Long-Range Transport of Continental Aerosols over the Atlantic Ocean and Their Effects on Cloud Structures

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ABSTRACT

Airborne measurements of aerosols and cloud microstructures were made in the vicinity of the Azores Islands. Two dichotomous cases are examined: a clean marine air mass and a continually influenced air mass. The clean marine air mass had relatively low Aitken nucleus (CN) and cloud condensation nucleus (CCN) concentrations, whereas the continually influenced air mass had high CN and CCN concentrations. Also, black carbon, sulfate, and SO$_2$ concentrations were significantly higher in the continually influenced air mass. The continually influenced air mass had a monomodal aerosol number distribution with a peak at about 0.05-μm radius, whereas the clean marine air mass had a bimodal aerosol number distribution with peaks at about 0.02 and 0.08 μm radius. The formation of the aerosol mode at 0.08 μm is attributed to aerosols left behind when drizzle drops (which grow efficiently by the collision–coalescence mechanism in clean marine air) evaporate. Stratocumulus clouds in the continually influenced air mass had a mean cloud droplet number concentration about 160% higher, and a cloud droplet effective radius about 27% lower, than for stratocumulus clouds in the clean marine air mass.

These measurements demonstrate that polluted continental air can affect aerosol concentrations and cloud microstructures up to at least 2500 km from land.

1. Introduction

The effects of aerosols on cloud droplet spectra were recognized by Squires (1958), who showed that trade wind cumulus clouds off the coast of Hawaii have significantly fewer droplets, and wider droplet spectra, than cumulus clouds in continental air masses over eastern Australia. These differences were attributed to higher cloud condensation nucleus (CCN) concentrations in continental air. Since then there have been many observations documenting the effects of pollution on CCN, cloud microstructures, and precipitation (e.g., Squires 1966; Warner and Twomey 1967; Warner 1968; Hobbs et al. 1970; Eagan et al. 1974a,b; Radke and Hobbs 1976; Hindman et al. 1977a,b,c).

Twomey (1974) suggested that the effects of anthropogenic CCN on cloud microstructures should increase the albedo of clouds, and Twomey et al. (1984) pointed out that marine stratiform clouds should be particularly susceptible to such modification. Subsequently, the phenomenon of “ship tracks” (Conover 1966; Coakley et al. 1987; Radke et al. 1989; King et al. 1993) provided excellent verification of these suggestions. Since the earth’s radiation balance is sensitive to the albedo of marine stratiform clouds (Randall et al. 1984), the extent of anthropogenic pollution over the oceans and its effects on the microstructures and radiative properties of these clouds is of particular interest.

From measurements of CCN over the oceans, Twomey and Wojciechowski (1969) deduced that the lifetime of CCN of continental origin as they move over the oceans is several days. Evidence that the microstructures of marine stratiform clouds can be modified by continental aerosols was provided by Noonkester (1984), Nicholls and Leighton (1986), and Stephens and Platt (1987). However, in these cases, the clouds were located within about 230 km from the land. In this paper we show that continental aerosols can modify the microstructures of marine stratocumulus clouds even after they have traversed about 2500 km from land. We also present data that support Hoppel et al.’s (1990) conclusion that continentally derived aerosol over the oceans has a distinct size spectral shape, and we discuss possible reasons for this.

2. Measurement techniques

The measurements described here were obtained in June 1992 during the Atlantic Stratocumulus Transition Experiment (ASTEX) in the vicinity of the Azores Islands in the northeast Atlantic Ocean. Unless mentioned otherwise, all of the measurements were obtained aboard the University of Washington’s Convair
C-131A research aircraft. Brief descriptions of the instruments relevant to this study are given below; a detailed description of the integrated airborne facility has been given by Hobbs et al. (1991).

A Rosemount, platinum-wire resistance, temperature probe was used to measure static air temperatures. For the range of liquid water contents (LWC) encountered in this study (<0.6 g m\(^{-3}\)) the errors in the temperature measurements were about 0.3°C in both clear and cloudy air (Lawson and Cooper 1990). Vapor mixing ratios were obtained using an Ophir-2000 infrared optical hygrometer; the accuracy of these measurements was about 10% for temperatures greater than 0°C.

