The Vertical Distribution of Cloud and Aitken Nuclei Downwind of Urban Pollution Sources

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(Manuscript received 20 May 1971, in revised form 24 September 1971)

ABSTRACT

Simultaneous airborne observations of Aitken and cloud nuclei upwind and downwind of Buffalo, N. Y., were made in an effort to determine the effects of air pollution on condensation nucleus concentration. The data show significant increases over background concentrations of both Aitken and cloud nuclei (at 0.3% supersaturation) immediately downwind of pollution sources, and, also, that a secondary maximum in cloud nucleus concentration usually occurs about 10–15 mi farther downwind. In spite of these large increases in the total number of nuclei downwind of an industrial-urban complex such as the Niagara Frontier, the concentration of cloud and Aitken particulates approaches upwind background levels within 20–50 mi of the sources.

Attempts to define the role (if any) of air pollution in cloud microstructure were confined to a single set of airborne observations. The limited drop-size distribution and cloud nucleus data were not sufficient to justify firm conclusions, and additional measurements are recommended.

1. Introduction

Because of the important role that condensation nuclei play in the formation of clouds and precipitation, there is a considerable need to know in some detail the concentration of nuclei which initially control the shape of the drop-size distribution in clouds. It is well known that not all atmospheric particulates are favorable condensation sites; and, in fact, depending on the type and size of the particulates and the cooling rate of the air, only a small fraction participate in the formation of cloud droplets. Those particulates that promote droplet growth at supersaturations (S) characteristic of natural clouds and fog (i.e., $\leqslant 1.0\%$) are known as cloud nuclei. Hence, it can be fairly said that cloud nuclei dictate the initial size and concentration of droplets in clouds and therefore help determine the ease with which precipitation forms in warm clouds, a fact that has been discussed in some detail by Squires and Twomey (1960).

Continental air masses, which are abundant in effective cloud nuclei, often produce clouds that consist of relatively high concentrations of very small droplets. The coalescence mechanism for precipitation formation is therefore frequently inefficient and the clouds are reluctant to precipitate. On the other hand, maritime atmospheres, which are rich in moisture but generally possess fewer cloud nuclei (Twomey, 1959; Jiusto, 1967), produce clouds which have larger droplets and which precipitate readily.

Since it is generally agreed that an inverse relation exists between the concentration of cloud nuclei and the average drop size in clouds, it is reasonable to ask what the result of large increases in atmospheric pollution might be on the cloud nucleus population. If most sources of atmospheric contaminants also produced effective cloud neuclei, the result could be the occurrence of clouds having extreme colloidal stability and hence a lower probability of producing precipitation. For example, after examining 60 years of rainfall records in Queensland, Warner (1968) showed that a reduction of rainfall had occurred in areas downwind of cane fires during the peak harvest season. Hobbs *et al.* (1970) determined that clouds frequently form downwind of paper mills in the state of Washington and suggested that inadvertent changes in precipitation due to anthropogenic activities may exceed those obtained in deliberate cloud seeding experiments. Earlier nucleus measurements by Pilié and Kocmond (1967) in central Pennsylvania revealed that paper mills are indeed prolific sources of cloud and Aitken nuclei. The question of pollution, therefore, and its effect on the cloud nucleus concentration and cloud microstructure is of considerable applied interest and continues to generate substantial discussion.

Other cloud nuclei measurements made at this Laboratory (Kocmond, 1965; Pilié and Kocmond, 1967; Kocmond and Jiusto, 1968) suggest that sources of pollution differ markedly in their ability to produce cloud nuclei. That is, high concentrations of particulates (due to pollution) can be present in the atmosphere while the cloud nucleus concentration remains low. This condition sometimes occurs when a very clean arctic air mass passes over an urban environment such as Buffalo, N.

