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TOMS Validation Based on Profiles of Aerosol Properties in the Lower Troposphere as Obtained with Light Aircraft Systems

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The goal of the University of Miami Aerosol Group (UMAG) in this project was to make measurements of vertical profiles of aerosol properties and aerosol optical depth using a light aircraft. The UMAG developed a light aircraft aerosol package (LAAP) that was used in light aircraft (Cessna 172) during the Puerto RIco Dust Experiment (PRIDE). This field campaign took place on Puerto Rico during July 2000. Design details and results from the use of the LAAP were presented at TOMS Science team meetings on April 1998, April 1999, and May 2000. Results from the LAAP collected during the PRIDE Experiment were presented at the Fall Meeting of the American Geophysical Union, December 2000. Some of the results from the LAAP collected during the PRIDE Experiment have been accepted for publication in the *Journal of Geophysical Research* in a "topical section" made up of papers from the PRIDE Program (that manuscript is appended to the end of this report.

The LAAP makes the following measurements:

- Aerosol light scattering (530 nm) of two aerosol size ranges measured using two single wavelength integrating nephelometers (Radiance Research® M903) with inlet impactors (one at 1 μm and the other at 10 μm).
- Aerosol light absorption (565 nm) measured using a Radiance Research® Particle/Soot Absorption Photometer.
- Aerosol size distributions measured by two instruments, a modified Met One® Model A237H Portable Airborne Particle Counter for aerosols from 0.1 to >1.0 μm and Met One® Model 237 for particles ranging from 0.5 to >5.0 μm.
- Total aerosol number concentration measured using a TSI® Model 3760 Condensation Particle Counter.
- Aerosol optical depth measured during the flights using a Microtops II photometer (Solar Light Co.) that makes measurements in five bands (various between 340-1020 nm).

- Aerosol samples for chemical analysis collected on precleaned 47 mm Teflon and/or Whatman® 41 filters.
- Temperature and relative humidity of the sampled air measured using a Vaisala® Humitter temperature and relative humidity probe; which enables us to correct the aerosol scattering data to adjust the data for aerosol growth due to relative humidity changes.
- The geographical location of the measurements and sample collections determined with a portable GPS system (i.e., Garmin GPS Pilot III).

Subsequent to the PRIDE Campaign, we added a light weight, low power, in situ ozone analyzer. This measurement was not in the original list of proposed instrumentation. However, this inexpensive instrument became available and given the value added of ozone profiles, we attempted to add this instrument to the LAAP in time for PRIDE. Unfortunately, the manufacturer of this analyzer could not deliver the instrument in time for the field deployment. Thus it was incorporated into the LAAP after the PRIDE field campaign.

Based on our experience during the PRIDE experiment, we upgraded some of the instrumentation in the LAAP to increase reliability and reduced electrical power requirements. Mechanically and instrumentally, the LAAP is ready for deployment. Out of other funds, we are in the process of optimizing the software to increase the speed and reliability of data handling and logging. Proposals have been submitted to use the LAAP in locations where local aircraft are available but US research aircraft cannot easily be deployed. In addition, we are planning to use the LAAP for air quality studies during the summer of 2003 along the eastern seaboard of North America.

The first publication of the results from the LAAP will be published in the *Journal of Geophysical Research – Atmospheres*. This paper deals with the LAAP itself and process studies based on data collected by the LAAP. Our next efforts will be directed at relating the quantitative vertical profiles of absorbing aerosols to the TOMS absorbing aerosol index.

Vertical distributions of dust and sea salt aerosols over Puerto Rico during PRIDE measured from a light aircraft

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Abstract

We developed the Light Aircraft Aerosol Package (LAAP) to make measurements of aerosol properties up to ~4000 m asl. The LAAP reliably measured, with empirical and theoretical corrections, aerosol micro-physical and chemical properties. During the Puerto RIco Dust Experiment (PRIDE), June - July 2000, we measured aerosol properties over the tropical North Atlantic. Based on aerosol filter samples and measured aerosol size distributions, we: a) modeled sea salt aerosol concentration with altitude, characterizing the rapid decrease in sea salt concentration with altitude; b) found no evidence of sea salt above the marine boundary layer (MBL); and c) observed mineral dust in the Saharan Air Layer (SAL) to exceed 170 $\mu g\ m^{-3}$. Substantial variability in the vertical distribution and small differences in the size distribution of dust between the MBL and SAL suggested dust properties were more dependent on the separate transport histories than the interaction between the MBL and SAL near Puerto Rico. Relatively constant dust size distributions with altitude suggested either: a) no relofting of dust aerosols once they get into the "deposition layer"; or b) dry deposition removes dust particles independent of particle size. Dust size distributions measured at low (~25 %) and high (~93 %) relative humidity were indistinguishable. A simple model calculation suggests dust particles over the tropical North Atlantic during summer increase their geometric diameter <6 % for a relative humidity change from 25 to 95 %. Thus optical properties of those dust particles should not change for relative humidities <95 %.

1. Introduction

The vertical distribution of aerosols in the atmosphere influences the importance of those aerosols in many respects. Atmospheric transport, utilization as cloud condensation nuclei, significance of light scattering and absorption, magnitude of wet and dry deposition fluxes, and the direct interaction of biological systems with aerosols all depend on the vertical distribution of those particles in the atmosphere. A number of research programs have measured the vertical distributions of sub-micrometer aerosols over the oceans. The First and Second Aerosol Characterization Experiments (ACE 1 & 2) included aircraft measurements in the vicinity of Tasmania [*Huebert et al,* 1998] and the Canary Islands [*Raes et al.,* 2000], respectively. Aerosol vertical profiles were also measured during the Tropospheric Aerosol Radiative Forcing Observational Experiment (TARFOX) [*Russell et al.,* 1999] and the Indian Ocean Experiment (INDOEX) [*de Reus et al.,* 2001] to name a few. However, relatively few direct measurements of super-micrometer aerosols with altitude have been made over the oceans mostly due to cost and the challenges associated with making measurements of large particles from aircraft. Passive remote sensing, both upward looking (i.e., sun photometers) and downward looking (i.e., satellites), can estimate the atmospheric burden of aerosol particles, but cannot determine concentration as a function of altitude. Lidars can measure the relative concentration of particles with altitude, but cannot identify the various types of aerosols present. Neither remote sensing nor lidars can measure the size distributions of aerosol populations without utilizing inversion techniques, which depend on assumptions as the refractive indices as well as to the shape of the aerosol size distributions present. In order to make more measurements of aerosols and aerosol properties with altitude at relatively low cost, we developed a package of instrumentation and samplers to be deployed in a light aircraft. Here we describe our Light Aircraft Aerosol Package (LAAP) and present results obtained using the LAAP during the Puerto RIco Dust Experiment (PRIDE) which took place during June and July 2000 on Puerto Rico.

One of the primary goals of PRIDE is to characterize the properties of airborne mineral dust over the tropical North Atlantic (TNAO) and Caribbean. Dust is important in this region because large quantities are known to be transported seasonally from North Africa westward over the tropical North Atlantic and Caribbean [*Delany et al.,* 1967; *Prospero and Carlson,* 1972]. Aircraft measurements made during the Barbados Oceanographic and Meteorological Experiment (Bomex) during May, June and July of 1969 showed dust aerosol mass concentrations peaked between 1.5 and 3.7 km asl in a relatively warm dry air layer dubbed the Saharan air layer (SAL) [*Prospero and Carlson,* 1972]. Relatively low concentrations of mineral dust that were nearly uniform with altitude were observed during May and the first half of June. High dust concentrations in the SAL were observed during the second half of June and July. The dust concentrations reported for the Bomex flights have substantial uncertainties due to filter efficiencies possibly as low as 80 %, dust mass determination by filter color which adds approximately a 25 % uncertainty and an unknown sampler nozzle efficiency. More recently, *Reid et al.,* [2002] reported higher concentrations of dust in the marine boundary layer (MBL) compared to the SAL during June and the first half of July 2000 at Puerto Rico. During the last half of July 2000 they observed vertical distributions of dust comparable to those described by *Prospero and Carlson,* [1972]. *Reid et al.,* [2002] suggest the change from the northeast to southwest monsoonal flow at the west coast of North Africa terminates dust transport in the MBL. Alternatively, *Colarco et al.,* [this issue] suggest regional subsidence may bring mineral dust from aloft into the MBL. Here we present additional vertical profiles of mineral dust over Puerto Rico as well as direct measurements of dust mass at altitude.

