INTERACTION OF SAHARAN DUST WITH LIQUID AND ICE CLOUDS

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1. INTRODUCTION

Crustal dust originates from Africa in the summer months. Dust liberation is anti-correlated with precipitation frequency in the Sahel region, and has increased in magnitude over the past few decades (Prospero, 1996). The impact of this dust on tropical convection is potentially large. If it contains soluble material, dust may act as a cloud condensation nucleus (CCN), decreasing mean droplet size and inhibiting precipitation (Rosenfeld et al., 2001; Mahowald and Kiehl, 2003; Koehler et al., 2007). Dust is also known to be an effective ice nucleus (DeMott et al, 2003; Twohy and Poellot, 2005; Field et al. 2006, etc). 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. However, dust nucleation impacts on microphysics, latent heat release and vertical transport (e.g., Khain et al., 2005; Van Den Heever et al. 2006) could also impact convection development in complex ways.

Analysis of microphysical properties of small cumulus clouds over the ocean reveal that in most cases, number concentrations were higher than expected for clean marine clouds, despite low liquid water contents. This suggests that a substantial fraction of dust and other non-marine particles may be acting as cloud condensation nuclei (CCN) in the region. Here, the chemical properties of dust and actual cloud residual nuclei in the tropical Eastern Atlantic, where few measurements exist, are presented. Modeling simulations are used to further study activation of dust as cloud condensation nuclei and its possible role in affecting microphysical properties and precipitation in deep convection.

2. OBSERVATIONS

In the NASA African Monsoon Multidisciplinary Activities (NAMMA) experiment, aerosol particle physiochemical characteristics and cloud size distributions were measured aboard the NASA/University of North Dakota DC-8 aircraft in summer of 2006. Both low-level small cumulus clouds, deep convection, and anvil cirrus outflow from mesoscale systems impacted by various amounts of dust were sampled.

Ambient aerosol and cloud residual particles were collected with a counterflow virtual impactor (CVI, Noone et al., 1988) to assess the percentage and size of dust particles actually incorporated into these clouds. The CVI removes interstitial aerosol and collects and evaporates droplets or ice crystals, while retaining their individual residual nuclei. It can also be used as an ambient aerosol inlet outside of cloud, by turning off the counterflow out the tip. Once collected by the CVI, residual particles were captured by a two-stage jet impactor. The small particle stage collected 0.17 to

0.65 μm diameter unit-density spherical particles, corresponding to 0.11 to 0.48 μm diameter for 1.7 g cm^{-3} density particles. The large particle stage collected larger particles up to several microns in size. For simplicity of display, percentages from both stages have been averaged, but when there are substantial differences as a function of size those differences are noted.

Particulate 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, industrial metals, sulfate, carbonaceous, or mixtures of these types as in Twohy and Poellot (2005). While sulfate aerosols can be routinely detected, volatile material like HNO_3 and volatile organics will be underrepresented by this technique.

3. RESULTS

3.1 Dust Near the Source

On 5 Sept 2006, a mission was flown directly over the Sahara Desert, and air was sampled at several altitudes near and downwind of the dust source.

The first ambient sample was collected at 2.1 km directly over Mauritania in Northwestern Africa. The top of the dust layer was at about 6 km. These particles from near the dust source were primarily unmixed crustal dust, with aluminosilicates being the most common type (Fig. 1). About 20% of the particles (by number) were dust mixed w/ soluble material like sulfur or chlorine, and a few particles were metals without significant silicon which could be different types of mineral dust. The HYSPLIT back trajectory may help explain the mixed particle types, as it showed some interaction of the air with the Atlantic and Mediterranean oceans four to seven days prior to sampling.

A typical X-ray spectrum (Fig. 2) reveals clay-like particles that contain not only insoluble elements like aluminum, silicon, and iron, but also soluble or slightly soluble elements like potassium, calcium and magnesium. As opposed to kaolinite which is more common closer to the equator (Prospero 1981), these smectitic clays are expected to adsorb water and other polar substances (organics, sulfuric and nitric acid).

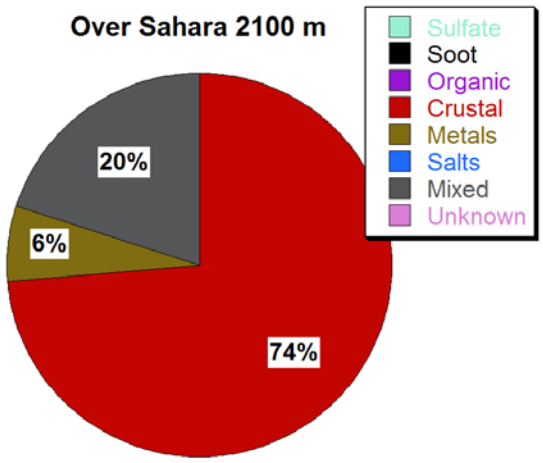

Fig. 1. Percentage of different particle types by number from a NAMMA aerosol sample over Mauritania (100 particles total on 2 size stages analyzed). About 20% of the dust particles were mixed with soluble material. Note that the "metals" category could include iron-containing crustal material with little or no silicon.

