Balloonborne Measurements of Condensation Nuclei

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ABSTRACT

A condensation nuclei (CN) counter has been developed for balloonborne use at ambient pressures in the troposphere and stratosphere. The instrument employs a thermal gradient diffusion cloud chamber to produce particle growth and a photoelectric particle counter for detection. After extensive laboratory tests, the instrument was successfully flown on several balloon soundings over Laramie. The results show a roughly constant mixing ratio in the stratosphere with a CN concentration of about 20 cm^{-3} at 15 km. The vertical profile of CN in the troposphere displayed concentration fluctuations ranging between 200 and 2000 cm^{-3} with a definite maximum in the mixing ratio just below the tropopause.

1. Introduction

Several aspects of condensation nuclei (CN) are relevant to atmospheric properties and processes. The role that these small particles play in the physics of cloud formation is of considerable interest. Perhaps the area of greatest potential importance is their influence on cloud development (which would affect the albedo of the earth) and rainfall. It is well known that the concentration of CN is greatly enhanced over background levels by human activity, and as a consequence these particles can be used as tracers of urban air masses. This is an extremely sensitive technique because a very negligible atmospheric mass loading can still be responsible for a high CN concentration.

The atmospheric chemistry of gas-to-particle conversion, whether in the troposphere or stratosphere, is obviously intimately related to and influenced by the concentration of CN. Although it may be argued that CN generally occur in concentration and size too small to affect the optical properties of the atmosphere, a measurement of this quantity is often very useful in establishing asymptotic limits for size distributions used in optical models. In addition, under high humidity conditions, small particles may grow and become optically important.

There has also been some concern over the amount and effect of CN injected into the upper troposphere by commercial air traffic and into the stratosphere by rockets.

Recent studies of CN tend to emphasize the type of particle that grows under very low supersaturations with respect to water ($<1\%$) because it is believed that these would be the most active in providing condensation sites in cloud formation, hence the most relevant. Little attention has been given to the vertical profile of total particle content which can be obtained from relatively large supersaturations. However, for reasons outlined above, they are still of considerable importance.

The first successful measurements of CN in the stratosphere were made from balloons by Junge *et al.* (1961) and were repeated by Käselau (1974) using a very similar detector. Other measurements have been made by Podzimek *et al.* (1975) up to 20 km using a high altitude aircraft. All of the above instruments work on the expansion chamber principle utilizing supersaturations in the range of 200–300% with respect to water. The purpose of this paper is to present the results of some recent measurements of the vertical profiles of CN in the troposphere and stratosphere using a different type of detector and compare these results with other available data.

2. Instrumentation

The device used to achieve supersaturation for the work reported here is a vertically oriented, cylindrical thermal gradient diffusion cloud chamber (TGDCC). The operating characteristics of this instrument type have been extensively described by several investigators (Fitzgerald, 1970; Saxena *et al.*, 1970; Saxena and Carstens, 1971; Katz and Kocmond, 1973; Sinnawalla and Alofs, 1973; Katz and Mirable, 1975). A description of the particular TGDCC used here, as well as the results of some preliminary testing, have been presented by Rosen *et al.* (1974). It is noteworthy that for this work a TGDCC operating at relatively high supersaturations over a large ambient pressure range was required. Glycol was chosen for the working fluid (which is responsible for the

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particle growth) rather than water because it will not boil or evaporate quickly at high altitude and will produce final particle sizes that are in a workable range. Other investigators have also used various types of alcohol for the working fluid but not necessarily for the same reasons (Sinclair and Hoopes, 1975; Bricard *et al.*, 1974). It should be noted that the basic operating principles of the TGDCC are essentially independent of the particular working fluid employed.

A schematic diagram of the flight instrument is shown in Fig. 1. An air sample dilution mechanism is used at low altitude because the CN concentration is too large for the instrument to handle directly. Above 15 km no dilution is required and the ambient air is drawn directly into the TGDCC through a ball valve. The outer cold cylindrical surface of the TGDCC is kept at 0°C by a hermetically sealed ice water bath, while the inner cylinder or rod, covered with filter paper saturated in glycol, is kept at a warmer constant temperature by an electronically controlled heater element.