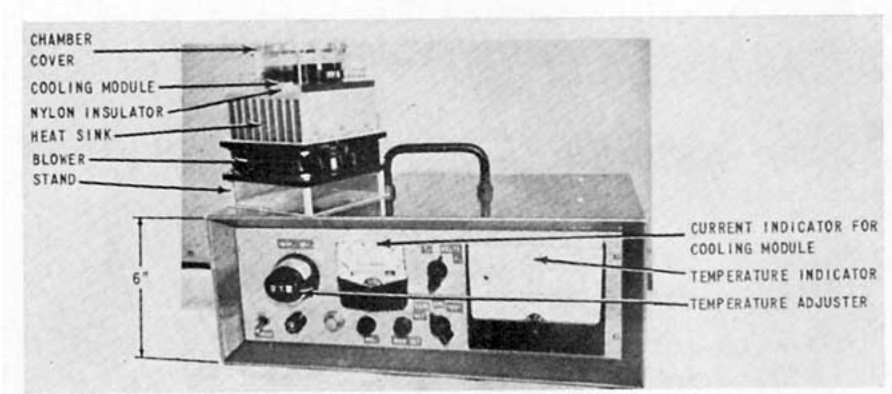

FIG. 1. CAL thermal diffusion chamber and temperature control unit.

Y., or the Niagara Frontier. Under these circumstances, there is a rapid increase in *total* nucleus concentration as a result of industrial and other man-made sources of pollution; however, the cloud nucleus count can remain below average if the pollutants are uncommonly small and hydrophobic. Recent measurements by Terliuc and Gagin (1971) near Jerusalem indicate that there is very little relationship between the Aitken and cloud nucleus concentration in that area, at least, implying that local contaminating sources are not mainly responsible for the observed cloud nucleus concentrations.

It was the purpose of this study to determine how the concentrations of cloud nuclei differed at cloud heights upwind and downwind of a large industrial area capable of producing substantial atmospheric pollution. Efforts to determine if changes in cloud nucleus concentration (due to pollution) affected cloud microstructure were not conclusive from the limited data we were able to acquire.

2. Apparatus

Measurements of cloud nuclei were made with the Cornell Aeronautical Laboratory (CAL) thermal gradient diffusion chamber. The chamber, shown in Fig. 1, was installed on an Aztec aircraft for measurement aloft.

The basic design of the chamber is patterned after that of Langsdorf (1936), Wieland (1956) and Twomey (1963). In brief, the unit consists of a cylindrical plexiglass chamber with upper and lower reservoirs, a collimated light beam to illuminate a small volume within the chamber, and a Polaroid camera for photographing droplets that have formed on cloud nuclei.

During operation, water vapor diffuses from the warmer upper surface to the lower reservoir, with the chamber supersaturation being a known function of temperature difference between the two reservoirs. A series of ten thermocouples (five on each surface) is used to measure ΔT. Air samples to be investigated are first allowed to reach temperature equilibrium by entering a conditioning chamber located ahead of the instrument. Transient high supersaturations due to cold outside air contacting the moist chamber walls are thereby avoided. When the desired temperature difference (supersaturation) has been achieved, the chamber is flushed with ambient air at a continuous rate for several seconds. The air sample is then allowed to reside in the chamber where in a few seconds droplet growth proceeds on the most active condensation nuclei. The growing droplets are illuminated in a well-defined volume of the chamber by a 100 W Osram mercury arc lamp and photographed at 90° to the light beam moments before sedimentation begins. Measurements of cloud nuclei made in this fashion are accurate to within ±20% of the actual count. An enlarged photo of droplets formed in the chamber is shown in Fig. 2.

In addition to the cloud chamber, a General Electric small particle detector was used on board the aircraft to make measurements of total nucleus concentration at

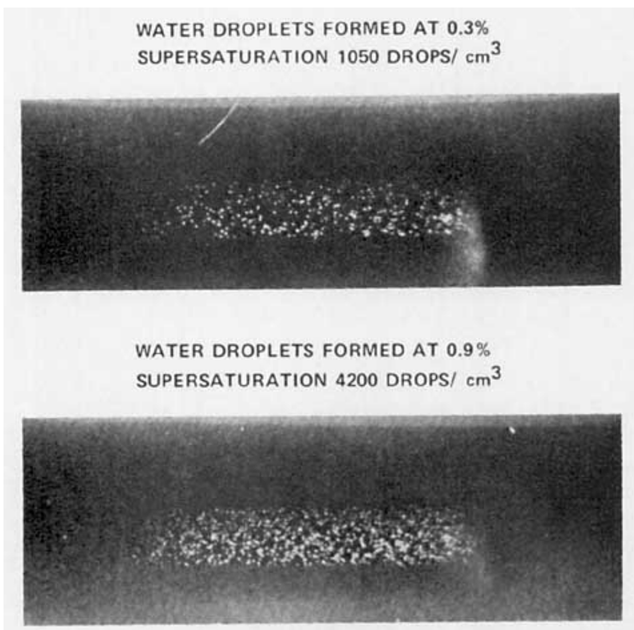

FIG. 2. Enlarged photographs of droplets formed in the thermal diffusion chamber.

