Aerosol and cloud microphysics measurements in Hawaii[1]

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ABSTRACT

In conjunction with a cooperative study of the warm rain process, selective measurements were made of the microphysical properties of Hawaiian clouds forming inland (orographic) versus those forming out to sea. Within the context of a brief field program, an attempt was made to establish certain bounds on associated aerosol concentration, cloud drop sizes, droplet concentration and liquid water content. Differences in the slopes of supersaturation-nucleus concentration spectra appear related to the continental character of the aerosol and the size distributions of nuclei involved. The data suggest that the size distribution of effective cloud nuclei can be approximated by an $r^{-3/4}$ to $r^{-3/2}$ radius function.

1. Introduction

The generation of rain in warm clouds is largely a function of mesoscale dynamic factors, necessitating an airmass with the proper combination of horizontal convergence, updraft intensity, moisture content, and thermal instability. Microscale variables, namely cloud nucleus concentration and initial cloud drop size distribution, can at times exert a considerable influence on a cloud's capacity to precipitate. Squires & Twomey (1960) were among the first to elucidate the relation between the spectra of condensation nuclei and cloud droplets and the resultant efficiency of the coalescence process. Other factors being equal, an airmass with relatively few effective nuclei should result in a cloud with larger droplets and a higher probability of reaching the precipitation stage. Maritime and continental clouds can often be differentiated on this basis.

There are less evident indications that the slopes of cloud nucleus spectra (concentration versus supersaturation) may vary depending upon the continental or maritime character of the aerosol. Junge (1960) has explained the absolute difference between these supersaturation spectra in terms of differences in the size distribution of the condensation nuclei.

Distinguishing features of the drop size distributions of maritime and continental clouds have been reported, among others, by Squires (1956) and Battan & Reitan (1957). More subtle differences in droplet size and concentration also occur between clouds imbedded within the same general airmass, such as observed by Squires & Warner (1957) in Hawaiian orographic and nearby trade-wind cumulus clouds.

Additional data on cloud microphysics and aerosol concentration in diverse regions of Hawaii were compared and contrasted with corresponding values in the northeastern United States. Emphasis was placed on studying the relation between nucleus concentration and initial cloud characteristics as well as the interpretation of supersaturation spectra obtainable with a thermal gradient diffusion chamber.

2. Field studies and equipment

A. *General*

Afternoon flights with a Cessna 336 airplane were conducted in which orographic and oceanic clouds were penetrated and numerous clear air soundings made. By tropical standards, weather conditions during the flights would probably be classified as good—partly cloudy skies, light trade winds (5–15 knots), and only occasional precipitation. The flights roughly paralleled the trade wind extending up to about 20 miles inland over the Mauna Loa–Mauna Kea slopes and a like distance over the ocean east of Hilo. The orographic clouds sampled typically commenced forming about 3 to 7 miles inland

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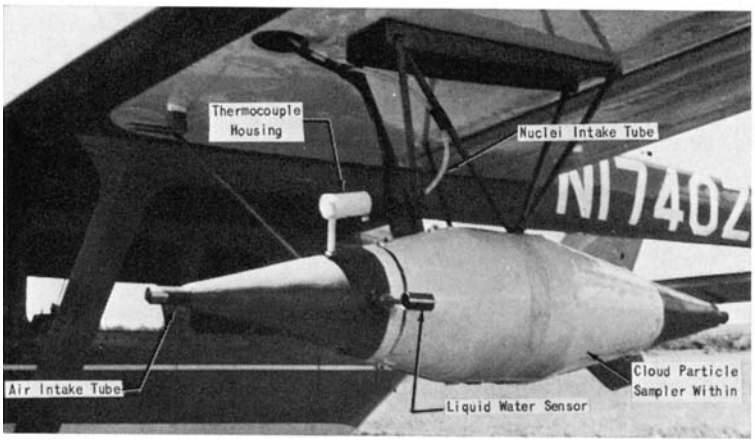


Fig. 1. Aircraft instrument pod. (Modified Illinois State Water Survey Instrument.)

and possessed the general structure described in detail by Mordy (1957). The following types of measurements were made:

1. drop size and liquid water content in orographic and oceanic clouds,
2. vertical distribution of Aitken nucleus concentration,
3. temperature profiles and trade wind inversion character.