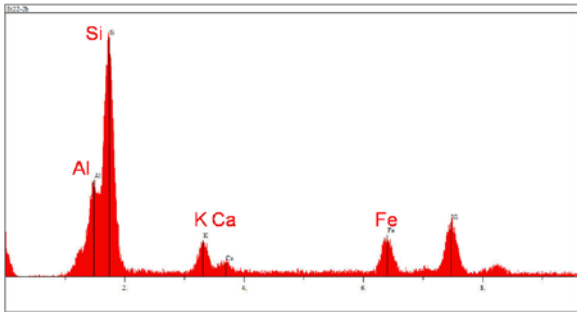

Fig. 2. X-ray spectrum of typical Saharan dust particle collected near the source. Horizontal axis is electron volts (in thousands) and vertical is intensity or counts. Predominant elements are silicon, aluminum, iron, potassium and calcium. The nickel peak is from the collection grid substrate material.

3.2 Interaction with Marine Air

The second ambient sample presented was collected just off the Mauritanian coast at low level (0.3 km). In contrast to the sample directly over the Sahara, Figs. 3, 4 and 5 show that a large percentage of particles in the marine boundary layer were internally mixed particle types. These were primarily dust with sulfate (in the small particle size range) and dust with sea-salt (in the larger size range.) The source of sulfur could either be pollution from Europe, or the ocean itself. The region just off the coast of northwestern Africa is an area of upwelling and high primary productivity, as evidenced by enhanced levels of chlorophyll, DMS and soluble nitrogen (Robinson et al., 2006). Others (Andreae et al, 1986; Levin et al., 1996) have also noted internally mixed dust particles in the marine atmosphere.

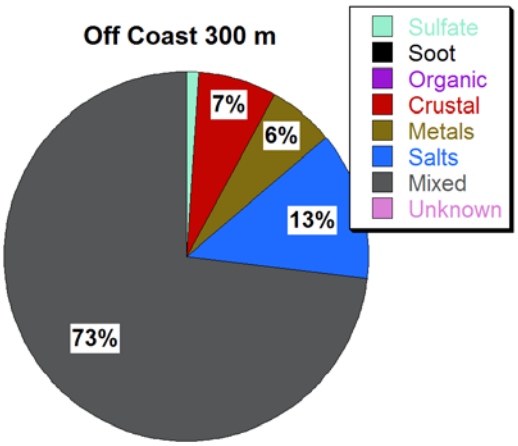

Fig. 3. Composition (% by number) of particles larger than 0.1 µm collected in the marine boundary layer off the coast of western Africa (Mauritania).

Thus small amounts of soluble material are not only naturally present in dust near the source, but increased amounts of soluble material are added through atmospheric processing. This material would make the dust more likely to act as cloud condensation nuclei and be assimilated into the lower and mid-section of convective systems where it may affect the microphysical, radiative and thermodynamic characteristics of these storms. Later, we show that dust and dust/salt/sulfate mixtures comprise a significant fraction of the droplet residual nuclei in small cumulus clouds in this region.

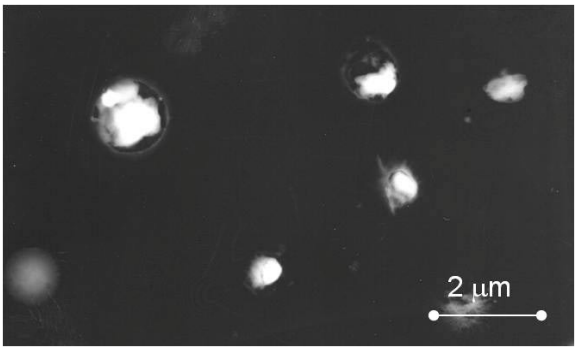

Fig. 4. Aerosol particles collected in the marine boundary layer just off the coast Africa. Particles are mixed with soluble material; an example of elemental analysis given in Fig. 5.

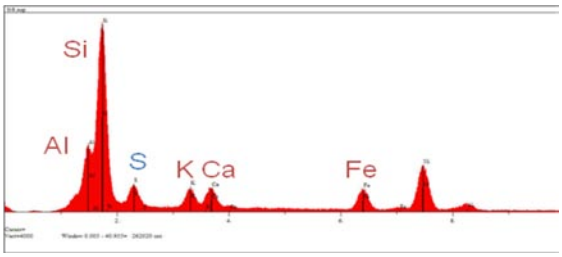

Fig. 5. X-ray spectrum of dust particles in the marine boundary layer mixed with sulfate. Horizontal axis is electron volts (in thousands) and vertical axis is intensity or counts.