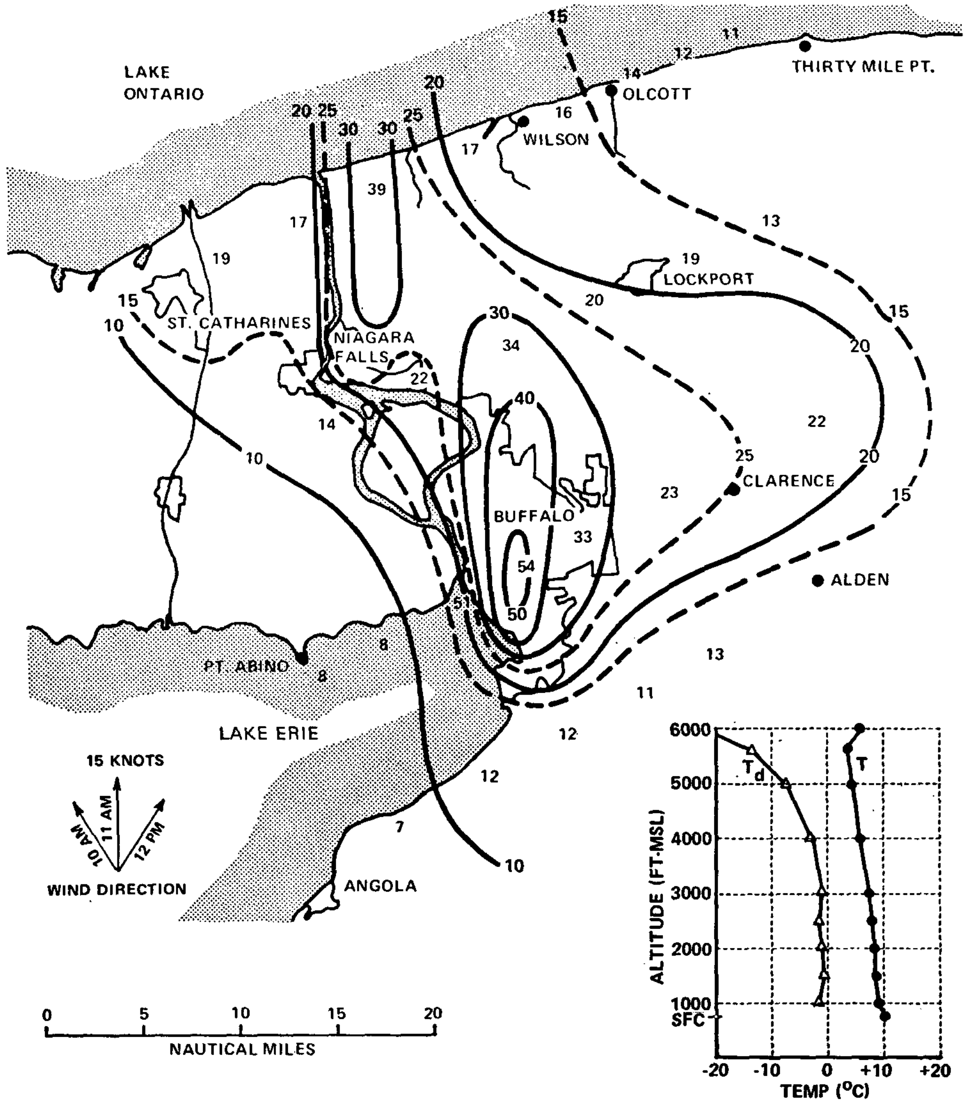

FIG. 3. Isopleths of Aitken nuclei (units 10^3 cm^{-3}) at 1500 ft above ground on 31 October 1969 for measurements made between 1000 and 1300. (Insert shows temperature and dew point profiles.)

selected altitudes and locations. The general level of atmospheric pollution could therefore be monitored.