B. *Aircraft sensors*

An instrumented pod, pictured in Figure 1, was suspended from the wing of the airplane. As shown, liquid water and temperature sensors were mounted on the front end of the pod, while a cloud droplet sampler with a 1.25 in. diameter intake duct was located within the unit. Air samples were channelled through a 3/8 in. diameter tube to an aerosol particle counter in the aircraft cabin.

The cloud particle sampler consisted of a modified slide projector, the optics compartment of which was replaced by the air duct mentioned above. Glass microscope slides of 8 mm width were contained in a slide tray and individually exposed to the airstream as desired. Slide exposure time could be varied down to a minimum of 0.04 second.

The slides are precoated with a 15% solution of gelatin and dried. As cloud droplets impact a treated slide, some of the gelatin is dissolved leaving well-defined circular images. True drop diameter is approximately one-half replica diameter. Slides with their permanent droplet imprints are stored for subsequent analysis under a phase contrast microscope. Overall drop size accuracy is estimated at 15% for drops larger than about 10 μ diameter and approximately 20% for smaller drops. Further details of the cloud particle sampler are given elsewhere (Jiusto, 1965).

The liquid water meter employed was the hot wire type manufactured by Johnson–Williams (Palo Alto, Calif.). As indicated in wind tunnel tests at the University of Chicago (Owens, 1957), instrument accuracy is approximately 5% when the drop sizes do not exceed about 30 μ. Larger drops tend to shatter when striking the wire and thus cause liquid water content to be underestimated. Further, if large droplets wet a velocity-compensating wire in the unit, spurious negative values of liquid water are temporarily recorded. Hence, it was expected that this instrument would be most reliable in non-precipitating clouds; in well developed clouds and rain situations, the data probably represent lower limits of cloud liquid water.

Air temperature was monitored with a copper-constantan thermo-couple mounted in a reverse flow housing. Overall accuracy is estimated at 0.5°C.

The "total" concentration of aerosol particulates or Aitken nuclei was determined with a General Electric Small Particle Detector (Type CN). This well known expansion-type device creates high supersaturations (up to 300% according to the manufacturer) and thus activates many nuclei that do not participate in

cloud droplet formation. Nevertheless, it can provide useful data on the upper limit of nucleus concentrations in the atmosphere.

C. *Cloud nuclei*

A thermal gradient diffusion chamber (cf. Kocmond, 1965) was used to measure condensation nucleus concentrations at two ground sites: (1) two miles inland at the University of Hawaii, Hilo Campus and (2) 50 m from the water's edge at Hilo Bay. The latter measurements were made during periods of sustained on-shore winds to minimize any land component of nuclei. Thus the two sites emphasized inland and oceanic aerosol characteristics, in general correspondence with the two cloud types sampled.

This portable chamber is similar in many respects to the equipment of Twomey (1963). The instrument was operated at supersaturations between 0.1 % and 6 % and thus provided information on effective "cloud nuclei".

3. Discussion of aircraft results

Approximately 60 penetrations of Hawaiian clouds over water and over land were made, with most clouds sampled at their mid-altitude. Detailed tabulations of cloud type, position, vertical extent, sampling altitude, liquid water content, and drop size properties for each sample are available in the project report (Rogers & Jiusto, 1966). Only summary analyses will be presented here.

A. *Liquid water content of Hawaiian clouds*

Liquid water content (LWC) data, obtained with the hot wire instrument, are summarized in Table 1. As shown, the average maximum LWC in cloud cells over land was approximately 0.5 g m^{-3} irrespective of inland distance (0 to 18 miles from Hilo). Corresponding values over water were about 50 % higher. Eight clouds were penetrated in which precipitation was known to be occurring at the time; the average maximum LWC of four of these rain clouds over land was 1 g m^{-3} while that of the four rain clouds over water averaged 1.1 g m^{-3}.

Despite the limited number of samples and the tendency for the instrument to underestimate liquid water when large drops are present, the data show general agreement with prior Hawaiian cloud measurements (Squires & Warner, 1957).

