated.

2. Methods

Data were obtained using the National Aeronautics and Space Administration (NASA) DC-8 research aircraft during the 2006 NASA African Monsoon Multidisciplinary Analyses (NAMMA) field program (Zipser et al. 2009). The aircraft was based about 700 km west of the Mauritania coastline in the Cape Verde Islands. Flights targeted convective clouds and developing storms over the ocean in this tropical northeastern Atlantic region, which is often impacted by Saharan dust. Sampling was conducted in August–September, when most emissions are from dust regions in northern Africa, rather than from the Sahel region where they may be mixed with biomass burning particles (Lieke et al. 2011; Rodríguez et al. 2011).

Cloud droplets larger than about $8\text{-}\mu\text{m}$ diameter and ice crystals larger than about $5\text{ }\mu\text{m}$ were sampled with an airborne counterflow virtual impactor (CVI) (Noone et al. 1988) to assess the amount of dust actually incorporated into these clouds. The CVI utilized a dry nitrogen counterflow and sample flow to remove particles smaller than the cut size and to evaporate or sublimate hydrometeors at about 60°C . Once collected by the CVI, dry, nonvolatile residual particles from hydrometeors were counted and sized with a Particle Measuring Systems 1001 LASAIR optical particle counter, which measured light scattered by particles and classified them into eight size bins between 0.1- and $>2\text{-}\mu\text{m}$ diameter. Particles in the same size range were captured by a two-stage jet impactor onto formvar-coated nickel grids and then analyzed by transmission electron microscopy

(TEM) and energy dispersive x-ray analysis. Individual particle types were classified based on their elemental composition and morphology (see Table S1 in the online supplement). Particles with an iron and chromium signature similar to the stainless steel inlet composition and mixed salt–metal types, as discussed by Cziczo and Froyd (2014), were presumed to be sampling artifacts and removed from the compositional results presented here. Additional information related to sampling, TEM analysis and calculations is given in the online supplement.

3. Results

a. Dust in marine boundary layer cumulus clouds

Individual droplet residual particles from three different small cumuli cloud fields within the marine boundary layer were collected during NAMMA. Droplets in these nonprecipitating, warm clouds were smaller than the size threshold for breakup within the CVI, and no ice was present, so one droplet can be assumed to produce one residual particle (Twohy et al. 2003). A large percentage of the droplet residual particles were salts—most derived from sea salt and some reacted with sulfate (Fig. 1). However, a significant fraction of the drops sampled contained dust. Dust was common in both particle diameter ranges ($0.12\text{--}0.43\text{-}$ and $>0.43\text{-}\mu\text{m}$ dust volume equivalent diameter), although larger dust particles were more often internally mixed with sea salt or sulfate, as observed by Levin et al. (2005). The percentage of dust plus internally mixed dust cloud residuals ranged from 16% to 52% by number of the total particles collected, with a number-weighted average of 32% across all samples and sizes.

These results can be combined with CVI sampling characteristics and peak droplet concentrations as described in the online supplement to estimate that between 23 and 209 cm^{-3} droplets in the MBL may contain dust particles. This is a relatively large number that could potentially act as freezing ice nuclei in deep convective clouds. Dust measurements in the anvils of these cloud types are described below.

b. Dust in anvil ice clouds

Tropical storms typically develop south of the SAL and can interact with it along their northern and western flanks as the dry air and dust wraps around them (Fig. 2). To assess how much dust actually reaches anvil levels in eastern Atlantic storms where it may influence cloud microphysical and radiative properties, five samples of ice crystal residual particles from anvil outflow were analyzed. The 1 and 12 September flights were associated with recent SAL outbreaks from the continent,