3. Nuclei sampling experiments

In a typical experiment, nuclei measurements were obtained from the Aztec aircraft at several selected altitudes first upwind of Buffalo, and then at varying distances downwind of the city. Normally, observations were made from the surface through 6000 ft at 1000-ft intervals. Occasionally, measurements were made at altitudes as high as 11,000 ft. The cloud chamber was operated at 0.3%S, a value considered representative of clouds having modest vertical development. Altogether, eleven flights were made over a one-year period. The flights were designed to take advantage of specific clear-weather situations that would allow meaningful comparisons of nucleus concentration in polluted and unpolluted atmospheres.

In Fig. 3, typical data are shown for the total particulate content at 1500 ft in and around the Buffalo area. The wind direction on this date, 31 October 1969, was initially from the southeast and slowly veered to southwest during the 3-hr measurement period. The temperature and dew point profiles through 6000 ft are shown in the insert. As expected, the data show a marked increase in total concentration of nuclei downwind of both the industrial areas of Buffalo and Niagara Falls.

Fig. 4 shows vertical isopleths of both Aitken and cloud nuclei on 2 July 1969 (from 0950 to 1140 EDT) upwind and downwind of pollution sources along the Buffalo waterfront. The Aitken counts are not corrected for changes in expansion ratio due to altitude changes or

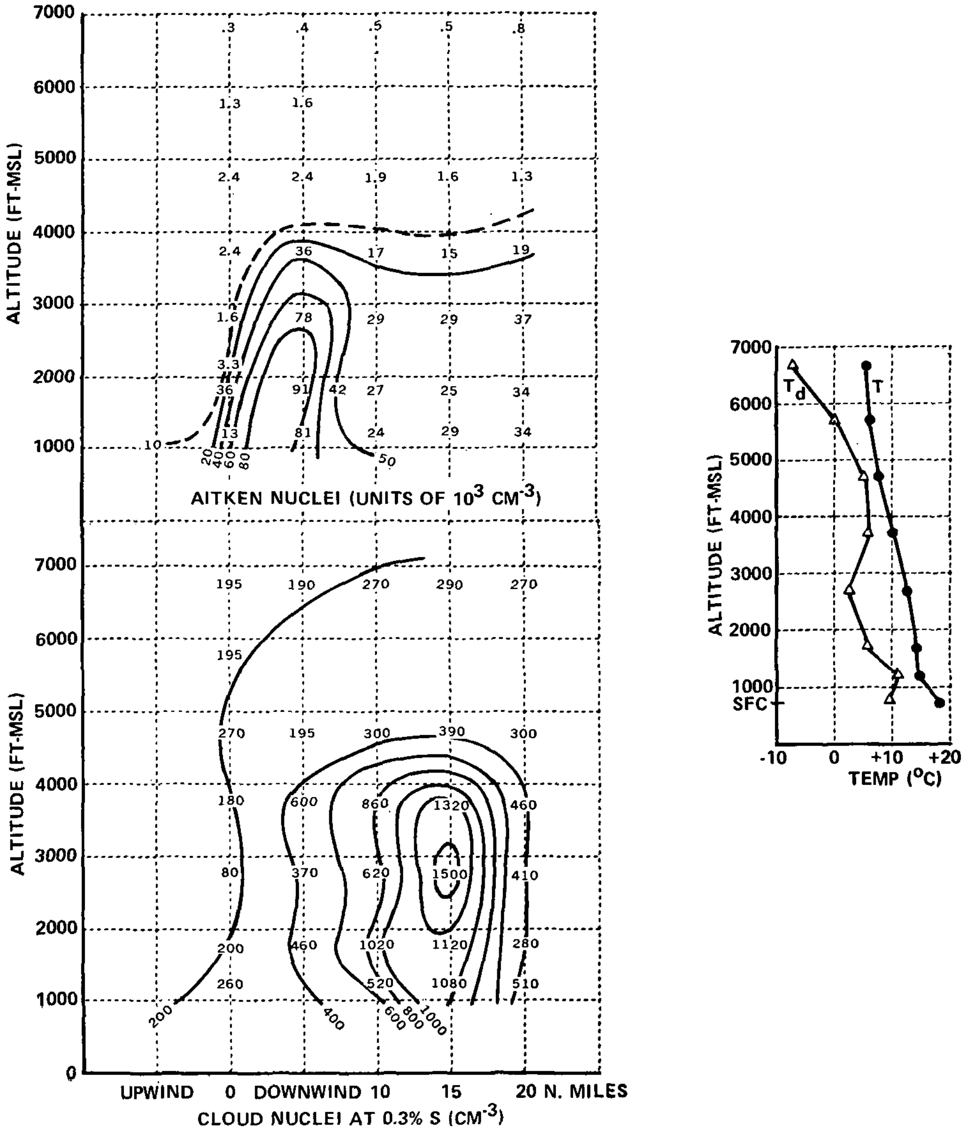

FIG. 4. Contours of Aitken and cloud nuclei upwind and downwind of Buffalo on 2 July 1969; wind speed and direction: 225°, 4 kt at 1000 ft MSL and 260°, 7 kt at 3000 ft MSL. (Insert shows temperature and dew point profiles.)