Table 1. *Liquid water content (LWC) in Hawaiian clouds, g m^{-3}*

Position	Inland			Over water
Distance from coast (Hilo) — miles	0–6	7–12	13–18	0–18
Av. max. LWC	0.44	0.54	0.49	0.76
Standard deviation (Av. max. LWC)	0.23	0.43	0.21	0.36
Max. recorded LWC	0.97	1.90	0.84	1.56
No. of cases	12	18	13	14

B. *Cloud droplet sizes and concentration*

Approximately 70 droplet samples were taken in various orographic and marine clouds. Two types of analyses were used to extract drop size and concentration information from the gelatin-coated slides. The first method involved scanning a portion of a slide to establish the minimum, maximum, and estimated average drop size present; selected slide-area counts were made in order to compute droplet concentration of the sampled cloud. With the analysis technique used, the average drop size corresponds most closely with a median value. The summary data in Table 2 resulted from this type of analysis.

The more-detailed second method involved the counting and microscopic measurement of each droplet replica on a portion of a slide. A complete drop size distribution is thereby obtained, which permits in addition the computation of liquid water content and the application of a correction factor for the collection efficiency of the slide (Langmuir & Blodgett, 1946). The results of this type of analysis are shown in Figures 2 and 3 with representative photomicrographs pictured in Figure 4.

Table 2. *Average diameter and concentration of Hawaiian cloud droplets*

	Clouds over land	Clouds over water
Average diameter, μ	14	22
(standard deviation)	(4)	(3)
Average drop concentration, cm^{-3}	100	45
(standard deviation)	(50)	(22)
No. of cases	46	12

