Vertical Profiles of CCN at Various Geographical Locations

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ABSTRACT

Measurements of the number density, volatility and supersaturation spectra of CCN during aircraft soundings to about 15,000 ft MSL over Arizona, Central Pacific, Alaska, and in a convective region over Florida, and on five flights to an altitude of 35,000 ft off the east and west coasts of the United States are presented. The results show that generally over continental areas away from strong anthropogenic sources the number of CCN is about ten times as great as in marine air masses and as found over snow-packed polar regions. Over continental areas the count decreases with altitude; whereas in the oceanic environments and polar regions the count sometimes increases above the inversion such that at higher altitudes there are no systematic differences between oceanic and continental environments. On several occasions, over the Pacific, anomalously high counts of several thousand CCN cm^{-3} have been encountered. Five soundings to 35,000 ft indicate no systematic increase or decrease in CCN with altitude above 20,000 ft. Above the inversion about 60% of the total aerosol population are CCN active at a supersaturation of 0.7%, the nuclei having an average radius $<2.6\times10^{-6}$ cm.

1. Introduction

The geographical distribution, source and physiochemical properties of cloud condensation nuclei (CCN) are of great importance in the study of natural clouds and fogs. Measurements of Dinger *et al.* (1970) and Twomey (1971a) have shown that the great majority of CCN are not "salt" particles or other involatile crustal matter, but rather consist of a volatile substance. Measurements over wide areas of the earth's surface have shown that the worldwide background concentration of CCN must be produced in the atmosphere itself and not primarily at the surface over the oceans (Dinger *et al.*, 1970; Twomey, 1971a; Hobbs, 1971; Twomey and Wojciechowski, 1969). Centers of population and industrialization may be local sources of CCN but probably do not play an important role in worldwide production of CCN at present (Squires, 1966; Twomey, 1971a; Kocmond and Mack, 1972; Hobbs and Radke, 1970; Ruskin, 1971; Fitzgerald, 1973). Aircraft measurements of the vertical distribution of CCN over the North Atlantic a few hundred miles southeast of Puerto Rico by Dinger *et al.* (1970) show that "sea-salt" nuclei (distinguished by their low volatility) comprise only about one-half the total CCN count at sea level and that a negligible number of sea salt nuclei penetrate above the trade wind inversion. Above the inversion essentially all nuclei are more volatile than sea-salt nuclei. An increase in the CCN count was observed just above the inversion. This increase was interpreted as evidence of the formation of CCN within the atmosphere by gas-to-particle conversion processes.

This paper presents a summary of the results of CCN measurements during aircraft soundings to about 15,000 ft MSL over Arizona, Central Pacific, Alaska, and in a convective region over Florida, and on five flights to an altitude of 35,000 ft off the east and west coasts of the United States. Representative values of CCN counts at different supersaturations over wide areas of the globe taken on transit flights are also presented. On all flights the volatility of the nuclei was measured.

2. Instrumentation

The CCN counts were taken with a thermal gradient diffusion cloud chamber (TGDCC). The equipment provided a means of passing the incoming air simultaneously through five parallel quartz tubes heated to different temperatures and then into holding chambers. In the bottom of each chamber is a metalized mylar diaphragm, the underside of which can be exposed to cabin pressure by opening a valve at the end of the sampling period. This compression is not adiabatic but occurs at a rate controlled by a metering valve. The sample can thus be brought to cabin pressure and temperature before entering the TGDCC. The air from the six holding chambers (one unheated sample and five samples previously heated) can then be sampled sequentially. The CCN count of the unheated sample was made at several values of supersaturation in the TGDCC ranging between 0.2 and 1.2%. The CCN count in the five heated samples was usually confined to a single value of supersatura-

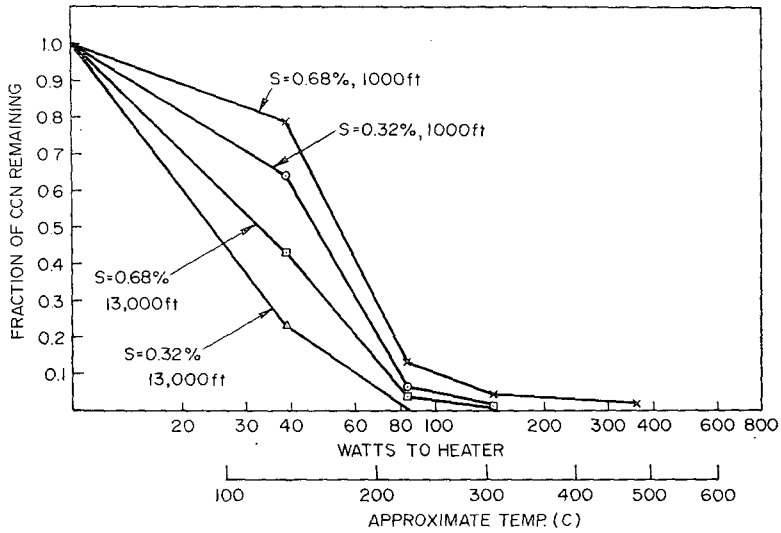

FIG. 1. Fraction of CCN which are not destroyed by heating to various temperatures at 0.32 and 0.68% supersaturations and at altitudes of 1000 and 13,000 ft MSL.

tion around 0.7%. The CCN instrumentation is described by Dinger *et al.* (1970).

