Mineral dust aerosol size distribution change during atmospheric transport

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Abstract. Airborne mineral dust can impact visibility, climate, biogeochemical processes and possibly human health. The magnitude of the impact of dust depends particle size. We measured the size distributions of airborne mineral dust over the Canary Islands during July 1995 and Puerto Rico during July 2000. Dust size distributions do not appear log normal. Stokes gravitational settling overestimates losses of large dust particles during atmospheric transport from North Africa over the tropical North Atlantic and Caribbean. Normalized mineral dust size distributions of particles smaller than 7.3 µm over the Canary Islands and Puerto Rico were indistinguishable indicating these particles were not preferentially removed during atmospheric transport. However, mineral dust aerosols larger than 7.3 µm were preferentially removed during atmospheric transport. Larger particles were more efficiently removed. A simple empirical model setting the vertical velocity of dust particles equal to the Stokes gravitational settling velocity minus an upward velocity of ~0.33 cm sec⁻¹ accurately predicts changes in dust size distribution during atmospheric transport. Thus it appears some atmospheric process(s) partially counteracts gravitational settling.
1. Introduction

Large amounts of mineral dust are transported through the atmosphere degrading visibility, modifying the earth’s energy budget [IPCC, 1996], affecting biogeochemical processes [Duce et al., 1991; Martin et al., 1994; Shinn et al., 2000], contributing significantly to deep-sea sediments [Pye, 1987], and possibly exacerbating symptoms in people with asthma. The size distribution of air borne dust affects radiative transfer in the atmosphere influencing weather and climate. In addition, studies of paleoclimate use size distribution measurements of aeolian dust in marine sediments to infer climatic conditions over geologic time [Rea et al., 1985].

The vast majority of the mass of mineral dust particles transported from arid continental areas through the atmosphere to remote marine regions range in size from <0.5 to >75 µm diameter (unless specified otherwise, “diameter” hereafter refers to geometric diameter) [Jaenicke and Schütz, 1978; Betzer et al., 1988; Maring et al., 2000]. During atmospheric transport, dust size distributions shift to smaller sizes likely because gravitational settling preferentially removes larger particles. However, measurements indicate the presence of dust particles in the marine troposphere larger than is consistent with Stokes gravitational settling alone controlling removal [Schütz et al., 1981]. Unexpectedly large dust particles (diameters > 50 µm) have been found over the remote ocean [Betzer et al., 1988; Westphal et al., 1986]. Also, a number of studies have been cited [Andreae et al., 1986], which noted surprisingly constant dust size distributions over a wide range of remote marine regions.

Cloud processing has been hypothesized to physically combine (i.e., internally mix) dust and sea salt particles [Schütz et al., 1981, Andreae et al., 1986]. Internal mixing of dust and sea salt may be important at very low dust concentrations (the samples on which one of those studies
[Andreae et al., 1986] based their conclusions had dust concentrations <1 µg m$^{-3}$) and in very aged aerosols. However, internal mixing is unlikely to be important in regions, such as the tropical North Atlantic and Caribbean, where dust concentrations are often elevated (i.e., >10 µg m$^{-3}$) and dust particle number concentrations are often much larger than those of sea salt. Additionally, if internal mixing were important, the size distribution of dust in the marine boundary layer would be broader extending toward larger sizes than that in the free troposphere. However, we observed indistinguishable dust size distributions in the boundary layer and free troposphere from aircraft over Puerto Rico during July 2000 [Reid et al., 2002; Maring et al., this issue].

So, if internal mixing does not stabilize the size distribution of dust particles, why doesn’t the dust size distribution change more during atmospheric transport? Here we address this question by presenting measured and derived dust size distributions from the Canary Islands only 350 km west of North Africa, a major dust source, and from Puerto Rico more than 5000 km further downwind, west across the tropical North Atlantic and Caribbean.

2. Methods

We measured aerosol microphysics and chemistry at Izaña, Tenerife, Canary Islands during July 1995 and Puerto Rico during July 2000. It is possible that aerosols measured during the two field campaigns originated from difference sources due to the time lapse between experiments. The most substantial shift in North Africa aerosol source regions is seasonal. Satellite observations show dust source regions to be located in the Sahara during summer. During the winter, dust originates south of the Sahara in the Sahel [Husar et al., 1997; Herman et al., 1997].
Since the measurements reported here were made during July, we avoided the seasonal change in dust source area. Aeolian dust transport is a geological process. Thus 5 years is likely not enough time for a significant change in dust source regions. Therefore, we believe the data from July 1995 and July 2000 can be profitably compared.

