

# Tests of the Salt-Nuclei Hypothesis of Rain Formation

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## *Abstract*

Atmospheric chlorides in sea-salt nuclei and the chlorides dissolved in shower rainwaters were recently measured in Hawaii. A comparison of these measurements reveals the remarkable fact that the weight of chloride present in a certain number of nuclei in a cubic meter of clear air tends to be equal to the weight of chloride dissolved in an equal number of raindrops in a cubic meter of rainy air. This result is explained as an indication that the raindrops grow on the salt nuclei in some manner which prevents a marked change in the distribution of these nuclei during the drop-growth process.

The data presented add new evidence in further support of the salt-nuclei raindrop hypothesis previously proposed by the first author (WOODCOCK, 1952).

## **1. Introduction**

The earlier study of atmospheric salt particles at cloud levels over Florida (WOODCOCK, 1952) showed that the number of the larger of these particles per cubic meter of air was similar to the average number of raindrops found during rain storms in other locations. This study also showed that the chloride in these nuclei, assuming that each raindrop forms around a single salt nucleus, was sufficient to account for the range of chloride concentrations usually observed in rains. This similarity of the number and chloride content of salt nuclei and the number of raindrops and the chloride content of rain waters led to the suggestion that raindrops form on the large salt nuclei. It was suggested that further studies should reveal a close relation between the salt in the clear air and salt in the rain falling from clouds subsequently formed in this air.

The purpose of this paper is to present the results of the first field tests of the salt-particle raindrop hypothesis. These tests were carried out during 1952 in Hawaii, where the salt nuclei content of the trade winds is well known and where shower rains occur with great regularity from orographic clouds over the mountains. These clouds form in the wind stream as it flows over the islands, and are usually restricted in vertical development to the lower atmosphere up to the trade-wind inversion at about 2,000 meters.

Rain samples were taken within the clouds at positions 1, 3, 4, and 5 (see fig. 1) on the island of Hawaii. Sampling of rain within the clouds made it possible to avoid the difficult problems of evaporation of raindrops and of the coalescence of drops with salt particles, which exists in rains which are sampled far below cloud base (MIYAKE, SUGUIRA, 1950). The sampling positions were selected because positions 1 and 4 are within the bases of the clouds near the area where many of the first shower rains reach the ground, and because

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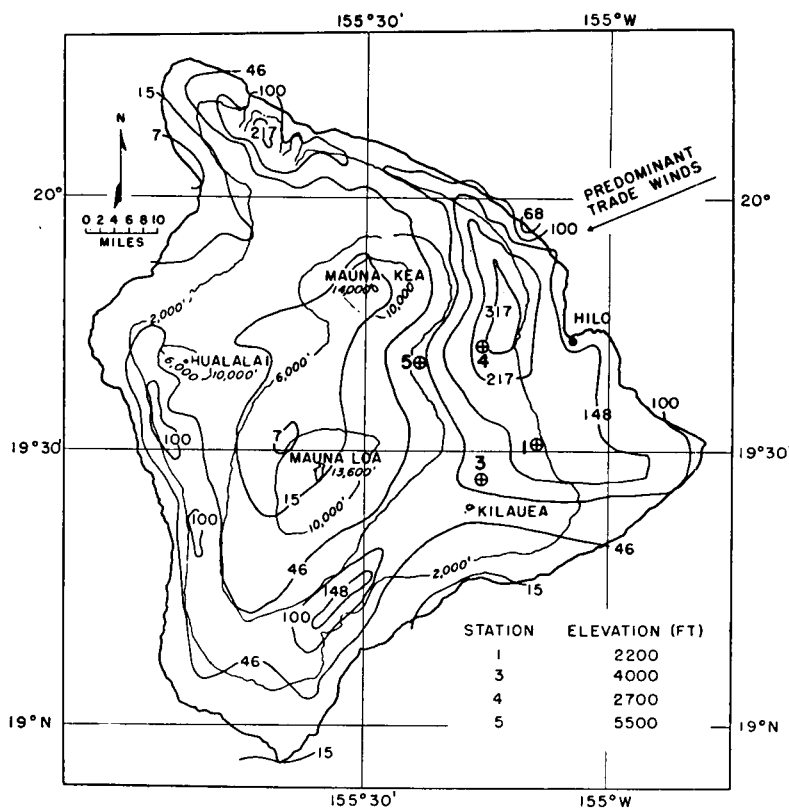


Fig. 1. Isohyetal map of the island of Hawaii, with the locations of the rain-sampling stations used in the present study. The rain-fall contours represent inches annually.

positions 3 and 5 are approximately downwind from 1 and 4 respectively and also within the clouds.

If the salt nuclei were growing to raindrop sizes in these shower clouds, we supposed that the larger less numerous nuclei, which are initially the larger of the cloud droplets (KEITH, ARONS, 1954), might become larger raindrops and fall out of the clouds at the windward stations 1 and 4. The remaining small and numerous drops, formed upon the smaller and more numerous nuclei, would be expected to fall more slowly and to reach the ground further downwind near positions 3 and 5. Thus it was anticipated that the raindrop number and the rain water chlorinity at stations 1 and 4 might be found to be related to the number of the larger salt nuclei in the air and that the rain at stations 3 and 5 might be similarly related to the smaller nuclei in the air.

The observations presented below give reasonably consistent support to this anticipated pattern of interrelationship of salt nuclei and rain.

## 2. Sampling rains and atmospheric salt in Hawaii

In this phase of our work in Hawaii the field problems were first, to measure from aircraft the number and the weight of the salt particles in the air stream before it arrived over the islands and second, to measure on the mountainside the drop size distribution, the salt content and intensities of the rains within the orographic clouds.

