Considerations in the Measurement of Pollution Effects on the Number Concentration of Cloud Condensation Nuclei

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ABSTRACT

Pollution effects on cloud condensation nucleus (CCN) concentration vary widely, sometimes even decreasing the CCN. The interpretation of these pollution effects may be invalid if the nuclei are measured by expansion-type Aitken nucleus counters operated at low expansion ratios to provide a readout (with natural nuclei) equal to that of a thermal-gradient diffusion cloud chamber (TGDCC) instrument operated at 1% supersaturation. Calculations indicate that the expansion chamber operated at a ΔP of 1.4 inch Hg produces a supersaturation of 26%, allowing for vapor depletion effects during the first few hundredths of a second after expansion. The maximum supersaturation in clouds is typically 0.1–1%; therefore, 26% causes a large positive error. Further, vapor depletion in the instrument during droplet growth limits the growth rate and size of droplets at supersaturations which are low compared to those for which the instrument is designed. Therefore, use of the standard calibration curve contributes a large negative error in the readout. The combination of a large positive error from excessive supersaturation and a large negative error in calibration appears to explain the systematic errors found at the second International Workshop on Condensation and Ice Nuclei (IWCIN). With NaCl nuclei the expansion instrument undercounted 90% of the time, averaging 0.6 of the average of five TGDCC instruments. On the other hand, with the fairly nonwettable nuclei of Teflon it read 25 times the TGDCC average.

An experimental development model of a continuous mixing-jet type instrument for CCN exhibited the same tendency, reading 0.04–0.33 of the TGDCC average with NaCl and 8 times too high with Teflon. If further development can correct the uncertainties in this NCAR instrument, it promises to be convenient and continuous.

Unless proven differently by future comparison tests which are more definitive than those of the IWCIN, interpretations of data on CCN in pollution should be evaluated in terms of the supersaturation at which the data are taken as compared to that in clouds. Future decisions on instrumentation for measurements of CCN should take into account the results of the IWCIN, particularly for measurements in polluted air where most Aitken nuclei do not act as CCN.

1. Introduction

The effects of air pollution on the increase of cloud condensation nuclei (CCN) have been the subject of conflicting results and conclusions. Some clarification of instrumentation problems contributing to the confusion is possible by considering results from the condensation nucleus section of the second International Workshop on Condensation and Ice Nuclei (IWCIN) summarized by Ruskin and Kocmond (1971). These results raise questions as to whether measurements which show increased CCN concentrations due to pollution effects are such that erroneously high values of the increase may often be reported; and, in fact, whether such errors may occasionally mask concentration decreases. The cause of systematically accentuated error in pollution data from some instruments lies in the fact that they activate nuclei at higher supersaturation than is present in clouds. In polluted air the CCN often constitute less than 1% of the Aitken nuclei, whereas in "natural" conditions the ratio may exceed 10% (Twomey, 1963). Therefore, CCN instruments must be capable of selecting only those nuclei which would be activated at the 0.1–1% supersaturation usually present in clouds.

The condensation nucleus portion of the second IWCIN held in Ft. Collins, Colo., in August 1970 provided probably the best opportunity to date for comparisons of expansion, mixing-jet, and thermal-gradient diffusion cloud chamber (TGDCC) types of instruments while counting a variety of types of both natural and artificial condensation nuclei. In several experiments one of the expansion-type counters was operated at an expansion ratio sufficiently below that normally used that it gave readings for natural nuclei which were equal to those of a TGDCC type instrument operating at 1% supersaturation. When used for counting artificial nuclei, considerable deviation from the thermal-diffusion type instruments was found. A continuous-mixing jet type instrument exhibited slightly less error, but in the same directions—low for NaCl and high for less easily activated nuclei of Teflon.

Calculations indicate that the expansion-type instrument had about 26% supersaturation at an expansion ratio such that its photoelectric readout indicated a

concentration of natural nuclei equal to the count from a diffusion chamber operating at 1% supersaturation.