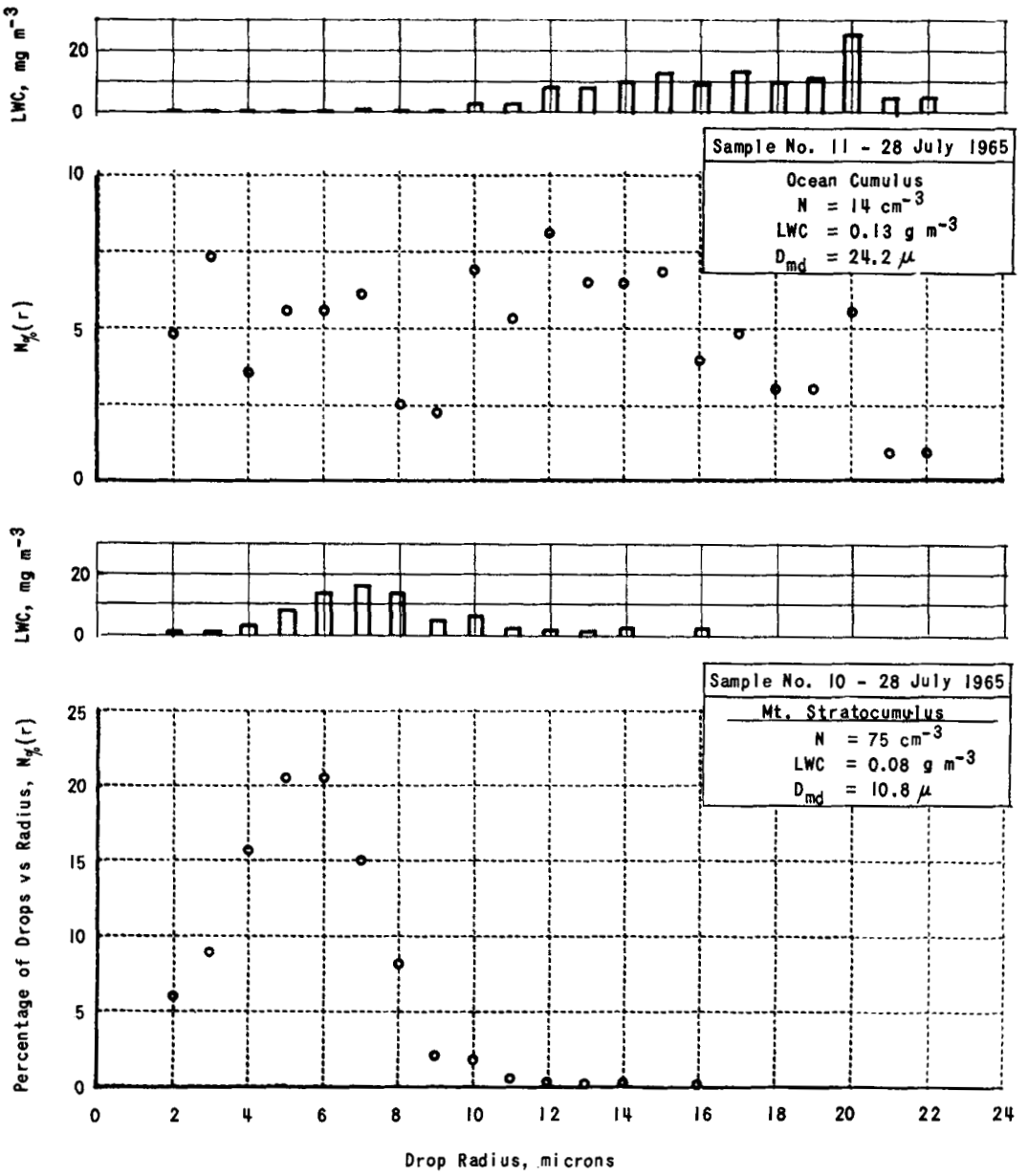


Fig. 2. Drop size and liquid water distributions—orographic and ocean cloud cases.

For the given sampler and aircraft velocities (50–60 m sec^{-1}) involved, each slide was exposed to approximately 1 liter of air. Of this, a slide area equivalent to an air sample of 1–10 cm^3 was selected for analysis; this corresponded to sizing 200 to 500 drops irrespective of which analysis technique was employed. Spot comparisons of both methods confirmed the validity of the more rapid scanning technique.

On the average, cloud droplets over land proved more numerous and smaller than cloud droplets over the ocean. This difference appears to be due mainly to a corresponding difference in condensation nucleus populations. The ap-