The filter-paper technique was used for measuring the sizes of the raindrops. Exposure of these filter papers was made at about the same time that bulk rain-water samples were

**Table 1. Intensities, chlorinities, and drop size distributions in rains at stations 1 and 4. Sampling by filter-paper and bulk-water techniques**

| Position                                       | Date (1952) | Time (LCT) | Bulk water samples       |                             | Rain drops sampled by filter paper technique |                                                                          |       |       |       |        |         |         |         |        |        |
|------------------------------------------------|-------------|------------|--------------------------|-----------------------------|----------------------------------------------|--------------------------------------------------------------------------|-------|-------|-------|--------|---------|---------|---------|--------|--------|
|                                                |             |            | Rate mm hr <sup>-1</sup> | Chloride mg l <sup>-1</sup> | Rate mm hr <sup>-1</sup>                     | Number of rain drops m <sup>-3</sup> within various diameter ranges (mm) |       |       |       |        |         |         |         |        |        |
|                                                |             |            |                          |                             |                                              | .2-.4                                                                    | .4-.6 | .6-.8 | .8-1. | 1.-1.2 | 1.2-1.4 | 1.4-1.6 | 1.6-1.8 | 1.8-2. | 2.-2.2 |
| Part 1—Rain intensity range 0.4 to 2.0 mm/hour |             |            |                          |                             |                                              |                                                                          |       |       |       |        |         |         |         |        |        |
| I                                              | May 4       | 1810       | 1.9                      | 6.3                         | 0.71                                         | 4320                                                                     | 855   | 21    |       |        |         |         |         |        |        |
| I                                              | » 4         | 1815       | 1.9                      | 6.2                         | 0.90                                         | 3200                                                                     | 1300  | 43    |       |        |         |         |         |        |        |
| I                                              | » 4         | 1827       | 0.5                      | 5.7                         | 0.21                                         | 2620                                                                     | 19    |       |       |        |         |         |         |        |        |
| I                                              | » 4         | 1832       | 1.8                      | 2.8                         | 1.00                                         | 6900                                                                     | 971   | 21    | 8     |        |         |         |         |        |        |
| 4                                              | » 6         | 1655       | 1.9                      | 13.7                        | 1.30                                         | 3210                                                                     | 890   | 287   | 20    |        |         |         |         |        |        |
| 4                                              | » 6         | 1706       | 0.4                      | 24.8                        | 0.71                                         | 4050                                                                     | 790   | 41    |       |        |         |         |         |        |        |
| 4                                              | » 6         | 1716       | 0.4                      | 22.9                        | 0.38                                         | 1600                                                                     | 580   |       |       |        |         |         |         |        |        |
|                                                |             |            | 1.26                     | 11.8                        | 0.74                                         | 3700                                                                     | 770   | 59    | 4     |        |         |         |         |        |        |
| Part 2—Intensity range 2.0 to 5.0 mm/hour      |             |            |                          |                             |                                              |                                                                          |       |       |       |        |         |         |         |        |        |
| I                                              | May 1       | 1339       | 2.9                      | 6.8                         | 3.4                                          | 1500                                                                     | 385   | 390   | 265   | 52     | 26      |         |         |        |        |
| I                                              | » 4         | 1835       | 2.3                      | 2.9                         | 3.2                                          | 1700                                                                     | 850   | 620   | 225   | 40     |         |         |         |        |        |
| I                                              | » 4         | 1842       | 2.4                      | 3.2                         | 2.0                                          | 1800                                                                     | 1770  | 390   | 41    | 6      |         |         |         |        |        |
| 4                                              | » 6         | 1722       | 3.2                      | 8.9                         | 2.3                                          | 2170                                                                     | 524   | 590   | 168   |        |         |         |         |        |        |
| 4                                              | » 6         | 1734       | 3.9                      | 8.0                         | 3.8                                          | 1080                                                                     | 137   | 355   | 550   | 31     |         |         |         |        |        |
|                                                |             |            | 2.6                      | 6.0                         | 2.9                                          | 1650                                                                     | 733   | 468   | 250   | 26     | 5       |         |         |        |        |
| Part 3—Intensity range 5 to 10 mm/hour         |             |            |                          |                             |                                              |                                                                          |       |       |       |        |         |         |         |        |        |
| I                                              | May 4       | 1848       | 7.0                      | 1.3                         | 8.0                                          | 111                                                                      | 520   | 690   | 670   | 194    | 54      |         |         |        |        |
| 4                                              | » 6         | 1735       | 7.9                      | 7.7                         | 8.4                                          | 790                                                                      | 280   | 82    | 87    | 121    | 65      | 125     | 14      |        |        |
|                                                |             |            | 7.45                     | 4.5                         | 8.2                                          | 450                                                                      | 400   | 386   | 379   | 158    | 60      | 63      | 7       |        |        |
| Part 4—Intensity range > 10 mm/hour            |             |            |                          |                             |                                              |                                                                          |       |       |       |        |         |         |         |        |        |
| I                                              | May 1       | 1344       |                          |                             | 20.6                                         | 1282                                                                     | 369   | 76    | 206   | 164    | 198     | 202     | 23      | 55     | 7      |
| I                                              | » 4         | 1846       | 12.9                     | 1.5                         | 25.0                                         | 290                                                                      | 196   | 251   | 710   | 850    | 410     | 85      | 11      |        |        |
|                                                |             |            | 12.9                     | 1.5                         | 22.8                                         | 786                                                                      | 282   | 163   | 458   | 507    | 304     | 144     | 17      | 28     | 4      |

taken for chloride analysis. One-half square meter stainless steel funnels were used to obtain these bulk-water samples. Adequate volumes of water for chloride analysis could be taken with these funnels in from 10 to 300 seconds, the time depending upon the rain intensity. The filter-paper and funnel samples were taken only at times when the rain rate appeared to be constant. During the sampling periods rain showers were not observed to be drifting in over the land from the windward sea. Thus the samples were taken in orographic showers, with little rain falling to windward of stations 1 and 4.