The following are examples in which the conclusions could have been reversed if the condensation nuclei had been measured by expansion- or mixing-type condensation nucleus counters which are incapable of operation at supersaturations low enough to be representative of clouds:

1) Two common pollutants, fuel oil smoke and automotive exhaust, were found by Twomey (1960) to be unimportant in increasing CCN. When measured at 0.5% supersaturation, the combustion of fuel oil produced CCN counts below the minimum of the instrument; whereas when measured by a Pollak-type instrument, fuel oil gave the highest condensation nucleus counts of any of the combustibles measured. At low supersaturation, auto exhaust fumes were found to contain fewer CCN than the environmental air entering the engine. Hobbs *et al.* (1970) also found that when measured at low supersaturation, "the exhaust from automobiles is a negligible source of CCN. Similarly, oil refineries and oil-fired power plants emit relatively few CCN into the atmosphere."

2) Smoke from a simulated forest fire was shown by Hobbs and Radke (1969) to produce no increase in concentration of those CCN which are active below 0.2% supersaturation.

3) One controversial "natural pollutant," terpene vapor (which forms "blue haze" from forests), was found by Twomey (1960) to reduce slightly the concentration of natural CCN when measured at low supersaturations.

4) From measurements by Squires (1966) at various locations in the United States the anthropogenic production of CCN active at 0.5% supersaturation was estimated to be 14% of the natural CCN production.

5) Another unexpected finding by Twomey and Wojciechowski (1969), considering the greater pollution in the United States, is that the "median-nucleus levels were higher for flights across Australia and Africa than for flights across North America."

2. Calculation of supersaturation

Supersaturation in a TGDCC is produced by the nonlinear relationship between temperature and saturation vapor pressure, combined with the linear gradients of both temperature and vapor pressure between two parallel plates which are at different temperatures and both saturated with water (Twomey, 1963). The degree of supersaturation which develops near the center of the chamber is approximately equal to the ratio of the average of the saturation vapor pressures at the temperatures of the top and bottom plates compared to the saturation vapor pressure at the average of the temperatures of the two plates. These calculated supersaturations using vapor pressure values

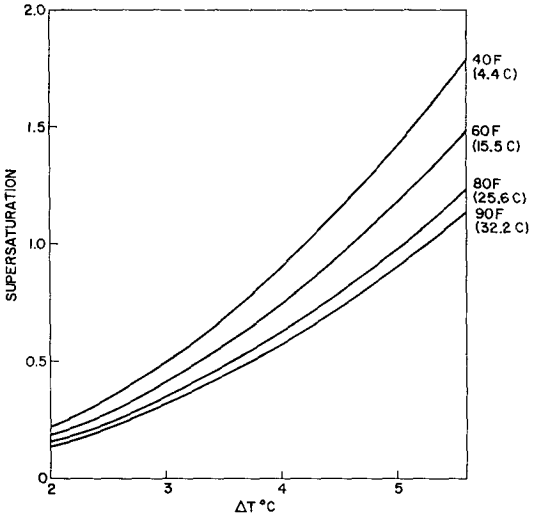

FIG. 1. Thermal-gradient diffusion cloud chamber (TGDCC) theoretical supersaturation (%) vs plate temperature difference (ΔT) plotted for the four warm-plate temperatures labeled on the right.

as tabulated by List (1966) are shown in Fig. 1 for several temperatures of the warmer plate. The same results are shown in Fig. 2 in terms of the temperature difference ΔT between plates, presenting for various warm (top) plate temperatures the value of ΔT needed to reach each of the three supersaturations (0.3, 0.75 and 1%) used at the IWCIN.

For expansion-type instruments the supersaturation is produced throughout a previously saturated air