Measurements of Aitken condensation nuclei (CN) (cummulative number of nuclei active up to 300%) were made with an Environment One Corporation CN monitor after the air sample had been brought to cabin pressure. These measurements (which are affected by changes in cabin pressure) have been corrected for cabin pressure changes at the various altitudes. Because of the small size of CN the diffusion loss was quite large and variable, depending upon the initial size distribution of the nuclei present. Diffusion loss in the conditioning chamber and inlet tube (exclusive of loss in the aircraft air inlet and manifold) was measured in the laboratory and a correction applied to the data. The CN count at higher altitudes and in unpolluted regions is usually very low and often below the sensitivity specified for the Environment One CN monitor. For these reasons all of the CN counts taken with the Environment One instrument are subject to considerable error and are certainly not reliable when the count falls below 500 cm^{-3}.

In addition to the nucleus measuring capability, air and dewpoint temperatures were measured with a vortex thermometer and dewpoint hygrometer. Concentrations of Rn 222 (3.8 days half-life) were obtained by measuring the radioactivity of radon daughter products collected on a filter. Radon 222 is a natural radioactive rare gas which emanates from the ground after its formation by the decay of radium 226. The radon measurements are described elsewhere (Larson and Hoppel, 1973) but will be referred to in this paper when the results of the random measurements can help to interpret the atmospheric dynamics. Most aircraft measurements discussed herein were made from a Naval Research Laboratory EC-121 aircraft which has an altitude ceiling of about 15,000 ft MSL. On one occasion it was possible to obtain five soundings with a Navy P-3 aircraft which has an altitude capability of about 35,000 ft MSL. The installation on the P-3 did not include the measurements of radon and dewpoint.

After a sample of air is admitted to the TGDCC (at the temperature of the warm top plate), the count is recorded on both video tape (30 frames per second) and on several frames of photographic film. The maximum number of CCN visible within the illuminated area of the TGDCC occurs a few seconds after closure of the inlet valve. This maximum count is assumed to be the total CCN count at the given supersaturation. The time after closure of the valve until this maximum occurs varies somewhat between different samples. It is customary to assume that the maximum count is the cumulative number of all CCN active below the given supersaturation. This latter assumption may not always be valid. Occasionally we have found evidence that some of the more active nuclei would grow and fall out before other less active nuclei were activated. For example, a bimodal distribution of sizes could result in an apparent bimodal supersaturation curve if large particles were activated at low supersaturations and fell out at higher supersaturations before the smaller CCN grow to visible sizes. Evidence of this type of spectra occurring on rare occasions in the atmosphere is presented later. If the CCN are more uniformly distributed with respect to their nucleating activity, then the error in the measured supersaturation spectrum may not be apparent but may still be significant.

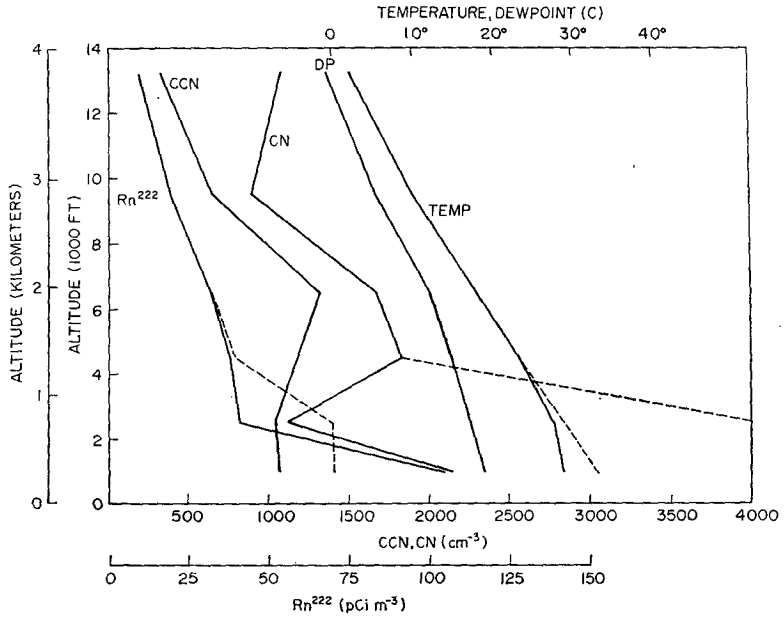

FIG. 2. Average values of CCN, CN, Rn 222, temperature and dewpoint from four daytime soundings over southwest Arizona. Dotted line denotes values during descent.

The results of volatility measurements and their meaning have been discussed by Twomey (1968, 1971a, b) and Dinger *et al.* (1970). As the nuclei traverse the heated tubes they evaporate. If they evaporate beyond some critical radius while traversing the tube, the nuclei will not act as CCN at a given supersaturation. The final size depends not only on the volatility but also on the original size. The interpretation of the volatility spectrum can further be complicated by mixed nuclei consisting partly of volatile and partly of nonvolatile substances. A coagulating aerosol can appear as an aerosol of decreasing volatility as is evident from the results of aging an aerosol formed by ultraviolet radiation (Hoppel and Dinger, 1973). For these reasons the volatility measurements cannot be used as a definitive measure of composition but can be used to distinguish between aerosols whose difference in volatility is great, as has been done by Twomey (1968, 1971a), Dinger *et al.* (1970) and Rosen (1971). Examples of normalized heating curves are shown in Fig. 1. The curves are the averages taken at altitudes of 1000 and 13,000 ft at supersaturations of 0.32 and 0.68% over southwest Arizona. The CCN of Fig. 1 are obviously much more volatile than sea-salt nuclei (see Dinger *et al.*, 1970), or other nonvolatile crustal material (but beyond that it is not possible to distinguish a definite composition). Note that at each altitude, heating inactivates a larger fraction of the CCN at 0.32% than at 0.68% supersaturation, and the CCN at higher altitudes are more easily destroyed by heating. This would suggest that on this particular occasion, at the higher altitude, the nuclei were either more volatile or smaller in size. Of course, if the size distribution were known, a more definitive statement regarding the composition of CCN from volatility measurements could be made as discussed by Twomey (1971b).