In an ideal TGDCC, sampled air would uniformly fill the volume between the hot and cold surfaces and experience no turbulence as it passes through the device. Under these circumstances the supersaturation experienced by a parcel of air, after it reaches temperature and vapor pressure equilibrium, can be calculated from well-known relationships. The degree to which our TGDCC approximates an ideal device will be discussed later. Particles of a critical size (r_c) and larger will nucleate and grow for a given supersaturation obtained by a transit air parcel. In general it is expected that the higher the supersaturation the smaller the value of r_c. Particles that nucleate and grow to the minimum detectable size (r_m) of the photoelectronic counter will be registered as CN. Thus the criteria for detection is that the particles, through some process, must grow at least to size r_m.

A complicating factor in the operation of the TGDCC is the non-uniform supersaturation that develops between the hot and cold surfaces. A particle passing through the center of the flow region would experience a higher supersaturation than a particle passing near the inner or outer wall. Therefore, it is not possible to associate a particular r_c with given operating parameters. Nevertheless, we have observed that much higher average supersaturations are required to detect small CN than to detect large CN.

In this application of the TGDCC to CN measurements, it is our intention to operate at supersaturations high enough for essentially all particles present in the natural atmosphere to grow to size r_m or larger. Thus the nonuniformity of the supersaturation between the walls of the TGDCC is of little consequence. The method used to determine the required operating parameters for natural atmospheric CN is discussed later.

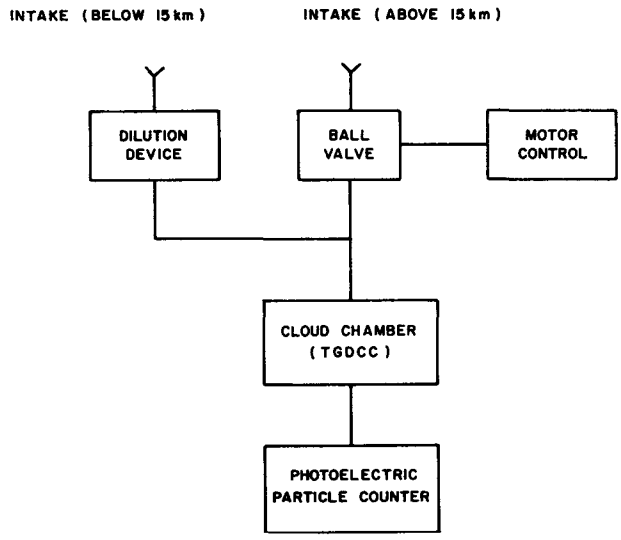

FIG. 1. Flight configuration schematic of the CN counter. Above 15 km the ball valve is open and the sample flows straight into the TGDCC.

The degree to which conditions in the actual TGDCC approach the ideal situation was studied by observing smoke streamlines as they passed through the device with the outer cylindrical wall replaced by a glass tube. In this manner it was possible to observe the flow as a function of pressure and operating parameters. At all altitudes the flow appeared to completely fill the space between the hot and cold walls. At ground level a very slight amount of turbulence was present but as the altitude increased the flow became rigorously laminar. At gound level with the TGDCC filled with smoke and the air sampling pumps off, a convection pattern developed that could be attributed to the difference in temperature of the walls. However, as soon as the pumps were turned on the convection pattern disappeared and the flow was completely dominated by the incoming air. In the high-altitude limit of this experiment, when the pumps were off, the single particles moved radially outward to the cold wall (presumably due to phoretic forces). The time for the TGDCC to be cleared of particles from this effect was considerably longer than the high-altitude transit time of the smoke when the pumps were on.

The time it takes an incoming parcel of air to reach vapor pressure and thermal equilibrium in the TGDCC can be calculated using well-known methods; in our case is found to be approximately 0.6 and 0.3 s, respectively. These time constants are roughly inversely proportional to the ambient pressure so that at high altitude the TGDCC comes into thermal and vapor pressure equilibrium much faster than at ground level. The time constants are to be compared with the measured transit time of about 8 s for altitudes between 0 and 14 km and about 1 s above 14 km (the variation in the transit time is due to a change in