The results of the rain sampling at stations 1 and 4 are shown on Table 1, and those at stations 3 and 5 on Table 2. In these tables averages are made from data which are separately tabulated into "parts" having similar rain

intensities. The average raindrop distribution from the different rain intensities shown on Table 1 were plotted cumulatively as shown on the lower left part of figure 2.

In the present study it is also necessary to know the liquid water content and the rate of fall of the rain sampled with the filter papers. These quantities were derived as illustrated on Table 4, where rate of fall and liquid water content (columns 6 and 7) are determined from the raindrop distributions found in the highest intensity rains at stations 1 and 4 (see Table 1, part 4).

The chlorinities of the rains, which are given on Tables 1 and 2, were determined by the standard Mohr method, using a microburette and a one-hundredth normal silver nitrate solution.

Atmospheric salt particles were sampled in

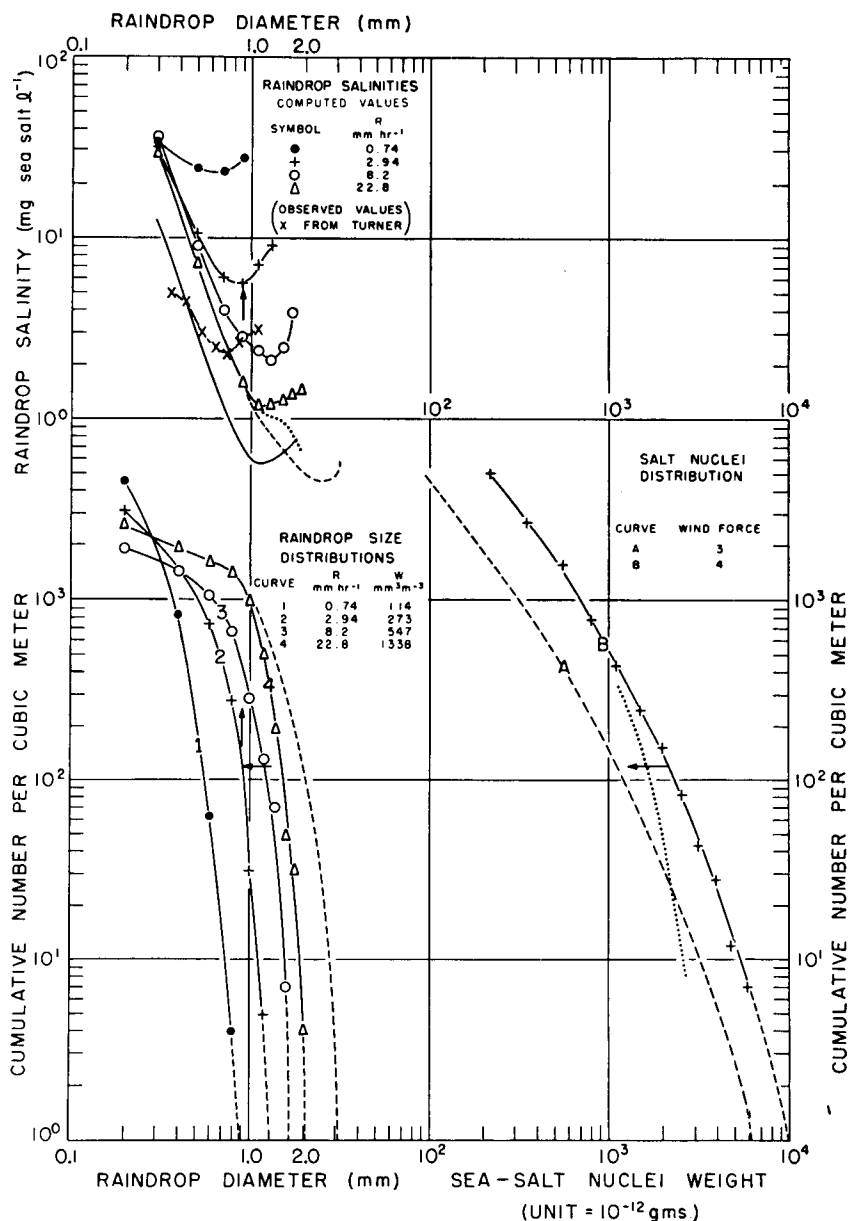


Fig. 2. The lower part of this figure shows cumulative average number distribution curves for salt nuclei and for drops in rains of various intensities ( $R$ ). These curves are used, as described in the text, to compute the average chlorinity of the rain waters (fig. 5) and to compute the raindrop salinity values which are given above.

clear air at cloud levels over the sea by exposing small glass slides from aircraft. The methods used to sample atmospheric salt and those used to sample the rains have been described previously (BLANCHARD, 1953; WOODCOCK,

GIFFORD, 1949; WOODCOCK, 1952), and will not be discussed further in this paper.