3. Results of measurements

a. Arizona soundings

In August 1971, seven aircraft soundings were made over the Gila River Valley area of southwest Arizona between Gila Bend and Mohawk. Data were taken on ascent and descent at 1000 ft MSL (500 ft above terrain), 1500, 4500, 6500, 9500 and ~13,000 ft MSL. The highest altitude for each sounding varied from 12,000 to 15,000 ft depending on the height of the haze layer as determined visually. (We were not always successful in getting above the haze layer.) About 15 min were required at each altitude to allow adequate time for flushing the holding chambers and for data aquisition. At the lowest and highest altitudes approximately 40 min were utilized in order that CCN counts could be taken at several supersaturations, whereas at intermediate altitudes the TGDCC was kept at a constant supersaturation of 0.35%. It should be noted that this value of supersaturation used on the Arizona series of flights is lower than the usual value of about 0.7% used on other NRL field trips. This, of course, results in a lower CCN count; the magnitude of this difference can be judged by the supersaturation spectra given in Fig. 11 by lines 4 and 5.

A total of seven soundings was taken. Four of the seven were taken at approximately the same time of

day beginning at 0700 and ending at 1100 local time (MST) of 24, 25, 28 and 29 August. Since these four flights had some basic similarities the measured variables have been averaged and plotted in Fig. 2. Three of the four flights showed CCN maximum in the altitude range between 4500 and 6500 ft. A high Aitken nucleus (CN) count at 1000 and 2500 ft on the descent on 28 August has weighted the lower altitude values on the graph considerably higher than the value found on the other three days. When the average values on descent were significantly different from those on ascent, the ascent is given by the solid line and the descent by the dotted line. More typical of the CN count (excluding the 28 August profile) would be 2500 and 2000 cm^{-3} at 1000 and 2500 ft MSL, respectively.

The patterns of local atmospheric circulations were extremely hard to decipher at the time of these measurements because the general circulation in the region was very weak. Mesoscale circulations probably resulted primarily from diurnal solar heating on the distant mountain ranges. Prior to our first sounding on 24 August there had been heavy rainfall as evidenced by large puddled areas, some of which were still evident on 25 August. At the beginning of the soundings, conditions were extremely stable on all four flights; and radon had accumulated during the night (Fig. 2). Upon descent near midday, considerable turbulence was usually experienced and upward dispersal of radon which had earlier accumulated near the surface was observed between time of ascent and descent. The CN count on three of the four days was considerably greater at the lowest altitude at the time of descent, indicating some upward mixing of Aitken nuclei during the day by convection.

As is evident from Fig. 1, nearly all CCN found on these soundings would be classified as volatile. One sounding was made in the afternoon of 26 August when there was considerable upward mixing as evidenced by numerous dust devils, some of which extended up to about 1000 ft. Even on this day the number of nonvolatile CCN found at 1000 ft was below the level of detection; however, the Aitken nucleus count rose to about 7000 cm^{-3} at the 1000–2500 ft level. The total CCN count remained below 2000 cm^{-3}. From these results we would suspect that even though crustal materials were visibly being dispersed into the atmosphere in the form of isolated dust devils, the overall number density of nonvolatile crustal particles acting as CCN was not sufficient to alter significantly the CCN count.

The number of CCN as a function of supersaturation is shown in Fig. 11 by curves 4 and 5. The number of nuclei at the 1000-ft level was greater and the supersaturation spectrum was much flatter than at 13,000 ft. This, taken together with the fact that the CCN were more readily destroyed upon heating at the 13,000-ft level (Fig. 1), suggests that the CCN at the higher level were smaller and of the same or greater volatility. The other alternative, that they were as large (or larger) but more volatile and at the same time less active, seems improbable.

b. Alaska soundings

Four soundings were taken over the Yukon Valley approximately 100 mi north of Fairbanks, on 15, 20,

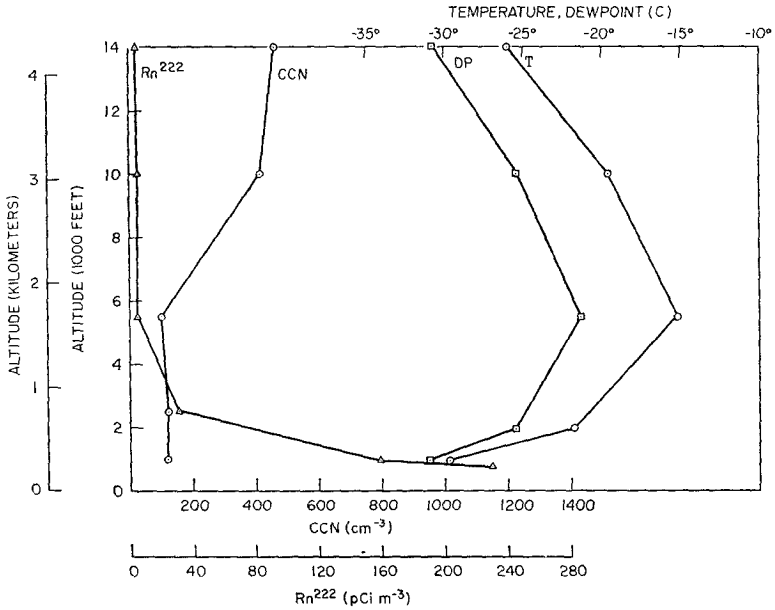

FIG. 3. Average values of CCN, CN, Rn 222, temperature and dewpoint from four daytime soundings over the Yukon Valley, Alaska.