3.3 Dust in a MidLevel Cloud

The third sample is representative of dust further offshore, but at higher altitudes with less interaction with marine aerosol. This 3.7 km sample was almost entirely composed of dust aerosol without detectable sulfate or chloride (Fig 6 and 7). A thin water cloud actually embedded in the SAL layer was sampled at 4.3 km. Residual nuclei were also almost entirely dust, with only small amounts of salts and mixed particles present (Fig. 8).

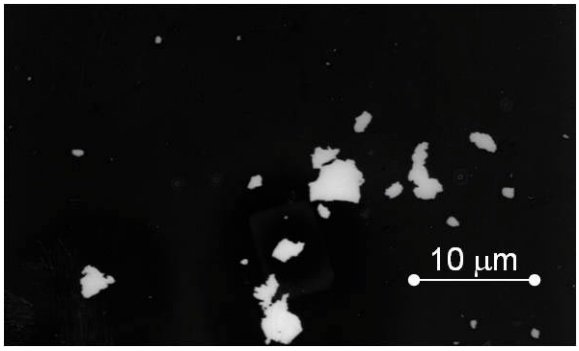

Fig. 6. Aerosol particles collected offshore in the SAL layer at 3.7 km showing dry with little or no non-volatile soluble material.

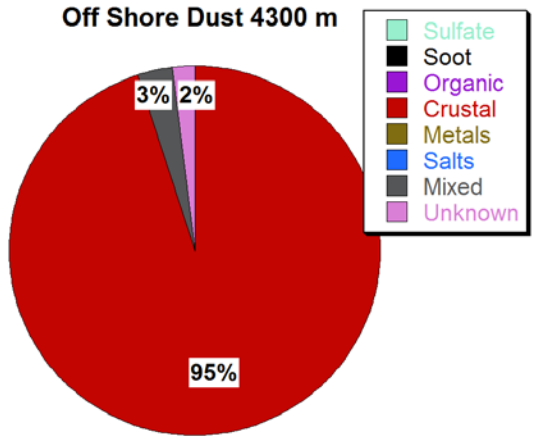

Fig. 7. Composition of aerosol in dust plume at high altitude offshore.

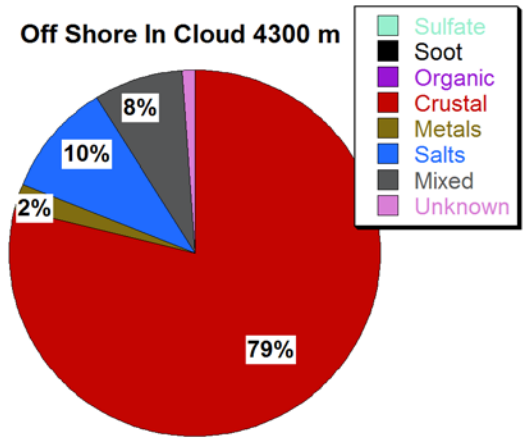

Fig. 8. Composition of residual particles from liquid cloud embedded in dust layer.

3.2 Interaction with Marine Cumulus Clouds

Small cumulus clouds were sampled in the marine boundary layer on several flights. In these cases, the bulk of the dust layer was actually above the low-level clouds, so it was of interest to study if and how the dust interacts with underlying clouds. Samples from three different cloud fields on two days were collected. The first day, shown in Fig. 9 (26 Aug 2006) had back-trajectories from over Africa, while air on the second day (30 Aug 2006) was more from the northeast over the ocean. As expected, high concentrations of salts, most derived from sea-salt and often reacted with sulfate, were observed. But over 40% of the drops sampled contained either solely dust or dust mixed with non-volatile soluble material (Fig. 9). For the three samples (and depending on particle size), the percentage of dust plus internally-mixed cloud residuals ranged from 14% to 54% by number of the total collected.

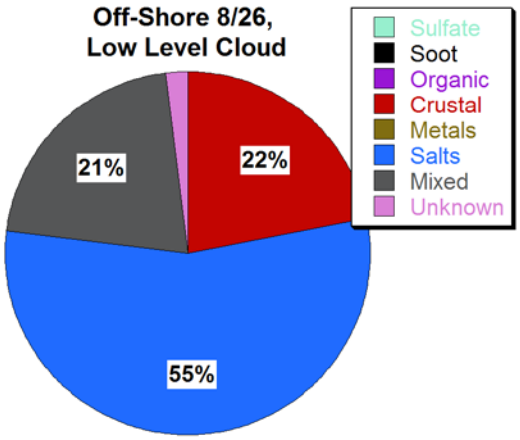

Fig. 9. Percent (by number) of various particles larger than 0.1 μm found in residual nuclei from small cumulus clouds downwind of the Sahara on 8/26/06. Similar results were observed on 8/30/06. "Mixed" particle types were usually crustal dust with soluble material like sulfate or sea-salt.