It proved impracticable, during the 1952 field trip, to obtain simultaneous observations of airborne salt and of rain. This difficulty was

**Table 2. Intensities, chlorinities and drop size distributions in rains at stations 3 and 5. Sampling by filter-paper and bulk-water techniques**

| Position                                  | Date<br>(1952) | Time<br>(LCT) | Bulk water<br>samples          |                                | Rain drops sampled by filter paper technique |                                                              |         |         |         |         |         |
|-------------------------------------------|----------------|---------------|--------------------------------|--------------------------------|----------------------------------------------|--------------------------------------------------------------|---------|---------|---------|---------|---------|
|                                           |                |               | Rate<br>mm<br>hr <sup>-1</sup> | Chloride<br>mg l <sup>-1</sup> | Rate<br>mm<br>hr <sup>-1</sup>               | Number drops m <sup>-3</sup> in various diameter ranges (mm) |         |         |         |         |         |
|                                           |                |               |                                |                                |                                              | < 0.2                                                        | 0.2—0.4 | 0.4—0.6 | 0.6—0.8 | 0.8—1.0 | 1.0—1.2 |
| Part 1—Intensity range 0.1 to 1.0 mm/hr   |                |               |                                |                                |                                              |                                                              |         |         |         |         |         |
| 5                                         | Apr. 28        | 1702          | 0.13                           | 2.10                           | 0.09                                         | 48,700                                                       | 85      |         |         |         |         |
| 5                                         | » 28           | 1740          | 0.22                           | 2.3                            | 0.19                                         | 41,500                                                       | 2,040   |         |         |         |         |
| 5                                         | » 28           | 1812          | 0.23                           | 6.8                            | 0.06                                         | 25,400                                                       | 243     |         |         |         |         |
| 5                                         | » 28           | 1819          | 0.23                           | 6.8                            | 0.19                                         | 78,500                                                       | 930     |         |         |         |         |
| 5                                         | » 28           | 1834          | 0.33                           | 2.9                            | 0.18                                         | 110,000                                                      |         |         |         |         |         |
| 3                                         | May 1          | 1513          | 1.00                           | 0.8                            | 0.58                                         | 800                                                          | 5,800   | 485     |         |         |         |
| 3                                         | » 4            | 1710          | 0.74                           | 0.8                            | 0.87                                         | 20,000                                                       | 8,600   | 56      |         |         |         |
| 3                                         | » 4            | 1737          | 0.29                           | 0.4                            | 0.37                                         | 28,500                                                       | 5,400   |         |         |         |         |
| 5                                         | » 5            | 2043          | 0.80                           | 0.6                            | 0.65                                         | 101,000                                                      | 7,030   | 144     |         |         |         |
| 5                                         | » 5            | 2116          | 1.00                           | 0.3                            | 0.53                                         | 80,800                                                       | 6,400   | 33      |         |         |         |
| 5                                         | » 5            | 2138          | 0.22                           | 0.4                            | 0.19                                         | 66,200                                                       | 1,360   |         |         |         |         |
| 5                                         | » 5            | 2149          | 0.11                           | 1.4                            | 0.25                                         | 149,000                                                      |         |         |         |         |         |
| 5                                         | » 5            | 2222          | 1.00                           | 0.2                            | 0.26                                         | 29,500                                                       | 3,600   |         |         |         |         |
| 5                                         | » 5            | 2235          | 0.92                           | 0.2                            | 0.45                                         | 51,000                                                       | 6,100   |         |         |         |         |
|                                           |                |               | 0.52                           | 1.9                            | 0.35                                         | 59,400                                                       | 3,400   | 51      |         |         |         |
| Part 2—Intensity range 1.0 to 3.0 mm/hour |                |               |                                |                                |                                              |                                                              |         |         |         |         |         |
| 5                                         | Apr. 28        | 1727          | 1.09                           | 2.00                           | 1.43                                         | 116,000                                                      | 11,500  | 1,150   |         |         |         |
| 5                                         | » 29           | 1759          | 2.28                           | 1.2                            | 1.95                                         | 2,450                                                        | 5,400   | 2,060   | 270     | 24      |         |
| 3                                         | May 1          | 1455          | 2.70                           | 0.7                            | 2.40                                         |                                                              | 8,300   | 3,860   | 31      |         |         |
| 5                                         | » 5            | 2053          | 1.10                           | 0.4                            | 1.34                                         | 65,000                                                       | 11,200  | 1,120   | 27      |         |         |
|                                           |                |               | 1.79                           | 1.1                            | 1.78                                         | 45,800                                                       | 9,100   | 2,050   | 82      | 6       |         |

eliminated because it had been found, during many previous salt-sampling airplane flights, that a clear relationship existed between the wind force and the quantity of salt nuclei in the air at cloud levels. Figure 3 shows the averaged results from the salt nuclei samples taken in Hawaii on twenty-three different days between June 1951 and July 1952. Differences in the distribution of mass of salt among the nuclei on these days are shown cumulatively on figure 4. This figure also shows that the smoothed curves on figure 3 are based upon from nine to twenty-seven measured size-range categories.

Thus by using these salt-nuclei data and the average wind force observations from U.S. Weather Bureau surface maps (see Table 3), we were able to make a quantitative estimate of the average weight of salt as nuclei in the air during the interval of days when we sampled rain. This average salt amount was compared to the average weight of salt in the rain on these days.

Tellus VII (1955), 4

### 3. Relating the salt in the nuclei to the salt in the rain at the windward stations 1 and 4

As previously stated, the purpose of the present study is to test the idea that a close relationship exists between the weight of the larger salt nuclei per cubic meter of clear air and the weight of salt dissolved in the rain in a cubic meter of rainy air. Expressed differently the above idea would mean that the largest salt nuclei form the raindrops, and that the individual drops which make up the first rains coming from a shower cloud should contain the salt found in an equal number of the largest salt nuclei.