FIG. 1. Composition of low-cloud residual particles, as a percentage by number of different particle types based on predominant elements as described in Table S1 in the online supplement. (a) Small residual particles (for these low-altitude conditions, between about 0.19- and 0.67- μm aerodynamic diameter or from 0.12- to 0.43- μm volume equivalent diameter for dust). (b) Residual particles larger than 0.67- μm aerodynamic diameter or 0.43- μm volume equivalent diameter. (c) Number-weighted average composition for all samples and sizes. Sample 8/26-7 is from 26 Aug 2006 (1937:05–1941:00 UTC) at 17.3°N, 24.7°W. Sample 8/30-3 is from 30 Aug 2006 (1645:55–1653:30 UTC) at 13.2°N, 24.0°W. Sample 8/30-5 is from 30 Aug 2006 (1848:20–1853:00 UTC) at 20.1°N, 23.6°W [previously published in Fig. 16 of Zipser et al. (2009) and shown here for aggregation purposes]. Fifty particles were analyzed for each size range per sample.

evidenced in Fig. 2 by the strong signal along the African coast north of about 16°N. On 12 September, a line of strong convection was encountered within the western sector of the storm immediately adjacent to the SAL

(Zipser et al. 2009; Heymsfield et al. 2009). Ice crystals in the anvil outflow were sampled just before the aircraft penetrated an updraft of about 23 m s^{-1} . Dust was less apparent near the storm sampled on 3 September.

FIG. 2. *Meteosat-9* Saharan-air-layer images for dates shown above each panel, with anvil sample locations in the green circles. The *Meteosat* SAL product shown is sensitive to not only suspended dust but also to dry air; thus, the true dust signal is attenuated in moist areas adjacent to convection. Green numbers refer to sample numbers for the dates shown, as given in Fig. 3 and Table 1. Sampling altitudes ranged from 8 to 11 km (from 243 to 227 K). (a) The two anvils sampled on 1 Sep were associated with an east–west trough producing a line of thunderstorm clusters. The first sample was close to the main system, whereas the later sample was associated with light cirrus along the coast, separated from the main storm system. (b) Samples from 3 Sep were associated with an easterly wave, with its circulation center over the Cape Verde Islands. (c) The 12 Sep sample was from the upper layers of a tropical depression that later became Hurricane Helene. Cooperative Institute for Meteorological Satellite Studies (CIMSS)/University of Wisconsin–Madison provided satellite images.

FIG. 3. Percentage by number of various residual particle types associated with ice crystals in NAMMA anvils, with artifacts removed as described in the text. (a) Small residual particles (for these high-altitude conditions, between about 0.10- and 0.54- μm aerodynamic diameter or from 0.064- to 0.35- μm volume equivalent diameter for dust). (b) Residual particles larger than 0.54- μm aerodynamic diameter or 0.35- μm volume equivalent diameter. (c) Number-weighted average composition for all samples and sizes. Sample 9/1-2 is from 1 Sep 2006 (1533:25–1535:45 UTC) at 9.8°N, 16.5°W. Sample 9/1-3 is from 1 Sep 2006 (1658:40–1701:00 UTC) at 15.2°N, 16.1°W. Sample 9/3-2 is from 3 Sep 2006 (1404:50–1406:40 UTC) at 15.0°N, 23.8°W. Sample 9/3-3 is from 3 Sep 2006 (1450:45–1453:45 UTC) at 11.7°N, 25.4°W. Sample 9/12-1 is from 12 Sep 2006 (1249:15–1251:40 UTC) at 13.0°N, 25.2°W, but an earlier time (1247:44–1249:20 UTC) was used for mass calculations (see Table 1 footnote). Fifty particles were analyzed for each size range per sample, except for 9/1-3 and 9/12-1, for which 25 and 80 particles were analyzed, respectively, from the large particle stage.

1) COMPOSITION OF ICE CRYSTAL RESIDUAL PARTICLES

The anvil samples exhibited a variety of residual particle compositions (Fig. 3), but all had substantial percentages of dust, ranging from 19% to 61% of the total particles analyzed. On average, dust was the predominant residual type (50% when dust particles mixed with sulfate or salts are included). Another 13% of particles were nonsilicate, metallic particles that could be naturally present in soil dust or released by mining or other industrial processes; they included elements such as iron, copper, aluminum, and titanium. Interestingly, these percentages of dust and metals are very similar to those found by Cziezo et al. (2013) in cirrus much farther west (North America, Central America, and nearby ocean areas), suggesting that Saharan dust may influence cirrus formation thousands of kilometers downwind of its origin.

