

Microphysics of INDOEX Clean and Polluted Trade Cumulus Clouds

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Short title:

Abstract. In this study, we examine how emissions from the Indian subcontinent modify the microphysical properties of trade wind cumulus clouds over the Indian Ocean. *In situ* microphysical data from 18 National Center for Atmospheric Research C130 research flights during the Indian Ocean Experiment (INDOEX) in February-March 1999, in polluted to pristine regions, as delineated from the concentrations of condensation nuclei (CN), were analyzed. Cloud properties were found to vary in a systematic way during the five gradient flights: i.e., those flights in which the C130 flew from about 4.5° N latitude in the high-CN regions, south across the intertropical convergence zone (ITCZ) into the clean regime, then farther south to about 8° S latitude. The high-CN regime contained relatively large concentrations of small droplets as compared to the low-CN regime, where low concentrations of large droplets, and more frequent drizzle, were measured. An analysis of the data from penetrations into thousands of clouds during all 18 flights supports these qualitative observations: In the polluted regions, the droplet concentrations were a factor of 3 higher, the droplet effective diameters were 35% smaller, and drizzle was observed 25% as often than in the pristine regions; in both polluted and pristine regions, the bulk cloud properties (liquid water content (LWC), vertical velocity, cloud horizontal dimensions) were approximately the same. Even larger differences in the microphysical properties between the high-CN, intermediate-CN, and low-CN regimes were noted when the data set was partitioned by the LWC. A high ratio offorward scattering spectrometer probe (FSSP) to CN concentrations was noted in the low-CN regime, whereas in the high-CN regime this ratio was small. Droplet growth calculations in an adiabatic, 1D parcel model over

a 300 m cloud depth support the droplet observations and indicate that the cloud optical depths in the high CN regime could have been as large as twice those in the low CN regime as a result of differences in the cloud condensation nucleus (CCN) population. The corresponding albedos of about 0.54 in the high CN regime and 0.47 in the low CN regime signify potentially large differences in albedo for the typical vertical velocities observed in INDOEX clouds, but it is unclear how entrainment would affect this difference. The calculations indicate that stronger updrafts associated with deep convection could lead to a larger difference between the microphysics of high-CN and low-CN regions.

1. Introduction

Over the past 15 years, theory, modeling, and satellite-based studies have suggested that enhanced aerosol concentrations resulting from anthropogenic emissions can enhance cloud albedos and cause a planetary cooling that could oppose the greenhouse effect of anthropogenic trace gases. This “indirect aerosol effect,” or “Twomey” effect, was proposed by *Twomey* [1974, 1977], and is based on the idea that increasing pollution generally increases cloud condensation nucleus (CCN) concentrations, hence increasing the number of cloud drops (N); for fixed liquid water content (LWC), increasing N increases the extinction coefficient and, for finite and fixed cloud thicknesses, increases the cloud albedo. Furthermore, increasing the droplet concentration decreases the cloud droplet sizes, and these changes constitute a second Twomey effect, which might inhibit coalescence precipitation and lead to colloidal stability. This would cause further planetary cooling by increasing the LWC, and extending cloud lifetimes [*Albrecht, 1989*]. On the other hand, the presence of absorbing aerosols within cloud layers in heavily polluted regions could reduce cloud lifetimes by warming and desiccating cloud layers [*Ackerman et al., 2000*]

Confirmation of the first Twomey effect, increases in cloud droplet concentrations and cloud albedo resulting from increases in CCN concentrations, comes from satellite and in situ observations. CCN are defined as a subset of the aerosol particles that are sufficiently large and hygroscopic to nucleate water droplets at a specified value of supersaturation. *Coakley et al.* [1987] showed that under stable meteorological

conditions, ship-stack exhaust enhanced cloud reflectivity at $3.7 \mu\text{m}$ in daytime satellite images. Ship-stack exhaust was found to be a source of CCN that increased the number of cloud droplets, reduced the mode radius, reduced the concentration of drizzle-sized droplets, and produced a narrow size distribution [King *et al.*, 1993; Ackerman *et al.*, 2000]. Han *et al.* [1994] conducted a near-global survey of effective droplet radii in liquid water clouds using International Satellite Cloud Climatology Project (ISCCP) data, noting that systematic differences in continental and maritime areas, as well as hemispheric contrasts that indicate the effects of aerosol concentrations; cloud droplet radii in continental water clouds were about $2\text{--}3 \mu\text{m}$ smaller than in maritime clouds, and droplet radii maritime clouds in the Northern Hemispheric (NH) were about $1 \mu\text{m}$ smaller than in the Southern Hemisphere (SH). Rosenfeld [1999] used radar measurements from the Tropical Rainfall Measuring Mission (TRMM) to show that rain production through the warm rain (coalescence) process was practically shut off in convective tropical clouds infected by heavy smoke. Higher CCN concentrations presumably produced greater numbers of cloud droplets and effectively eliminated the production of drizzle and raindrops. Pawlowska *et al.* [1999] examined the properties of clean and polluted clouds, using data from the Second Aerosol Characterization Experiment (ACE 2) in the northeast Atlantic; droplet concentrations were low in the clean clouds ($< 50 \text{ cm}^{-3}$) and much higher in the most polluted clouds (300 cm^{-3}), and the drops were nearly twice as large in the polluted clouds as in the clean clouds. Martinsson *et al.* [2000] observed cloud droplet concentrations up to 3000 cm^{-3} in orographic wave clouds forming in polluted air masses during ACE 2.

Our objectives in this study are twofold: (1) to characterize the properties of trade wind cumuli over the Indian Ocean and (2) to estimate the enhancement in droplet concentrations resulting from pollution. Our second objective, broadened to include the indirect radiative effect, was a primary objective of INDOEX. The basic idea behind this part of INDOEX was to compare the microphysical and radiative properties of clouds that formed over the Indian Ocean in polluted NH air with these properties of clouds in clean SH air. In situ measurements by the NCAR C130 during transits from the NH into the pristine SH air—the “gradient flights” were used to infer changes in the cloud microphysical properties resulting from pollution. INDOEX was conducted in February and March of 1999, during the northeast monsoon: a period when intense high-pressure systems centered over Mongolia force cool continental air over the whole of Southeast Asia. A systematic study of this type, especially for the populated regions of southeast Asia, had not previously been carried out. Although scattered instances of enhanced droplet concentrations because of increased CCN resulting from anthropogenic emissions have been reported, INDOEX conducted the first systematic examination of the microphysical properties of clouds which have been modified by pollution in the populated regions of Southeast Asia.