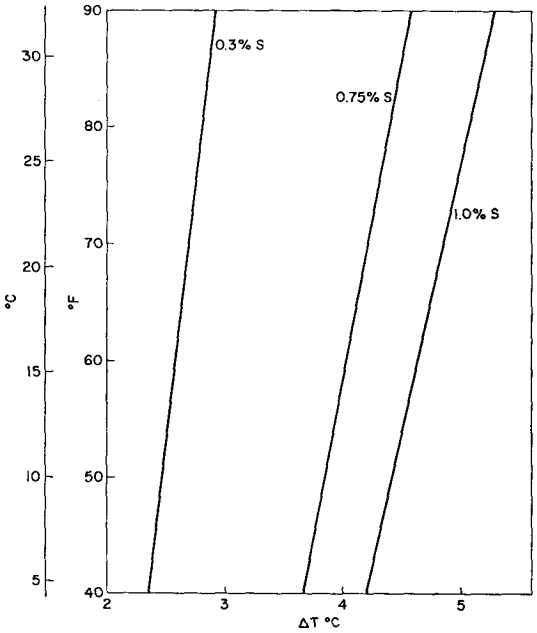

FIG. 2. Variations of diffusion chamber ΔT (abscissa) required to provide a constant supersaturation S in a TGDCC as a function of warm-plate temperature (ordinate). The three supersaturations shown are those used at the second International Workshop on Condensation and Ice Nuclei (IWCIN).

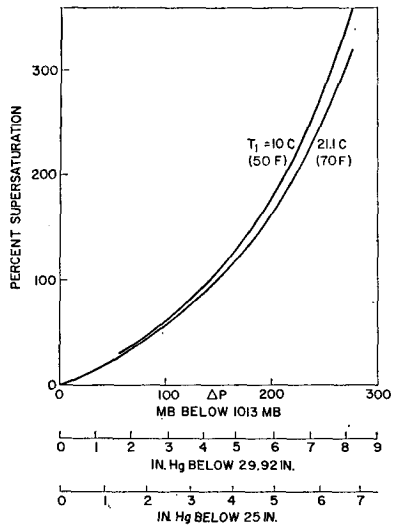

FIG. 3. Expansion cloud chamber supersaturation at two temperatures (T_1) before expansion as calculated from the adiabatic expansion equation with allowance for vapor reduction during expansion, but without corrections for vapor depletion by droplets. Pressure change (ΔP) during expansion is shown on abscissa for both standard sea level pressure and for 25 inches Hg (average value for the IWCIN at Ft. Collins, Colo.).

sample by a temperature drop during an adiabatic expansion. The temperature T_2 after expansion is calculated from

$$T_2=T_1(P_1/P_2)^{(1-\gamma)/\gamma}, \quad (1)$$

where T_1 is the temperature at the original pressure P_1, P_2 the pressure after expansion, and γ the ratio of specific heat of air at constant pressure to that at constant volume. For dry air $\gamma=1.403$. For air saturated at 21C the value of γ is reduced $\sim 0.2\%$, resulting in a 0.4% greater temperature drop during expansion. During expansion the vapor pressure is reduced [from the value for T_1 tabulated by List (1966)] by the ratio P_2/P_1. The ratio of this new vapor pressure to the saturation vapor pressure at T_2 is the saturation ratio. In Fig. 3 the resulting supersaturations [100×(saturation ratio−1)] are plotted as a function of the pressure change ΔP for 10 and 21.1C (50 and 70F) for starting pressures of 29.92 inches Hg (standard sea level) and 25 inches Hg (the nominal atmospheric pressure at Ft. Collins, Colo., at the IWCIN). From these curves the supersaturation is 27.6% at the ΔP of 1.4 inch Hg which was employed at the IWCIN to obtain readings of natural nuclei equal to the CCN concentration as read by a diffusion chamber operating at 1% supersaturation. From these curves it is interesting to note that pressure changes as low as 0.1 inch Hg in a wet pipe or duct can give a supersaturation of 1.5%. Therefore, precautions must be taken to avoid this much pressure drop in the sample duct when the sample flow rate is high while flying in clouds or rain, where moisture may enter the plane's air sampling inlet.

3. Supersaturation losses in expansion chambers

Questions have been raised as to whether the computed supersaturations as plotted in Fig. 3 may be reduced in practice by vapor diffusion to the chamber walls and heat diffusion from them. Along the center line of the long cylinder-type chamber used in the Gardner, Nolan and Pollak counters, this effect is shown by Kassner *et al.* (1968a) to cause a reduction of about 1% of the original supersaturation during the first tenth of a second and 35% by the end of 1 sec. However, they also indicate that the number of nuclei which can be activated is determined by the supersaturation present during the first few hundredths of a second. Thereafter, the supersaturation decrease affects mainly the rate of growth of the droplets formed on the nuclei.