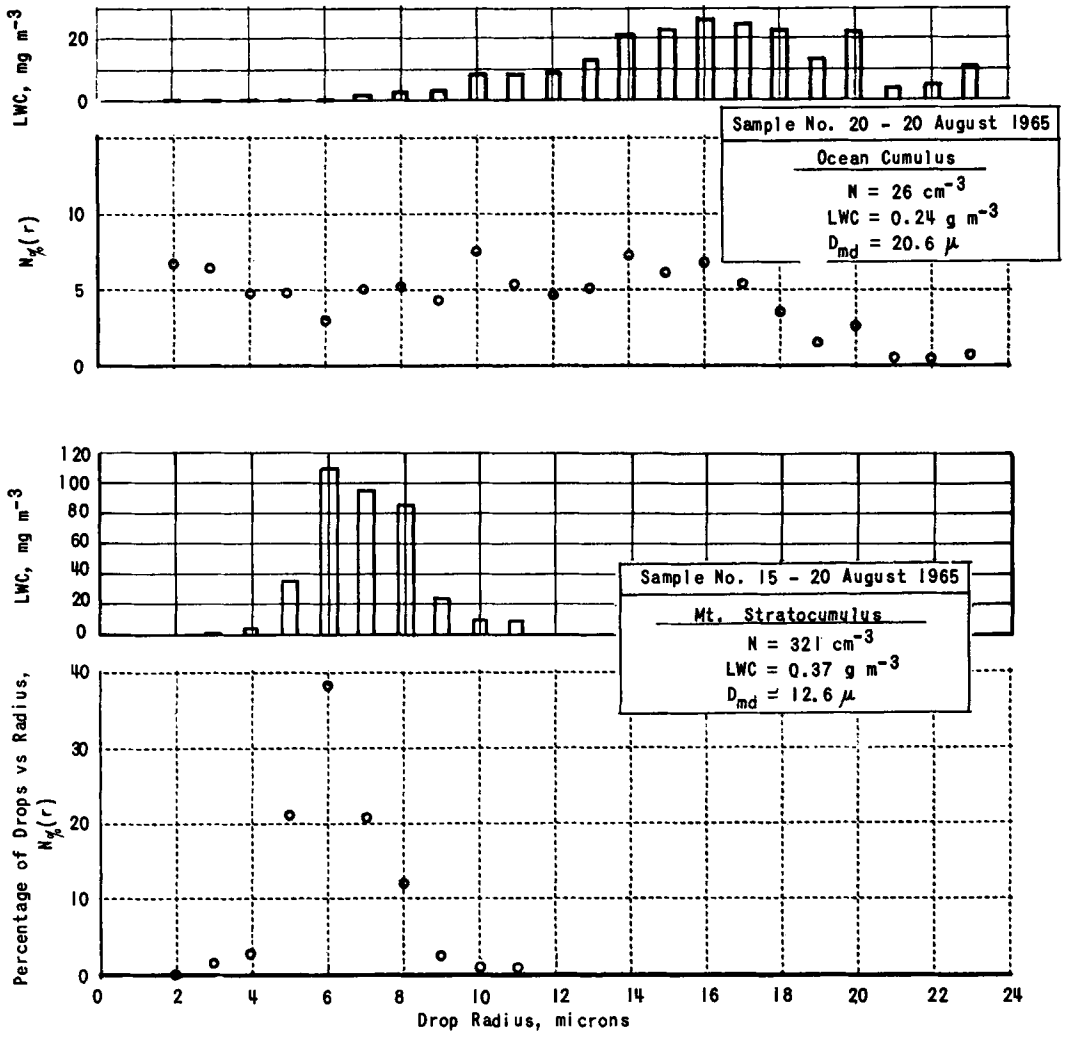
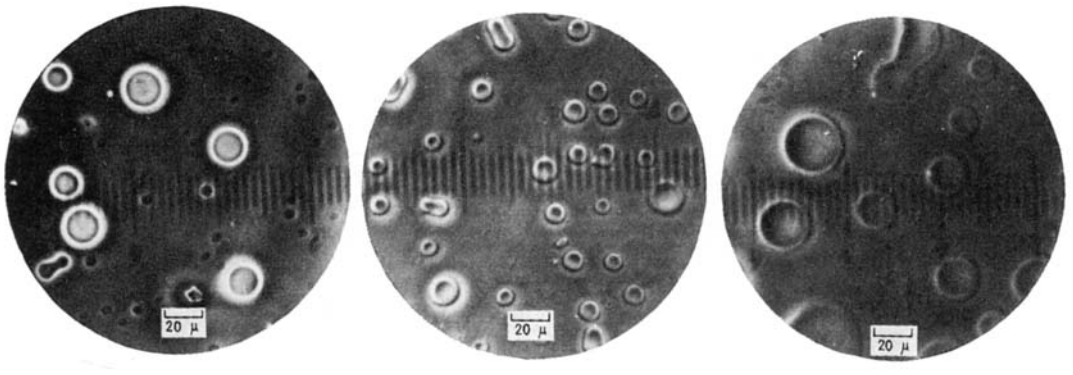


Fig. 3. Drop size and liquid water distributions—orographic and ocean cloud cases.

proximate factor of two increase in droplet concentration of clouds over land versus water showed considerable similarity with the inland and oceanside measurements of condensation nuclei. Droplet concentrations calculated from nucleus-supersaturation measurements (Section 4A) compared favorably with the values measured in clouds. Thus the nuclei and cloud microphysics data appear mutually consistent and in accord with the nuclei-drop distribution hypothesis.

The above results contrast somewhat with measurements of Squires & Warner (1597), who generally found larger cloud drops inland. One possible explanation may be offered by differences in prevailing synoptic conditions and cloud types during the two programs. The aircraft measurements reported herein were associated with weak trade winds and slight precipitation; the orographic clouds that were sampled formed inland and remained separate from ocean clouds. By contrast, during periods of more widespread precipitation and stronger trades, cloud cells were sometimes observed to form over water and then progress inland. In such transitional cases or in more intense oro-



a b c

Fig. 4. Cloud drop samples, 20 August 1965.
(*a*) Mountain stratus (slide No. 10). (*b*) Mt. stratocumulus (slide No. 15). (*c*) Ocean cumulus (slide No. 20).

graphic systems, one might expect substantial differences in the droplet spectra. Another possibility is that increased industrialization in Hilo over the intervening 10 years has increased the background nucleus concentration and changed the microphysical characteristics of certain clouds in the area.

As indicated in Table 2, variations in average drop size and concentration were greater in land clouds than in ocean clouds. This may be due to the greater homogeneity in conditions over the ocean both with respect to nucleus concentrations and dynamic factors such as heat, water vapor, and momentum transport. No significant correlation was found between average drop size and cloud liquid water.