In section 2 of this article, we discuss the instrumentation and the flight patterns used to collect these data. The microphysical data from the gradient flights are presented in section 3, and data from all flights are combined in section 4. The implications of this study are discussed in section 5 and summarized in section 6.

2. Instrumentation and Flight Patterns

Microphysical measurements were acquired from the NCAR C130 based at Hulule, Maldives, during the intensive field phase (IFP) of INDOEX. The IFP was conducted from February 16 to March 25, 1999, when the C130 sampled many trade wind cumuli. Trade wind cumuli are scattered cumulus clouds in the trade wind belts. The classic trade wind cloud structure is characterized by a base near 700 m, with detrainment under the base of a strong trade inversion near 1500 m (S. Krueger, personal communication, 2000).

Various aspects of the 18 IFP research missions flown by the C130 are summarized in Table 1. Shown in the table are the flight number, the date, the number of seconds the C130 flew in-cloud at various latitude ranges, a subjective rating of the microphysical data based on the duration of in-cloud sampling in polluted and pristine regions, and a general comment on the type of mission. The five gradient flights were the most useful for acquiring microphysical data in NH and SH air on single flights; these flights were low-level transits, in and below cloud, from Male, Maldives (approximately 4.5° N), to as far south as possible, with a reciprocal track through the middle troposphere to collect solar and infrared radiometer data. Relatively long periods of in-cloud sampling in both NH and SH clouds were obtained for gradient flights 3, 4, and 8, whereas only SH clouds were penetrated in gradient flight 11, and a shorter period of in situ sampling was obtained during flight 17. Clouds in the latitude range 2° to 6° N were almost certainly located in polluted, NH air, while those to the south of the intertropical convergence

Table 1

zone (approximately 2° S) were usually located in the pristine regimes. Aircraft wind data usually did not reveal the air mass origin because it usually contained an easterly component in the SH; accurate trajectory calculations, requiring an accurate knowledge of winds across the ITCZ, would be needed to define the location of the ITCZ. Trade wind cumuli were sampled in visually polluted regions off the coast of India on flights 9, 15, and 16 (rating of 7–8); more limited sets of data for visually polluted regions were obtained on other flights. Microphysical measurements were obtained in both low and midlevels in a cumulonimbus (Cb) cloud that was sampled during flight 14. Comparatively little microphysical data were obtained for the clear air closure (CAC) flights, and no rating has been ascribed to these flights, even though some microphysical data were collected.

A comprehensive set of aerosol concentration measurements and cloud droplet size distributions were acquired from the C130 flights. A standard set of NCAR Research Aviation Facility (RAF) instruments were used to acquire the data. Condensation nuclei were measured with a Thermo-Systems Inc (TSI) 3760 counter, which has a lower size cutoff of $0.013 \mu\text{m}$. Cloud droplet size distributions were measured with two Particle Measuring Systems Inc (PMS) FSSP-100s, which sized from approximately 3.5 to $60 \mu\text{m}$ in 15, uneven-size bins (see *McFarquhar and Heymsfield*, [this issue]). The FSSPs were located on opposite wings of the aircraft, making comparison between the probes difficult, as the clouds sampled were small horizontally (see Section 4). We used the FSSP-100 data from the left-wing probe for flights 1 through 3 and from the right-wing probe from flights 4 through 18, based on a thorough analysis of FSSP data quality, as reported in

Baumgardner (NSF/NCAR EC-130Q Hercules data quality report: Cloud microphysical measurements, available from <http://raf.atd.ucar.edu/Projects/INDOEX/QA2.html>).

Housekeeping data for the FSSP-100 were unavailable for INDOEX, and the resulting assumptions about sampling volume led to uncertainty in the concentration and FSSP-derived LWC of 15% and 35%, respectively. A PMS 260-X (1D-C) probe, measuring from about 50 to 300 μm in 15 size bins, and a PMS 2D-C probe, measuring from 25 μm to more than 800 μm , provided information on the drizzle and raindrop size distributions. Liquid water contents were measured from two King hot-wire probes. Both probes functioned well throughout the project and compared well with each other. Given that few drops were observed above 40 μm (at which size the hot-wire probe does not completely vaporize drops), there was minimal underestimation in the cloud LWC from the hot-wire probes. Vertical winds were measured using radome gust probe sensors and an inertial navigation system (INS) with an absolute accuracy of ± 0.5 m s^{-1} in straightflight, which encompassed most of the INDOEX sampling periods. State parameter measurements were acquired using the standard RAF complement of temperature, dew point, and pressure (altitude) sensors. The PMS probe data were collected at a rate of 10 Hz, whereas the King probe and state parameter data were collected at a rate of 25 Hz.

3. Gradient Flight Observations

In this section, we present the microphysical observations from the flights 3, 4, and 8, during which the C130 collected extensive in situ microphysical data in both NH and

SH clouds. The in situ data were obtained on the outbound portions of the flights, due south from Male to as far south as possible, with a few variations. The aircraft flight paths were generally not moved, either vertically or laterally, to sample cloud. Cloud penetrations in most instances occurred at approximately the same altitudes throughout the outbound leg on each day. Examination of the data also confirmed that clouds in the three regions were sampled at approximately the same times on average, indicating that diurnal variations should not complicate the analysis.

The microphysical observations from the southbound leg of flight 3 on February 20, 1999 are presented in Figure 1. The average penetration altitudes for the NH clouds were 671 ± 23 m, transition clouds were 652 ± 3 m, and SH clouds were 676 ± 21 m. Since cloud base heights in each regime are likely to be approximately the same, differences in the microphysical properties between regimes are significant.