Mineral dust aerosols transported through the atmosphere over the TNAO and Caribbean from arid regions in North Africa range in size from <0.5 to >50 μm geometric diameter [*Jaenicke and Schütz,* 1978; *Westphal et al.,* 1986; *Maring et al.,* 2000]. *Maring et al.,* [this issue] have shown that the size distribution of North African dust shifts to smaller sizes during atmospheric transport due to preferential settling of larger particles. However, they, like others [*Jaenicke and Schütz,* 1978], found that Stokes gravitational settling would remove particles >10 μm geometric diameter too rapidly to be consistent with observed dust aerosol size distributions. They proposed a model defining the vertical velocity of dust particles in the SAL to be the Stokes settling velocity minus 0.33 cm sec^{-1}. This model is consistent with observed changes in the dust size distribution and with observations of nearly uniform dust size distribution over much of the remote ocean [*Andreae et al.,* 1986].

The dust size distribution in the MBL is dependent on the dust supplied to it, often from the SAL, and the deposition processes that remove dust from the atmosphere. Any change in size distribution with altitude in the remote marine atmosphere would be a consequence of particle dry deposition or hygroscopic growth to form condensation nuclei. The size distribution of aerosols containing dust particles would change as a function of relative humidity if dust acts as condensation nuclei [*Levin et al.,* 1996]. Literature estimates for the hygroscopic growth of mineral dust range from 0 to 90 % [*Twomey,* 1977; *Zimon,* 1982; *Hänel,* 1976]. Measurements of scattering by dust particles as a function of relative humidity up to 83 % at Barbados suggest mineral dust particles over the TNAO may not act as condensation nuclei [*Li-Jones et al.,* 1998]. However, measurements at relative humidities >83 % are needed to characterize water uptake by mineral dust particles of the TNAO. Here we present dust aerosol size distributions at various altitudes and relative humidities to help characterize removal processes.

Bubbles, produced by breaking waves, bursting at the surface of the ocean inject sea salt aerosol particles into the atmosphere. Thus in regions with stable wind fields, such as the trades, the concentration of sea salt aerosols at altitude in the MBL should be related to the concentration of sea salt aerosols at sea level. Early measurements of sea salt aerosols with altitude over the ocean generally show a rapid decrease in concentration with altitude [*Woodcock,* 1953; *Durbin and White,* 1961]. However, neither of these studies related measurements of sea salt aerosol concentration at sea level to their measurements aloft. Here we present measurements of sea salt at various altitudes as well as at sea level and develop a simple empirical model of sea salt aerosol mass concentration as a function of altitude.

2. Experimental

The LAAP was developed by the University of Miami Aerosol Group to augment our ground-based aerosol measurement capacity. The purpose of the LAAP is to extend our detailed, high resolution, high sensitivity ground-based aerosol measurements into the vertical with less extensive and lower resolution but nonetheless comparable measurements made with the LAAP.

The LAAP is a self-contained inlet, instrument/sampler, data-logging package designed to be deployed in a light aircraft to make measurements of aerosol properties with altitude in the troposphere. The whole LAAP weighs less than 100 kg and can be easily shipped to a location of interest and installed in a rented aircraft. We typically deploy in a Cessna 172, a small, commonly available, single engine, overhead wing aircraft that is relatively inexpensive to operate. The LAAP consists of a cubical instrument rack approximately 0.7 m on a side, which is clamped to a base that bolts to the plane's floor replacing the back seat, a laptop computer, and a sample air nozzle/inlet tube. The 3.8 cm outside diameter stainless steel inlet tube clamps onto the wing strut extending forward of the wing, ending in a pitot tube-like nozzle with a 6 mm diameter hole. The inlet tube extends back from the nozzle approximately 1 m, makes a 90° turn extending approximately 1 m toward the fuselage, passing through the co-pilot side door window and connecting to the instrument rack. A custom bracket fastened to the screw holes of a courtesy light panel in the underside of the wing supports the inlet tube at the right angle bend. The nozzle was designed to be isokinetic at our standard sampling air speed of 31 m $\sec^{-1}$ (60 knots). The LAAP maintains a sample-air volumetric flow rate at 50 l $\min^{-1}$ to maintain isokinetic sampling at the nozzle and laminar flow in the intake tube, automatically compensating for changes in altitude. The LAAP draws approximately 400 watts from the plane's electrical system.

Table 1 lists the measurements and instrumentation of the LAAP. The sample air flow rate to the TSI® Model 3760 condensation particle counter was controlled by its critical orifice at 1.41 volumetric l $\min^{-1}$. The uncertainty in the measurement of aerosol number concentration of particles ranging in size from approximately 15 nm to 1 μm depended on the variability in the sample flow rate, particle losses at the nozzle and in the intake tube, and counting statistics. Generally, the variability in sample flow rate was comparable to or smaller than the uncertainty due to counting statistics. The uncertainty due to particle losses at the nozzle (see below) as well as those caused by particle diffusion and impaction were negligable [*Willeke and Baron,* 1993]. In all cases, ambient variability was large compared to measurement uncertainty. The sample air flow rate through the The MetOne® 237B optical particle counter was 2.83 volumetric l min^{-1} controlled by a flow restrictor and the instrument's carbon vane pump. Spot checks of that flow rate indicate the uncertainty to be on the order of 5 %. The uncertainty in optical particle counter concentration measurements stems from variability in the sample flow rate, counting statistics, coincidence undercounting, and particle losses at the nozzle and in the intake tubing. We estimated the corrections for coincidence undercounting and particle losses due to diffusion, impaction and gravitational settling using theoretical calculations [*Willeke and Baron,* 1993]. We determined losses at the nozzle empirically (see below). The MetOne® 237H optical particle counter malfunctioned during this experiment. The sample flow rate through the two Radiance Research® nephelometers was held at 10 volumetric l min^{-1} using measurements of air mass flow rate, ambient atmospheric pressure and temperature, and computer-controlled variable speed vacuum pumps. The sample air flow rate through the Radiance Research® Particle/Soot Absorption Photometer (PSAP) was held at 2.0 volumetric l min^{-1} in a fashion identical to the nephelometers. A more detailed description of the aerosol optical measurements will be presented elsewhere. The optical particle counters, nephelometers, PSAP, and aerosol filter all operated at the temperature and pressure of the aircraft cabin.

The aerosol sampling filter for chemical analysis was actually two separate filter holders containing filter media. During taxi, takeoff, landing, and much of the flight; air flow was directed through a "dummy" filter. Only during constant altitude flight was sample air flow directed through the "sample" filter (see below). In either case, the flow rate through the aerosol filter was controlled so the total flow through the system was 50 volumetric l min^{-1}, ensuring isokinetic conditions at the nozzle and laminar flow in the intake tube.

PRIDE took place 28 June to 24 July 2000, at Naval Station Roosevelt Roads, located on the eastern end of the island of Puerto Rico. We made detailed high-resolution measurements of aerosol chemical, micro-physical and optical properties from a ground-based portable laboratory built inside a standard 20 ft. shipping container on Cabras Island (Lat. ~18.21° N, Long. ~65.60° W) located just east of the main island of Puerto Rico. Our ground based measurements and the analytical methods used for chemical analysis have already been described [*Maring et al.,* 2000; *Maring et al.,* this issue]. Specifically, aluminum, an indicator element for mineral dust, was determined by instrumental neutron activation analysis. Sodium, an indicator element for sea salt, was determined by flame atomic absorption. Sulfate was measured by ion chromatography.