The average numbers and sizes of raindrops for various rain intensities at stations 1 and 4 are plotted cumulatively on figure 2 (see curves, lower left), starting with the largest drops observed. On the same figure the average distribution of number and weight of salt nuclei is similarly presented at the lower right. This salt nuclei curve, taken from the data shown on figure 3 is used because the

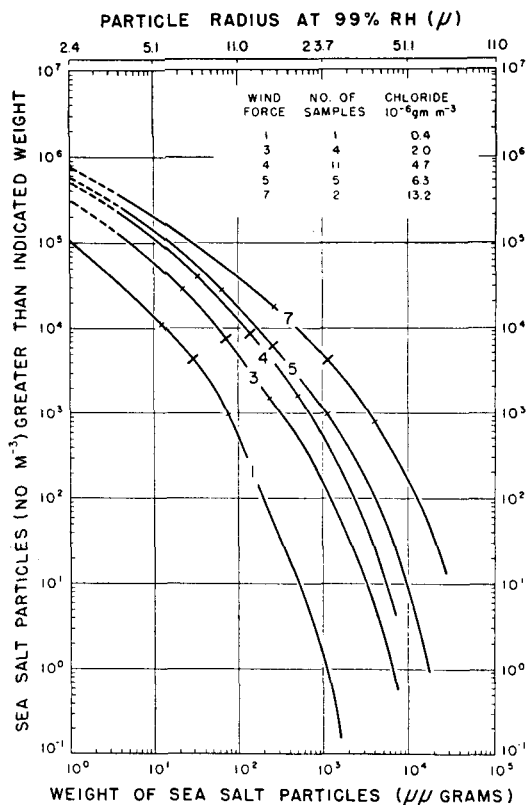


Fig. 3. Cumulative distribution curves for averaged salt nuclei number during winds of various forces in Hawaii. (Taken from WOODCOCK, 1954.)

observed average wind force during the days when the rains samples were taken was very nearly force 4 (see Table 3).

Figure 2 and Table 4 contain the information required to compute the chloride concentration which should be observed in rains if each of the larger salt particles has become a raindrop and if, in the process, little change has occurred in their distribution in the air. For example: rains averaging  $6,328 \text{ mm}^3 \text{ m}^{-2} \text{ sec}^{-1}$  intensity were made up of a definite number of drops of each size range falling per square meter per second (see Table 4, columns 4 and 5). The approximate weight of sea salt in the nucleus of each of these raindrops is taken from the force 4 cumulative salt nuclei curve on figure 2, using the appropriate cumulative raindrop curve to establish, in each case, this nucleus weight. These weights are shown on Table 4, column 8. As an indication of the derivation of these weights, note on salt curve B (fig. 2)

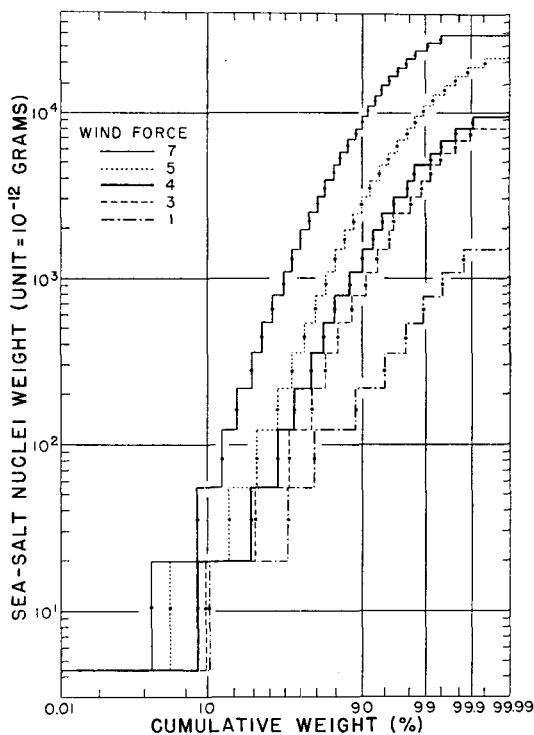


Fig. 4. Averaged cumulative per cent weight distribution curves for salt nuclei present in the atmosphere in Hawaii under varying wind conditions (forces 1 through 7).

Table 3. Surface winds over the sea, during the days when rain was sampled, as reported by ships and planes within 400 nautical miles of Hilo, Hawaii. Taken from the four daily surface maps of the North Pacific, which were obtained from the US Weather Bureau, Honolulu, Hawaii

| Date 1952     | Number of<br>servations<br>reported | Average wind<br>force |
|---------------|-------------------------------------|-----------------------|
| April 28..... | 14                                  | 4.9                   |
| April 29..... | 5                                   | 3.6                   |
| May 1.....    | 11                                  | 3.3                   |
| May 4.....    | 8                                   | 3.8                   |
| May 5.....    | 14                                  | 3.8                   |
| May 6.....    | 9                                   | 3.8                   |
|               |                                     | av. = 3.9             |

that salt particles weighing  $420 \mu\mu\text{g}$ , occur at the cumulative number 2150, which corresponds to the same number of 0.3 mm drops on raindrop curve number 4. Thus  $420 \mu\mu\text{g}$  is entered on Table 4, column 8, as the weight