differences in pressure. [Pollak and Metnieks (1960) have shown that the percentage correction (reduction) of the measured concentration of Aitken nuclei increases with decreasing pressure, amounting to about 25% at 5000 ft and 34% at 10,000 ft. Nearer the surface where most of these measurements were made, the differences are small (*ibid.*; Auer, 1965) and within the accuracy of the instrument.] The data show that on this date the maximum concentration of Aitken nuclei occurred within 1–2 mi of the waterfront industrial complex. The highest concentration of cloud nuclei, however, did not occur until 15 mi downwind of the pollution source area. This example is typical of the manner in which cloud and Aitken nuclei vary downwind of the waterfront. It is thought that photochemical processes were one of the mechanisms responsible for producing additional cloud nuclei on this date. Aggregation of smaller and less effective nuclei may also have contributed to the observed increases in cloud nucleus concentration downwind of the city.

In another comparison, Fig. 5 shows cloud and Aitken nuclei data obtained during a period when there was a temperature inversion at about 3500 ft. The data were obtained over a 2-hr period on 29 October 1969. Note, first of all, the sharp demarcation between high counts below the inversion and much lower values above 3500 ft. This is particularly apparent from the Aitken counts. As in the previous example, the area of maximum total nucleus concentration was very near the pollution sources near the waterfront. Areas of maximum cloud