LAAP flights originated from a small private airstrip roughly 20 km south of Roosevelt Roads. This air strip was oriented east-west and was separated from the east coast of Puerto Rica by 3.5 km of farm fields and a narrow uninhabited beach. LAAP research flights consisted of the following: a) 10 to 15 minutes of measurements while on the ground with the plane facing into the prevailing easterlies, b) take off, c) low level (<10 m) run at 31 m sec^{-1} over the runway and farm fields to the coast lasting 2 to 3 minutes, d) spiral up to the operational ceiling of the aircraft (3500 to 4000 m asl), e) cruise for 1.5 to 2 hours at constant altitude for collection of a filter sample for chemical analysis, f) fly immediately adjacent to and upwind of our ground site on Cabras Island at the same elevation as the sample air inlets (15 to 20 m asl) for 10 to 20 minutes, and g) land. All LAAP flights took place over water and upwind of the main island of Puerto Rico within a 60 km radius of the Cabras Island site. During PRIDE we flew 6 research flights, each lasting approximately 5 hours.

Particle losses, especially losses of super-micrometer particles, at the nozzle and in the inlet tube are an important concern relative to aircraft measurements. Because the sample-air flow in the inlet tube was always laminar, we were able to calculate particle losses in the inlet tube [*Willeke and Baron,* 1993]. We determined particle losses at the nozzle empirically. We compared the size distribution of particles measured while the plane was on the ground facing into the wind just prior to takeoff to the size distribution measured while the plane flew at low level (~3 m) over the runway and farm fields at our standard sampling air speed of 31 m sec^{-1}. Thus we were able to determine the nozzle efficiency as a function of particle size (Figure 1). We used the empirically determined nozzle efficiency, the calculated intake tube particle penetration efficiency, and the optical particle counter coincidence correction to adjust measured aerosol size distributions to ambient particle size distributions. This correction was also used to correct measured dust and sea salt aerosol mass to ambient aerosol mass concentrations.

3. Results and Interpretation

3.1 LAAP Data Verification

In order to verify the aerosol measurements made by the LAAP, we compared the aircraft measurements to those made at our ground site on Cabras Island during the low-level flybys. The comparison of condensation particle concentrations (Figure 2) shows that the measurements made by the LAAP were indistinguishable from those made on the ground, except on 22 July 2000. On that day, we ran low on fuel when 30 km away from the Cabras Island site. So we made the low-level measurements at that location and then landed. Thus the difference observed on 22 July was likely a result of horizontal heterogeneity in the condensation particle concentration. The good agreement between the LAAP and ground measurements is not surprising because the condensation particle number concentration is dominated by sub-micrometer aerosols and the nozzle/inlet efficiency for those particles is essentially 100 %.

Figure 3 compares aerosol size distributions measured by the optical particle counter in the LAAP to those measured on the ground using a TSI® Scanning Mobility Particles Sizer (Model 3934L) and a TSI® Aerodynamic Particle Sizer (Model 3310) for 12 July 2000. Aerodynamic diameters were converted to geometric diameters assuming a density of 2.0 g cm^{-3} [*Maring et al.*, 2000]. The ground-based measurements were made on sample air heated so the relative humidity was approximately 50 %. For this comparison, we assumed the particles measured by the optical particle counter in the LAAP were composed of sea salt and mineral dust. In converting size distributions measured by the LAAP at ambient relative humidity to a relative humidity of 50 %, we assumed any mineral dust present did not take up any water. We dried sea salt aerosols from a reasonable estimation of the relative humidity in the optical particle counters to 50 % using the aerosol model of *Fitzgerald et al.,* [1998] using the ratio of sea salt to mineral dust in the bulk aerosol during that sampling period for all size ranges. We performed these comparisons for all six flights. Figure 3 is typical of all the comparisons. Thus we believe corrected aerosol size distributions from the LAAP provide a reasonably accurate measure of the ambient aerosol size distribution.

3.2 Temperature and Relative Humidity with Altitude

The temperature and relative humidity profiles (Figure 4) show weak temperature inversions (change in the rate of change of temperature with altitude) and significant reductions in the relative humidity at altitudes ranging from ~1800 m asl (10 July) to ~2300 m asl (24 July). A more substantial temperature inversion is evident at ~3700 m asl (22 and 23 July) which is probably the top of the SAL. Here we consider the lower marine troposphere to be made up of the MBL, which extends from sea level to the altitude of the first inversion and or rapid decrease in relative humidity, and the SAL, an approximately 2000 m thick relatively dry air layer which overlays the MBL.

The operational ceiling of the Cessna 172 is approximately 4000 m asl. Since nearly all of the sea salt and mineral dust aerosols at Puerto Rico are found below 3500 m asl [*Reid et al.*, 2002], the maximum altitude of the Cessna 172 is not a significant limitation in the study of dust and sea salt particles in the marine troposphere.

3.3 Aerosol Aluminum and Sodium Concentration with Altitude

Measurements of aerosol chemical composition from samples collected using the LAAP are presented in Table 2. We assume all aluminum is associated with mineral dust and that 8 % of the mass of dust particles is aluminum [*Taylor and McLennan,* 1985; *Taylor and McLennan,* 1995] to calculate the mineral dust mass concentration. The sodium to dust mass ratios in samples collected above the MBL are less than in crustal material [*Taylor and McLennan,* 1985; *Taylor and McLennan,* 1995]. Thus sodium in aerosols above the MBL was likely associated with mineral dust. In other words, no sea salt aerosols were detected above the MBL. Similarly, measurements of aerosol size distribution and chemistry in the free troposphere over the eastern TNAO show that the super-micrometer aerosol to be periodically dominated by mineral dust particles, while there is no evidence of sea salt [*Maring et al.,* 2000].

We used the average sodium to dust mass ratio in the SAL samples (10, 21, 23, 24 July) collected using the LAAP to calculate the amount of crustal sodium in the aerosol. That amount of crustal sodium was subtracted from total sodium, leaving sea salt sodium. We assumed sodium to sea salt mass ratio in sea salt aerosols is identical to that in seawater, 0.308 [*Wilson,* 1975] to calculate the sea salt aerosol mass in the samples collected in the MBL (12, 22 July).

In order to estimate ambient dust concentrations from the dust mass measured on the sample filter, it is necessary to compensate for the loss of dust aerosol mass in the nozzle and intake tube of the LAAP. We determined a correction factor by calculating the ratio of the corrected total aerosol volume (i.e., based on particle size distribution correction described above) to the measured total aerosol volume for aerosols whose diameters were >0.5 μm for measurements made in the SAL. Similarly, we determined a correction factor for the loss of sea salt aerosol mass in the nozzle and intake tube of the LAAP. Using the model of *Fitzgerald et al.,* [1998], we increased the size of the dry sea salt aerosol size distribution [*Maring et al.,* this issue] determined at sea level to what they were at 70 % relative humidity, the ambient relative humidity when the aerosol samples were collected in the MBL. Using the method described above for correcting measured aerosol size distributions, we determined the ratio of ambient sea salt aerosol volume to an estimate of the sea salt aerosol volume adjusted for nozzle and intake tube losses. This method will likely overestimate the correction factor because the ambient sea salt aerosol size distribution at altitude should be shifted to smaller sizes due to the greater settling velocity of larger particles. Unfortunately, we could not measure the sea salt only size distribution as a function of altitude because, even when their concentrations were low, mineral dust particles dominated the ambient aerosol size distribution for aerosols with diameters >0.5 μm.

3.4 Vertical Distribution of Sea Salt

It is well known that bubble bursting at the surface of the ocean produces sea salt aerosols. Surface winds and associated turbulence carry these aerosols upward in the MBL. Many sea salt aerosols are sufficiently large to have substantial downward settling velocities. Consequently, the concentration of sea salt should decrease with altitude. *Woodcock,* [1953] observed a rapid decrease in sea salt mass concentration with increasing altitude in samples collected from a light aircraft. However, those measurements were not associated with comparable measurements at sea level.

Atmospheric conditions in tropical regions tend to be quite stable most of the time (i.e., relatively constant wind speed, wind direction, sea state and relative humidity). During the PRIDE expedition, hourly averages of the surface wind speed averaged 6 ± 1 m sec^{-1}, the surface wind direction averaged 98° ± 15°, and sea level relative humidity averaged 77 ± 3 %. Therefore, the vertical distribution of sea salt aerosols should be relatively constant as well.