During this study we carried out the first airborne field tests of a new cloud microphysics probe (the PVM-100A) developed by Gerber Scientific Inc. (Gerber et al. 1994). Liquid water contents measured by the PVM-100A were reported to have a precision of about 2% with a response of 10 Hz. To permit comparisons with other cloud microphysical instruments aboard the Convair C-131A, the PVM-100A was mounted close to a Johnson—Williams hot-wire probe, a King hot-wire probe, and a Particle Measuring Systems FSSP-100. With the exception of the King hot-wire probe, which tended to drift in cloud, and excluding clouds with LWC > 0.6 g m\(^{-3}\) and heavy drizzle, the measurements of LWC by all four of these instruments were consistently within 10% of each other.

The cloud droplet number distributions for droplets between 2 and 320 μm in diameter and the area-weighted cloud droplet radius (i.e., the effective radius) were obtained from the Particle Measurement Systems (PMS) FSSP-100 and OAP-100X 1D cloud probes. (The first two channels of the 1D cloud probe, from 20 to 60 μm, were discarded due to systematic undercounting.) The FSSP-100 was calibrated on several occasions during the field project using glass beads of known sizes. The validity of these tests were confirmed using a Droplet Measurement Technologies "micropositioner" tool. These calibrations led to an adjustment of bin width from the standard 3.0–3.3–μm diameter; this increased the maximum possible FSSP-100 droplet size measured by the FSSP-100 from 47- to 51.5–μm diameter. For probe intercomparison, the concentrations measured by the FSSP-100 were adjusted using a correction for coincidence and dead-time losses (Baumgardner 1982; Mossop 1983). An intercomparison flight of the Convair C-131A and the British Meteorological Office’s C-130 aircraft in a stratocumulus cloud layer over the Azores on 23 June 1992 showed agreement to within 10% of the effective radius derived from FSSP-100 probes on each of the two aircraft. The OAP-100X was calibrated with lead shot of various sizes, dropped through the field of view of the probe. Liquid water contents derived from this probe were compared to those obtained from the King and Johnson–Williams LWC probes over the appropriate droplet size range. These various calibration and tests indicated that a high degree of confidence can be placed in droplet spectra measurements from both the FSSP-100 and the OAP-100X probes aboard the Convair C-131A. However, for purposes of comparability, the droplet concentrations in each size bin of the two PMS probes were scaled (by ≈10%) so that the LWC derived from the PMS probes were the same as that measured from the PVM-100A.

Aitken nucleus concentrations were measured with a General Electric CNC 1 condensation nucleus (CN) counter. This probe measures the total concentration of particles with diameters between 0.005 and 6 μm with an approximate 2-s time resolution. Aerosol size spectra were measured using a TSI Differential Mobility Analyzer (DMA) coupled to a TSI 3760 CN counter; we refer to this system as the Differential Mobility Particle Sizer (DMPS). The DMPS measured particles with diameters from 0.02 to 0.6 μm. A description of the DMPS has been given by Hegg et al. (1993). Cloud condensation nucleus spectra were measured aboard the National Center for Atmospheric Research (NCAR) Electra aircraft using the CCN spectrometer described by Hudson (1989).

3. Criteria for choice of case studies

The C-131A aircraft flew 16 research flights during ASTEX, for a total of approximately 75 hours of flight time. For the present study, two flights were chosen that highlight the dichotomy between aerosol and cloud droplet number distributions in continentally influenced boundary layer air, which was transported for a large distance over the ocean, and in marine boundary layer air, which had not encountered a continent for at least four days.

For a direct correlation to exist between CCN and cloud droplet concentrations and size distributions, similar cloud LWC are required. Therefore, in both cases, the clouds chosen were of a similar type (stratocumulus) and they had similar dynamic and thermodynamic properties and similar LWC. Airborne measurements in complete vertical profiles and long horizontal transects of the clouds were also required in order to properly characterize the cloud microstructures.