Table 4. Averaged rain data derived from filter paper samples, table 1, part 4

| 1         | 2               | 3               | 4                   | 5                                 | 6                                 | 7                               | 8                                | 9                                                     |
|-----------|-----------------|-----------------|---------------------|-----------------------------------|-----------------------------------|---------------------------------|----------------------------------|-------------------------------------------------------|
| Drop dia. | Drop no.        | Drop vol.       | Drop fall rate      | Drop no. falling                  | Rain rate mm <sup>3</sup>         | Rain water in air               | Wt. sea salt in each drop. Unit= | Total sea salt falling in drops                       |
| mm        | m <sup>-3</sup> | mm <sup>3</sup> | m sec <sup>-1</sup> | m <sup>-2</sup> sec <sup>-1</sup> | m <sup>-2</sup> sec <sup>-1</sup> | mm <sup>3</sup> m <sup>-3</sup> | 10 <sup>-12</sup> g              | 10 <sup>-6</sup> gm m <sup>-2</sup> sec <sup>-1</sup> |
| 0.3       | 786             | .0141           | 1.17                | 920                               | 13                                | 11.1                            | 420                              | 0.386                                                 |
| 0.5       | 282             | .065            | 2.06                | 581                               | 38                                | 18.3                            | 485                              | 0.282                                                 |
| 0.7       | 163             | .180            | 2.90                | 473                               | 85                                | 29.4                            | 530                              | 0.251                                                 |
| 0.9       | 458             | .375            | 3.65                | 1,670                             | 628                               | 172                             | 600                              | 1.000                                                 |
| 1.1       | 507             | .700            | 4.30                | 2,180                             | 1,523                             | 354                             | 830                              | 1.810                                                 |
| 1.3       | 304             | 1.14            | 4.90                | 1,490                             | 1,700                             | 347                             | 1,350                            | 2.010                                                 |
| 1.5       | 143             | 1.73            | 5.45                | 780                               | 1,348                             | 248                             | 2,270                            | 1.770                                                 |
| 1.7       | 17              | 2.56            | 5.95                | 100                               | 256                               | 43                              | 3,500                            | 0.350                                                 |
| 1.9       | 28              | 3.59            | 6.35                | 175                               | 628                               | 99                              | 5,100                            | 0.892                                                 |
| 2.1       | 3               | 4.85            | 6.75                | 22                                | 108                               | 16                              | 10,000                           | 0.220                                                 |
|           |                 |                 |                     |                                   | 6,328                             | 1,338                           |                                  | 8.971                                                 |

of sea salt to be found in a 0.3 mm raindrop and in rains of the indicated average intensity. Column 9 gives the product of these nucleus weights and the numbers of raindrops (column 5). The computed salinity of the rain (*S<sub>c</sub>*) is now simply the ratio of the total weight of sea salt falling, to the total amount of water falling.

$$S_c = \frac{Ms}{W} \tag{1}$$

where *S<sub>c</sub>* = computed sea-salt concentration of rain (mg l<sup>-1</sup>)

*Ms* = sea salt falling m<sup>-2</sup> sec<sup>-1</sup> (mg)  
*W* = liquid water falling m<sup>-2</sup> sec<sup>-1</sup> (liters)

For the high-intensity rains averaged on Table 4, the amounts of sea salt and water falling are 8.97 μg m<sup>-2</sup> sec<sup>-1</sup> and 6,328 mm<sup>3</sup> m<sup>-2</sup> sec<sup>-1</sup> respectively. The ratio of these quantities, with the units adjusted, yields a computed sea-salt concentration of 1.42 mg l<sup>-1</sup>, or a chloride concentration 0.8 mg l<sup>-1</sup>.

The above computed value for the average chloride concentration, plus other concentrations similarly computed from the averaged data concerning rains of lower intensity (see Table 1) are plotted on figure 5<sup>1</sup>. This also shows the observed average chlorinities of the rains

<sup>1</sup> These computed chloride concentrations for the rains of other intensities were derived in the manner shown on Table 4, but are not tabulated here in order to save space.

Tellus VII (1955), 4

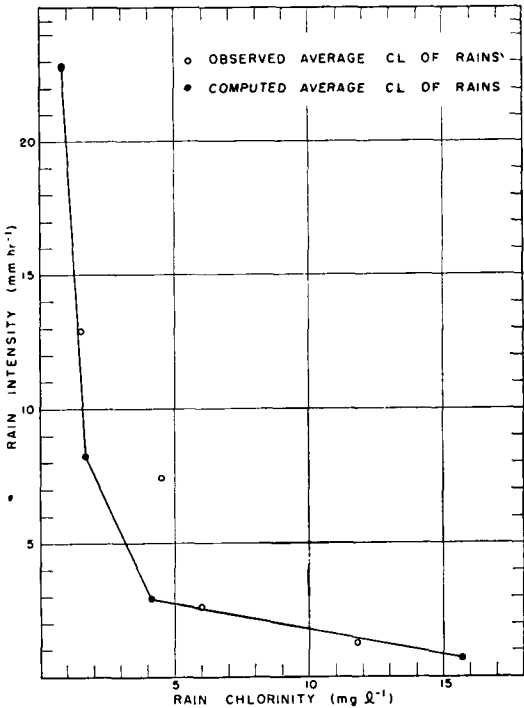


Fig. 5. Observed and computed chlorinities of rains related to rain intensity.

at stations 1 and 4. These observed chloride concentrations are remarkably similar to the computed concentrations, and this similarity is regarded as suggesting that each large salt particle has formed a raindrop.