Referring to the drop-size distributions of Figures 2 and 3, one can observe not only the discussed drop size disparity between ocean and land clouds but also the difference in spectrum shape noted by previous investigators. In short, ocean spectra are broader, more irregular, and the liquid water peak is shifted to larger drop sizes.

The mountain stratus cloud sample at 8900 feet (Figure 4) is interesting in that it had the highest droplet concentration (340 cm^{-3}) and the smallest average droplet diameter (4 μ) of all samples taken. This stratus deck was merging in places with lower stratocumulus clouds. At the time an inversion (4.5°C)—the strongest encountered during the flight program—existed with its base at 8300 ft. The high concentration of small droplets may have been associated with a corresponding high concentration of nuclei often encountered above the inversion base.

4. Spectra of cloud nuclei

The results of the cloud nuclei and Aitken nuclei (aloft) measurements are described elsewhere (Jiusto, 1966). This discussion is restricted to certain aspects of the data that have developed subsequently.

The cloud nucleus spectra measured at the two ground sites are shown in Figure 5. Each straight line on the logarithmic plot of nucleus concentration N (cm^{-3}) activated as a function of supersaturation S (%) represents a least-squares fit to approximately 50 data points. Comparative data of Kocmond (1965) for the industrial city of Buffalo, New York, are also shown. The resulting expressions are:

$N = 53\ S^{0.46}$ (Hawaii—ocean side),

$N = 105\ S^{0.63}$ (Hawaii—two miles inland),

$N = 3500\ S^{0.90}$ (Buffalo, New York).

Squires & Twomey (1960) have briefly discussed the use of such an approximating function of the form $N = cS^k$. There is justification for its use provided the supersaturation interval is limited. For example, it can be shown that at supersaturations under about 0.1 %, the slopes flatten and k approaches zero (Jiusto, 1967). Above about 1 or 2 % supersaturation, spectral interpretation becomes more tenuous owing to instrument difficulties in maintaining desired supersaturations, possible activation of non-hygroscopic wettable particles, fluctuations in the nucleus size distribution, and more pronounced aging (agglomeration) effects. In the supersaturation interval 0.1 to 1.0 % the approximation appears reasonable.

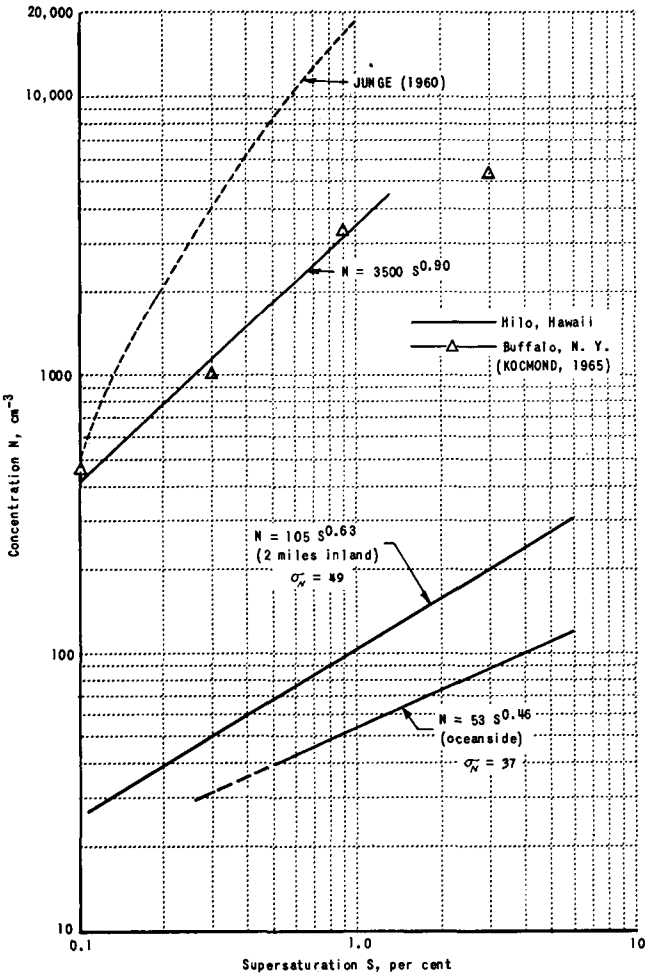


Fig. 5. Concentrations of cloud nuclei at Hilo, Hawaii and Buffalo, New York.