In addition to measurements of sea salt aerosol concentrations at altitude, we also collected and analyzed daily bulk aerosol samples to measure sea salt aerosol concentrations near sea level (~10 m asl) [*Maring et al.,* this issue]. On the two days we collected aerosol samples for chemical analysis from the LAAP (12 and 22 July), the sea level sea salt aerosol concentrations were 20 and 8.9 μg m^{-3}. To compare these measurements and relate them to our sea level measurements, we normalized these sea salt aerosol concentrations to the sea level concentrations (Figure 5). We modeled the vertical distribution of sea salt and determined the following logarithmic relationship: normalized sea salt aerosol mass concentration = - 0.206 * ln (altitude) + 1.473, $R^2 = 0.9996$; where the sea salt aerosol mass is in μg m^{-3} at standard temperature and pressure and altitude is in meters above sea level. Thus the sea salt aerosol concentration at a particular altitude can be estimated by calculating the normalized sea salt concentration at the altitude of interest and multiplying by the sea salt aerosol mass concentration measured at sea level.

We tested the validity of our model by comparing it to the measurements of *Woodcock,* [1953]. To relate Woodcock's aircraft measurements of sea salt concentration to our model, we adjusted his data so that the data point at the lowest altitude fell on our model curve; the data for higher altitudes varied proportionately from that lowest altitude. Those profiles are presented in Figure 5 and are identified by their date of collection. That data does not exactly overlay our model. The difference between our model and Woodcock's measurements averaged 1.3 ± 1.7 μg m^{-3}. Those differences may be due to differences in cloudiness, wind speed and possibly other local meteorological factors. Clearly, more measurements at a variety of altitudes, especially 50 to 200 m asl, are necessary to refine this model. Nevertheless, this approach may provide a useful first approximation of sea salt aerosol concentration with altitude in meteorologically stable environments such as the trade winds.

3.5 Vertical Distribution of Mineral Dust

Our measurements of dust vertical profiles (Figure 6) are completely consistent with *Reid et al.*, [2002] who have shown that while high concentrations of dust particles are often found in the SAL, the majority of atmospheric dust can sometimes be found in the MBL. Prior to the middle of July 2000 we observed the majority of dust in the MBL. After mid July, African dust was primarily located in the SAL. *Reid et al.,* [2002] suggest the change from the northeast to southwest monsoon at the coast of North Africa during mid July terminates export of dust particles in the planetary boundary layer limiting long-range transport of dust to the SAL.

At times the concentration of dust did not vary substantially over the course of the LAAP flight (e.g., 21 July) indicating an air mass with a relatively constant dust concentration. However, at other times, the dust concentration varied by more than a factor of 2 within 3 to 4 hours (e.g., 10 July in the MBL). Since the height of the MBL did not change substantially during that flight, the observed change in dust concentration in the MBL reflects the magnitude of horizontal heterogeneity.

The vertical distribution of dust was quite complex, especially in the SAL. Sometimes, the dust was contained in a single layer that seemed to fill up the whole SAL (e.g., 10 July). At other times, we found dust aerosols in multiple distinct layers (e.g., 21 July). At times, the dust layers were quite stable. For example, on 21 July, the dust layers stayed at the same altitude as can be seen from the nearly identical dust vertical profiles during both ascent and descent. Further, the tight clustering of the data points generated during the collection of the filter sample for chemical analysis shows the dust concentration on that day was relatively constant in the horizontal (i.e., varying <20 %). On that day, the dust layers were relatively thick, on the order of hundreds of meters. In contrast, on 24 July, the dust layer in the SAL was much more variable. The substantial differences between ascent and decent suggests significant horizontal variability. While collecting the filter sample on that day, we had to constantly adjust the altitude of the aircraft to stay in the dust layer. The varying altitude of the dust layer combined with our inability to stay in the layer resulted, at least in part, in the wide range of dust particle concentrations recorded during the collection of the aerosol filter sample. The thickness of the dust layer in the SAL at any given time appeared to be on the order of meters to tens of meters. The rapidly changing altitude of the dust layer on 24 July may indicate turbulent processes affecting the SAL.

The dust concentrations we measured in the SAL, corrected for nozzle and intake tube losses, ranged from 63 to 170 $\mu g\ m^{-3}$. The concentrations derived from the aerosol filter samples collected during 1.5 to 2 hour periods. The dust particle concentration varied substantially during those sampling periods. Therefore, the maximum dust concentrations were likely substantially higher than the integrated values. We used the ratios of the maximum and minimum total aerosol volumes to average total aerosol volume for particles with diameters >0.5 μm along with the integrated dust mass concentration of 160 $\mu g\ m^{-3}$ to estimate the maximum and minimum dust mass concentration on 21 July. The dust concentration in the dust layer appeared to range from 120 to 190 $\mu g\ m^{-3}$ during the 21 July flight. The dust concentrations estimated here for the SAL are on the order of 3 times greater than those reported by *Prospero and Carlson*, [1972] measured during June and July 1969. The differences may be due to variability in the ambient concentrations, but may also be due, in part, to intake losses and filter inefficiency.

3.6 Dust Aerosol Size Distribution

The dust size distributions measured by the LAAP were made at temperatures slightly greater than ambient because the air inside the aircraft cabin was warmer than outside. Consequently, the measured relative humidity of the sample air inside the instrument package was <50 %. The combination of nozzle, intake tube and optical particle counter used in the LAAP made it impossible to measure particles larger than about 10 μm in diameter. Consequently, our measurements cannot be used to characterize the whole dust particle size distribution. For example, the volume median diameters (VMD) calculated from our data cannot be directly compared to estimates based on other more comprehensive measurements. Nevertheless, our measurements can be profitably compared to each other to identify changes and differences.

The size distributions measured during the collections of the aerosol samples for chemical analysis are presented in Figure 7. The dust size distributions in the SAL are much more similar to each other than to the spectra measured in the MBL, with the spectrum measured at 328 m asl

(22 July) being made up of smaller particles than the dust particles in the SAL. The spectrum measured at 643 m asl (12 July) is intermediate between the extremes. Dried sea salt aerosols appear to be smaller than mineral dust particles at Puerto Rico [*Maring et al.,* this issue]. This range of spectra is consistent with the measurements in the MBL being a variable, altitude dependent combination of dried sea salt aerosols and mineral dust particles.

The SAL and MBL appear to be at least somewhat uncoupled. The relative concentrations of dust (Figure 6) in the SAL and MBL do not appear systematically linked. The VMD's of dust in the SAL are statistically distinct from day to day. The same is true of VMD's of dust in the MBL. The VMD's of dust in the SAL is more often than not statistically different from that in the MBL on any given day. However, the mean VMD of dust in the SAL averaged over all the flight days is not statistically distinct from that in the MBL. Further, on half of the days, the VMD of dust in the SAL was larger than that in the MBL; on the other days, the VMD of dust was larger in the MBL. Therefore, no clear relationship exists between dust in the SAL and the MBL. If the SAL and MBL are uncoupled, then it may be that dust in the SAL and in the MBL over the same geographic location at the same time could have quite different transport histories.

This lack of systematic change between the SAL and MBL in dust concentration and size distribution also seems to imply that deposition processes do not affect the size distribution of dust in the MBL. During July 2000, trade cumulus clouds were common, tending to build up each afternoon. However, rain showers were infrequent and small. Thus dry deposition was likely the dominant removal process at that place and time.

Measurements and models predict that the ratio of deposition velocities for the dry deposition of 0.6 μm and 7.0 μm particles should range from nearly 20 to 1000 depending on ambient wind speed and the particular measurement or model [*Sehmel and Sutter,* 1974; *Slinn et al.,* 1978; *Lin et al.,* 1994; *Noll et al.,* 2001]. Therefore, dry deposition should cause a shift in the dust particle size distribution toward smaller particles. The lack of change in dust size distribution may imply that no dust particles are re-lofted once they get into the "deposition layer" [*Slinn et al.,* 1978]. In other words, despite the wide range in deposition velocities of large and small dust particles, dry removal takes essentially all dust particles out of the air before they can be re-lofted. Alternatively, if some fraction of particles in the deposition layer do get re-lofted, then the dry removal process must take dust particles out of the air without regard to size. Perhaps a moving, rough water surface with breaking waves captures dust particles independent of size. In either case, atmospheric transport appears to be more important in terms of defining dust aerosol size distributions in the SAL and most of the MBL than dry deposition.