Figure 1

Cloud droplet and CN concentrations, along with a smoothed curve showing latitude as a function of time, are given in the top panel. From from the beginning of the flight to 3° S latitude, the CN concentrations decrease steadily from above 1500 to 300 cm^{-3} , and remain relatively constant thereafter. The region south of 5° S latitude, where CN concentrations are consistently less than 300 cm^{-3} , therefore marks the point of pristine, SH air. A CN concentration of 300 cm^{-3} is consistent with previous observations of CN for pristine air; the regions where CN concentrations exceed $1,500 \text{ cm}^{-3}$ are clearly in polluted, NH air (see Appendix A). Between these regions is a transition zone, where CN concentrations are from 500 to $1,500 \text{ cm}^{-3}$. The spikes noted in the CN trace are artifacts and coincide with penetrations into cloud; they are shown

in Figure 1 for completeness but were filtered out in later analyses.

Photographs taken at two times during the transit of flight 3, one in polluted NH air and the other in clean SH air, are shown in Plate 1. The photographs were taken at approximately the same altitude and show the dramatic differences in visibility that are associated with high and low CN concentrations (top and bottom photographs, respectively). The ocean surface is not visible in the top photograph, whereas windrows are clearly visible on the ocean surface in the lower photograph. Note that the clouds in the photographs are typical of those sampled during flight 3.

Plate 1

The in-cloud updraft velocities throughout the transit (Figure 1, middle panel) were typically within the measurement uncertainty range of $\pm 0.5 \text{ m s}^{-1}$. Several updrafts of 2 m s^{-1} were measured in clouds throughout the transit.

On average, cloud droplet concentrations were considerably higher in the NH air than in the SH air. In the NH region, concentrations typically peaked at 500 cm^{-3} ; whereas in the SH air, they typically peaked at $200\text{--}300 \text{ cm}^{-3}$. The ratio of the concentration of drops to concentrations of CN varied dramatically between the NH and SH regions. In the NH region, the ratio was less than 0.3; in the SH region, it was as large as 0.8. Judging from the widths of the concentration traces associated with each cloud region, cloud widths appeared comparable in the NH and SH clouds.

Cloud droplet size distributions are summarized in Plate 2, where the concentrations in each size bin are represented by the color coding plotted on the right side of the plate. As shown in the plate, in the NH latitudes, where CN concentrations are high, large concentrations of relatively small droplets were observed. Conversely, in the pristine

Plate 2

SH regime south of 4° latitude, the droplet concentrations were low, the mean size was large, and the size distributions were broad. Several clouds in the transition and SH regions contained drizzle-size drops ($>55 \mu\text{m}$), as denoted by asterisks in the top panel of Figure 1. These regions were small and isolated.

The LWCs, derived from the FSSP probes, were approximately 25% larger after 0450 than earlier (Figure. 1, middle panel). The regions of higher LWCs were in NH, transition, and SH regimes. LWCs were typically only a few tenths of a gram per cubic meter, and no major differences in these were noted throughout the transit, suggesting that there was no major difference in cloud dynamics throughout the transit.

Because LWC was approximately constant and the concentrations were larger in the NH clouds than in the SH clouds, it is not surprising that the cloud droplet median mass-weighted diameter (D_m) was almost twice as large in the SH clouds as in the NH clouds (Figure 1, bottom panel). What is striking in this figure is the change in the diameter from about 6 to 11 μm in the transition region from 1° to 3°S latitude.

Trends in the microphysical properties observed for gradient flight 4 (February 24, 1999) are shown in Figure 2. These data displayed features that were qualitatively similar to those observed for flight 3, but with some important differences resulting from more vigorous clouds:

1. The cumulus congestus clouds penetrated between 0400 and 0415 UTC in the region of intermediate-CN concentrations contained updrafts up to 5 m s^{-1} . It is not surprising that these clouds contained drizzle, rain, and relatively large LWC and D_m . Drop concentrations were nevertheless high—300 to 400 cm^{-3} —even though the drop

Figure 2

coalescence process, which would reduce the number of cloud droplets, was active.

2. The trade cumuli sampled between 0500 and 0515 UTC were deeper and contained higher LWC than the clouds sampled during flight 3. Updraft velocities as high as 4 m s^{-1} were measured. Quite a few of these clouds contained drizzle.

3. The heights of the cloud penetrations on this flight spanned a wider range than for flight 3, because the clouds were deeper.

The measurements for gradient flight 8 (March 4, 1999) were similar to those observed for flight 3 and are shown in Figure. 3. The relatively low ratio of FSSP to CN concentrations in the high-CN regions and the large ratio of FSSP to CN concentrations in the low-CN regions are striking (top panel). As viewed from the aircraft cockpit and from photographs, the clouds sampled on this were deeper than those sampled during flight 3 (see Plate 3), which may explain why the LWCs were higher (middle panel). Peak updraft velocities were in the range 2 to 3 m s^{-1} . Drizzle was observed more frequently in clouds in the clean regime (top panel, Figure 3).

Imagery from the NCAR scanning aerosol backscatter lidar (SABL) in the high-CN and low-CN regimes is shown in Plate 4. Both images in the figure were taken with SABL looking directly upward from approximately 100 m above the ocean surface. Relative backscatter values in the range 5 to 15 dB were observed in clear air at altitudes up to 2 km in the high-CN region (top panel) near 3° N . Note the presence of a few clouds with higher backscatter values in the boundary layer. Conversely, above a few hundred meters from the ocean surface, the relative backscatter values of 5–10 were observed in clear air in the boundary layer in the low CN regime south of 2° S . Note

Figure. 3

Plate 3

Plate 4

the presence of a few small clouds in the imagery. This comparison strengthens the argument that the boundary layer in the NH air is polluted. Note the presence of a few small clouds in the imagery.