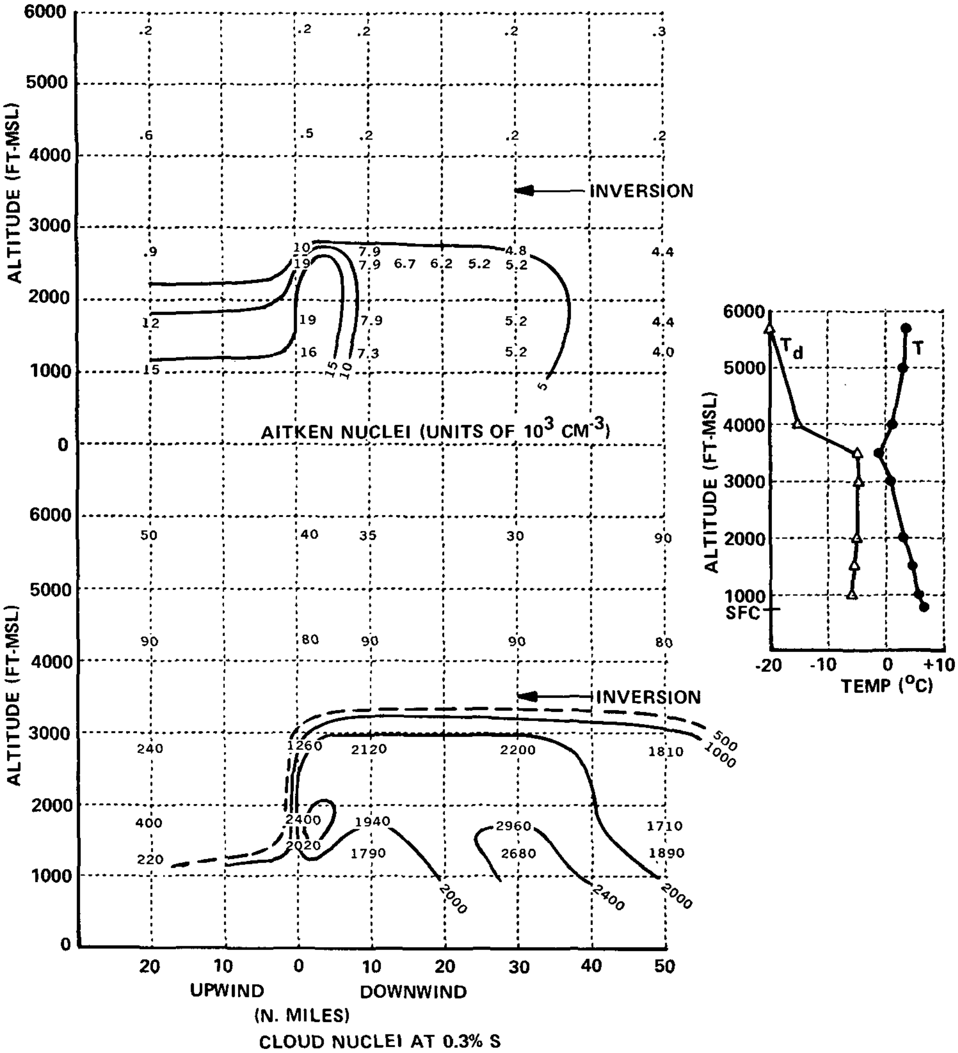

FIG. 5. Contours of Aitken and cloud nuclei upwind and downwind (over Lake Erie) of Buffalo on 29 October 1969; wind direction and speed: 045°, 7 kt, all levels. (Insert shows temperature and dew point profiles.)

nuclei in this case were found near the pollution sources and also about 30 mi downwind of the city where sources are negligible. On this date, the wind direction was from the east-northeast, parallel to the axis of Lake Erie. Since the flight track downwind of the city was over the center of the lake, no additional contaminants could have influenced the nucleus count downwind of the waterfront. Again, photochemical processes and particle aggregation were probably principal mechanisms in producing cloud nuclei far downwind of the city. In this example, the cloud nuclei count was very high in the same area that the Aitken concentration was highest. Evidently, many of the pollution sources were also effective in producing cloud nuclei.

In each of the cases shown, both the cloud and Aitken nucleus concentrations are only temporarily affected by atmospheric pollutants. Thus, while there is a large increase in the total particulate matter in the immediate vicinity of the city, the increases do not persist for great distances downwind. This result is in agreement with measurements by Twomey (1959) and Squires (1966), which suggest that urban pollution is not likely to contribute greatly to the background level of cloud nuclei.

Averages of the data are displayed in Fig. 6 where the increase in the cloud and Aitken nucleus concentrations over background levels are shown on the eight flights in which vertical profiles were obtained upwind and downwind of the city of Buffalo. Again, increases in average Aitken nucleus concentration are highest approximately 5 mi downwind of shoreline sources of pollution. The averages of cloud nucleus measurements, however, show two distinct maxima: one in the immediate vicinity of the pollution sources and another 15 mi downwind of these major sources. It is thought that, initially, diffu-