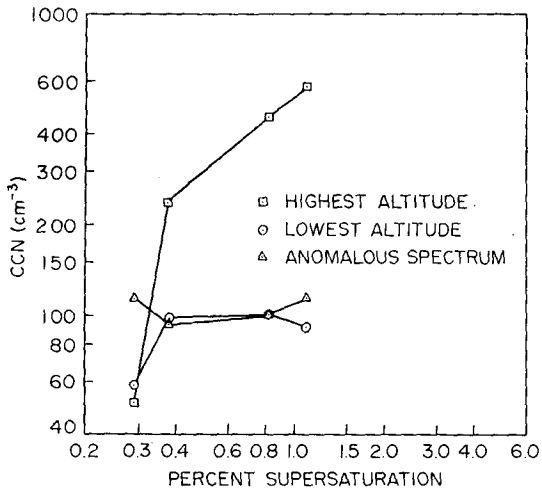

FIG. 4. Average supersaturation spectra taken over the Yukon Valley, Alaska.

21 and 22 February 1972. Here measurements were made only during ascent at altitudes of 1000 ft MSL (approximately 500 ft above terrain), 2500, 5500, 10,000 and 14,000 ft (on one flight the top altitude was 17,000 instead of 14,000 ft). Again the four profiles varied from day to day but the general characteristics of the four profiles can be obtained from the profile of the average values shown in Fig. 3. The soundings were taken roughly between 1100 and 1400 local time. At this time of year this area is characterized by a strong temperature inversion as shown in Fig. 3. Even though there was considerable snow pack, radon was escaping from the ground and being trapped under this strong inversion. The CCN profile reflects the increase in concentration found at the highest altitudes on three out of four days. No CN counts are shown because they were very low (< 700 cm^{-3}) and thus usually below the reliable sensitivity of our Aitken nucleus counter.

The average supersaturation spectra are shown in Fig. 4 for the highest and lowest altitudes, together with the average of three anomalous spectra obtained on 20 February. The number of CCN at the highest altitude ($\sim$14,000 ft) increased much more rapidly with increasing supersaturation than did the average sample at 1000 ft. In fact, at three altitudes [700 ft MSL (300 above terrain), 2500 and 5500 ft MSL] on 20 February anomalous supersaturation spectra occurred, indicating that there were some extremely active CCN present which were falling out in the TGDCC at the higher supersaturations before other less active nuclei could be activated. This apparent bimodal distribution of nucleating activity casts some doubt on the validity of always interpreting the count in the TGDCC as the cumulative number active below the given supersaturation as discussed earlier. The anomalous supersaturation spectra were investigated further by determining fall rates of the activated CCN in the chamber from the video recording. It was found that the particles nucleated at 0.28, 0.37 and 0.80% supersaturations were falling at an average rate of about 0.5, 0.6 and 0.8 cm sec^{-1}. However, a few nuclei falling at rates of about 1.5 cm sec^{-1} were observed at the 0.80% supersaturation.

The average heating curves at 0.80% supersaturation for the lowest and highest altitudes are shown

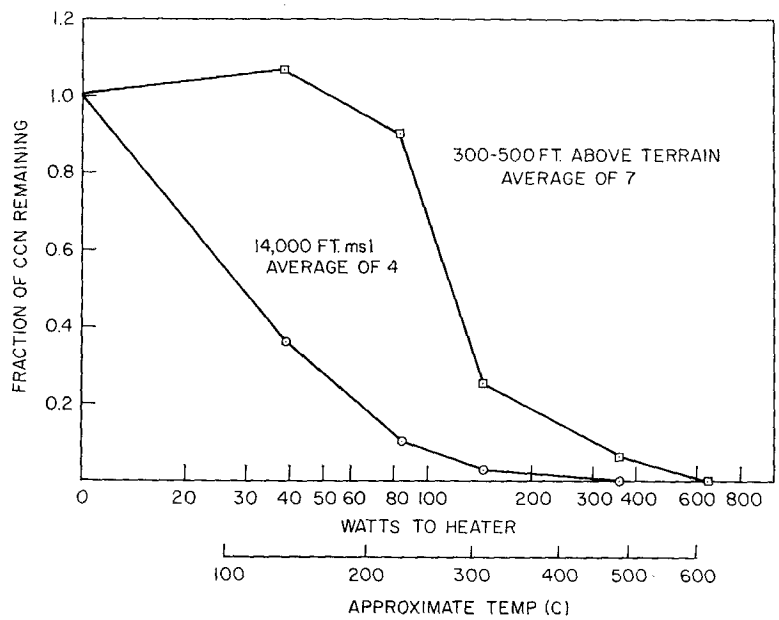

FIG. 5. Average fraction of CCN remaining after various degrees of heating from data taken over Yukon Valley, Alaska.