Complete descriptions of the Izaña sampling site and experimental methods have been presented [Maring et al., 2000]. Briefly, aerosols were measured in the free troposphere at 2360 m asl at the Global Atmospheric Watch (GAW) Station (28.30°N, 16.48°W). Aerosol samples for chemical analysis were collected on upward facing 20x25 cm Whatman-41® filters changed after 12 hours of sampling. Sodium, a tracer for sea salt, was measured by flame atomic absorption spectrophotometry in deionized water extracts of sample filters. Dust concentrations were measured two ways; by measurement of aerosol mass after filter combustion and by instrumental neutron activation analysis (INAA) of aluminum, a tracer for mineral dust. A TSI® Aerodynamic Particle Sizer (APS) Model APS33 measured aerosols from 0.8 to >15 µm aerodynamic diameter and a TSI® Scanning Mobility Particle Sizer (SMPS) Model 3934L measured aerosols from 13 to 850 nm geometric diameter.

Our measurements in Puerto Rico were part of the Puerto Rico Dust Experiment (PRIDE). We deployed samplers and instruments at the Naval Station Roosevelt Roads at sea level on Cabras Island (Lat. ~18.21 N, Long. ~65.60 W) east (up wind) of the main island. Identical sampling and analytical protocols were used during PRIDE as in Izaña for bulk aerosol samples, except we did not utilize INAA. The omni-directional inlet we used during PRIDE was identical to the inlet we used in Izaña [Liu et al., 1983] except scaled larger to handle a higher flow rate. This inlet was mounted atop a 13 cm internal diameter (ID), 10 m tall vertical polyvinylchloride tube, which extended from the top of our portable laboratory (a standard 20 ft. aluminum
shipping container). The flow rate in the 13 cm ID tube was 200 l min\(^{-1}\), the maximum laminar flow rate. Approximately 90 l min\(^{-1}\) of sample air passed into a 4.8 cm ID thin wall aluminum tube. This air was supplied to our aerosol instrumentation. We used the same instrumentation to measure aerosol number size distribution at Puerto Rico as in Izaña. The PRIDE measurements were made at sea level in the marine boundary layer where the relative humidity often exceeded 80%. To make our measurements in Puerto Rico directly comparable to those in Izaña, we heated the sample air intake and instrumentation so measurements were made at relative humidities lower than 50%.

3. Results and Interpretation

3.1. Determination of Mineral Dust Aerosol Size Distribution

At Izaña in the free troposphere, aerosols with diameters >0.6 µm appeared to be almost exclusively mineral dust \cite{Maring et al., 2000}. A dust mass closure calculation indicated a dry dust aerosol density of 2.0 g cm\(^{-3}\). This density was used here to convert all aerodynamic to geometric diameters. Nearly the whole dust size spectrum was measured by the APS. Since the measurement technique is aerodynamic and the aerodynamic to geometric conversion used here was consistent, the Izaña and Puerto Rico dust size distributions can be compared without regard to particle shape or density. In addition, Stokes settling velocities can be calculated without regard of particle shape.

During PRIDE, aerosols with diameters >0.6 µm at Puerto Rico consisted of dust and sea salt. Typical surface area size distributions from both Izaña and Puerto Rico confirm the 0.6 µm
division between the accumulation mode versus dust and sea salt (Fig. 1). Thus hereafter we deal exclusively with particles whose diameters are >0.6 µm.

Since mineral dust particles were essentially the only particles with diameters >0.6 µm present in the free troposphere at Izaña, measurements of aerosol size distributions with diameters >0.6 µm are direct measurements of dust particles at that site. However, the size distributions of dust and sea salt particles overlap. Therefore, at Puerto Rico, we discriminated between dust and sea salt aerosols using the multi-step iterative process. We averaged individual, 20 min aerosol size distribution measurements over the 24 hr sampling periods of the bulk aerosol samples, enabling the association of aerosol chemical composition to mean aerosol size distributions. The discrimination procedure used data from sampling periods when dust concentrations were very low and sea salt concentrations were moderate to estimate the sea salt aerosol size distribution. Using that estimated sea salt aerosol size distribution, we estimated the dust aerosol size distribution by subtracting the sea salt component of the aerosol from sample period size distributions when sea salt concentrations were moderate and dust concentrations were high. The resulting dust size distribution was then used to refine the sea salt size distribution derived from sampling periods with moderate sea salt and low dust concentrations. This process was repeated until the dust and sea salt size distributions did not change significantly. Both the sea salt and dust size distributions produced by the second and third iterations were different by less than their standard deviations.