The vapor depletion ratio from this cause is the ratio of the total mass of water in the droplets at any given time to the original vapor density excess over that needed for saturation at T_2. This vapor depletion per 1000 droplets, expressed as percent of the original supersaturation, is plotted as dashed lines in Fig. 4 for three supersaturations.

In the same figure solid lines show the total reduction (as a percent of the original supersaturation) caused by vapor depletion and warming of the air by release of the latent heat of condensation during growth of the droplets. Assuming 1000 droplets cm^{-3}, the temperature rise ΔT from this source is given approximately by

$$\Delta T=\frac{L_v M_L}{C_v \rho_A}, \quad (2)$$

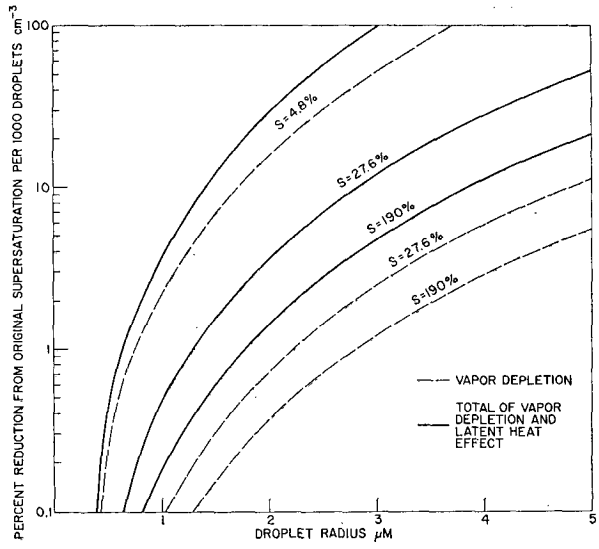

FIG. 4. Bulk vapor depletion and reduction of supersaturation by release of latent heat of condensation for each 1000 droplets cm^{-3} plotted as a function of droplet radius for typical Aitken nucleus instruments: 1) at 190% supersaturation, 2) at 27.6%, a value used in the expansion instrument to simulate TGDCC at the second IWCIN, and 3) at 4.8%, a value which would result from a ΔP of 0.3 inch. Hg (1 inch Hg on the Gardner gage).

where L_v is latent heat of vaporization (or condensation) of water at T_2, M_L the mass of liquid water in 1000 droplets of a given radius, C_v the specific heat of saturated air at constant volume, and ρ_A the density of the saturated air at T_2 and P_2. As in the case of the vapor depletion, ΔT varies approximately in direct proportion to droplet number concentrations for other than 1000 cm^{-3}. Because of slight changes in L_v, C_v and ρ_A with temperature and pressure, this proportionality is only approximate. The resulting change in saturation vapor pressure is not constant with ΔT and therefore causes a greater deviation from linearity. However, for concentrations of 1000 ± 1000 cm^{-3} the errors from scaling the values shown in Fig. 4 are generally less than the uncertainties introduced by inaccuracies of the pressure gage on the Gardner counter at low readings. The latent heat effect is generally 1–4 times the vapor depletion effect.

From these curves it is evident that the vapor depletion and latent heat effects are negligible for 27.6% or higher supersaturations until the droplets have grown to be larger than 2–3 μm radius. At 4 μm radius 1000 droplets cm^{-3} reduce the 27.6% supersaturation to 22.6%. At 4.8% ($\Delta P = 0.3$ inch Hg) 1000 droplets cm^{-3} would condense 100% of the supersaturation vapor if they grow to 3 μm radius. Therefore, the droplets cannot reach this radius if their concentration is 1000 cm^{-3} or more. This limitation points up two of the problems of using an expansion-type instrument at low supersaturations: 1) the droplets cannot grow large enough for the photoelectric detector to provide the signal per droplet for which it was originally calibrated, and 2) combining this growth limitation and the wall diffusion effects discussed above, the slowly growing droplets typical of small supersaturations will not have time to grow appreciably before they begin to reevaporate. This latter problem is avoided in a thermal diffusion chamber by a continual replenishment of the vapor from the warm plate to maintain approximately the original supersaturation throughout the period of droplet growth.