4. Overview of the two case studies

The two cases chosen for this study occurred on the mornings of 12 June 1992 (UW Flight 1561) and 22 June 1992 (UW Flight 1569) in the vicinity of Santa Maria Island (37°N, 25°W) in the Azores. The 12 June case (hereafter case 1) was characterized by a single layer of stratocumulus about 300 m thick with its base at about 800 m. The 22 June case (hereafter case 2) was a stratocumulus layer about 200 m thick with its base at about 1200 m. In case 1 the aircraft first ascended through the depth of the cloud, followed by an approximate 15-km horizontal flight at about 100 m
below cloud top. In case 2 the aircraft climbed through the cloud layer and then flew a 4-km horizontal leg about 25 m below cloud top.

Airmass back trajectories for the two cases were computed from the 6-h operational analyses of the European Centre for Medium-Range Weather Forecasts (ECMWF), which were archived for the ASTEX region at 25 hPa in the vertical and 1.25° horizontal spacing. For improved regional accuracy, the ECMWF analyses incorporated most of the special soundings taken during ASTEX (Bretherton et al. 1995). Boundary layer trajectories were computed using 1000-hPa horizontal winds; 3D winds were used to determine trajectories in the free troposphere.

Trajectories for cases 1 and 2 are shown in Fig. 1. The exact courses of these trajectories must be interpreted with caution since data were scarce over the Atlantic Ocean. However, it is clear that the boundary layer air mass associated with case 1 was still far from land four days prior to 12 June. The trajectories in both the boundary layer and free troposphere associated with case 2 were in the vicinity of the British Isles on 19 June, and then after traveling about 2500 km over the Atlantic Ocean, the air reached the Azores two to three days later.

The atmospheric boundary layers were similar for cases 1 and 2 (Fig. 2). Blended global sea surface temperature (SST) from the ECMWF indicate that in both cases the SST temperature was about 18.5°C. Also, in both cases, the subcloud mixed layer had a nearly dry adiabatic temperature profile with no measurable inversion at cloud base. The equivalent potential temperature \( \theta_e \) and wind profiles are suggestive of a well-mixed cloud deck above a weakly convectively unstable subcloud layer for both cases 1 and 2. In the cloud layers, \( d \theta_e / dz \) is constant with height and the wind shear is low; below the cloud layers, \( d \theta_e / dz < 0 \) and the wind shear is moderate. Therefore, mixing was weakly inhibited between the subcloud layers and the stratocumulus clouds. Scattered cumulus clouds were present below the stratocumulus decks, but in neither case were the cumulus clouds large enough to penetrate the bases of the stratocumulus. Also, surface observations along both of the boundary layer back trajectories in Fig. 1 showed cloud coverages and cloud types similar to those measured at the location of our airborne measurements.

5. Results

Airborne chemistry measurements made in the vicinity of the Azores confirm the different natures of the air masses for cases 1 and 2. Black carbon data (from the NCAR Electra aircraft) show boundary layer concentrations of less than 5 ng m\(^{-3}\) on 12 June and 15 ± 5 ng m\(^{-3}\) on 22 June. Boundary layer sulfate and SO\(_2\) concentrations measured from the C-131A were, respectively, 0.03 ± 0.03 μg m\(^{-3}\) and 12 ± 2 pptv in the maritime airmass of 13 June, and 3.0 ± 0.5 μg m\(^{-3}\) and 25 ± 10 pptv in the continental airmass of 22 June. High concentrations of black carbon, sulfate, and SO\(_2\) concentrations are good indicators of continental (anthropogenic) pollution.

Average CN concentrations measured below and above the cloud layer for case 1 were 210 and 310 cm\(^{-3}\), respectively, and for case 2 they were 900

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**Fig. 1.** Back trajectories ending at the Azores on 0930 UTC on 12 June (clean marine air: case 1) and at 0845 UTC on 22 June (continentally influenced air: case 2). Solid lines show boundary layer trajectories (1000 hPa), and dashed lines free-troposphere trajectories with associated pressures in hPa. Arrows indicate positions of air parcels at 12-h intervals.
and 1180 cm$^{-3}$, respectively. Aerosol number distributions measured from individual “grab” samples in the boundary layer and in the free troposphere in the two air masses are shown in Fig. 3. For case 1 the aerosol number distributions are bimodal, with maxima at particle radii of about 0.02 and 0.08 μm (Fig. 3a). In case 2 the aerosol number distributions are monomodal, with maxima at about 0.05-μm radius (Fig. 3b). The aerosol number distribution confirm the CN measurements that the total particle concentrations were much higher in case 2 than in case 1. However, for particles between 0.01 and about 0.03 μm the reverse was true.