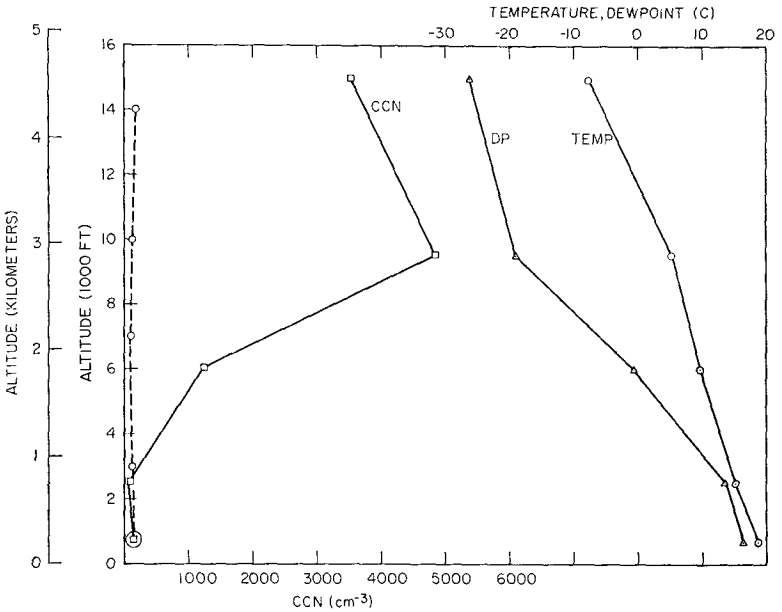

FIG. 6. Average of two anomalous soundings of CCN taken over the Pacific in February 1972. Dotted line gives typical values found over the Pacific.

in Fig. 5. The average heating curve for the highest altitude is not unusual but the average curve for the lowest altitude is unusual. Six of the seven CCN samples showed slight increases after passing through the tube with 39 W applied to it. A possible explanation for this increase may be the partial evaporation of the larger nuclei which had fallen out at 0.8% supersaturation in the unheated sample as discussed in the last paragraph. At the reduced radius (after partial evaporation) the condensational growth and subsequent fallout would be retarded in the sample heated by 39 W. In any case, the nuclei encountered at the lowest altitudes were more difficult to destroy by heating than previously encountered volatile nuclei, but at the same time did not survive passage through the 650 W heated tube and therefore cannot be classified as "salt nuclei."

c. Pacific soundings

Since 1965 CCN data on various flights over the Pacific Ocean have been taken with the NRL TGDCC. On several occasions it was possible not only to obtain data in transit but also to take a vertical sounding of CCN. The dotted line in Fig. 6 shows the average of three soundings taken in 1966, two of which were off the Island of Tasmania in extremely clear antarctic air; the third was near Wake Island. On both these occasions there were widely scattered fair weather cumulus. Since the altitude steps were not exactly the same on each day, the altitudes plotted are average altitudes. The values on the Wake Island soundings were somewhat higher than the Tasmania profiles. On the Wake Island profile the CCN count jumped from 192 cm^{-3} at 13,000 ft to the anomalously high value of 4300 cm^{-3} at 15,000 ft. This value has not been included in the average profile shown by the dotted line in Fig. 5. After the anomalously high value was obtained, the system was checked for leaks by bypassing the incoming air through a sterile cotton filter which removes all CCN. No leaks were found and the measurement was repeated with atmospheric air. Again a high count was obtained at 15,000 ft. Subsequent measurements after descent to 13,000 ft yielded low values again. The transit flights over the Pacific were at constant altitude but the flight altitude varied from flight to flight within the range of 6,000 to 14,000 ft. On these flights hundreds of CCN counts were taken, yielding values consistent with the general magnitude given by the dotted line. However, on several occasions when transiting at the higher altitudes, anomalously high counts of several thousand CCN per cubic centimeter have been encountered at 0.7% supersaturation. Since no instrumental reason for discounting these readings has been found, we must conclude that upon certain occasions large CCN counts are found over the oceans. These large counts are not associated with any known anthropogenic source or nearby land mass.

On 6 and 7 February 1972 two soundings were made north of the Hawaiian Islands. The average of these two soundings together with the average temperature and dewpoint is given in Fig. 6. Below the trade wind inversion (below 3000 ft) the relative humidity is high and the number of CCN is low. Above the inversion anomalously high readings were encountered. When the high count was encountered

on 6 February at 9500 ft, the system was checked for leaks by admitting an air sample through the sterile cotton filter and a zero count was recorded in the TGDCC, thus indicating no instrumental problems. When outside air was again admitted directly to the chamber a high CCN count was encountered. The pilots were careful to maneuver in an upwind direction such that there was no danger of contaminating the air sample with the aircraft's own exhaust; and no other aircraft was operating in the vicinity. The results found on 6 February were so unusual that a second (previously unscheduled) sounding was requested for the next day; again unusually high counts were encountered above the tradewind inversion.