A diagram illustrating the changes in the droplet size distribution during flight 8 is shown in Plate 5. The higher concentrations of smaller cloud droplets, with narrow size distributions, were noted in the higher latitudes, whereas the lower latitudes contained lower concentrations of larger drops with broader size distributions. This result was noted even though the cloud penetrations in the NH air were slightly higher: 722 ± 102 m versus 613 ± 89 m in the SH clouds.

Plate 5

4. Composite Data Set

Our purpose in this section is to synthesize the cloud microphysical data collected by the C130 in 18 INDOEX research flights and to determine whether the observed variations in cloud microphysical properties can be attributed to differences in the CCN spectrum between the NH and SH regimes or because of differences in cloud dynamics, entrainment, and drizzle. The C130 microphysical and state parameter data have been measured and analyzed at 10 Hz, yielding a horizontal resolution of approximately 14 m.

The data presented are partitioned into one of three regions based on CN concentrations: clean, intermediate, and polluted. In Appendix A, we give earlier measurements of CN and CCN concentrations in the unpolluted marine boundary layer, and infer background concentrations for continental boundary layer regions for reasons

discussed in Appendix A, based primarily on a large set of data from Australia and the Southern Ocean. On the basis of the evidence given in Appendix A, following are typical concentrations in places where sources from populated landmasses are likely to have little effect: (1) Remote oceans, CN: 300 cm^{-3} ; CCN at 1% supersaturation: 200 cm^{-3} and (2) remote continental surface CN: $< 700 \text{ cm}^{-3}$; CCN at 1% supersaturation: $200\text{--}300 \text{ cm}^{-3}$. On the basis of these data, CN concentrations may be taken to be less than 500 cm^{-3} in the unpolluted regime; greater than 1500 cm^{-3} in the polluted regime; and between these limits in the intermediate region. Spikes in the CN concentrations observed during penetrations into cloud, as shown in the gradient flight figures have been removed using data immediately before and after cloud penetration and applying the procedure described by *McFarquhar and Heymsfield*, [this issue].

Frequency distributions of cloud droplet LWC, concentration, and effective radius (droplet volume divided by cross-sectional area) and of drizzle, are plotted in Figures 4–7.

Figures 4–7

The essential findings from these data are as follows: (1) The LWC was essentially the same in all regimes (Figure 4). (2) The median droplet concentration in the high-CN regime was 40% larger than in the intermediate-CN regime and 250% larger than in the low-CN regime (Figure 5). The median is clearly skewed to high concentrations in the high-CN regime and to low concentrations in the low-CN regime. (3) The effective radius in the low-CN regime was 36% larger than in the intermediate-CN and high-CN regimes (Figure 6). (4) Because the LWCs are approximately the same in the clean and intermediate regimes, and only slightly higher in the high CN regime, the results

shown in Figures 5 and 6 are consistent with the premise that variations in droplet concentration and effective radius result from differences in the CCN spectra in the various regimes and not from variations in the LWC. (5) Drizzle and raindrops were infrequently found in the various regimes (Figure 7; see also and Figures 1, 2, and 3) and have a negligible effect on the distribution of LWC. However, drizzle was found four times more frequently in the low-CN regime than in the high-CN regime.

Differences in the cloud microphysical properties in the various regimes may not be the results of variations in populations of CN and CCN. The extent of entrainment and the magnitudes of the vertical velocities can also have a profound influence on cloud microphysical properties. The former can reduce LWC and modulate the strengths of updrafts and vertical cloud extent, whereas cloud-base updraft strength directly controls the numbers of droplets activated for a given CCN supersaturation spectrum and the corresponding mean droplet diameter. In the remaining portion of this section, we will examine whether there was any significant differences in the width and vertical velocities of clouds between the three regimes, using the composite data set. Insufficient data were available from the INDOEX data set to quantify the magnitude of entrainment from conserved parameters.

Cloud widths, defined here as the horizontal extents of the regions where the C130 encountered LWCs in excess of 0.01 g m^{-3} , using the high-rate (10 Hz) microphysical data, are shown in Figure 8 for each CN regime. Also shown in the figure are the widths of the liquid water cores, taken to be the horizontal extents of the regions where $\text{LWC} > 0.1 \text{ g m}^{-3}$. The clouds sampled during INDOEX, and the liquid water cores,

Figure 8

were small horizontally, although this is not surprising based on visual observations of the clouds penetrated (see Plates 1 and 3). There were no substantial differences between the cloud widths, the widths of the cores, and the vertical velocities in any of the regimes; hence there probably were no significant differences in the extents of entrainment in clouds in the three regimes.

Frequency distributions of vertical velocities in the liquid water regions of clouds in each of the three regimes are presented in Figure 9. Most of the data points fall within the accuracy of the measurement $\pm 0.5 \text{ m s}^{-1}$. The mean vertical winds $\approx 0.4 \text{ m s}^{-1}$, and the peak vertical winds were $+3$ and -2 m s^{-1} . The median vertical velocities, and the shapes of the frequency distributions, were comparable in all three regimes.

Figure 9

The variations of the microphysical properties in the three regimes and the similarities in the cloud widths and strengths lead to the conclusion that the differences in cloud microphysical properties resulted from differences in the aerosol properties producing the clouds sampled during INDOEX.

5. Discussion

A review of earlier studies of the microphysical properties of marine cumuli, and trade wind cumuli may aid in understanding the observations from INDOEX. Cloud droplet spectra have been measured in maritime cumuli

by *Squires* [1956, 1958a, 1958b], *Squires and Warner* [1957]. These measurements were taken off the eastern and southern coasts of Australia and off the coast of the island of Hawaii. The median droplet concentration was 42 cm^{-3} , with the first and

third quartiles averaging 19 and 66 cm^{-3} , respectively. Small seaward cumuli penetrated within 1 km of the ocean surface had pass-averaged median diameters of 13-20 microns and pass-averaged LWCs of 0.08 to 0.5 g m^{-3} . *Raga and Jonas* (1993) measured the microphysical properties of small cumuli over the sea areas around the British Isles. Mean droplet diameter increased, on average, from 6–8 μm near base to 12–20 μm near top, and penetration-averaged concentrations from 20–140 cm^{-3} . In the clean SH stratocumulus clouds studied by *Boers et al.* [1998], cloud droplet concentrations varied from 50 to 180 cm^{-3} , and an important role for droplet coalescence in reducing the number of large cloud droplets and the production of drizzle was inferred from the data. *Squires* [1958a, 1958b] noted that, in general, the continental cumuli had about 7 times higher concentrations of droplets than those found in the maritime cumuli.