We tested the validity of this approach by comparing total aerosol volumes of particles with diameters >0.6 µm estimated from measured number size distributions, assuming spherical particles, versus that calculated using derived sea salt and dust mass normalized volume size distributions scaled by measured sea salt and dust mass concentrations. The absolute value of the
total aerosol volume differences averaged 12% of the total aerosol volume with a standard deviation of 7%, ranging from 2 to 22%. Given the errors inherent in the measurements and the uncertainties added during the derivation of the dust and sea salt size distributions, we believe this represents reasonable agreement.

The size distributions of sea salt and mineral dust at Puerto Rico during July 2000 overlap, but are not identical. Volume size distributions of dry sea salt and dust, normalized to peak value, show the dust size spectra to include larger particles (Fig. 2) than the dry sea salt spectra. The volume median diameter (VMD) of dry sea salt aerosols was 3.2 µm while that for dust was 3.6 µm with standard deviations of 0.3 µm. A t-test indicates the probability of the sea salt and dust VMD’s at Puerto Rico being different is 96%. The distribution of sea salt particles appears log normal. However, the mineral dust spectrum at Puerto Rico does not appear log normal.

Aircraft measurements of dust particles over Puerto Rico during July 2000 indicate dust size distributions to be uniform with altitude [Reid et al., 2002]. These measurements range from just above sea level to >4000 m asl and include the Saharan Air Layer (SAL) where most dust transport takes place [Carlson and Prospero, 1972]. Thus, we assume here that our sea level measurements represent the size distribution of dust particles both at the surface and aloft. Consequently, we can profitably compare measurements of dust size distribution in the free troposphere at Izaña and at sea level at Puerto Rico.

3.2. Characterization of Mineral Dust Aerosol Size Distribution Change

The dust size distribution changed during atmospheric transport (Fig. 3). Dust aerosol volume size distributions from Izaña and Puerto Rico are indistinguishable for particles <8 µm
diameter. However, the spectra diverge for larger particles, becoming substantially different for dust particles >10 µm. The dust aerosol spectrum at Izaña is even less log normal-like than that at Puerto Rico. The VMD for dust was 4.1 µm at Izaña, while at Puerto Rico the dust VMD was 3.6 µm with standard deviations of 0.3 µm. A t-test indicates the probability the VMD’s of dust particles at Izaña and Puerto Rico are different is >99%.

We characterized the change in dust aerosol size distribution during atmospheric transport across the tropical North Atlantic and Caribbean by calculating the fraction of particles lost between Izaña and Puerto Rico versus particle size (Fig. 4). Particles <7.3 µm appear not to be preferentially removed. The fraction of particles removed increases rapidly until at ~12 µm essentially all particles have been lost. The preferential removal of larger dust particles suggests gravitational settling is important in controlling the removal of dust particles from the atmosphere.

In an effort to quantify the processes controlling the removal of dust particles, we modeled the losses of dust aerosols. Our models assume a 2000 m tall air parcel located in the atmosphere between 2000 and 4000 m asl, typical of the SAL over the tropical North Atlantic and Caribbean [Carlson and Prospero, 1972; Karyampudi et al., 1999]. Inside the air parcel, dust particles start with the size distribution we observed at Izaña and are uniformly distributed in the vertical. We assumed the air parcel was externally vertically stable (i.e., no convective mixing from above or below), initially internally neutrally stable [Westphal et al., 1987], and that once a particle leaves the air parcel it is permanently lost. Further, we assumed any precipitation removal of dust particles did not change the dust aerosol size distribution. In each of the cases presented here (i.e., Models 1 & 2), we ran the models for 5.5 days, a reasonable approximation of the transport
time required for aerosols to travel from the west coast of North Africa to Puerto Rico [Reid et al., 2002]. Thus we are modeling processes taking place in the SAL.

Model 1 (Fig. 4) represents removal by Stokes settling alone and shows too great a loss of larger particles. In addition, our observations indicate a more abrupt transition from essentially no losses to total removal than is suggested by Stokes settling alone. These findings are consistent with previous comparisons of measured and modeled dust aerosol size distributions [Schütz et al., 1981].