A tacit assumption usually made is that these spectra relate to effective cloud nuclei, i.e., those that are able to achieve droplet sizes on the unstable branch of the well known droplet growth curves. These nuclei are generally the ones that a thermal gradient diffusion chamber detects; however, depending on the supersaturations and given optics system involved, some chambers can and do record "haze" particles. These are solution droplets large enough to be detected but limited in growth on the stable side of the growth curves. At supersaturations of and below approximately 0.1%, haze particles constitute an increasing percentage of the droplet population.

A. *Calculated cloud droplet concentrations*

From measured nucleus-supersaturation spectra, the concentration of resultant cloud droplets can be calculated using a derivation of Twomey's (1959). His expression, converted to percentage supersaturations, takes the following form:

$$N_d = c^{2/(k+2)} \left[\frac{68.7 \times 10^{-3} V^{\frac{3}{2}}}{k B(3/2, k/2)} \right]^{k/(k+2)},$$

where N_d is cloud droplet concentration (cm^{-3}), V is updraft intensity (cm sec^{-1}), $B(3/2, k/2)$ is a beta-function, and c and k are the described nucleus function constants.

Table 3. *Cloud droplet concentrations* (cm^{-3}) *calculated from nucleus-supersaturation data and measured*

Updraft (m sec^{-1})	0.5	1.0	2.0	Measured conc.
Hawaii — Ocean side	40	50	60	45
Hawaii — Inland	65	85	105	100

Substituting measured values for c and k in the above expression and assuming varying updrafts, the calculated droplet concentrations of Table 3 result. Updrafts in Hawaiian rain as measured with a Doppler radar 9 miles inland by Rogers (paper in this issue) never exceeded 7 m sec^{-1}, and in more stable orographic clouds did not exceed 1 m sec^{-1}. It is likely that the type of clouds penetrated by aircraft generally possessed maximum updrafts within a factor of two of 1 m sec^{-1}. As shown, the calculated and measured cloud droplet concentrations in Hawaii agree reasonably well.

B. *Size distributions cumulative of cloud nuclei*

Junge (1960) has calculated supersaturation spectra for cloud nuclei using the droplet growth equation and empirical data on particle size distribution. His calculated supersaturation spectrum for continental air is shown in Figure 5. It is assumed that the particles are soluble and that the effects of chemical composition differences between soluble nuclei are minor. Where particle concentration n varies inversely as radius r cubed, this approach leads to the cloud nucleus-supersaturation relation $N \alpha S^2$.

From diffusion chamber data obtained thus far, it is apparent that average k values upwards of 2 over the supersaturation range involved are uncommon. Values of k approaching unity, for urban areas such as Buffalo, may represent an upper limit.

Starting with measured supersaturation spectra, one may reverse Junge's approach and calculate the cloud-nucleus size distribution. Making the same assumptions, and rounding off the k exponents for simplicity, the calculated cloud-nucleus size distributions of Table 4 apply.

Thus these data support previously observed differences between size distributions of cloud nuclei and "total" particle population for which the inverse-cube rule is generally valid. Differences arise since not all of the atmospheric particles are soluble or sufficiently hygroscopic to promote droplet growth.

C. *Airmass influence on slopes of nucleus-supersaturation spectra*

The data in Figure 5 suggest that the exponent k increases in magnitude as the airmass varies from fresh maritime to mixed continental–maritime to continental. Values of approximately 0.4 to 0.9 are involved. Twomey (1959) reported a typical k range of 0.2 to 0.5 for data obtained in Australia and assigned values of 1/4 and 1/3 to maritime spectra and 2/5 to continental spectra. Thus the same general trend was observed, and the differences are similar if only the Hawaiian measurements are compared with his data. The northeastern United States, because of air trajectory and urbanization, undoubtedly represents a continental character not realized in Australia. In terms of the absolute values of k, equipment and optical resolution differences alluded to previously dictate that no closer agreement should be expected.