A third contribution to the reduction of supersaturation is by the formation of a "dead space" around each droplet caused by the release of the latent heat of condensation at the droplet boundary and by the depletion of the vapor more rapidly, both within a distance of a few molecular mean-free paths from the droplet. This "dead volume" was investigated by Carstens *et al.* (1966) and found to be generally less than 3% of the bulk volume. It would be mainly within this volume that other nuclei would fail to be activated because of a decreased supersaturation during the first few hundredths of a second.

Combining the four effects of wall diffusion, bulk vapor depletion, temperature rise from latent heat of condensation, and "dead space" around the droplets, the reduction in supersaturation during the nucleus activation period appears to be approximately 6% of the original supersaturation (a reduction from 27.6% to 26%) for a nucleus concentration of 1000 cm^{-3}. Combining the curve for $S = 190\%$ in Fig. 4 and the "dead space" effects, for 10^4 droplets cm^{-3}, the reduction appears to be roughly in agreement with the finding at the IWCIN (Ruskin and Kocmond, 1971) that with nucleus concentrations of 10^4 cm^{-3} the commercial Aitken nucleus instruments read approximately 50% lower than a research-type expansion chamber (Kassner *et al.*, 1968b) in which the samples were diluted with filtered air to provide low droplet concentrations in the instruments for all measurements.

4. Comparisons of condensation-nucleus instruments at the second IWCIN

In addition to the Aitken nucleus comparisons, data were obtained at the IWCIN on five laboratory models of TGDCC instruments, each of a different design from a different laboratory, all operated by experienced personnel from the various laboratories. These five instruments, operated at supersaturations of 0.3, 0.75, and 1%, generally agreed within ±30% for natural nuclei and a factor of 2 for artificial nuclei with a wide range of ease of activation. These included soluble (NaCl) and insoluble, fairly nonwettable (Teflon) types. Individual descriptions of these instruments are included in the *Proceedings* edited by Grant (1971).

Also described in the same *Proceedings* is an experimental development model instrument from the National Center for Atmospheric Research (NCAR) intended for continuous CCN monitoring aboard aircraft. This instrument employed a continuous jet of moist warm air impinging into moist cold air. The resulting supersaturation was calculated by NCAR to be about 2%. The chamber volume was sufficiently large to allow 15–20 sec for the droplets formed on nuclei to grow to 20–30 μm before passing through an acoustic detector. The readout counts were multiplied by 5 to allow for droplets found to collect on the walls during previous laboratory tests. For natural nuclei this instrument varied from 0.07–3.6 times the thermal-diffusion chamber average curves extrapolated to 2% supersaturation, the calculated maximum in the mixing jet. With NaCl the readings were 0.04–0.33 of the diffusion chamber values; and with the less-easily-activated nuclei of Teflon the readings were 8 times higher than the TGDCC. Examples of the NaCl and Teflon data are plotted in Figs. 5 and 6, respectively.

In Figs. 5 and 6 are also plotted the values from measurements made with an Aitken nucleus counter of the expansion (Gardner) type. In this instrument a small chamber is partially evacuated; then the main sampling chamber is expanded into the combined volume of the two chambers. The resultant pressure drop is ~0.29 of the previous ΔP of the smaller volume. This system provides a convenient method of instantaneously ex-