**Table 5. Summary of averaged rain data from windward stations 1 and 4, Hawaii. May 1, 4, and 6, 1952**

| A                           | B                              | C                              | D                                    | E                                                | F                                                  | G                                                          | H                                                                                    | J                                         | K                               | L                                             |
|-----------------------------|--------------------------------|--------------------------------|--------------------------------------|--------------------------------------------------|----------------------------------------------------|------------------------------------------------------------|--------------------------------------------------------------------------------------|-------------------------------------------|---------------------------------|-----------------------------------------------|
| Rain from funnel sampler    |                                | Rain from filter paper samples |                                      |                                                  | Rain Cl computed from nuclei<br>mg l <sup>-1</sup> | Rain Cl m <sup>-3</sup> of air (ExF)<br>10 <sup>-6</sup> g | Wt. of Cl falling in rain<br>10 <sup>-6</sup> g<br>m <sup>-2</sup> sec <sup>-1</sup> | Ratio of Cl in rain to Cl in air<br>G/4.7 | Rain-drop size range<br>mm dia. | Total number of rain-drops<br>m <sup>-3</sup> |
| Rate<br>mm hr <sup>-1</sup> | Cl conc.<br>mg l <sup>-1</sup> | Rate                           |                                      | Water content<br>mm <sup>3</sup> m <sup>-3</sup> |                                                    |                                                            |                                                                                      |                                           |                                 |                                               |
|                             |                                | mm hr <sup>-1</sup>            | mm m <sup>-2</sup> sec <sup>-1</sup> |                                                  |                                                    |                                                            |                                                                                      |                                           |                                 |                                               |
| 1.26                        | 11.8                           | 0.74                           | 206                                  | 114                                              | 15.7                                               | 1.79                                                       | 5.68                                                                                 | 0.38                                      | 0.3—0.9                         | 4,533                                         |
| 2.60                        | 6.0                            | 2.94                           | 821                                  | 273                                              | 4.1                                                | 1.12                                                       | 6.09                                                                                 | 0.24                                      | 0.3—1.3                         | 3,132                                         |
| 7.45                        | 4.5                            | 8.20                           | 2,279                                | 547                                              | 1.7                                                | 0.93                                                       | 6.80                                                                                 | 0.19                                      | 0.3—1.7                         | 1,903                                         |
| 12.90                       | 1.5                            | 22.80                          | 6,329                                | 1,338                                            | 0.8                                                | 1.07                                                       | 9.0                                                                                  | 0.23                                      | 0.3—2.1                         | 2,693                                         |

#### 4. Relating the salt nuclei to the salinity of the rains at the leeward stations 3 and 5.

As previously suggested on pages 437 and 438 the rains falling at the leeward stations 3 and 5 were found to be made up of great numbers of small drops containing relatively little chloride (see Table 2). This character of these leeward rains tends to support the idea that they are composed of drops which have formed on the very numerous salt nuclei found among those particles weighing from about 5 to 20  $\mu\text{g}$ . The average chloride in these rains (see Table 6, column G) equals 6.2 per cent of the total chlorides found during force 4 winds (total =  $4.7 \times 10^{-6}$  g m<sup>-3</sup>). On figure 4 it can be seen that during force 4 winds about 7.5 per cent of the chlorides in the air are between 4.5 and 20  $\mu\text{g}$  in weight, and in figure 3 the salt nuclei curve for force 4 winds shows that there are about  $1.4 \times 10^5$  particles present in this weight range. Thus the chloride content and the numbers of the small salt nuclei are quite adequate to account for the chloride content and the numbers of the raindrops in the leeward rains.

Due to the nature of the techniques used to

measure these small raindrops and small nuclei, it is not possible, with the present data, to relate the raindrops to the nuclei more precisely. Such precision could be obtained in future measurements by dividing the smaller raindrop and salt nuclei number distributions during the measuring process into greater numbers of size-range categories. Note on Table 2 that the greatest numbers of the raindrops at stations 3 and 5 are in the first size category, while figure 4 shows that all of the appropriate salt nuclei fall within a single measured size-range category.

Most of the raindrops sampled at stations 3 and 5 are so small that due to evaporation it is very unlikely that they would survive a fall through many hundreds of meters of clear air. This probably explains why these small drops have not been reported in such great numbers by other observers.

#### 5. Computed raindrop salinity compared to observed salinity

During the spring of 1954, TURNER (1955) measured the salinity of individual raindrop size ranges in Hawaii. For obtaining rain waters

**Table 6. Summary of averaged rain data from leeward stations 3 and 5, Hawaii. April 28 and 29, and May 1, 4, and 5, 1952**

| A                               | B                                  | C                        | D                                                    | E                                                  | F                                                                    | G                                                   | H                                     | J                                               |
|---------------------------------|------------------------------------|--------------------------|------------------------------------------------------|----------------------------------------------------|----------------------------------------------------------------------|-----------------------------------------------------|---------------------------------------|-------------------------------------------------|
| Rain-water samples              |                                    |                          |                                                      |                                                    | Rain Cl<br>m <sup>-3</sup> of air<br>(BxE)<br><br>10 <sup>-6</sup> g | Ratio of<br>Cl in rain<br>to Cl in air<br><br>F/4.7 | Raindrop<br>size range<br><br>mm dia. | Raindrop<br>number<br>m <sup>-3</sup> of<br>air |
| funnel collector                |                                    | filter-paper samples     |                                                      |                                                    |                                                                      |                                                     |                                       |                                                 |
| Rate<br><br>mm hr <sup>-1</sup> | Chloride<br><br>mg l <sup>-1</sup> | Rate                     |                                                      | Rain in air<br><br>mm <sup>3</sup> m <sup>-3</sup> |                                                                      |                                                     |                                       |                                                 |
|                                 |                                    | mm hr <sup>-1</sup>      | mm <sup>3</sup> m <sup>-2</sup><br>sec <sup>-1</sup> |                                                    |                                                                      |                                                     |                                       |                                                 |
| 0.52<br>1.79                    | 1.9<br>1.1                         | 0.35<br>1.7 <sup>8</sup> | 97<br>494                                            | 119<br>331                                         | 0.22<br>0.36<br>av. = 0.29                                           | .047<br>.077                                        | 0.1—0.3<br>0.1—0.9                    | 62,850<br>57,038                                |

from different drop size-range categories, he used a simple modification of the raindrop spectrograph which has been described by BOWEN and DAVIDSON (1951). Turner determined the sodium content of drops in many rain samples at cloud base near station 4, and it is useful here to compare his interesting observations to the raindrop salinities computed from our raindrop size distributions obtained at the same location. For this purpose his sodium chloride concentration values<sup>1</sup> were converted to total sea-salt concentration by multiplying by 1.3, the approximate ratio of total salts in the sea to the sodium chloride present.