The results from the INDOEX clean regime are quite close to the values found in these earlier observations. In the low-CN regime, the median LWC was 0.145 g cm^{-3} ; the median concentration N equaled 89 cm^{-3} ; and the median droplet effective radius was 7.9 μm . However, the observations from the INDOEX high-CN regime differ markedly from results of these earlier studies. Although LWC was approximately the same, N was 316 cm^{-3} and the median effective radius was 5.8 μm . The clouds sampled in the high-CN regime were trade cumuli, but their properties were similar to those found for continental cumuli in these earlier studies.

Insight into the microphysical processes operative in the INDOEX clouds can be gained by an examination of the cloud droplet size distributions. In Figure 10, the size distributions normalized by binwidth are partitioned according to the CN regime

Figure 10

and LWC. The size distributions are truncated at $0.01 \text{ cm}^{-3}/\mu\text{m}$ where the sampling statistics become poor. The following physical processes can be ascertained from the size distributions.

1. For each CN regime, the width of the droplet spectrum, the median concentration, and the median diameter each increase with increasing LWC.

2. The differences in the droplet concentrations between the various CN regimes are much larger when partitioned by LWC than those found in the combined data set for all LWC (Figure 5). Likewise, the differences in median diameter are much larger.

3. Droplet concentrations in the high-CN, highest-LWC category approaches the concentrations found in continental cumulus clouds.

4. The median diameter can be expected to increase with increasing LWC. However, droplet concentrations depend on the strength of the cloud-base vertical velocity, not the LWC, for a given CCN spectrum [Twomey, 1959].

5. Lognormal type size distributions were fitted to the data, with the functional form and fit coefficients identified in Table 2. The fit coefficients included the intercept coefficient N_0 , and the standard deviation and mean of the logarithm, σ and d_{mean} , respectively. The N_0 coefficients increase with LWC and CN in a predictable manner. The σ do not vary much with either LWC or CN. Modal diameters from the fits agree quite closely with the mean diameters in Figure 10. The lognormal distribution can be used to derive the size distribution for a given LWC. One would go to a given LWC and CN range in Table 2, and adjust N_0 upward or downward to produce the desired LWC.

Table 2

Inspection of the INDOEX data set indicates that LWC and (local) vertical velocity are related: the average LWC is 0.125 g m^{-3} for vertical velocities that fall within the range 0 to 1 m s^{-1} ; 0.274 g m^{-3} for vertical velocities from 1 to 2 m s^{-1} ; and 0.83 g m^{-3} for vertical velocities from 2 to 10 m s^{-1} . Although the extent of entrainment, the magnitude of the local vertical velocity, and the LWC are probably related, insufficient data are available to evaluate these relationships.

Aircraft-and ground-based studies have shown that droplet concentrations and concentrations of CN or CCN in nonprecipitating maritime clouds are related. *Hudson* [1983] showed a positive correlation in these concentrations in a study conducted west of San Diego, *Hegg et al.* [1991] and *Vong and Covert* [1998] showed a similar correlation near the Washington coast, and *Yum et al.* [1998] found such a relationship in ACE 1 project data from nearly adiabatic cores south of Tasmania. *Martin et al.* [1994] reported a one-to-one relationship between aerosol ($0.05 \mu\text{m} < D_p < 3 \mu\text{m}$) and droplet concentrations in clean maritime continental air masses from several aircraft studies over the Pacific and Atlantic oceans. However, the proportionality was much smaller in polluted continental air masses that were also over the sea, consistent with our findings.

The INDOEX data set has been examined to ascertain whether there is a relationship between CN and droplet concentrations and to learn more about the droplet activation process in the INDOEX clouds. Frequency distributions of the ratios of FSSP to CN concentrations are partitioned into several vertical velocity ranges in Figure 11. It is not surprising that a great deal of scatter is evident, in part resulting from the effects of entrainment of subsaturated air from near the cloud boundaries; the effect

Figure 11

of drizzle on the ratios is minimal (see the discussion at the end of section 3). The frequency distributions are considerably broader for the low-CN region than for the intermediate-CN and high-CN regions at all vertical velocities. For the highest vertical velocities, a high proportion of the CN in the low-CN regime produced cloud droplets. However, in the high-CN regime, a limiting ratio of approximately 0.4 was reached. The issue is partially one of activation; the activated droplets remove the excess water vapor and reduce the supersaturations relative to what might be achieved with the same vertical velocity if there were fewer CCN. The droplet/aerosol ratio probably depends on aerosol type, with less hygroscopic particles found in the polluted than clean air. In any event, higher supersaturations, with larger proportions of droplets to CN, are likely to occur in association with deep convection.

Calculations of droplet growth in a one-dimensional (1-D) model based on the droplet growth parameterization developed by *Feingold and Heymsfield* [1992] are used to learn more about the microphysical properties of the INDOEX clouds. The purpose of these calculations is to provide insight into how the numbers and sizes of the droplets vary according to CN regime and not to replicate the observations. The model assumes a moist adiabatic lapse rate, and does not derive the acceleration of a parcel due to latent heat release. While entrainment could have been considered in the model by reducing the LWC, the effect on the droplet concentrations, and hence size, would have involved unsupportable assumptions about how mixing affects droplet size distributions.