3.7 Dust Size Distribution and Relative Humidity

A modeling study by *Dentener et al.,* [1996] indicated sulfur dioxide gas may react with calcium rich mineral dust particles coating the dust with sulfate. Sulfate on the surface of mineral dust may cause the dust particle to become hygroscopic. The hygroscopic character of dust is important for two reasons. First, a change in the size of dust aerosols due to absorption of water would modify the amount of light scattering caused by those aerosols. Second, a hygroscopic coating covering dust particles would increase the likelihood that those dust particles would act as cloud condensation nuclei. Indeed, over the eastern Mediterranean, *Levin et al.,* [1996] found mineral dust particles to be coated with sulfate causing them to be effective as giant cloud condensation nuclei.

The relative humidity in the MBL over Puerto Rico often peaked above 90 %, while the in the SAL relative humidities were often <30 %. For example, on 21 July the relative humidity averaged 97 ± 1 % from 1080 to 1260 m asl in the MBL and in the SAL from 1480 to 1860 m asl the relative humidity averaged 25 ± 1 %. Similarly, on 23 July the relative humidity averaged 91 ± 1 % from 790 to 1600 m asl in the MBL, while in the SAL the relative humidity averaged 22 ± 3 % from 2000 to 3690 m asl. If the dust particles are coated with sulfate, they should absorb water and grow when exposed to relative humidities >90 %.

The optical particle counter in the LAAP was located inside the cabin of the airplane, the temperature of the sample air inside that instrument was a few degrees warmer than the ambient air. Consequently, the relative humidity of the sample air inside the LAAP's optical particle counter was lower than the ambient air. The relative humidity of very humid air changes rapidly with changes in temperature (e.g., at 95 % relative humidity and increase of 1°C will drop the relative humidity 5 %). Thus LAAP measurements of aerosol size distribution do not measure wet aerosols. However, the other aircraft that flew as part of PRIDE (Piper Navajo) measured aerosol size distribution from 30 to >5000 m asl with an optical particle counters mounted under the wing (PMS® Forward Scattering Spectrometer Probe: FSSP-100 and Passive Cavity Aerosol Spectrometer Probe: PCASP-100X) which made measurements at ambient relative humidity [*Reid et al.,* 2002].

LAAP measurements of dust size distribution in the MBL and SAL on 21 and 23 July show statistically significant differences in the VMD. However, the differences are relatively small averaging <4 % on those two days. Measurements of aerosol size distribution by the PCASP-100X and FSSP-100 on 21 and 23 July over the altitude ranges specified above for these dates contrast similar dust size distributions at significantly different relative humidities. The VMD's of dust aerosols at high relative humidity in the MBL were statistically indistinguishable from the VMD's of dust particles at low relative humidity in the SAL on both 21 and 23 July. Thus it appears that dust particles over the TNAO and Caribbean do not change size significantly due to the absorption of water up to a relative humidity of 97 %.

To further investigate the growth of dust particles internally mixed with sulfate, we estimated particle growth for an increase in relative humidity from 25 to 95 %. During PRIDE, we made measurements of sulfate and dust in size segregated aerosols collected using a MSP® Corp. Micro-Orifice Uniform Deposit Impactor (MOUDI) at the surface site on Cabras Island. The dust to non-sea salt sulfate mass ratio at Puerto Rico during the period 21 to 24 July 2000 was 36, 63, and 98 for 1.7, 4.4, and 7.8 μm geometric diameter particles, respectively. This range of particle size includes more than 70 % of the aerosol dust mass. We assumed all the super-micrometer non-sea salt sulfate was associated with dust aerosol particles. If any of the non-sea salt sulfate were contained in sea salt aerosols, then the following calculation will overestimate the hygroscopic growth of mixed dust and sulfate particles. Further, we assumed all non-sea salt sulfate associated with dust particles was in the form of ammonium sulfate, the most soluble of the expected sulfate salts. Assuming the sulfate coated the surface of the dust particle, we estimated water absorption by assuming sulfate on dust particles absorbs the same amount of water as a pure ammonium sulfate particle [*Seinfeld and Pandis,* 1998] and that the absorbed water also coated the dust particle. We found that the geometric diameters of the 1.7, 4.4, and 7.8 μm particles increased 0.17, 0.15, 0.13 μm, respectively. Thus the diameters of these particles increased by <6 % going from 25 to 95 % relative humidity. If any of the non-sea salt sulfate is bound up in a salt less soluble than ammonium sulfate (e.g., calcium sulfate) then the amount of water absorbed would be proportionately smaller. Therefore, both our observations and a simple calculation of dust particle growth indicate that very little growth with increasing relative humidity up to at least 95 %. Over the TNAO light scattering by dust particles are likely to remain constant for relative humidities lower than ~99 %. However, if dust particles are coated with relatively thick layers of sulfate salts, those particles may grow substantially with increasing relative humidity. Since all of our measurements were made at relative humidities lower than 100 %, we cannot address the issue of dust particles as cloud condensation nuclei in supersaturated air.

4. Summary

1. The Light Aircraft Aerosol Package (LAAP) can measure, with appropriate empirically and theoretically determined corrections, aerosol micro-physical and chemical properties as a function of altitude from near the surface to ~4000 m asl.
2. Based on aerosol filter samples collected by the LAAP as well as measured aerosol size distributions, we:
   a. developed an empirical model of sea salt aerosol concentration with altitude over the ocean in the area trade winds,
   b. found no evidence of sea salt aerosols above the marine boundary layer (MBL), and
   c. observed concentrations of mineral dust in the Saharan Air Layer (SAL) to range up to 170 $\mu g\ m^{-3}$ for the 1.5 to 2 hour aerosol sampling periods of the LAAP; instantaneous dust concentrations exceeded 200 $\mu g\ m^{-3}$.
3. We observed substantial variability in the vertical distribution of dust as well as small but statistically significant differences in the dust size distribution between the MBL and SAL. This variability indicates dust properties in the SAL and MBL well downwind of the North African source are more dependent on the separate transport histories of dust particles in the SAL and MBL than on the interaction between the MBL and SAL at or near that location.
4. The lack of substantial change in dust size distribution with altitude suggests either:
   a. no relofting of dust aerosols once they get into the "deposition layer", or
   b. whatever dry deposition process dominates dry removal at that surface removes dust particles independent of particle size.
5. Dust size distributions measured at ambient relative humidity showed no statistical difference between low (~ 25 %) and high (~95 %) relative humidity. Further, a simple model calculation suggested dust particles over the tropical North Atlantic Ocean during summer increase their geometric diameter <6 % for a relative humidity change from 25 to 95 %. Thus, the optical properties of those dust particles should not change for relative humidities <95 %.

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Table 1. Measurements and instrumentation of the Light Aircraft Aerosol Package (LAAP)

Measurement	Instrument
Aerosol number concentration from 14 to 1000 nm	TSI® 3760 Condensation Particle Counter
Aerosol number size distribution 0.1 - 0.2; 0.2 - 0.3; 0.3 - 0.5; 0.5 - 0.7; 0.7 - 1.0; >1.0 μm	MetOne® 237H Optical Particle Counter
Aerosol number size distribution 0.3 - 0.5; 0.5 - 0.7; 0.7 - 1.0 1.0 - 2.0; 2.0 - 5.0; >5.0 μm	MetOne® 237B Optical Particle Counters
Aerosol light scattering at 530 nm for particles <10 μm and <1 μm aerodynamic diameters	Radiance Research® Model 903M Integrating Nephelometers (2x)
Aerosol light absorption at 565 nm	Radiance Research® Particle/Soot Absorption Photometer
Sulfate, nitrate, chloride, sodium, potassium, ammonium, and mineral dust in bulk aerosols	47 mm bulk filter sampler Whatman 41
Solar radiation at 340, 500, 675 870, 936 nm	MicroTops® II Sun Photometer
Atmospheric pressure	Vaisala® Model PTB101B Pressure Sensor
Temperature and relative humidity	Vaisala® Model HMP45A T/RH Probe
GPS location	Garmin® GPS Pilot III

Table 2. Measured concentrations (μg m^{-3}) of indicator elements in samples collected using the LAAP during July 2000, sodium to dust mass ratios in dust samples, and aerosol dust and sea salt concentrations (μg m^{-3}) corrected for nozzle and inlet efficiency.