The surface weather charts showed that the two Hawaii flights were made on the eastern edge of a mid-Pacific subtropical high. Above 14,000 ft there was a strong westerly flow. From the temperature and dewpoint soundings shown in Fig. 6, there was evidently subsidence which caused the air at 10,000 ft to be extremely dry (15% R.H.). On both the 6th and 7th there were fair weather cumulus between 1500 and 5000 ft, and on the sixth there was a broken stratus deck at about 7500–8000 ft.

d. Florida soundings

In Fig. 7 are shown the results of profiles made over Florida in the general area of Miami. In contrast to the other profiles herein presented, these measurements were made when conditions were quite convective. The right-hand curve presents measurements made on two days when the wind was from the north, this wind resulting from circulation around a high pressure area in the Gulf of Mexico. The trajectory of the air was such that it had probably crossed the Gulf States prior to flowing over the Florida peninsula; that is, it had a long path over land areas and could be expected to have nucleus concentrations typical of continental air. At the times the profiles were taken the air was quite convective over the peninsula with cumulus clouds growing to 20,000–30,000 ft. After these two days the wind shifted from north to generally west or south. During this period there was a tropical disturbance in the area. The left-hand curve of Fig. 7 presents measurements made during the time these conditions prevailed. The CCN count at cloud base appears to have been influenced by a mixture of continental and maritime air. The higher degree of mixing by convection associated with the disturbed conditions during this period produced considerably less percentage drop in CCN concentration with altitude than had been the case during the period when the measurements for the profiles of the right-hand curve were made.

Measurements made of nucleus volatility at the various altitudes also show the effect of convective mixing in that the number of nuclei exhibiting low volatility (sea salt) remains approximately constant with altitude ($\sim$50 cm^{-3}).

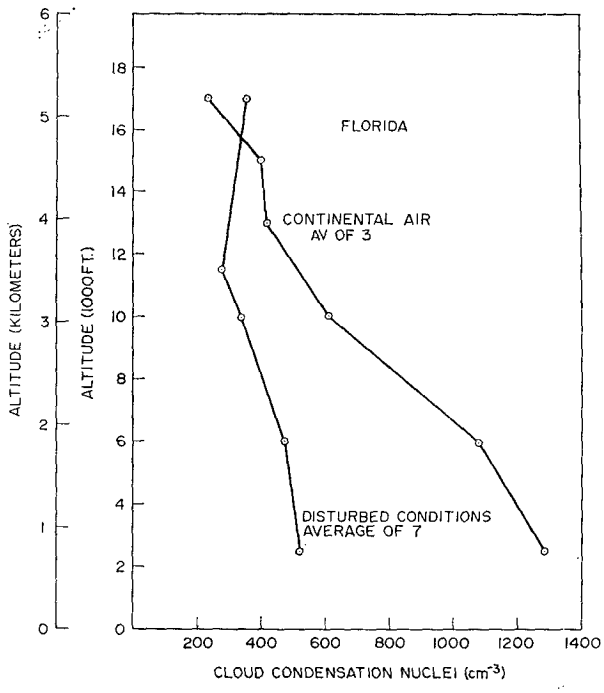

FIG. 7. Average values of CCN from soundings taken in a continental air mass and during a tropical disturbance over southern Florida.

e. Soundings of CCN to 35,000 ft

Soundings of CCN over oceans and polar regions often revealed an increase in the CCN concentration with altitude above 10,000 ft. This, of course, raises the question as to whether or not the CCN count may increase even further at altitudes greater than 15,000 ft. In June 1972 it was possible to obtain five soundings to about 35,000 ft MSL in a Navy P-3 aircraft. Two soundings (14 and 16 June) were taken about 50–100 mi off the eastern coast of the United States and the other three off the western coast. These soundings are shown in Fig. 8, where the CCN counts have been increased by the ratio of the density of the atmosphere at sea level to the density of the atmosphere in the aircraft, i.e., as if the sample had been compressed to sea-level density. The results of these soundings would indicate that there is no systematic increase or decrease of CCN with altitude above 10,000 ft by which we might characterize these five profiles. However, the failure to find a marked decrease in CCN with altitude, when plotted at constant density as in Fig. 8, would indicate that CCN at the higher altitude did not diffuse upward from the earth's surface but rather are formed in the atmosphere itself. The increase in CCN between 10,000 and 14,000 ft often observed in the earlier flights

appears in Fig. 8 as a general trend of increasing CCN counts between 10,000 to 20,000 ft.

On this series of flights, a parallel plate condenser was installed in the aircraft and some of the incoming air was passed through the condenser and into a conditioning chamber. The voltage across this condenser was chosen such that all particles carrying one or more electronic charges and having radii $< 10^{-4}$ cm were removed by the electric field. Before the air passed through the condenser it was passed through a region of high ion density produced by a Po^{210} ionization source. In this manner the CN were brought to charge equilibrium (if they were not already in charge equilibrium before entering the ionizer). From the ratio of uncharged to total number of nuclei it is possible to determine the average size of the nuclei (Keefe *et al.*, 1959; Metnieks and Pollack, 1961; Hoppel, 1969). The ratio of uncharged to total CCN varied considerably between individual measurements and the statistical errors are such as to make any single reading of questionable value, but the average ratio from 36 measurements at 0.8% supersaturation was 0.78 ± 0.17; likewise, the average value of 17 measurements at 0.28% supersaturation was 0.59 ± 0.25. This corresponds to an average CCN radius somewhere between 0.85×10^{-6} and 2.6×10^{-6} cm for those active at 0.8%, and a radius somewhere between 1.3×10^{-6} and 8.0×10^{-6} cm for those active at 0.28%. These values can be compared to those of Twomey (1972) who found by measuring diffusion loss of CCN in Nuclepore filters that the nuclei active at 0.75% supersaturation were in the size range $1\text{–}2.5\times10^{-6}$ cm radius. The small size found here for CCN would reinforce the conclusion of Twomey (1972) and Fitzgerald (1973) that these nuclei are highly soluble.