Airborne measurements (from the NCAR Electra aircraft) of CCN over the Azores on 12 June and 22 June 1992 gave spatially varying concentrations at 0.8% supersaturation of about 30–100 cm$^{-3}$ and about 100–800 cm$^{-3}$, respectively. Hence, the continental influence on aerosols on 22 June was clearly reflected in the CCN.

Vertical profiles of various parameters measured in the stratocumulus clouds for case 1 and case 2 are shown in Fig. 4. In both cases, the LWC are close to adiabatic and they increase with height at about the same rate. Consequently, the influences of the different ambient aerosols on the cloud structures in case 1...
and case 2 were quite apparent, as summarized in Table 1. In case 2 (the continentally influenced air mass) the cloud droplet concentrations were higher (by $\sim 160\%$), and the effective cloud droplet radius lower (by $\sim 27\%$), than in case 1 (the clean marine air mass).

Horizontal transects of the clouds were flown at an altitude of 992 m for case 1 and 1376 m for case 2. Table 2 shows the averages and standard deviations of the cloud microphysical properties measured in each case. At approximately the same height above cloud base, the horizontally averaged LWC for case 2 was about 20% lower than for case 1. It can be seen from Fig. 5 that the stratus cumulus cloud that formed in the clean, marine air (case 1) had a wider droplet spectrum than the cloud that formed in the continentally influenced air (case 2). The higher droplet number concentrations in case 2 were due entirely to droplets less than 7.5-µm radius. Particularly notable is the absence of a drizzle mode in case 2. For example, at 65-µm radius the droplet number concentrations for case 1 are about 50 times greater than for case 2.

6. Discussion

We will consider first the shapes of the aerosol number distributions shown in Fig. 3. On 12 June, in the clean marine air mass, the aerosol number distributions were bimodal (Fig. 3a) with peaks at about 0.02- and 0.08-µm radius. On 22 June, in continentally influenced air, the aerosol number distributions were monomodal with a peak at about 0.05-µm radius (Fig. 3b). These spectral shapes are similar to those reported for marine and continentally influenced air by Hoppel et al. (1990). By considering the sources and sinks for aerosols in a clean marine air mass, Hoppel et al. showed that the peak in the aerosol number distribution near 0.02-µm radius was likely due to condensational growth of recently nucleated particles. The peak at about 0.08-µm radius was attributed to aerosols left behind by evaporated droplet in which aqueous-phase chemical reactions had taken place. The formation of a particle concentration mode at 0.02 µm is inhibited in continental airmasses by (a) the large concentration of particles at 0.05 µm, which provide a high surface area sink for available condensate, and (b) the high surface area of droplets in continental clouds, which permit relatively high rates of scavenging of the interstitial aerosol between cloud droplets. Chemical reactions in cloud droplets may be less effective in continental than in marine air masses in producing the peak at about 0.08 µm. This is because a plausible combination of low pH cloud water and low H$_2$O$_2$ concentra-
Fig. 4. Microphysical measurements in the stratocumulus clouds for cases 1 and 2 for the same vertical profiles shown in Fig. 2. The two panels on the left side are cloud droplet concentrations from the FSSP-100 probe, the two center panels are LWC from the Gerber PVM-100A probe (the dashed lines are the adiabatic LWC profiles), and the two right-hand panels show the surface-area-weighted mean (or effective) cloud droplet radius \( r_e \) and the mean volume cloud droplet radius \( r_v \) derived from the FSSP-100 measurements.

Sections in continentally influenced air masses could result in minimal aqueous-phase sulfate production. In addition, sulfate production by aqueous phase \( O_3 \) and \( H_2O_2 \) oxidation is a strong function of cloud droplet radius (Hoppel et al. 1994). Since the mean volume radius of the droplets in the case 2 cloud was about 29% lower than in case 1, aqueous-phase particle growth would have been significantly slower in the continentally influenced air mass.