Comparing the Buffalo spectrum and Junge's calculated spectrum for continents, the concentrations are similar at 0.1% supersaturation. As supersaturation is increased to 1.0% and progressively more smaller-sized particles are activated, the curves depart considerably. This suggests that as particle size decreases, a greater percentage of the nucleus population consists of nonhygroscopic or less hygroscopic particles. Recognizing that combustion particles of low solubility can contribute substantially to the Aitken nucleus population, such a trend appears intuitively reasonable.

Two individual runs made within 5 hours of one another at the oceanside site are interesting in that they represent, respectively, the highest and some of the lowest nucleus concentrations obtained during the summer. The high concen-

Table 4. *Calculated cloud-nucleus size distributions:* $n \alpha r^b$

	k (Meas.)	b(Calc.)
Hawaii — Oceanside	$\frac{1}{2}$	$-\frac{3}{4}$
Hawaii — Inland	$\frac{2}{3}$	-1
Buffalo, New York	1	$-\frac{3}{2}$

trations measured between 0950 and 1050 L were associated with a NW flow of air from the Hilo industrial area; sugar refining plants and cane fire contamination lay in the upwind direction. By 1430–1530 L, the NE trade wind had been established, and the resultant oceanic air trajectory was accompanied by very low nucleus concentrations. At 3% supersaturation, for example, the morning nucleus concentration was approximately 1200 cm^{-3} while the afternoon value measured 60 cm^{-3}. Corresponding Aitken counts were 6×10^4 cm^{-3} and 200–300 cm^{-3}, respectively. Below 3% supersaturation, the two sets of data tended to converge.

These individual observations and the summary data of Figure 5 suggest that slope characteristics of supersaturation spectra for given aerosols are particularly sensitive to the abundance of Aitken nuclei present. Fresh maritime and continental aerosols differ markedly in this component, a major difference appearing to be the relative absence of industrial or combustion nuclei in oceanic air.

5. Summary

Comparisons of orographic and nearby oceanic clouds in the vicinity of Hilo, Hawaii indicated a higher drop concentration, smaller drop sizes, and a narrower droplet spectrum for the inland clouds. The observations were consistent with measured cloud nucleus concentrations made inland and at oceanside. Average cloud water content values of approximately 0.5 g m^{-3} for inland clouds, 0.75 g m^{-3} for trade wind cumuli, and 1 g m^{-3} for precipitating cells agreed with previous measurements (Squires & Warner, 1957).

Cloud-nucleus supersaturation spectra obtained with a thermal gradient diffusion chamber indicated k values of 0.4 to 0.9 for the approximation function $N = cS^k$. The exponent appears to be proportional to the continental character of the aerosol, with $k = 1$ for industrial continental areas perhaps representing an average upper limit.

Converting these measured spectra to nucleus size distributions indicates that cloud nucleus concentration varied inversely with radius from approximately $r^{-3/4}$ to $r^{-3/2}$. The larger negative exponent for Buffalo, New York, still departs from Junge's accepted r^{-3} expression for continental aerosols because (as is well known) not all atmospheric particles promote droplet growth. A related implication is that as particle size decreases, a larger percentage of the nucleus population consists of nonhygroscopic or low solubility particles.

The above approximate relations apply primarily to the supersaturation range 0.1 to 1.0%. For supersaturations below about 0.1%, k approaches zero; above about 1%, uncertainties in instrument and nucleus behavior presently render spectral interpretations tenuous.

Acknowledgements

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АЭРОЗОЛЬНЫЕ И ОБЛАЧНЫЕ МИКРОФИЗИЧЕСКИЕ ИЗМЕРЕНИЯ НАД ГАВАЙЯМИ

В связи с комплексным изучением теплых дождей были проведены выборочные измерения микрофизических свойств гавайских облаков, формирующихся орографически над внутренними частями острова по сравнению со свойствами облаков, возникающих над морем. В рамках короткой полевой программы была сделана попытка установить определенные границы для концентрации аэрозоля, размеров облачных капель, концентрации капель и содержания жидкой воды. Различия в наклонах спектров концентраций пересыщенных ядер, по-видимому, объясняются континентальным характером аэрозоля и различными распределениями по расмерам ядер.

Результаты измерений распределения по размерам эффективных облачных ядер могут быть аппроксимированы функцией от радиуса r в степени от $-3/4$ do $-3/2$.