The process(es) controlling dust aerosol removal from the SAL, and hence size distribution, remove particles more slowly than Stokes settling predicts. Further, the shift from virtually no preferential loss to essentially total removal versus particle size appears more like a step function than the gradual transition predicted by Stokes settling. Because many airborne dust particles, especially larger ones, have significant gravitational settling velocities, we assumed Stokes settling partially controls dust aerosol removal. However, we hypothesized an upward velocity inside the air parcel partially counteracting Stokes settling. This upward velocity reduces the dust removal rate and maintains a relatively uniform dust aerosol size distribution in the vertical. We estimated the magnitude of the upward velocity to be equal to the Stokes settling velocity of the particles located at the point where preferential losses become observable (i.e., 7.3 µm diameter with Stokes settling velocity of 0.33 cm sec\(^{-1}\)). In Model 2 we hypothesized a vertical velocity for dust particles equal to the Stokes gravitational settling velocity minus 0.33 cm sec\(^{-1}\). Calculated vertical velocities less than zero were assumed to be zero. Model 2 more closely mimics our observations than Model 1 and appears to reasonably approximate the time dependent change in dust size distribution during atmospheric transport across the tropical North Atlantic and Caribbean in terms of both aerosol size distribution and timing.
Studies have been cited [Andreae et al., 1986] that found dust aerosol size distributions varied little in the remote marine atmosphere. Starting with the dust size distribution measured at Izaña, we calculated the VMD of dust aerosol particles as a function of atmospheric transport time (Fig. 5) using Model 2. Modeled dust VMD decreased by only 20% after 10 days atmospheric transport. Initially, VMD decreased fairly rapidly with ~80% of the 10 day reduction taking place in the first 2 days. Given the short term ambient variability and uncertainties associated with measurements of dust aerosol size distributions, changes would likely be undetectable after 1 to 2 days of atmospheric transport away from a source region.

4. Conclusions

Measurements of mineral dust size distribution close to the North African coast and >5000 km down wind in the Caribbean show non-log normal size distributions. Preferential removal of particles >8 µm diameter indicates that removal driven by Stokes settling is too great to account for the observed changes in dust particle size distribution. A simple model with a vertical velocity for dust particles defined by the Stokes gravitational settling velocity minus 0.33 cm sec\(^{-1}\) accurately reproduces observed losses of dust particles as a function of aerosol size. Further, consistent with observations, the model predicts a relatively invariant dust size distribution after the first 1 to 2 days of atmospheric transport. We are not sure which physical process(es) are responsible for the upward velocity hypothesized here. However, buoyancy caused by the solar heating of the dust [Prospero and Carlson, 1981] and/or intermittent turbulence in the SAL similar to that in the planetary boundary layer [Pye, 1987] could be important. Our model does not account for the presence of very large dust particles (>50 µm diameter) over remote ocean
regions. Clearly, additional systematic representative measurements of those very large dust aerosols are necessary before the atmospheric transport of those particles can be understood. Nevertheless, the empirical model presented here should be helpful in the development of a theoretical model describing the removal of dust from the atmosphere as well as to help quantify the transport time dependent change in dust aerosol properties (e.g., dust mass scattering efficiency).

Caveats on the conclusions of this study extend beyond the uncertainties in the measurements. The measurements at Izaña and Puerto Rico are essentially “end members” of the atmospheric transport of dust. In addition, we expect the magnitude of any upward velocity to be dependent on meteorology, which in turn depends on, among other things, geography, season, and climate variability. Therefore, additional measurements should be made at multiple locations downwind of North Africa as well as other important dust sources (e.g., China) before this model of dust aerosol removal should be generally used.

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References


Figure Captions

**Figure 1.** Typical aerosol surface area size distributions from Izaña, Tenerife, Canary Islands measured in the free troposphere during the night of 23 to 24 July 1995 (solid line) and from Puerto Rico measured in the marine boundary layer during a 24 hour period from 9 to 10 July 2000 (dashed line).

**Figure 2.** Peak height normalized average dry sea salt (solid line) and dust (dashed line) aerosol volume size distributions measured in the marine boundary layer during July 2000 at Puerto Rico.

**Figure 3.** Peak height normalized dust aerosol volume size distributions from the free troposphere at Izaña, Tenerife, Canary Islands (solid line with ▲) and from the marine boundary layer at Puerto Rico (dashed line with ●). Error bars represent one standard deviation.

**Figure 4.** Fraction of dust aerosol particles removed or lost from SAL during atmospheric transport as a function of particle size. “Measurements” is an estimate of the change in dust aerosol size distribution that takes place between a few hundred kilometers west of the North African coast and more than 5000 km further west at Puerto Rico (solid line with ●). The error bars are estimates of one standard deviation as propagated through calculations. Very large uncertainties at or near 100% particle removal are a consequence of proportionately larger uncertainties for lower particle concentrations. “Model 1” is the particle removal by Stokes gravitational settling alone after 5.5 days of atmospheric transport (solid line). “Model 2” is the
particle removal by gravitational settling minus an upward velocity of 0.33 cm sec$^{-1}$ after 5.5 days of atmospheric transport (dashed line).

**Figure 5.** Dust aerosol volume median diameter as a function of atmospheric transport time according to “Model 2”. 
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