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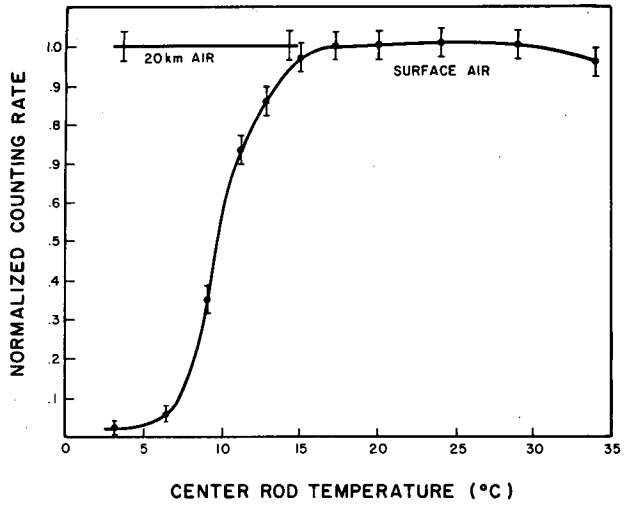

FIG. 2. Typical nuclei saturation curves for atmospheric air. See text for explanation.

pumping rate). Experimentally the point at which an incoming air parcel reaches approximate vapor pressure equilibrium in the TGDCC can be estimated from the distance from the inlet at which glycol vapor begins to condense on the cold surface. Condensation is observed on at least the last three-fourths of the TGDCC. Thus there is good theoretical and experimental evidence that an incoming parcel of air has adequate time to reach thermal and vapor pressure equilibrium before exiting.

These observations suggest that our TGDCC approximates an ideal device reasonably well and that the theoretical calculations of supersaturation can be used as a general guide to actual supersaturations developed.

In order to be sure that all particles passing through the TGDCC grow at least to size r_m it is necessary to investigate the influence of the center rod temperature on the counting rate for the particular CN of interest. As the temperature of the center rod is increased the average supersaturation in the TGDCC increases and smaller and smaller particles will nucleate and grow to a detectable size. Finally, at some point, the supersaturation will become so large throughout most of the volume between the hot and cold surfaces that practically all CN present will grow to a detectable size. A well-defined plateau in the counting rate versus center rod temperature curve could thus be interpreted as evidence that the instrument is counting essentially the total number of CN present. Typical curves of this type are shown in Fig. 2 for surface air and stratospheric air. The high-altitude measurements were obtained on three separate flights with the balloons floating at constant altitude. The center rod temperature was cycled from hot to cold several times during each float period. The TGDCC is usually operated with a center rod temperature of about 15°C which corresponds to a peak theoretical supersaturation with respect to glycol of about 20% or an average value between the surfaces of about 10%.

In the limit of high center rod temperature the particles can grow so large that under the influence of gravitational settling they cannot pass out of the TGDCC and through the particle counter. Thus it is desirable to operate the center rod temperature no higher than necessary since settling becomes an increasing problem at high altitude. The instrument employs a multi-channel size analyzer which is used to determine if the particles ever grow too large for proper detection.

Ideally, curves such as those presented in Fig. 2 should be obtained at frequent altitude intervals and on every flight. However, we feel that the physical nature of atmospheric CN does not change substantially with time and the determination of curves such as those in Fig. 2 on every flight is not strictly necessary. The fact that the curves in Fig. 2 were repeatable on several occasions lends some credibility to this argument.

In order to determine whether or not an average supersaturation of 10% (center rod temperature ~15°C) is sufficient to count essentially all CN present throughout the troposphere a special sounding was made. Two practically identical CN counters were flown on the same balloon: one instrument was operated at an average supersaturation of 10% and the other at an average supersaturations of about 90%. The counting rates of the two instruments were practically identical up to an altitude of about 17 km. Beyond this level the instrument with the larger supersaturation produced a counting rate smaller than the other one. We attribute this effect to the particles growing excessively large due to the higher supersaturation, and not being counted by the detector. These results indicate that a center rod temperature of 15°C is probably sufficiently high for the instrument to count most of the CN present in the troposphere.

In a similar experiment two CN counters were again flown, one with a center rod temperature of 15°C and the other with a center rod temperature of 0.5°C (corresponding to a supersaturation of about 0.02%). Throughout the troposphere the instrument with the lower supersaturation had a considerably lower counting rate than the other but at altitudes between 18 and 25 km the two counting rates were essentially the same. Beyond 25 km there appeared to be some losses in the high-supersaturation unit due to gravitational settling. The results of this experiment strongly suggest that the well-defined plateau in the counting rate versus center rod temperature curve shown in Fig. 2 for 20 km is representative of altitudes from 18 to 25 km.