The NAMMA anvil dust particles had primarily aluminosilicate clay compositions and were as small as 0.1 μm in diameter—similar to dust measured directly in the SAL (Twohy et al. 2009). While dust mixed with

hygroscopic salts or sulfate was common in clouds in the marine boundary layer, on average only about one-fifth of the anvil dust particles were mixed with hygroscopic material. This suggests that much of the anvil dust originates from the midlevel SAL rather than the marine boundary layer (e.g., Avery et al. 2010) and may also be indicative of rapid upward transport of dust inside droplets, with relatively little time for chemical processing (Yin et al. 2005).

2) DUST NUMBER AND MASS CONCENTRATIONS IN ANVIL ICE

The number concentration of dust particles larger than 0.1 μm in anvil ice was estimated from the optical particle counter and the TEM composition results. Residual particles collected by the CVI represent actual ice nuclei in single or aggregate crystals, plus (if present) material in frozen droplets and nonvolatile particles scavenged subsequent to droplet or ice formation. Thus, residual concentrations/composition should not be assumed to represent ice nuclei alone but, instead, the aerosol material present in the condensed phase. Residual number concentrations were similar to ice crystal

TABLE 1. Mean number and mass concentrations for the five anvil cases.

Sample date-No.	Total residual number ^a (cm ⁻³)		Dust residual number ^b (cm ⁻³)		Dust mass in ice ^c (μg m ⁻³ air)		Submicron dust mass (percentage of total)	Ice water content ^d (g m ⁻³) (σ)	Dust mass/ice mass (μg g ⁻¹)
	Ambient	STP ^e	Ambient	STP ^e	Ambient	STP ^e		Ambient	
9/1-2	0.66	2.1	0.23	0.74	0.066	0.21	17	0.056 (0.044)	1.2
9/1-3 ^f	0.13	0.46	0.08	0.28	0.008	0.03	28	0.022 (0.016)	0.38
9/3-2 ^f	0.65	1.6	0.15	0.35	0.046	0.11	16	0.17 (0.23)	0.26
9/3-3	0.14	0.46	0.06	0.22	0.014	0.05	29	0.036 (0.037)	0.39
9/12-1	6.0	20	3.8	13	0.85	2.9	15	0.13 (0.099)	6.5

^a Number concentrations of residual particles larger than 0.1-μm diameter as measured by LASAIR.

^b Number concentrations of dust particles larger than 0.1-μm diameter as determined by LASAIR and TEM.

^c Dust mass calculated based on LASAIR size distributions and compositions in Fig. 3. See online supplement for details.

^d Mean ice water content from CVI measurements if 1-Hz data are greater than 0.002 g m⁻³ (σ is standard deviation).

^e STP is the standard temperature and pressure of 288 K and 1013 hPa.

^f Because of clear-air gaps during these sample periods, concentrations were corrected for time in cloud using CVI ice water content greater than 0.002 g m⁻³ to define “in cloud.”

number concentrations measured by a 2D-S probe in the two 3 September cases, 6–34 times larger for the two 1 September cases, and could not be reliably determined for the 12 September case. While interstitial aerosol measurements were not available, dust particles reaching the anvil were likely mostly in the condensed phase. See the online supplement for reasons and more sampling details.

Results for the five cases are given in Table 1. In four cases, dust residual concentrations in ice were approximately 0.2–0.8 cm⁻³ [values given here are at standard temperature and pressure (STP) for comparison to lower-altitude concentrations]. The 12 September sample near the vigorous updraft impacted by dust (Zipser et al. 2009; Heymsfield et al. 2009) had a higher dust concentration of 13 cm⁻³ (at STP). In contrast, dust concentrations in the SAL layer are typically about 200 cm⁻³ (Chen et al. 2011), and as shown earlier, about 100 cm⁻³ of boundary layer cloud droplets may contain dust. Thus, although dust is more prevalent than other particle types in anvils, it constitutes a relatively small number percentage (~0.1%–10%) of the dust present at low altitudes.