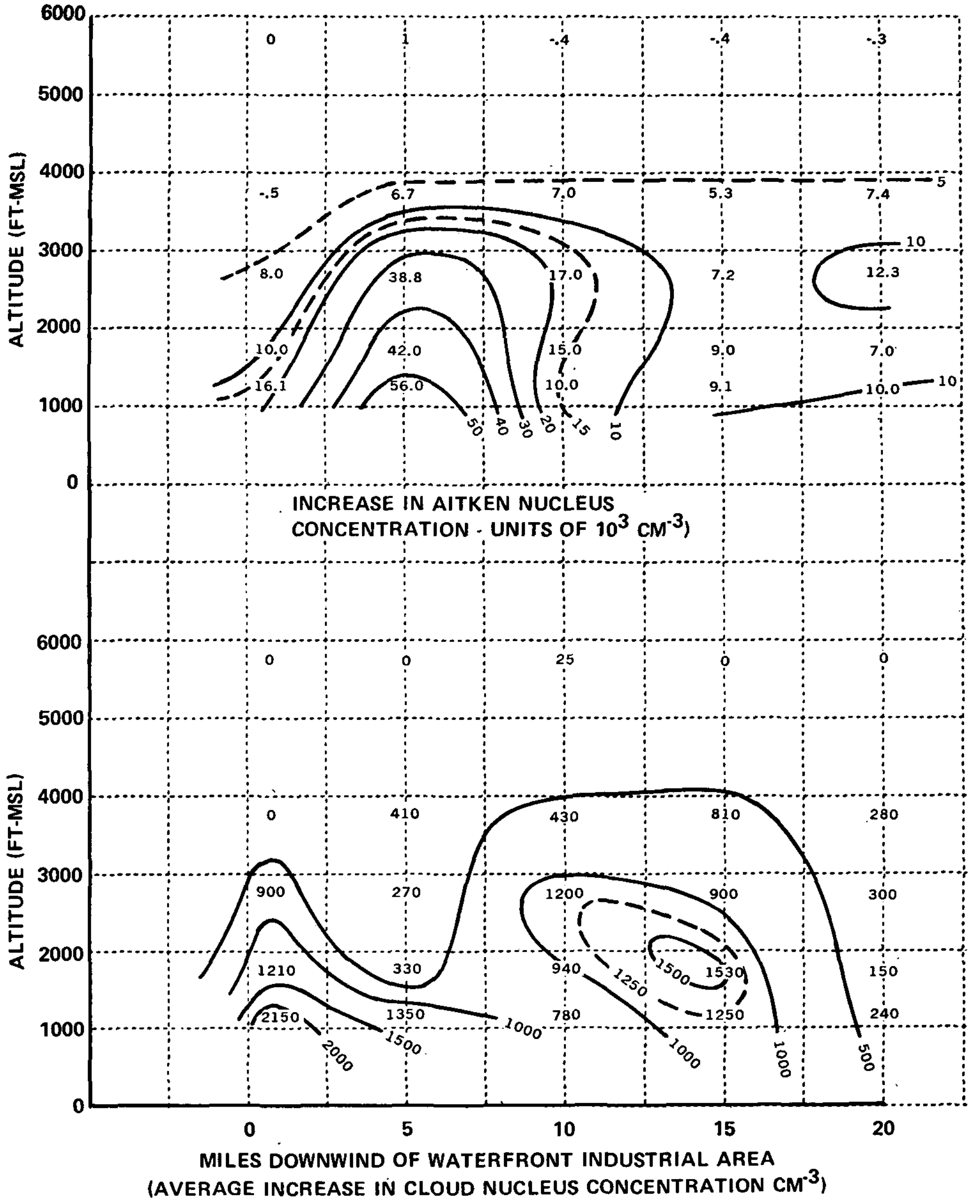

FIG. 6. Contours of average increase in Aitken and cloud nucleus concentrations downwind of Lake Erie waterfront, Buffalo, N. Y.

sion and mixing cause the cloud nucleus concentration to decrease to the observed minimum within a few miles of the city. Farther downwind, interaction of gaseous constituents with water vapor in the atmosphere, photochemical reactions, and aggregation of the smaller and less effective Aitken nuclei may be responsible for producing the observed increase in cloud nucleus concentration at 15 mi. As in other specific examples, both the cloud and Aitken nucleus concentrations appear to return to background levels within ~20 mi of the pollution sources.

In view of the demonstrated increase in cloud nucleus concentrations downwind of nearby industrial areas, an attempt was made to determine if these changes in nucleus concentration also brought about differences in cloud microstructure. Plans were made to obtain drop samples in clouds formed upwind and downwind of Buffalo. The drop sampler used for making these measurements employs a gelatin replication technique (Jiusto, 1965) that has been used extensively at CAL for the past five years. Nucleus measurements were to be obtained at cloud base in order to locate areas of maximum cloud nucleus concentration. Because of time limitations, only one flight was made in which weather conditions were suitable and in which both cloud and Aitken nucleus data were obtained.

Three clouds were sampled on 7 May 1970. The data are summarized in Fig. 7 together with the drop-size distributions that were observed during the penetration.

As indicated, both the cloud and Aitken nucleus concentrations were highest at the base of clouds 2 and 3 downwind of the city. The clouds were small cumuli and, in general, merged with stratus at 4500 ft. Observations of cloud nuclei were made at 2000 and 3600 ft (cloud base) and droplet samples were obtained in clouds at 4000 ft. Cloud 1 samples were obtained over the Niagara Peninsula in Canada ~10 mi upwind of Buffalo. Cloud 2 observations were made ~8 mi downwind (east-southeast) of the shoreline steel mills. Because of a slight shift in wind direction, cloud 3 was sampled directly to the east of Buffalo and ~20 mi downwind of shoreline pollution sources. Pollution plumes (i.e., areas of maximum particulate concentration) were located by means of the Aitken counter.