The ratio of CCN to the total number of CN was measured in the following manner. The CCN formed in the thermal diffusion chamber were recorded on video tape and then allowed to fall out; immediately following gravitational settling of the CCN, an expansion of the air from the TGDCC into a partial vacuum produced a supersaturation of about 300%. The CN activated at this supersaturation were then recorded on video tape at 30 frames per second. The maximum observable CN count in the chamber occurred about 3 or 4 frames after the expansion when the hydrodynamic motion induced by the expansion subsided. The average ratio of CCN to CN (including all altitudes above 2000 ft) was 0.60 ± 0.20. This value is considerably larger than ordinarily found at ground levels or in polluted areas. The average values of CN found on the five soundings are given by the dotted line in Fig. 8.

The average value of CN found here is somewhat larger but the same order of magnitude as found by Junge *et al.* (1961) with the balloon-borne Aitken nucleus counter at comparable altitudes. In a recent report Remsberg (1972) discusses the errors in CN

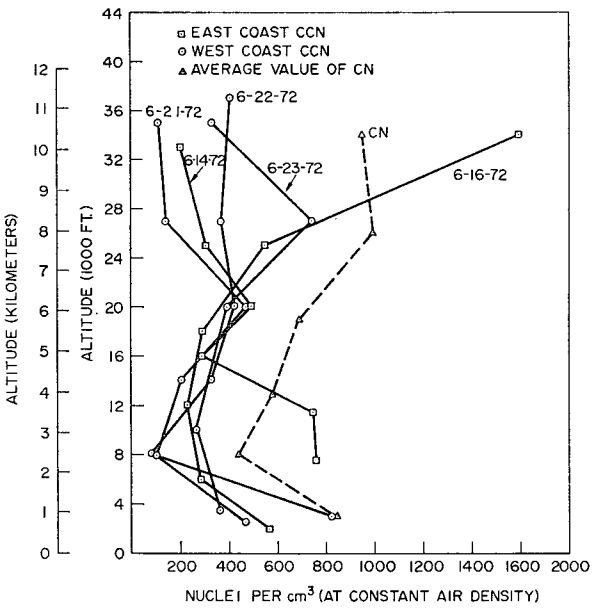

FIG. 8. Profiles of CCN from five soundings to altitudes of about 35,000 ft. Dotted line gives average value of CN during these five soundings.

counting resulting from adiabatic heating during compression. Our air sample is compressed from ambient to cabin pressure as mentioned earlier but this compression in our case is slow and the time required is of the same order of magnitude as the time required for the air in the conditioning chamber to come to thermal equilibrium so that the temperature increase during compression should not be a factor. Volatility curves at the different altitudes and at a supersaturation of 0.7% are shown in Fig. 9. From this figure it can be seen that nuclei which are lost by heating the air sample to temperatures greater than 100C are not being lost in the unheated sample.

The dotted lines in Fig. 9 are the average heating curves taken from the Arizona trip. It is interesting to note that the volatility of CCN at 13,000 ft over Arizona is consistent with the curves found off the east and west coasts. In general, the normalized heating curves of samples taken well above the mixing layer are similar and do not depend critically upon geographic location.

Since the air passes through the heated tubes at ambient pressure, the decrease in the apparent volatility with altitude at higher altitudes could be accounted for by a decrease in the boiling point with decrease in pressure. This has been discussed by Rosen (1971) in interpreting data on the boiling point of stratospheric aerosols. However, the large difference often encountered in the volatility curves below and above the inversion as evidenced in Fig. 5 and the 1000-ft characteristic of Fig. 9 would indicate a change in the physical characteristic (either size or composition) of the aerosol.

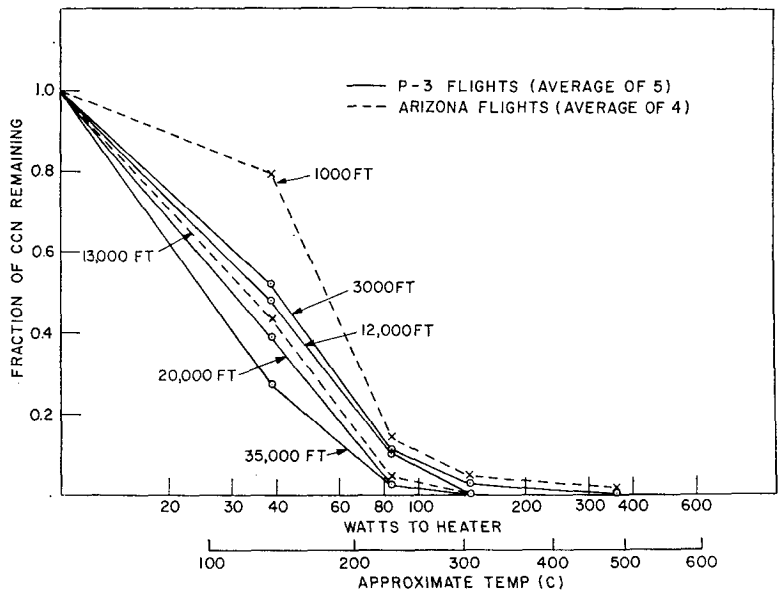

FIG. 9. Average fraction of CCN remaining after heating to various temperatures at different altitudes from the five sounding to 35,000 ft (Fig. 8). Dotted lines give average of Arizona data.