The fraction of dust particles shown in Fig. 3 and the number concentration measured by the optical particle counter in corresponding size ranges were used to estimate the mass concentrations of dust in cirrus residuals for each sample period (Table 1). Because of potential errors in deriving mass concentrations from size distributions, uncertainty in calculated dust mass concentrations may be at least a factor of 2. However, an estimate of dust mass in thunderstorm anvils based on actual data over the eastern Atlantic has not been available before, and important conclusions can still be made based on approximate values calculated here.

Estimated anvil dust mass concentrations ranged from about 0.03 to 0.2 μg m⁻³ at STP for the 1 and 3 September

samples, with higher values of about 2.9 μg m⁻³ at STP for the 12 September anvil sample where lidar showed dust entraining near the vigorous updraft (Zipser et al. 2009). These values are only about 0.1%–1% of those within the SAL itself, which can contain hundreds to thousands of micrograms of dust per cubic meter of air (Weinzierl et al. 2009; Chen et al. 2011). Since anvil samples presented here represent a range of anvil ages and thicknesses, results were also normalized by ice water content. Normalized concentrations were similar for the 1 and 3 September samples, ranging from about 0.3 to 1.2 μg of dust per gram ice, with higher concentrations of 6.5 μg of dust per gram of ice for the 12 September cloud. While submicron particles dominated residual number concentration in all cases (96%–99% of total), dust mass was predominately in the coarse mode—consistent with clear-air SAL data (e.g., Chen et al. 2011).

4. Discussion

Given the ability of dust particles to act as both CCN and freezing nuclei discussed earlier, the relatively low concentrations of dust in eastern Atlantic storm anvils are somewhat surprising. Yet this is consistent with Lawson et al. (2010), who found that anvil ice crystal number concentrations measured directly during the NAMMA project were similar to those measured in anvils over the tropical eastern Pacific during another project. This indicates no widespread enhancement of ice concentrations in anvils by dust particles in the eastern Atlantic. The generally low dust concentrations in NAMMA anvils could result from a number of factors, which are being further studied by modeling: 1) Dust within the SAL may infrequently penetrate storm centers where it can be lofted to anvil altitudes or may penetrate only in specific storm regions (Rutherford and

Montgomery 2012). The much higher dust concentrations for the 12 September case suggest sampling location is important. 2) Dust particles that do act as nucleating agents within the updraft may be efficiently precipitated out. In fact, “muddy rain” is observed in dust-impacted regions (Prodi and Fea 1979), and high-resolution modeling studies that include dust as CCN predict that large amounts of dust can be removed to the ocean surface in precipitation (Twohy et al. 2012). 3) Simple dilution of anvil ice concentrations after entrainment of environmental air, sublimation of smaller crystals, and sedimentation of larger ones (Jensen et al. 2009) can reduce ice concentrations by about a factor of 10 between convective turrets and attached anvils (Lawson et al. 2010).

5. Summary and conclusions

Mineral dust was found in both liquid cloud droplets and ice crystals over the Atlantic downwind of the Sahara–Sahel region. While dust in the boundary layer was often mixed with sea salt or sulfate, small cloud droplets sometimes contained unmixed mineral dust, confirming that dust particles can act as CCN. When dust particles within droplets are lifted in deep convection, they have the potential to provide many freezing nuclei at mid- and upper levels of tropical storms. Measurements show that mineral dust is the dominant aerosol type sampled in anvil ice from three storms over the eastern tropical Atlantic with varying degrees of Saharan-air-layer influence. However, estimated dust number and mass concentrations in anvil ice were typically two to three orders of magnitude less than those within the SAL itself. Further measurements and modeling studies are needed to fully understand how much dust is lofted by tropical storms and how much is transferred from the atmosphere to the ocean in precipitation.

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