Average droplet concentrations in these clouds ranged from about 50 to 500 cm^{-3}, with peak concentrations much higher. We

estimate, based on residual particle size distributions, that about half or more of the droplet nuclei were measured by this technique. Even if the smaller, unmeasured nuclei contain no dust, this is still a potentially large number of droplets containing dust that may later act as ice nuclei in deep convection.

4. MODELING

A Lagrangian parcel model based on Feingold and Heymsfield (1992) was used to simulate activation of dust as CCN in this environment for a range of different updraft velocities. The original model was modified to parameterize water activity and hygroscopic growth with a single parameter Kappa, κ (Petters and Kreidenweis, 2007). Kappa scales with the fraction of soluble material and is about 1.3 for sodium chloride and about 0.6 for ammonium sulfate. Kappa is 0.00 for a completely insoluble and wettable particle and has been measured to be 0.054 for Saharan dust (Koehler, 2008). This is in general agreement with the typical fraction of soluble and partially soluble material (Ca, K) in fresh Saharan dust as measured in our electron microscope analysis.

The input aerosol distribution was bimodal and based on typical accumulation-mode measurements in the tropical marine boundary layer for the soluble mode (Heintzenberg et al., 2000) and on NAMMA-measured size distributions for the dust mode. Two cases were studied, both with a soluble (sea-salt or sulfate) mode with κ=1.0, N_a=240 cm^{-3}, d_g=0.16, and σ_g=1.5. For the dust mode, N_a=46 cm^{-3}, d_g=0.56, and σ_g=2.0, but with two different Kappa values: κ=0.00 for Case 1, and κ=0.05 for Case 2.

Fig. 10 shows that activation characteristics for the soluble mode are expected to be similar in both Case 1 and 2. For Case 1 with κ=0.00, only a small fraction of dust particles (the largest ones) activate at low updraft velocities. However, the small amount of soluble material present naturally in the dust in Case 2 allows it to be activated with high efficiencies, even at relatively small updraft velocities. In fact, it activates in similar fractions as soluble particles like salt or sulfate.

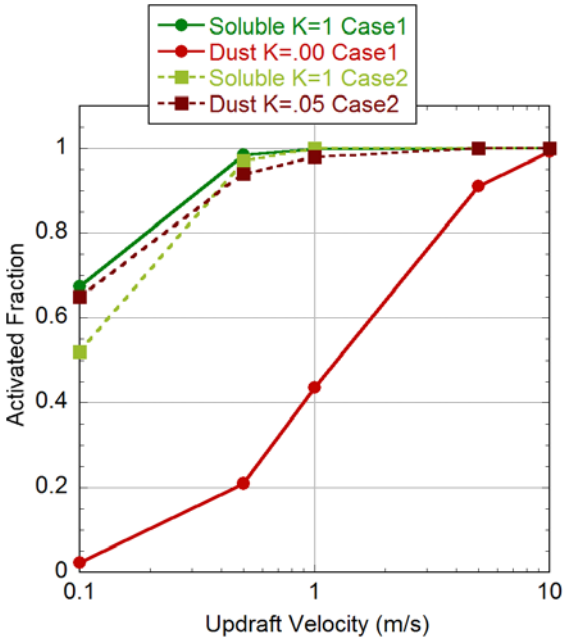

Fig. 10. Fraction of aerosol particles, of each individual mode, activated as a function of modeled updraft velocity. Case 1: Soluble smaller mode with κ =1.0 and larger dust mode with κ =0.00. Case 2: As in Case 1, but with κ =0.05 in the dust mode. Ambient conditions were 900 mb and 292 K. Size distributions described in the text.

Other simulations (not shown) demonstrate that nearly all dust with κ =0.05 would also be activated in the case shown in Fig. 7 and 8, where a cloud formed in an embedded dust layer at mid-levels.

5. DISCUSSION AND CONCLUSIONS

Saharan dust has soluble components that contribute to its ability to act as a cloud condensation nucleus in liquid clouds. This material may be present naturally in the dust at the source, as well as added afterward by interaction with atmospheric gases and particles. Using direct measurements

of cloud residual nuclei, we have demonstrated that Saharan dust acts as CCN. In high dust situations such as in the Eastern Atlantic, dust can contribute substantially to the number of droplets in small cumulus clouds.

If even a small fraction of the dust submerged in droplets is lifted to cold temperatures in deep convection, a substantial change in ice concentration is likely. Additional impacts may occur through changes in vertical profiles of latent heat. Interestingly, modeling studies have shown that increased CCN and ice nuclei can increase or decrease precipitation and convective intensity, depending on environmental conditions (Khain et al., 2008). Future modeling work will examine this effect for the Eastern Pacific convective environment.

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