As previously stated, the cloud and Aitken nucleus concentrations were highest beneath clouds 2 and 3. The average drop radius in cloud 3 was significantly smaller than in the upwind cloud and the drop-size distribution was narrower. Cloud 3, therefore, may have been considerably more stable than cloud 1 (all other things being equal) and, hence, less likely to precipitate.

The characteristics of cloud 2, on the other hand, do not appear to be consistent with the observed nucleus increases. In spite of an increased nucleus concentration at cloud base, the average drop size is larger in this cloud than in the "unpolluted" cloud upwind of the city. Interpretation of the data is complicated, however, by the fact that the upwind and downwind clouds were almost certainly sampled at different times during their life cycle. Thus, it is quite possible that in both examples the downwind cloud microstructure was altered by the increased number of cloud nuclei but the differences may have been slight and completely masked by natural changes in drop spectra occurring during the lifetime of the cloud. Furthermore, there is no way of knowing precisely where the observed clouds originally formed and hence what fraction of the cloud nucleus population was involved. It is obvious that additional flights and data are needed in order to draw conclusions relative to the hypothesis that increased urban pollution results in smaller sized droplets in clouds forming downwind of the sources.

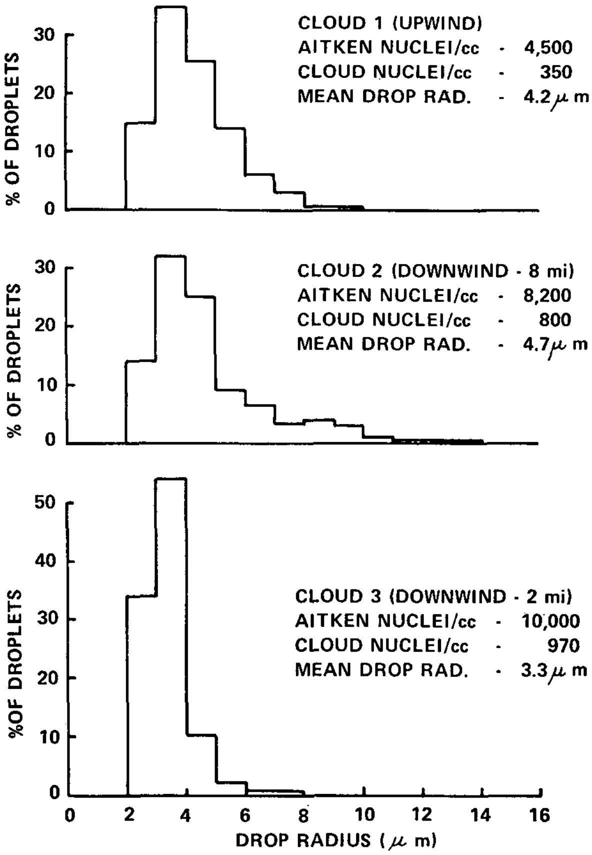

FIG. 7. Drop-size distributions in three clouds on 7 May 1970. Cloud nuclei measurements were made at 0.3% S.

4. Conclusion

Results of eleven flights near Buffalo, N. Y., in which cloud and Aitken nucleus observations were obtained, show that significant increases occur in the cloud and Aitken nucleus concentrations downwind of pollution sources. The data show that the cloud nucleus concentration is frequently high in two locations: 1) in the immediate vicinity of pollution sources where the Aitken count is also very high; and 2) 10 or more miles farther downwind of sources where the Aitken count is quite low. Photochemical reactions in the atmosphere and aggregation of particulates are thought to be primarily responsible for the observed secondary maximum in cloud nucleus concentration.

The data further show that in spite of large increases in the total number of nuclei downwind of pollution sources, the concentration of particulates returns to lower values and soon approaches the background level within 20–50 mi of the pollution sources. Under most weather situations, therefore, dilution of the high concentrations of particulates with cleaner background air results in near normal counts within a few tens of miles of the source. Under near stagnant conditions, the ability of the atmosphere to accommodate large increases in pollution is severely restricted and widespread local increases in nuclei can occur.

Attempts to study the effects of increased cloud nucleus concentration on cloud microstructure did not yield sufficient data to permit valid comparisons of polluted and unpolluted clouds. Additional flights to acquire these data are recommended.

Acknowledgments. This investigation was supported by Cornell Aeronautical Laboratory. The authors wish to express special thanks to Mr Roland J. Pilié for his helpful contributions during the course of this investigation.

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