4. Discussion and conclusions

Fig. 10 shows a comparison of the average profiles taken at five different locations. All profiles were taken at about 0.7% supersaturation in the TGDCC except for the Arizona profiles which were taken at about 0.35% supersaturation. From curves (4) and (5) of Fig. 11 it can be seen that an increase by a factor of 1.4 at the lowest altitude and a factor of about 2 at the highest altitude would be necessary to obtain the number of CCN active at 0.7% on the Arizona profile. Several conclusions are immediately evident from Fig. 10:

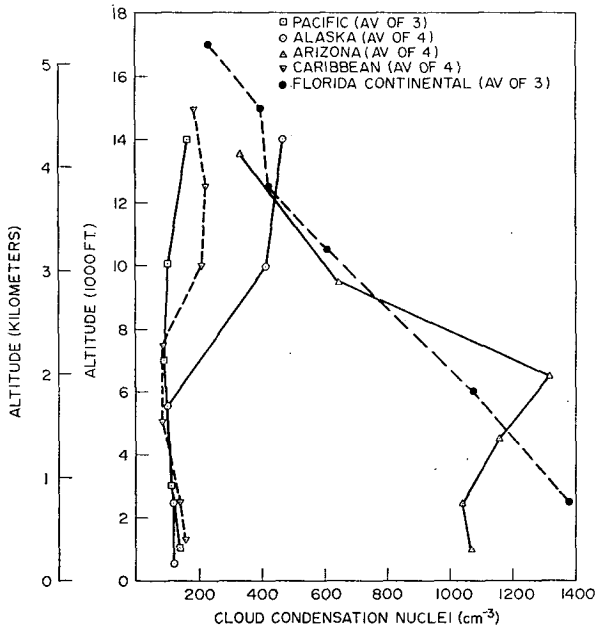

FIG. 10. Comparison of the average soundings of CCN from five different geographical locations.

1) At low altitudes over continental areas away from strong anthropogenic sources the number of CCN is about ten times as great as is found in marine air masses and over snow-packed polar regions.

2) Over continental areas the count decreases with altitude; whereas in oceanic environments and polar regions the count sometimes increases above the inversion such that at higher altitudes there are no systematic differences between oceanic and continental environments.

3) In comparing the continental air measurements over Florida with those taken over Arizona, we see that the convective conditions which prevailed in Florida removed the effects of an inversion on the profile of nucleus concentration and distributed the sea-salt portion nearly uniformly throughout the convective levels.

In terms of a purely diffusive model the continental profiles would indicate an upward movement of CCN with the possibility of CCN being produced at or near the earth's surface. The oceanic and arctic profiles would indicate a downward movement of CCN with a possible source in the upper troposphere (or stratosphere) as suggested by Dinger *et al.* (1970). The oceanic profiles might also be explained in terms of horizontal advection of CCN into a region with washout of CCN by precipitation scavenging at the lower altitudes. At higher altitudes the CCN count

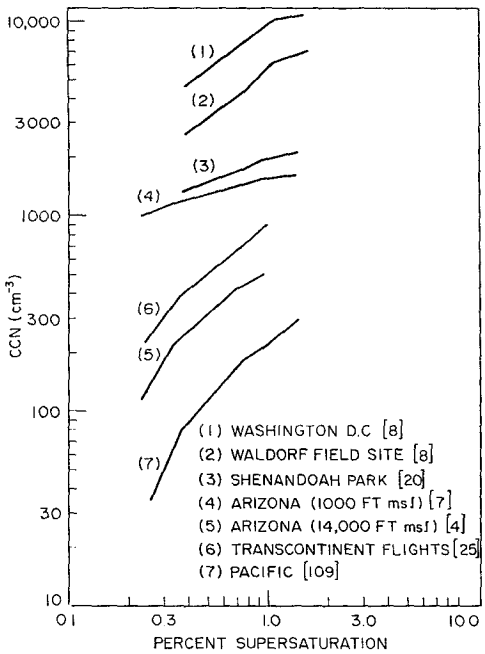

FIG. 11. Average supersaturation spectra from seven different geographical locations.

does not decrease as one might expect if the source of CCN were in the lower troposphere; rather, the five profiles to 35,000 ft indicate no systematic increase or decrease with altitude. The data obtained here would fit the speculation of Twomey (1971a) that the earth's surface plays an important role in the production of volatile CCN possibly through the release of gases which subsequently react and convert to particulates. In addition to this conversion near the earth's surface there appears to be a weaker source of particles in the atmosphere itself perhaps associated with gas-to-particle reactions.

Fig. 11 shows a comparison between the average supersaturation spectra of CCN at seven different locations. In order that each individual spectrum contribute equally to the shape of the average spectrum, the plotted spectra were calculated by normalizing each spectrum with respect to the number of CCN at 0.7% supersaturation. The average of the normalized spectra was then multiplied by the average number of CCN observed at 0.7% supersaturation to obtain the spectra plotted in Fig. 11. The highest count was found at the Naval Research Laboratory in Washington, D. C., and the lowest over the Pacific Ocean [curve (7)]. The Waldorf Field site (2) is located about 20 mi south of downtown Washington but is still greatly influenced by the pollution from the suburbs. The Shenandoah Park site is located on a mountain ridge at Big Meadows, Va. The numbers in brackets in Fig. 11 give the number of spectra included in the average. The increase of CCN caused by land surfaces and the further increase resulting from urban pollution are obvious from Fig. 11.

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