The model uses as input a CCN-supersaturation S spectrum of the form $N = cS^k$, where c and k are constants, to predict the number of droplets activated as a function of

the cloud base temperature, vertical velocity, and pressure, then calculates the evolution of the droplet size distribution as a function of the LWC. Droplets grow through diffusion, which is not a limitation for the INDOOX clouds since drizzle, signifying appreciable growth through coalescence, was rarely observed. The effective radius, mean mass-weighted diameter, and extinction coefficient are derived using a lognormal type droplet size distribution. The cloud base temperature in the calculations is specified to be 21.5°C and cloud base height of 660 m based on the penetrations through cloud base (*McFarquhar and Heymsfield*, [this issue]). Cloud thickness is taken to be 300 m based on an examination of lidar imagery from INDOEX. Two sets of c, k coefficients were used to approximate the activation of droplets: one set for the clean regime, $c = 200$, $k = 0.6$; and the other for polluted regimes, $c = 1000$, $k = 0.7$ (P. Chuang, NCAR, and J. Hudson, Desert Research Institute, private communications, 2000).

The calculated LWC and various calculated properties of the particle size distribution are compared over a wide range of constant vertical velocities in Figure 12. The peak LWC through the 300 m depth of 0.68 g m^{-3} (Figure 12a) is about 4 times the observed median LWC and about twice the LWC at the peak of the frequency distribution shown from the synthesized data set in Figure 4. Since the height above cloud base for the various C130 penetrations is not known, this comparison does not necessarily imply entrainment from the observations. The calculated concentrations are consistent with the observed concentrations (Figure 12) for the observed median vertical velocities of approximately 40 cm s^{-1} shown in Figure 9, although this may be a fortuitous consequence of the choice of CCN spectra. The droplet concentrations

Figure 12

between the low and high CN regimes vary dramatically (Figure 12b). Because the low CN regime is characterized by CN concentrations below 500 cm^{-3} and more reliably below 300 cm^{-3} (Appendix A), a relatively large percentage of the CN become droplets. Conversely, for the high CN ($> 1500 \text{ cm}^{-3}$) regime, a low percentage of the CN become droplets. While supersaturation is intentionally not plotted in Figure 12, a maximum supersaturation of 1.0%, which activates droplet concentrations of 200 cm^{-3} in the clean regime, requires a vertical velocity of only 260 cm s^{-1} , whereas a similar supersaturation in the polluted regime requires a vertical velocity above 500 cm s^{-1} . Therefore the concentrations in the high CN regime are limited by the strength of the cloud base vertical velocities.

Calculated properties of the particle size distributions differed substantially for the low-CN and high-CN regimes. The effective radii and the median mass diameters were much smaller in the high-CN than low-CN regime, and this difference increased with increasing vertical velocity (Figures 12c and 12d). Note that the effective radii at an updraft velocity of 50 cm s^{-1} were larger than the observed median values (Figure 6) both in the low and high CN regimes at most heights, presumably as a result of entrainment. Also note that the higher vertical velocities produce smaller mean diameters. This effect is opposite to that surmised from Figure 10, which showed that increases in the LWC were associated with increases in the droplet size. Entrainment, or some contribution to droplet growth by coalescence not considered in the model, could account for this difference. Extinction coefficients (Figure 12e) were appreciably higher in the high-CN regime than in the low-CN regime, and this difference widens

with vertical velocity. The net effect is that the high-CN regime could have had almost a factor of 2 larger optical depth than the low-CN regime (Figure 12f) for a given vertical velocity.

The higher extinction coefficients in the modeled high CN regime converts to higher albedos. Albedos for the model clouds were derived using the broadband radiative transfer model described by *McFarquhar and Heymsfield*, [this issue], along with the modeled droplet size distributions used to obtain the results in Figure 12. At 50 cm s^{-1} the albedo increases from 0.47 for the low CN regime to 0.54 for the high CN regime. At 100 cm s^{-1} the albedos are 0.49 and 0.56 for the low- and high- CN regimes, respectively. These results show that the clouds in the high-CN regime should have larger albedos than in the low-CN regime, all else being the same. While entrainment will lower the extinction coefficients and albedos, it is unclear how the differences in albedos between the low and high regimes would be impacted.

6. Summary and Conclusions

In this study, we have examined the microphysical data obtained in more than 13,000 clouds during 151 min. of sampling of primarily trade wind cumuli over the Indian Ocean by the NCAR C130 during the intensive field phase of INDOEX. By partitioning the data according to local CN concentrations, with less than 500 cm^{-3} denoting the clean regimes; between 500 and 1500 cm^{-3} denoting the intermediate regimes; and more than 1500 cm^{-3} denoting the polluted regimes, we found large effects of anthropogenic emissions on cloud microphysical properties. The largest effect

was on cloud droplet concentrations: 3 times larger concentrations were found in the polluted versus the clean regime; and 4 times larger concentrations were found in the regions with large LWCs. We were fortunate to find that cloud bulk properties (cloud widths, vertical velocities, LWCs) were approximately the same in each regime, thereby making a comparison between the various CN regimes meaningful. Calculations of droplet growth in a 1 D model capture the differences in the microphysical properties between the low-CN and high-CN regimes and demonstrate that clouds in the high-CN regime could have had up to a factor of 2 larger optical depth for a given LWC. From the observations and calculations, we can conclude that anthropogenically produced pollutants appreciably modify cloud droplet size distributions, with corresponding effects on the extinction coefficient and optical depth, in clouds over the Indian Ocean. With stronger cloud base updrafts associated with deep convection, an even larger difference would be found because most of the CN already produce droplets in the low-CN regime, whereas, at most, 40% of the CN produce droplets in the high-CN regime.

Cloud droplet effective radius was, on average, 37% larger in the low-CN than in the high-CN regime, and the median diameter was 58% larger when the higher LWCs were considered. Drizzle is infrequently found in any of the INDOEX clouds: 8% of the time in the low-CN regime and only 2% of the time in the high CN-regime. Thus, because drizzle is infrequent in clouds in both the low-CN and high-CN regimes, the second Twomey effect, that increasing the lifetimes of clouds that form in high CN regimes are increased [Albrecht 1989], was not a major factor for the INDOEX trade wind cumuli.