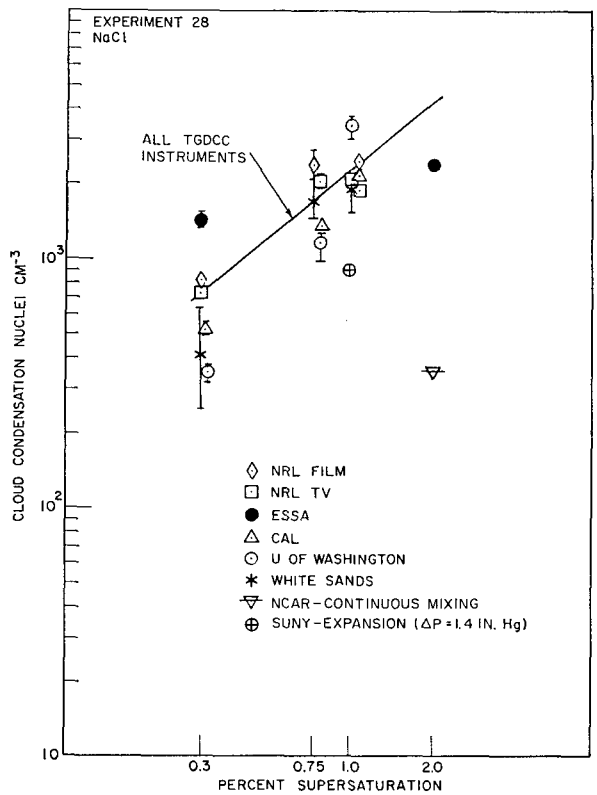

FIG. 5. IWCIN data from one NaCl nucleus experiment. The straight line represents the average of all TGDCC instruments. The readings from the expansion (Gardner) counter operated at a $\Delta P = 1.4$ inch Hg while those from the NCAR mixing-jet CCN instrument are 0.25 and 0.08, respectively, of the average of the TGDCC instruments.

panding by predetermined small amounts. At expansion a cloud is formed with one droplet on each of those condensation nuclei which are activated at whatever supersaturation is present during the first few hundredths of a second after expansion. The optical density of this cloud is measured by the reduction in light transmission to a photoelectric detector. This photoelectric readout is calibrated at the factory by reference to another Aitken-nucleus counting instrument, both operating at 200–300% supersaturation. In addition to its standard application to Aitken nucleus (total aerosol) counting, one of the Gardner instruments was operated at the IWCIN at a small expansion. A value of ΔP (1.4 inch Hg) was found at which the photoelectric readout referred to its original Aitken nucleus calibration curve gave values of nucleus counts equal to those from a TGDCC instrument operated at 1% supersaturation when counting natural nuclei. The natural nucleus data can be seen in Fig. 7 to fall generally between 0.5 and 2 times the TGDCC concentration, only one case being a factor of 6 high.

In 33 readings of NaCl nuclei, 90% of the readings were low, averaging 0.6 of the TGDCC instrument. One of the lower NaCl readings is plotted in each of Figs. 5 and 7.

Artificial nuclei composed of Teflon powder were included in the IWCIN to study whether nonwettable nuclei can act as CCN. The powder apparently included impurities which altered its surface chemistry from that of a Teflon flat surface to such a degree that the results were not definitive for the original purpose. Fortuitously, however, these particles provided an activation spectrum comparable to that of those pollution constituants which are not easily activated; thus, they do not act as CCN at low supersaturations, but are activated in large concentrations at higher supersaturation. Unfortunately, only one Teflon experiment (No. 35) had a sufficiently high concentration to provide reliable data. The data from all the instruments are plotted in Fig. 6 and the comparison between the TGDCC and expansion chamber at $\Delta P = 1.4$ inch Hg is also included in Fig. 7. In this case the Gardner counter at $\Delta P = 1.4$ inch Hg gave a reading 17 times the highest CCN instrument reading at 1% supersaturation and 25 times the average of all TGDCC instruments.

5. Discussion

From Fig. 6 it can be seen that many of the nuclei which are not activated by clouds having a maximum supersaturation less than 0.75% (most continental and