Flight Corrected Date	Altitude (m asl)	Measured		Mineral Dust	Na / Dust	Sea Salt	Nozzle and Inlet	
		Al	Na				Dust	Sea Salt
10 July 2000	2796	3.0 ± 0.3	0.3 ± 0.3	38	0.0074		63	
12 July 2000	643	0.6 ± 0.1	0.9 ± 0.2	7.8	0.1171	1.7	13	2.8
21 July 2000	3059	7.8 ± 0.7	0.5 ± 0.3	97	0.0051		160	
22 July 2000	328	0.4 ± 0.1	0.7 ± 0.3	4.4	0.1486	1.4	7.3	2.3
23 July 2000	3121	8.2 ± 0.7	1.0 ± 0.4	102	0.0094		170	
24 July 2000	2749	7.6 ± 0.7	0.6 ± 0.3	95	0.0062		157	

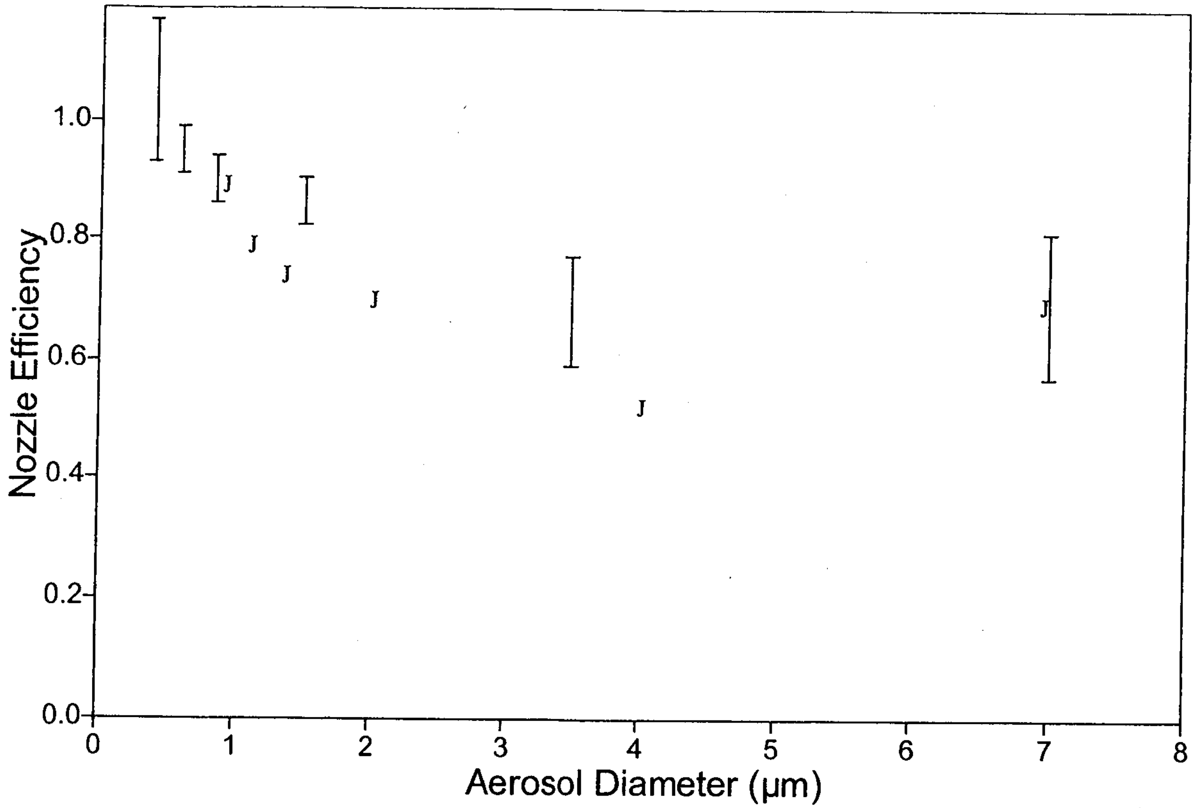

Figure 1. LAAP nozzle efficiency as a function of particle size. The error bars represent one standard deviation.

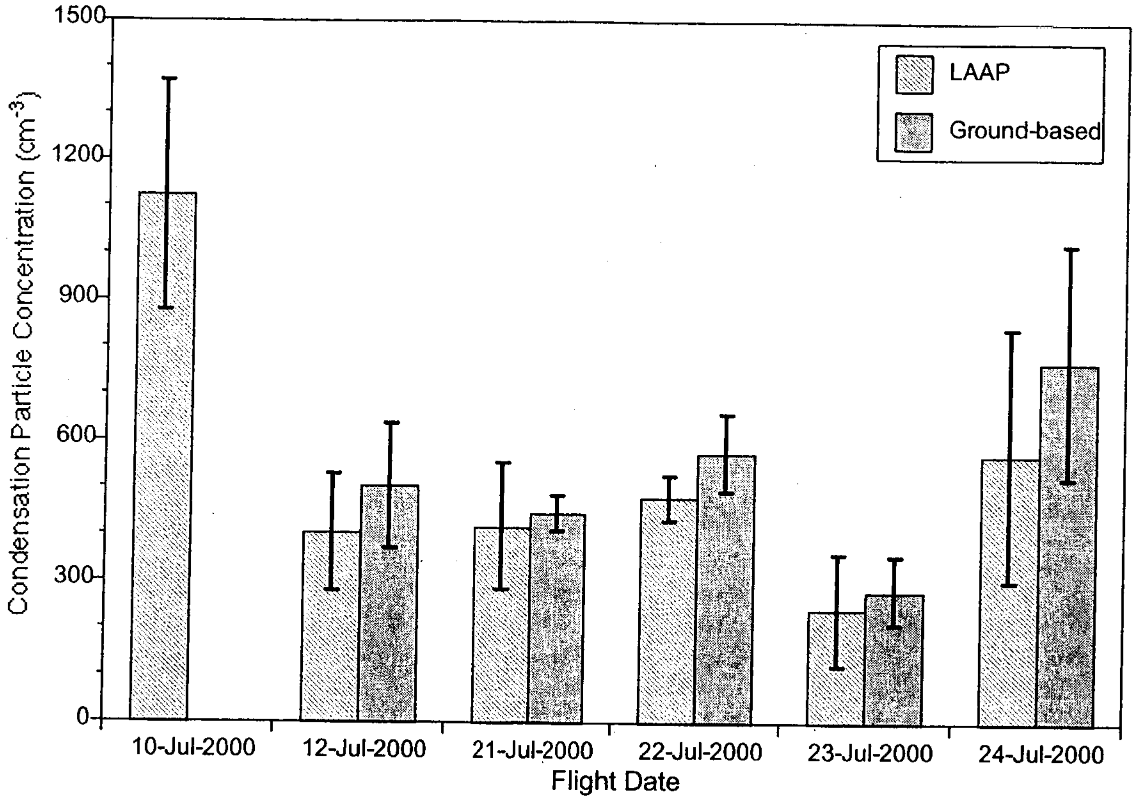

Figure 2. Comparison of LAAP and ground-based measurements of condensation particle number concentrations during Cabras Island ground site flybys. The error bars represent one standard deviation.