Thus a center rod temperature of 15°C appears to

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be adequate for proper operation of the instrument between ground level and 25 km. At higher altitudes a reduction in the center rod temperature is required to prevent excessive growth of the particles. However, proper operation on a plateau of the counting rate versus center rod temperature curve can probably still be maintained. Although the minimum supersaturation required to operate on the plateau of the curve is apparantly quite low for stratospheric CN, it is not unreasonable and is probably a reflection on the size of the CN. It should be pointed out that at 0% supersaturation (with respect to glycol) a 0.03 μm radius particle (soluble in glycol) will not nucleate but will grow to a new equilibrium size greater than r_m. Thus the characteristic low supersaturation needed to operate on the plateau of the curve may be explained if most of the stratospheric CN had sizes of about 0.03 μm radius and larger.

It should be noted that this work does not eliminate the possibility of the existence of a class of CN above 18 km that will only nucleate and grow under supersaturations in excess of that used here (20% maximum). However, it is clear from the curves shown in Fig. 2 that such a hypothetical class of CN would be well separated from the types of CN measured in this program.

A significant effort was given to the testing of this method of CN counting at low ambient pressures. The instrument was operated in a vacuum chamber that covered pressure equivalent altitudes ranging from sea level to 30 km. As the air is removed from the chamber the counting rate is expected to be proportional to the pressure and to remain constant as clean air is let back into the chamber. In general it was found that the counting rate decreased somewhat faster than the pressure but held fairly constant when the chamber was repressurized with clean air. This discrepancy was attributed to partially volatile CN and diffusional deposition of CN on the walls of the vacuum chamber during the relatively slow pump down. The final results of the vacuum chamber testing indicated that there may be a slight loss of efficiency above 20 km altitude and an unreliable performance above 27 km if the center rod temperature is in excess of 13°C. However, as indicated above, good repeatable results were hard to achieve due to the difficulty in controlling the nature of the CN and our inability to accurately account for the diffusional losses to the walls. None of the data presented here have been corrected for the possible drop in efficiency above 20 km since it appears to be a relatively small correction of somewhat doubtful magnitude. The unreliable performance of the instrument above 27 km at center rod operating temperatures above 13°C has been attributed to counting efficiency problems arising from excess particle growth.

In the overall view we feel that the type of instrument used in this work shows considerable promise but there still remain some unanswered questions concerning its operation over the entire altitude range considered here. The technique has several advantages over other methods. The instrument can operate throughout large ambient pressure ranges and inherently has good sensitivity at the low CN concentrations occurring in the stratosphere. It is small, light-weight (~7 kg) and the data can be telemetered to a ground receiving station so that instrument recovery is not necessary. Since individual particles are counted, no calibration in principle is required to relate the output of the device to the ambient CN concentration. In practice, uncertainties associated with the pumping rate, temperature corrections to the volume flow rate, efficiency losses due to gravitational settling, and the dilution factor in the troposphere could possibly contribute to a total uncertainty in the absolute concentration of as much as a factor of 2. However, at the present time even a factor of 2 uncertainty is relatively unimportant for most considerations. The absolute concentration as obtained with our instrument was compared with that obtained from a Nolan-Pollack counter as well as several other counters for surface-level air (Cadle *et al.*, 1975). Although there appeared to be almost exact agreement with the Nolan-Pollack counter, experimental uncertainties limited the accuracy of the agreement to about 30%. The repeatability of the instrument can be assessed by comparing the ascent and descent profiles. This will be discussed in a later section.

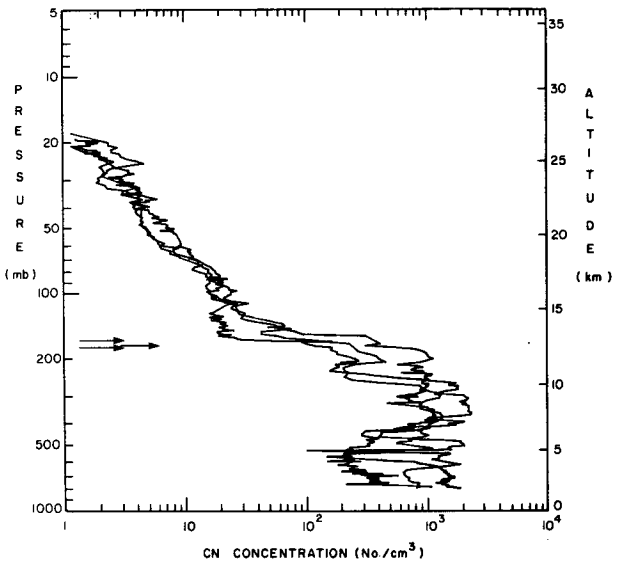

FIG. 3. A comparison of three typical CN soundings made over Laramie. The flights were made on 27 and 30 May 1974 and 24 May 1975. The arrows mark the height of the tropopause.