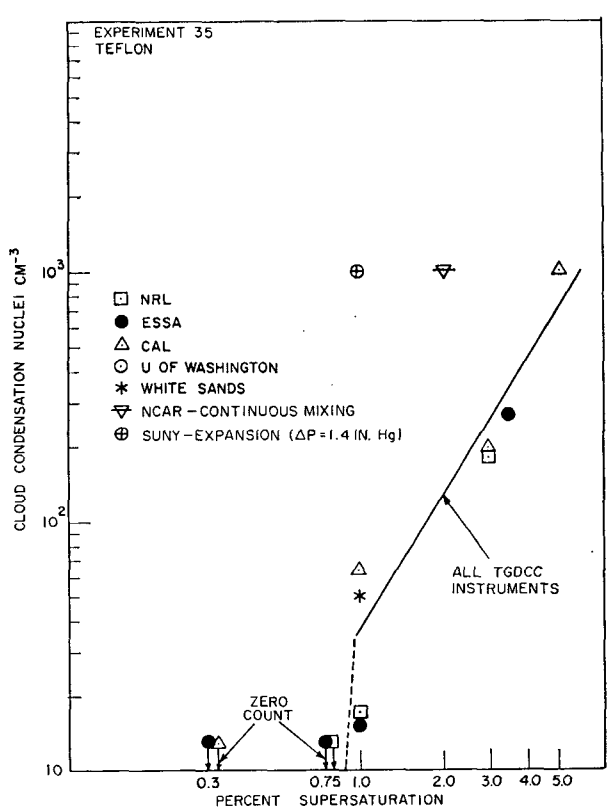

FIG. 6. IWCIN data from the Teflon nucleus experiment. The Gardner counter at same ΔP as in Fig. 5 gave a reading 25 times the average of the TGDCC instruments; the NCAR instrument read 8 times the TGDCC average.

small maritime clouds) may still appear to be CCN if measured by an expansion or mixing-jet type instrument which subjects the nuclei to excessive supersaturation. Even in cases where the concentration of CCN is decreased (e.g., in some auto exhaust or by terpenes), it may appear to increase by a large factor if measured with these instruments. Considering the large positive error with fairly nonwettable nuclei and the negative error with NaCl shown in Fig. 7, it appears that each of these errors results from a combination of two errors which can compensate at only one ratio of CCN to total Aitken nuclei. One explanation which fits the data and calculation of expansion chamber supersaturations is as follows. When either the mixing jet or expansion instrument is "calibrated" (with natural nuclei) against a TGDCC instrument operating at 1% supersaturation, the readings from the expansion or mixing-jet instruments are affected by a combination of a large positive error because of the greater number of droplets actually formed (in excessive supersaturation) and a negative error in the readout. With NaCl a lower reading will be indicated than for natural nuclei if the negative readout error is constant and the positive error is reduced by virtue of the concentration of NaCl nuclei being nearly the same whether measured at a low or high supersaturation (less slope in the spectrum). The opposite effect applies for Teflon with its steeper slope (Fig. 6). An explanation for the NaCl error being less for the expansion than for the mixing jet is that the readout error for the NCAR acoustic detector is probably nearly constant for various activities of nuclei (assuming a constant fraction lost to the walls); whereas for the expansion instrument the photoelectric readout must give an indication of higher counts for larger droplet diameters (from faster growth rates). In observing the cloud droplets growing in the Naval Research Laboratory TGDCC it is apparent from the video screen that the growth is considerably faster with NaCl than with natural CCN or Teflon.

If the mixing-jet readout error is constant, it is possible to estimate that this error is about 3–8 times the correction used at the IWCIN. The supersaturation to compensate would be 5–10 times higher than the 2% value used. Since the expansion chamber readout error changes with droplet size, it is only possible to estimate from the NaCl and Teflon data that the supersaturation is considerably higher than that in the mixing jet. On the other hand, the theoretical calculations of supersaturation are reasonably well verified for expansion, but are quite uncertain for the mixing jet. The uncertainty in the mixing-jet supersaturation may be improved in the future if less difference in vapor pressure could not be used between the wall and the jet air.

In the light of Warner's (1968) conclusion that the median supersaturation for small-to-medium continental cumuli is only 0.1% in their initial condensation region (which determines the droplet concentration), a further question is raised as to whether even the 0.2–0.3% lower limit of supersaturation obtainable with thermal-diffusion type CCN instruments is realistic for many clouds. For example, the finding of Hobbs *et al.* (1970) that increased rainfall develops within 10 min after "fumulus" clouds form downwind of anthropogenic sources of copious CCN active at 1% supersaturation may actually be explained as they suggest (p. 84) by continuous emission of a small number of large, very efficient CCN. In that case the number of cloud droplets formed would be small, and would thus grow to precipitation size drops sooner. In other words, the actual cause may be the opposite of the obvious conclusion based on even the thermal diffusion chamber measurements because 1% supersaturation is too high in this instance.