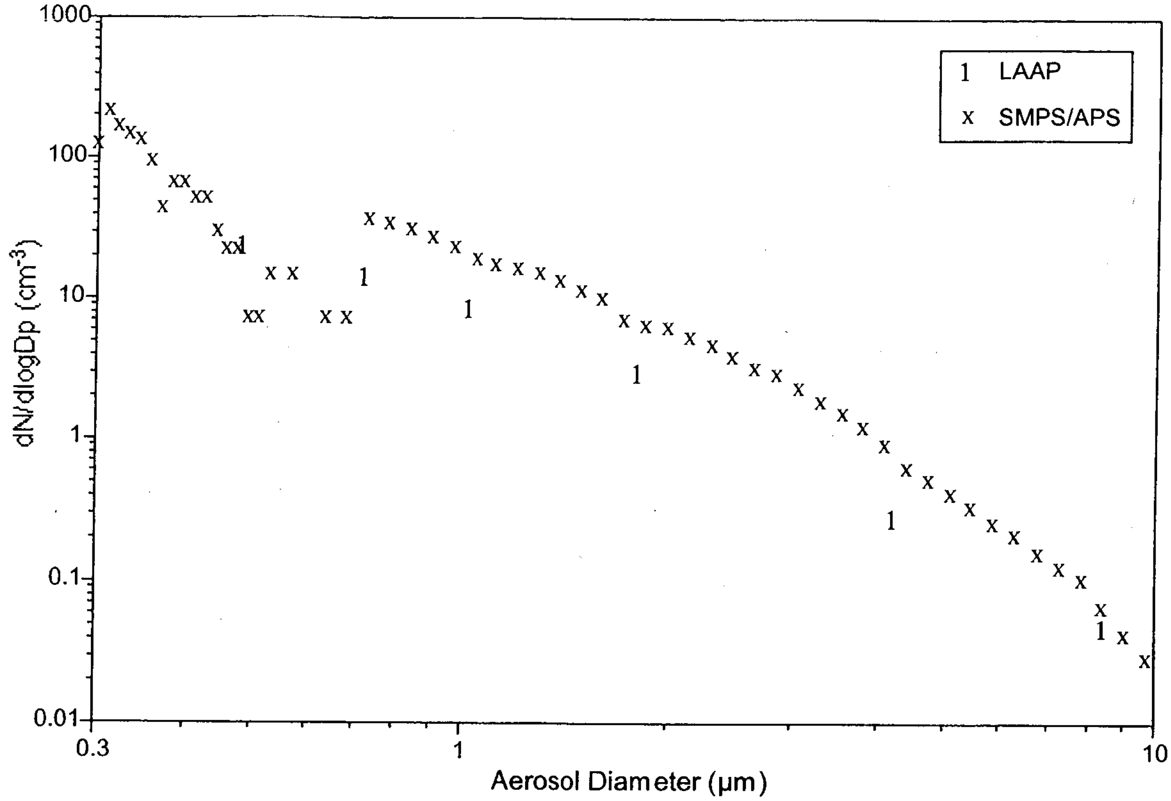

Figure 3. Comparison of aerosol number size distributions measured on 12 July 2000 from an aircraft using the LAAP and on the ground using a TSI Scanning Mobility Particle Sizer (SMPS) and a TSI Aerodynamic Particle Sizer (APS). The scatter in the SMPS/APS data from 0.4 to 0.7 µm is a consequence of being at the upper end of the SMPS measurement range where analytical uncertainties are large.

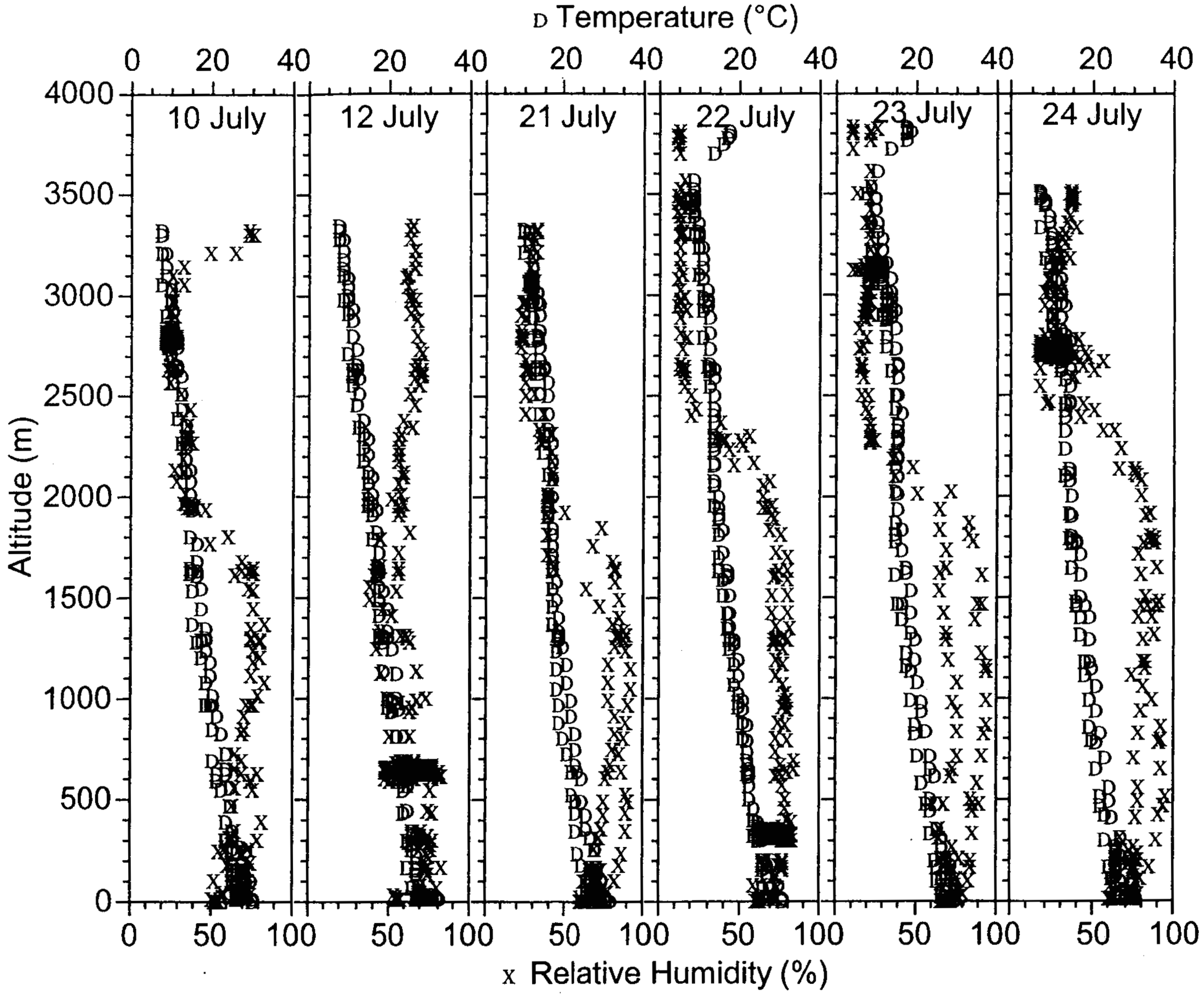

Figure 4. Temperature and relative humidity profiles measured by the LAAP near Puerto Rico during the PRIDE field experiment during July 2000. Measurements were made during both ascent and descent. The clusters of data points indicate where samples for chemical analysis were collected.

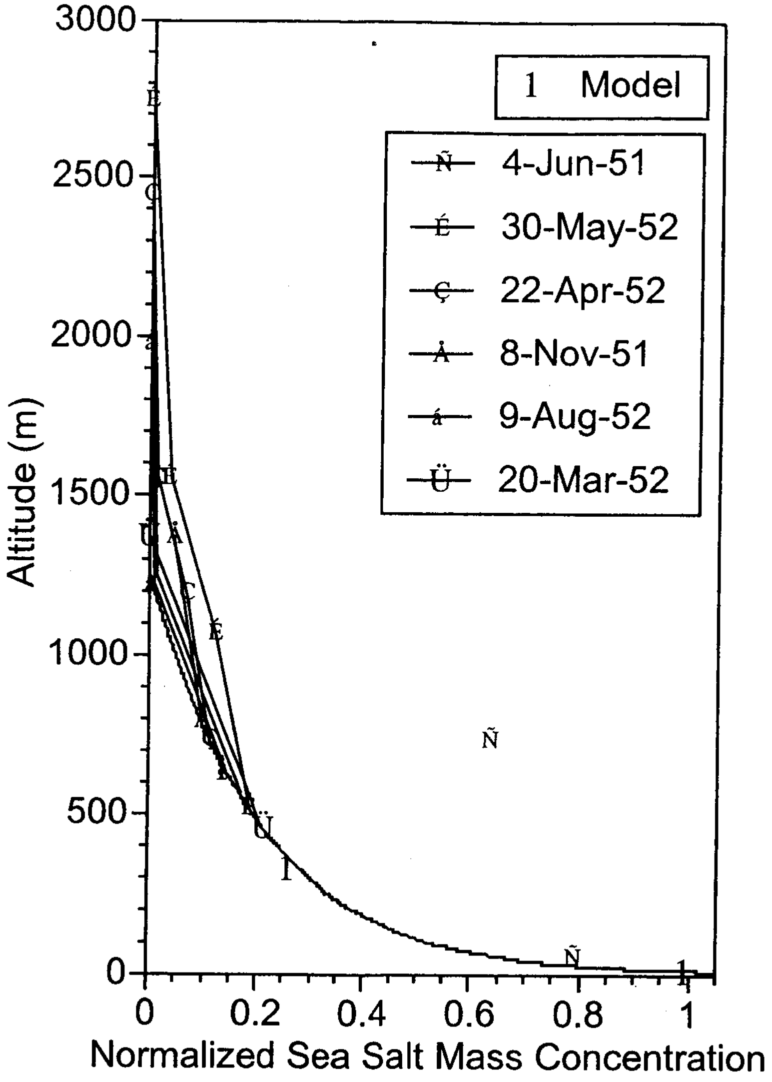

Figure 5. Surface normalized sea salt aerosol mass concentration measured during PRIDE with altitude are identified with the solid "x". The curve is the model described in section 3.3. The open symbols are the data of *Woodcock* [1953] normalized to the model described in section 3.3 at their lowest altitude.