3. Results

The CN profiles reported here are the results of several soundings over Laramie covering the time

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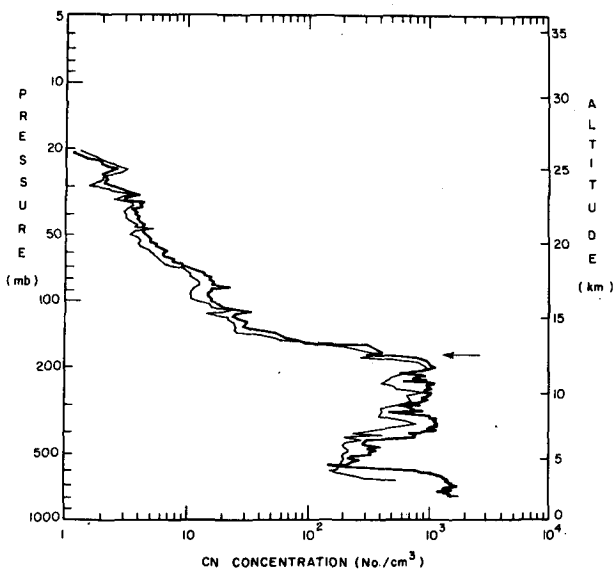

FIG. 4. A comparison of CN concentration on ascent and descent. The sounding was made over Laramie on 27 May 1974 and the arrow marks the height of the tropopause.

period December 1973 to May 1975 and are representative of soundings made through September 1976. A composite chart of three typical profiles is presented in Fig. 3 and is useful for comparing the consistency as well as the variation of the measurements. Examples of comparisons between ascent and descent profiles can be found in Figs. 4 and 5. Also shown in Fig. 5 is the concentration profile of atmospheric particles having diameter >0.30 μm as measured on the same flight using a separate particle counter identical to that described by Hofmann *et al.* (1975). The increase above the tropopause is the start of the well-known 20 km layer. Note that the CN concentration is about 10 times that of the larger particle concentration in the stratosphere, above 15 km. The CN profile above 15 km is missing on ascent due to a telemetry problem that developed. Fig. 6 is a comparison of the CN and large-particle profile before and after the eruption of the volcano Fuego, in Guatemala, in October 1974. It appears that the large particle concentration increased by about a factor of 10 while any increase in the CN concentration is lost in the natural fluctuations.

4. Discussion

The self-consistency checks of the in-flight data tend to indicate that the instrument produces credible measurements. The agreement between ascent and descent, even in the finestructure, is good and attests to the repeatability of the measurement. The similarity of the CN profiles in the stratosphere from flight to flight at one location (see Fig. 3) attests to the long-term consistency of the technique, since under normal circumstances the stratospheric CN concentration would probably not be expected to change very rapidly.

A comparison of the profiles presented here with those from other studies is shown in Fig. 7. Only the average profile from each work is shown and no attempt was made to illustrate the range of values encountered by the observers. In some cases the range of values at one altitude obtained by a single technique varied by more than a factor of 10. The absolute errors associated with each measurement in Fig. 7 are generally not available. All of the profiles, we feel, show remarkable agreement in the troposphere.

The possible agreement of the profiles, shown in Fig. 7, for the stratosphere is less certain. The results of two very similar techniques (Junge *et al.*, 1961; Käselau, 1974) show a disagreement approaching two orders of magnitude. The agreement between the profile of Käselau and that of Podzimek *et al.* appears quite good in the 15–20 km region. However, the latter profits shows very large fluctuations in this region, whereas the former does not. Thus this agreement may be fortuitous. Although these discrepancies may be attributed to natural sources (time, location, normal fluctuations), serious consideration must also be given to the reliability of the data and instrument operation at high altitude. Limiting the discussion to recent data, only two profiles above 20 km are available for comparison: that from this work and that from Käselau. The disagreement in the CN concentrations between these two is about a factor of 3 to 8, which could be considered fairly reasonable in view of the difference in techniques and particularly the super-