Entrainment may have had an important effect on the microphysical properties of

the clouds sampled during INDOEX, especially since the average cloud width (where $LWC > 0.01 \text{ g m}^{-3}$), and the width of the liquid water cores (where $LWC > 0.10 \text{ g m}^{-3}$), was 200 m or less on average for any of the regimes. The correlation noted between increases in LWC, median diameter, and number concentration in Figure 10 may be a manifestation of entrainment, although the flight patterns used to collect the data were not designed to ascertain the effects of entrainment. A better understanding of the microphysical properties of the clouds that form over the Indian Ocean, and their associated radiative properties and effects of anthropogenic emissions, can be gained in future experiments through efforts to characterize the extent of entrainment.

INDOEX was conducted in the months of February and March, during the northeast monsoon when trade wind cumuli partially cover the high CN regions off the coast of India. As the ITCZ moves northward in April and May, deep convective clouds are likely to ingest aerosols from the high-CN regions. We speculate that even larger effects on the cloud microphysical and radiative properties may occur in convective clouds that form off the coast of India: greater concentrations of droplets; mitigation of the drizzle and rain production process [*Rosenfeld, 1999*]; mitigation of the ice production process via the Hallett-Mossop effect [*Hallett and Mossop, 1974*], which is thought to require the presence of droplets $>22 \mu\text{m}$ diameter; the homogeneous nucleation of large numbers of unfrozen droplets that are transported through the -37°C level in deep convection [*Rosenfeld and Woodley, 2000*]; and, consequently, reduced precipitation and anvils with potentially enhanced brightness. Further insight into this possible range of effects can be gained through measurements in deep convection during the late northeast monsoon

and into the summer monsoon.

Appendix A: Delineation of Clean, Intermediate, and Polluted Regions From CN and CCN Concentrations

A1. Marine Boundary Layer

The background levels of condensation nuclei and cloud condensation nuclei concentrations can be estimated from a number of studies in clean maritime regions, and these estimates provide some means of distinguishing clean from polluted regions. Background aerosol levels are those that have natural origins. *Hudson et al.* [1998] examined the vertical distributions of CN and CCN concentrations over the summertime southern ocean from aircraft measurements during ACE 1. These data should be representative of clean maritime conditions. In the boundary layer, CN concentrations in the clean air several hundred kilometers south of Tasmania were approximately 300 cm^{-3} ; CCN concentrations active at 0.2% supersaturation were in the range 50–70 cm^{-3} and at 1.2% supersaturation were from 180–220 cm^{-3} . These concentrations increased to slightly above 50 cm^{-3} at 0.2% supersaturation and 200–300 cm^{-3} at 1.2% supersaturation, evidently as a result of a small amount of pollution from Tasmania. *Bigg and Turvey* [1978] measured particle concentrations from ground level to 6 km over more than 100,000 km of flight path in the Australian region. The median CN concentration was 220 cm^{-3} in the boundary layer over the maritime air masses, roughly comparable to observations by *Hudson et al.* [1998] in the clean marine boundary layer

in the Southern Ocean.

A2. Clean Continental Boundary Layer

The area downwind of India over the Indian Ocean may have elevated background CN and CCN levels resulting from aerosols transported from the continent, either from desiccation or denudation of soil or from natural fires. The reason for considering CN concentrations in clean continental regions was that nonanthropogenic CN (e.g., dust) from the Indian subcontinent could have potentially produced high CN readings and incorrect demarcations between clean and polluted regions. The data from *Bigg and Turvey* [1978] for the unpolluted regions of the continental boundary layer in Australia should be useful for assessing natural concentrations of CN for clean land-based surfaces. Median concentrations were in the range 350—524 cm^{-3} in clean regions. When both clean regions and regions suspected of being mildly polluted were combined, the median CN increased to 680 cm^{-3} . Higher concentrations of CN were measured in the vicinity of naturally produced forest fires. Measurements of CCN concentrations over New South Wales, Australia, over a 5 year period by *Twomey et al.* [1978] provide some indication of background CCN levels over continental regions; CCN concentrations were of the order of 250—300 cm^{-3} when wind direction was from the south and southeast, both relatively unpolluted areas.

A3. Summary

Typical CN and CCN concentrations in places where sources from populated land masses are likely to have little effect are as follows:

1. remote oceans: CN: 300 cm^{-3} ; CCN at 1% supersaturation: 200 cm^{-3}
2. continental surface: $< 700 \text{ cm}^{-3}$; CCN at 1% supersaturation: $200\text{--}300 \text{ cm}^{-3}$.

Acknowledgments. We are grateful to Darrel Baumgardner, Sean Webb, Cynthia Twohy, Paul Willis, Beverly Armstrong, and members of the NCAR Research Aviation Facility, especially Ron Ruth, for their help during the course of this study. This research was supported by the Center for Clouds, Chemistry, and Climate (C4) at the Scripps Institute of Oceanography and by the National Science Foundation.

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Received July 12, 2000; revised October 20, 2000; accepted November 21, 2000.

Figure 1. Microphysical measurements from gradient flight 3, February 20, 1999. (top): Concentrations of condensation nuclei from the TSI 3760 CN counter, cloud droplets from the FSSP-100, and a smoothed plot of the latitude. Asterisks * denotes regions of drizzle (diameters larger than $55 \mu\text{m}$). (middle) Liquid water content from the King probe, and vertical velocity. (bottom) Median mass-weighted diameter.

Figure 2. Microphysical measurements from gradient flight 4, February 24, 1999. Data in each panel are the same as for Figure 1.

Figure 3. Microphysical measurements from gradient flight 8, March 4, 1999. Data in each panel are the same as for Figure 1.

Plate 1. Photos from the NCAR C130 aircraft during gradient flight 3 on February 20, 1999. (top) Latitude 0.1°N , altitude equal to 665 m. (bottom) Latitude 4.9°S , altitude equal to 6522 m.

Plate 2. Plot of cloud droplet concentration as a function of size in several ranges of latitude during gradient flight 3 on February, 20 1999. The concentration per size bin is color-coded, using the scale on the right side of the figure.