**Table 1.** Vertically averaged values of cloud droplet concentrations \( \bar{N} \), liquid water content (LWC), cloud droplet effective radius \( \bar{r}_c \), and mean volume cloud droplet radius \( \bar{r}_v \) for the stratocumulus clouds that formed in clean marine air (case 1) and in continentally influenced air (case 2). The data for case 1 were averaged over the lower 200 m of the cloud layer and for case 2 over the entire cloud depth (200 m).

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<tr>
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<th>Case 1</th>
<th>Case 2</th>
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<tbody>
<tr>
<td>( \bar{N} ) (cm(^{-3}))</td>
<td>174</td>
<td>457</td>
</tr>
<tr>
<td>LWC (g m(^{-3}))</td>
<td>0.23</td>
<td>0.21</td>
</tr>
<tr>
<td>( \bar{r}_c ) ((\mu)m)</td>
<td>7.3</td>
<td>5.3</td>
</tr>
<tr>
<td>( \bar{r}_v ) ((\mu)m)</td>
<td>6.8</td>
<td>4.8</td>
</tr>
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**Table 2.** Horizontal averages \( \bar{x} \) and standard deviations \( \sigma \) of the parameters shown in Table 1 for case 1 (clean marine air) and case 2 (continentally influenced air).

<table>
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<tr>
<th></th>
<th>Case 1</th>
<th>Case 2</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \bar{N} ) (cm(^{-3}))</td>
<td>161</td>
<td>605</td>
</tr>
<tr>
<td>LWC (g m(^{-3}))</td>
<td>0.42</td>
<td>0.34</td>
</tr>
<tr>
<td>( \bar{r}_c ) ((\mu)m)</td>
<td>9.7</td>
<td>5.8</td>
</tr>
<tr>
<td>( \bar{r}_v ) ((\mu)m)</td>
<td>8.5</td>
<td>5.1</td>
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There is, however, a mechanism that can explain the particle mode at 0.08 μm that does not involve aqueous-phase cloud chemistry. After two cloud droplets collide, the combined material within them is released into the air when the resulting drop evaporates. Since droplets grow more efficiently by the collision–coalescence mechanism in marine clouds than in continental clouds (note that a drizzle mode was present on 12 June—see Fig. 5), larger particles from evaporating cloud drops should be more common in marine-type air masses than in continental-type air masses. Numerical modeling studies by Flossman (1994) and Flossman et al. (1987) show the formation of a peak in the aerosol number distribution near 0.08-μm radius due to material left behind by evaporated droplets, even in the absence of aqueous-phase chemical reactions. Hence, residuals left behind by the evaporation of large cloud drops provides a viable explanation for the peak in the aerosol number distribution near 0.08 μm in the marine air masses on 12 June. Hoppel et al.’s measurements of monomodal and bimodal aerosol number distributions were obtained in the marine boundary layer. Our measurements show that on 12 June 1992 the same modal shapes were present in both the marine boundary layer and the free troposphere (Fig. 3). This indicates that aerosol exchange occurred across the marine boundary layer inversion. In the 36 h prior to 1200 UTC on 12 June heavy overcasts of stratuscumulus clouds mixed with cumulus were observed at surface stations aligned along the free-troposphere back trajectories. Thus, the transport of aerosol across the marine boundary layer might well have been due to cumulus pumping.

The higher concentrations of aerosols and CCN in the air over the Azores on 22 June 1992 resulted in stratuscumulus clouds that had higher droplet concentrations and a smaller effective radius than otherwise similar clouds that were present in the same region on 12 June 1992 when the air was much cleaner. Moreover, because the LWC was similar in both cases, and the effective radius \( r_e \) does not differ greatly from the mean volume radius \( r_v \), the difference in the effective radius was almost exactly that expected from the difference in droplet concentrations (i.e., \( r_e \alpha N^{-1/3} \)).

The earth’s radiation budget is sensitive to both the direct effects of aerosols and the indirect effects of aerosols on cloud microstructure (e.g., Charlson et al. 1992). The observations presented here demonstrate that polluted air from continents can penetrate several thousands of kilometers over the oceans, where they can have both a direct and an indirect effect on the radiation balance. The magnitudes of these effects on local, regional, and global scales remain to be established.

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