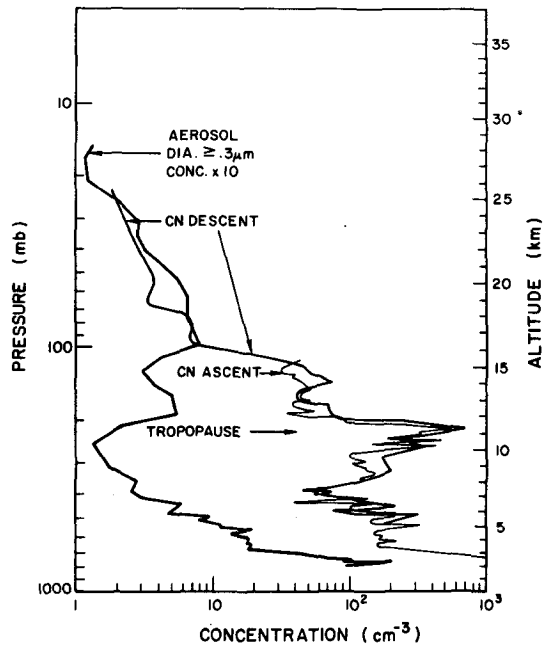

FIG. 5. A comparison of the CN profile and the profile of particles with diameters greater than 0.3 μm. The data were obtained during a single flight on 19 December 1973. Note that the large particle concentration has been multiplied by a factor of 10 before being plotted on this chart. The arrows mark the height of the tropopause.

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saturation. However, the implications derived from the shape of the two profiles are not easily reconcilable. The profile of Käselau indicates a CN mixing ratio increasing with altitude, implying a source above 25 km, while this work indicates a constant or decreasing mixing ratio above the tropopause which may be more consistent with a tropospheric source. Obviously, it will be necessary to determine the correct source of stratospheric CN before even a first step in their understanding can be made. For this reason no profiles of CN derived from theoretical models will be presented here.

The volcanic eruption of Fuego during October 1974 does not seem to have affected the CN concentration significantly. This is in contrast to the changes seen in the concentration of larger stratospheric particles (Fig. 6). By virtue of the technique, the larger particles are included in the measurement of CN concentration and it may, therefore, be argued that there must also be some increase in the CN concentration as presented in the figures. However, since the large particles are not as numerous, the corresponding percentage change in the CN profile is small and may be lost in the natural fluctuations. This could be the case for the data shown in Fig. 6. Another explanation of these results is that particles were not injected directly, but that SO_2 was injected into the stratosphere by the volcano. Subsequently, this gas was converted to H_2SO_4 and condensed on the preexisting particles causing them to grow but conserving the total particle concentration. Both explanations seem plausible at the present time and perhaps the only definite conclusion that can be drawn is that the volcanic eruption did not appear to significantly increase the average stratospheric CN concentration.

All direct measurements of CN in the stratosphere are consistent on one important point: there are relatively few CN. This fact has far-reaching implications concerning the effective lower limit of the size distribution and consequently for optical models of stratospheric aerosols. For instance, a model requiring large amounts of very small particles to explain a quasi-Rayleigh scattering function would not be consistent with any direct measurement. The first attempt to develop an optical model of stratospheric aerosols based on a size distribution consistent with the observed CN concentrations has recently been presented by Pinnick *et al.* (1976).

A knowledge of the CN concentration also has an important application in the interpretation of total mass measurements as obtained by filter samples. A relatively low CN level, for instance, indicates that most of the mass is in larger particles.

The results shown in Fig. 7 indicate that the CN concentration remains approximately constant for nonpolluted air over an altitude range from a few kilometers above the ground to the region of the tropopause. The only measurement not consistent with this statement is that of Cadle and Langer. The discrepancy here could be attributed to the low supersaturation with respect to water (~2%) that was used which may not be sufficient to nucleate all of the particles present.