Plate 3. Photos from the C130 aircraft during gradient flight 8, March 4, 1999. (top) The polluted NH regime. (bottom) The clean SH regime.

Plate 4. Imagery from the SABL looking up from approximately 100 m above sea level at two times on March 4, 1999. The top panel is from the polluted regime; the lower panel is from the clean regime.

Figure 4. Frequency distributions of cloud liquid water content ($\text{LWC} \geq 0.01 \text{ g m}^{-3}$) from 10 Hz measurements from the King probe. The data are segregated into three regions. (top) Relatively clean regime, CN concentrations $< 500 \text{ cm}^{-3}$. (middle) CN concentrations are between 500 and 1500 cm^{-3} . (bottom) Polluted regime, CN concentrations $> 1500 \text{ cm}^{-3}$. Within each panel, median LWC (g m^{-3}) is shown.

Figure 5. Frequency distribution of cloud droplet concentrations from 10 Hz measurements from the FSSP, segregated into three regimes (see Figure 4 caption).

Figure 6. Frequency distributions of cloud droplet effective radius from the FSSP, segregated into three regimes (see Figure 4 caption).

Figure 7. Frequency distributions of drizzle from 10 Hz measurements from the 260X probe, segregated into three regimes (see Fig. 9 caption).

Figure 8. Frequency distributions of cloud widths where the 10 Hz FSSP LWC is $> 0.01 \text{ g m}^{-3}$ (top panel), and widths of regions where LWC is $> 0.1 \text{ g m}^{-3}$ (bottom panel).

Figure 9. Frequency distributions of vertical velocities where the LWC exceeds 0.01 g m^{-3} in each of the three aerosol regimes. Median values of the vertical velocities are shown in each panel.

Plate 5. Plot of cloud droplet concentration as a function of size in several ranges of latitude during gradient flight 8, March 4, 1999. The concentration per size bin is color-coded, using the scale on the right side of the figure.

Figure 10. Average cloud droplet size distributions, partitioned by CN concentrations and LWC. The left, center, and right columns represent the distributions for CN concentrations in the three aerosol regimes. The rows top to bottom represent the distributions for LWC over the indicated range. Within each panel the median droplet concentration (cm^{-3}) and diameter μm , are shown.

Figure 11. Ratio of FSSP to CN concentrations, subdivided according to CN regimes (columns) and the vertical velocities (rows).

Figure 12. Growth of cloud droplets in low and high CN regimes associated with INDOEX clouds using the model of *Feingold and Heymsfield* [1992]. Different CCN-supersaturation relationships are used for low- and high-CN regimes (see text). (a) Height versus LWC; the LWC is essentially independent of vertical velocity. (b) Concentration of activated droplets as a function of vertical velocity. (c) Dependence of r_e , (d) Median-mass diameter, and (e) Extinction coefficient on height above cloud base for vertical velocities shown. (f) Optical depth as a function of vertical velocity.

Table 1. Flight Summary

Flight	Date	Time In-Cloud,					Rating ^a	Comments
		<2°S	2°S—2°N	2°S—6°N	6°S—10°N	>10°N		
1	Feb. 16, 1999	0	0	15	66.8	0	—	CAC ^b ,
2	Feb. 18, 1999	0	0	5.5	68.8	0	—	CAC,
3	Feb. 20, 1999	170.3	181.0	403.9	0	0	10	gradient flight
4	Feb. 24, 1999	647.6	286.9	81.8	0	0	10	gradient flight
5	Feb. 25, 1999	0	0	20.6	81.6	0	—	CAC, some p
	Feb. 27, 1999	0	0	3.8	299.2	0	7	polluted clouds
7	Feb. 28, 1999	0	125.2	213.8	0	0	—	
8	Mar. 4, 1999	489.8	132.7	342.0	0	0	10	gradient
9	Mar. 7, 1999	0	0	11.8	0.7	0	8	moderately p
10	Mar. 9, 1999	0	0	7.5	30.9	0	—	
11	Mar. 11, 1999	601.4	0	55.3	0	0	9	gradient flight
12	Mar. 13, 1999	0	0	82.7	614.9	0	6	polluted
13	Mar. 16, 1999	0	0	8.8	276.8	0	7	pollution, few p
14	Mar. 18, 1999	0	295.8	2125.0	0	0	7	Cb o
15	Mar. 19, 1999	0	0	0	256.8	0	7	moderately p
16	Mar. 21, 1999	0	0	33.2	194.1	0	8	p

Table 1. (continued)

Flight	Date	Time In-Cloud,					Rating ^a	Comments
		<2°S	2°S—2°N	2°S—6°N	6°S—10°N	>10°N		
17	Mar. 24, 1999	291.6	192.5	34.5	0	0	9	gradient flight few polluted clouds
18	Mar. 25, 1999	0	0	316.5	23.2	0	5	polluted

^aQualitative assessment of the data collected during the flight: duration of the in-cloud measurements, and whether both polluted and/or clean clouds were sampled.

^bClear air closure flight

Table 2. Droplet Size Distribution Fits

LWC Range, g m ⁻³	CN Range, cm ⁻³	N_0 , cm ⁻³	σ	D_{mode} , μm
0.00 – 0.05	< 500	0.20	0.37	9.89
	500 < 1500	0.35	0.35	7.64
	> 1500	0.60	0.40	6.51
0.05 – 0.15	< 500	0.28	0.30	11.91
	500 < 1500	0.42	0.32	8.36
	> 1500	0.41	0.37	7.35
0.15 – 0.25	< 500	0.19	0.29	13.27
	500 < 1500	0.36	0.36	9.61
	> 1500	0.10	0.40	8.41
0.25 – 0.35	< 500	0.07	0.27	15.17
	500 < 1500	0.06	0.21	11.58
	> 1500	0.06	0.44	9.67

$$\frac{dN}{dD} = \frac{N_0}{\sqrt{2\pi} \sigma D} e^{-\left(\frac{[\ln D - \ln(D_{\text{mode}})]^2}{2\sigma^2}\right)}$$