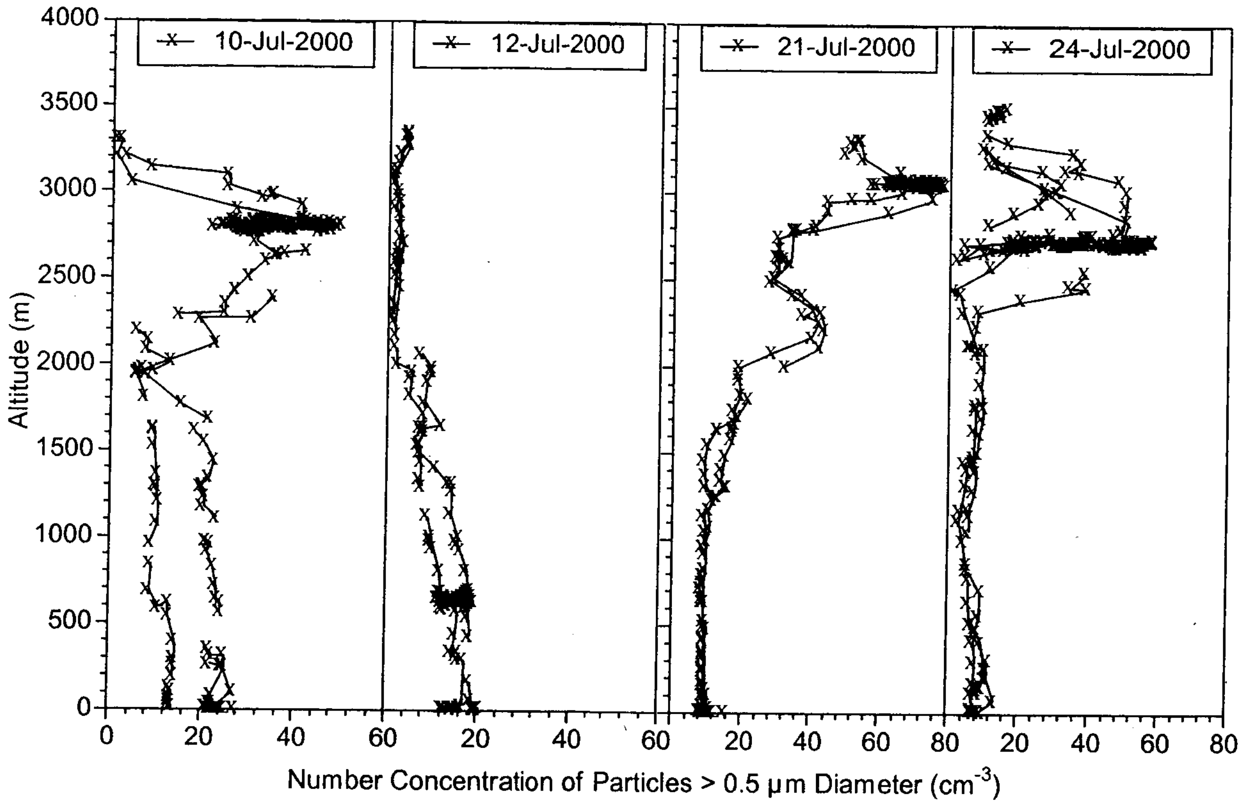

Figure 6. Number concentration of particles with diameters larger than 0.5 μm with altitude over Puerto Rico during July 2000. Except for altitudes below 100 to 200 m, these particle measurements reflect almost exclusively dust aerosols. These representative profiles exemplify the features evident during the PRIDE expedition.

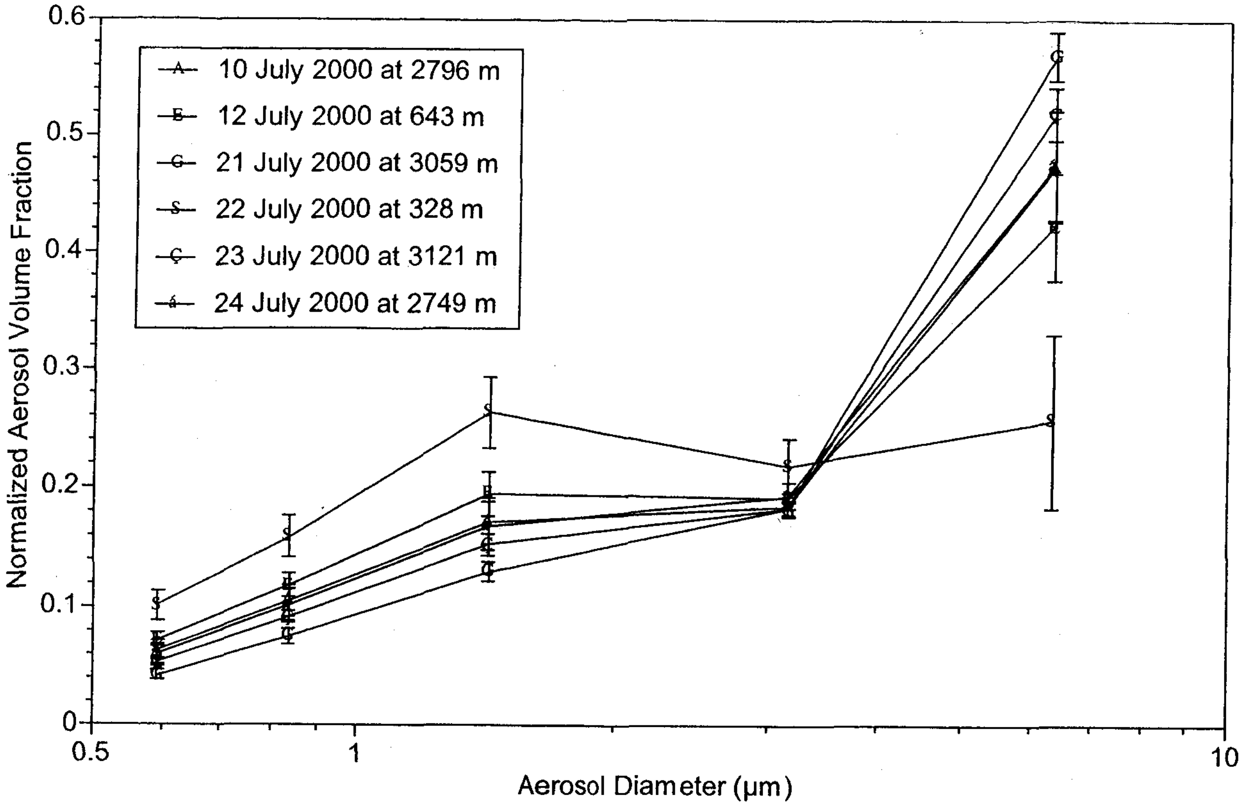

Figure 7. The normalized aerosol volume size distributions for measurements made during the collection of aerosol filter samples, each spectra represents between one and a half and two hours of sampling a nearly constant altitude. The spectra for 12 and 22 July are measurements in the MBL; the rest of the size distributions were measured in the SAL.