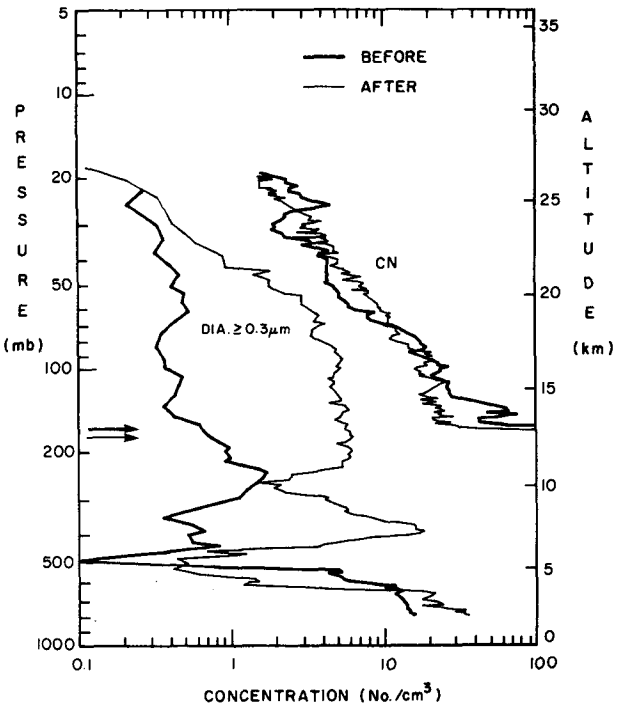

FIG. 6. A comparison of the CN and large particle (>0.3 μm diameter) profiles before (May 1974) and after (May 1975) the eruption of the volcano Fuego in October 1974. The arrows mark the height of the tropopause.

The average CN mixing ratio implied from Fig. 7, therefore, has a maximum around the region of the tropopause. This characteristic profile is in direct contrast with the concentration profile of particles whose diameters are greater than 0.3 μm and would seem to require a source of CN in the upper troposphere. Attempts have been made to attribute this source to commercial jet air traffic (Rosen *et al.*, 1974) but a measurement of the jet engine CN emission index will be necessary before a realistic evaluation can be made.

5. Conclusions

There seems to be general agreement that the concentration of CN in the stratosphere is relatively low. The results of the various measurements are also in good agreement in the upper troposphere and indicate a maximum in the CN mixing ratio near the tropopause.

The results of the measurements made in this study indicate that the stratospheric CN concentration profile is relatively constant in time and was not measurably affected by the volcanic eruption in Guatemala during October 1974.

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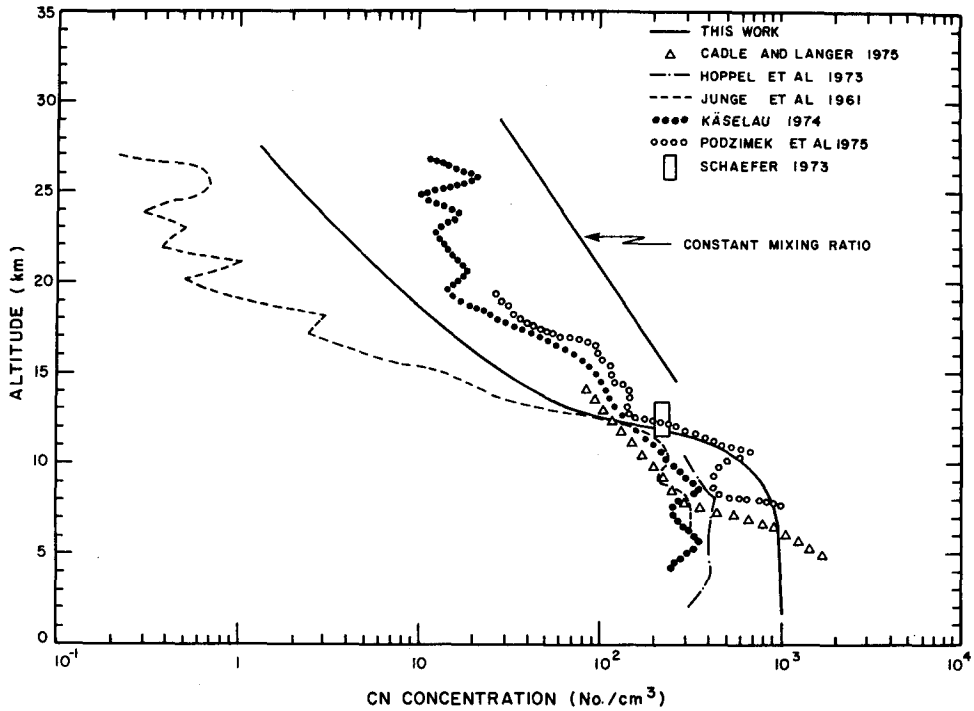

FIG. 7. A comparison of the available CN profiles for the upper troposphere and stratosphere. All data have been averaged and smoothed.

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