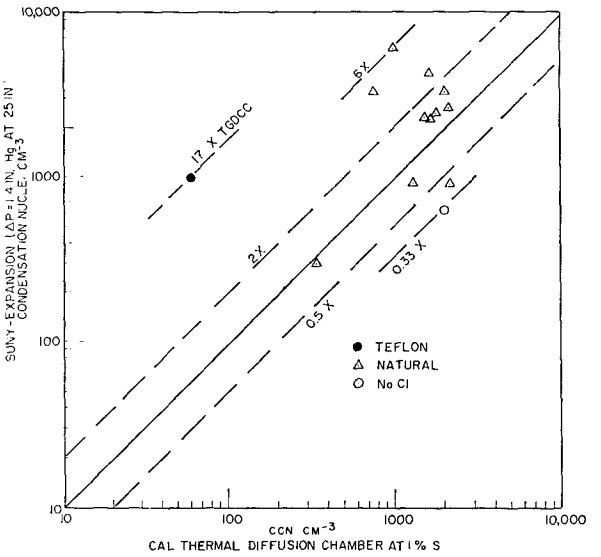

FIG. 7. Data from State University of New York (SUNY) expansion (Garner) counter at $\Delta P = 1.4$ inch Hg vs TGDCC data from the Cornell Aeronatical Laboratory (CAL) instrument. The numerals on the dashed isopleths indicate the ratio of the expansion instrument readings to those of the TGDCC instruments.

A maritime example in which it is indicated that CCN should be measured at lower supersaturations is discussed by Dinger *et al.* (1970). They found that in a subsident air mass the fraction of CCN from sea salt generally decreased with altitude, reading essentially zero above an inversion; however, in one case over the Atlantic the fraction of CCN composed of sea salt was greater in the residual air from an evaporated cloud than was the case even in the surf at sea level.

Apparently the updrafts were slow enough so that, for these maritime clouds growing under suppressed conditions, measurements of CCN should have been made at a supersaturation considerably lower than 0.75% in order to be representative of the nuclei actually being used by these clouds.

6. Conclusions

Calculations indicate that the expansion (Gardner) counter had a supersaturation of 26% when it was used at an expansion ratio which gave (with natural nuclei) output readings equal to the nucleus concentration from a thermal-diffusion type CCN instrument operated at 1% supersaturation. With natural nuclei a compensating error may have been due to slower droplet growth, hence lower readout, than the instrument was designed to provide at 300% supersaturation.

Measurements made at supersaturations as high as 26% are not applicable to clouds. They are particularly inapplicable to clouds when measuring the effects due to pollution where often most of the condensation nuclei present are active only at supersaturations higher than could be reached in clouds.

In pollution conditions measurements made at supersaturations typical of clouds would be likely to lead to conclusions which are opposite to those reached from measurements made with an expansion type counter which has been adjusted to operate in a mode which gives natural nucleus readings comparable to a thermal diffusion instrument operating at 1% supersaturation. Examples by other investigators in which CCN measured at 0.2–0.5% supersaturation were increased negligibly are the CCN from forest fire smoke and fuel oil fumes. Slight decreases in CCN were found in auto exhaust and in air exposed to terpene vapors such as those forming "blue haze" from forests.

The applicability of TGDCC instrument data to natural cloud conditions has been verified by comparing activity spectra taken by such an instrument with droplet number concentration in clouds by Squires and Twomey (1961) and by Twomey and Warner (1967).

Some diffusion-chamber measurements at 0.6–1% supersaturations have indicated sizeable increases in CCN in pollution (Alkezweeny and Green, 1970). However, conclusions from measurements made with expansion-type Aitken nucleus instruments should be evaluated in terms of the actual supersaturations used, particularly under those pollution conditions in which a large fraction of the nuclei are less easily activated than true CCN. The same precaution is needed to a slightly lesser degree for data from the NCAR continuous-mixing-jet nucleus instrument until improvements can be made to reduce its supersaturation. If the supersaturation can be reliably maintained at values of 0.3–1%, this instrument has potential as a continuous instrument with an automatic digital readout.

Further efforts are needed to discover methods for reliably measuring CCN at supersaturations $<0.2\%$ in order to obtain data applicable to fog, stratus clouds and small-to-medium cumuli.

Further efforts would also be welcomed in verifying the error or lack thereof in using the more convenient expansion chambers to count CCN. However, considerable effort will be required to obtain more comprehensive comparisons than were achieved at the second IWCIN. In the interim, decisions being made on instrumentation for CCN studies, particularly in polluted air, should take into account the large errors indicated at the IWCIN and confirmed by calculations to be inherent in expansion-type instruments.

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