On figure 2 (top) his average raindrop salinity values for 39 samples are compared to similar values computed from the average rain and salt nuclei data (lower curves on figure 2) used in the present paper. The method used to derive these latter raindrop salinities is given in equation (1) and is applied, for example, as follows. A nucleus, having a mass of approximately  $2,100 \mu\text{g}$ , corresponds in cumulative number per cubic meter of air, to the number of 0.9 mm diameter raindrops shown on one of the raindrop distribution curves (see arrow marks near lower curves on fig. 2). The ratio of this nucleus mass ( $2.1 \times 10^{-6} \text{ mg}$ ) to the volume of a 0.9 mm raindrop ( $.375 \times 10^{-6} \text{ l}$ ), equals a concentration of  $5.6 \text{ mg l}^{-1}$ . This drop salinity is indicated by the arrow mark beside the second raindrop salinity curve at the top of figure 2. The other computed values for raindrop salinity were similarly derived.

It is interesting that the form of Turner's average observed raindrop salinity curve and those computed from our raindrop and salt nuclei data is similar, despite the differences in the sampling times, sampling durations, and in the methods used to derive the salinity values.

The occurrence of the pronounced salinity minimum in Turner's results and in our computed values is apparently a reflection of the average slopes and relative values of the cumulative raindrop and salt nuclei curves. For instance, if the observed cumulative average raindrop distribution curve represented by

curve 4 (see figure 2, lower left) had actually followed the adjacent dashed line, then the computed raindrop salinity minimum would not have occurred at a drop diameter of 1.1 mm (see triangular symbols, upper diagram, fig. 2). Instead a different salinity curve would have occurred (see extended dashed line), with the salinity minimum falling at a drop diameter of about 2.5 mm. As an indication of the effects of changing nuclei distribution, the assumption of a force 3 salt-nuclei curve instead of the average force 4 curve (see curve A, fig. 2), simply shifts the computed drop salinity curves down. The unbroken drop salinity curve on figure 2 shows the dilution effects of the reduced nucleus sizes upon drop salt concentrations for the highest intensity rains. As a final example of the effects of the shape of the salt nuclei curve, note that the use of the dotted line modification of curve B in computing raindrop salinity, removes the salinity minimum from the drop salinity curve (see dotted line at top of fig. 2). This removal of the salinity minimum is mentioned here because Turner has remarked that about one-third of his results showed no salinity minimum. Thus the form and, of course, the slope of the computed raindrop salinity curves is clearly dependent upon the relative slopes and magnitudes of the cumulative raindrop and salt-nuclei curves.

Turner's raindrop salinity curve is based upon many hours of sampling, during which time the rain intensity probably varied greatly. Had he been able to collect his rain water during short periods of time (i.e., in about 100 seconds) and, in each case during rains of nearly constant intensity, it is supposed that he would have obtained different salinity curves for different rain rates similar to those which we compute from our observed raindrop and salt nuclei curves.

Turner's average values for the salinity of raindrops of each size range, probably represent an integrated quantity. Note for instance on figure 2 that the salinity of the 0.9 mm diameter raindrops computed from our curves may vary from 1.6 to  $28 \text{ mg l}^{-1}$ , depending upon the rain intensity in which they are sampled. The raindrop salinity values which will be obtained during a prolonged sampling period in orographic rains at station 4 would seem to depend, in part at least, upon the relative

<sup>1</sup> Sodium concentrations, measured by flame photometry, were converted by Turner to equivalent NaCl values.

frequency of occurrence of rains of different intensities. They will also presumably depend upon the extent to which the samples are taken in the first rains from the showers, or in rains which are the winnowed remnants of showers occurring to windward of the raindrop spectrograph sampling position.

In future physical-chemical studies of rain, it would be very useful to combine raindrop spectrograph measurements of drop salinity, with simultaneous determinations of raindrop size distribution and of salt nucleus size distributions.

## 6. Discussion and conclusions

Clearly many more observations, such as those given on Tables 1 and 2, are needed in order to thoroughly test the consistency of the pattern of interrelationship of nuclei to raindrops which has evolved from the present study. Data deficiencies are especially great among the higher intensity rains (see Table 1), and the lack of simultaneity of the rain and salt-nuclei observations introduces a further element of uncertainty. However, the authors feel that the data and ideas presented here, when considered as a whole, are sufficiently useful to justify publication at this time.

Some of the above observational deficiencies were eliminated in further measurements recently made (1954) in Hawaii in connection with the co-operative cloud physics work of "Project Shower". These numerous measurements, which will require much time to completely analyse, will be presented in a later study of this problem.

A rather obvious result of the present study is the emphasis which is given to the importance, in the understanding of Hawaiian shower rain formation, of detailed knowledge of the salt particle size distribution in the clear air and the distribution of raindrop size and raindrop salinity within the clouds. It is also important to know where the rain samples are taken in relation to the stage of development of the showers. In the present study an effort was made to sample the first and the last rains falling from showers moving up the mountain slope. The authors feel that this detailed knowledge of the nuclei, the raindrops and the rain-sampling position, is essential to a further understanding of the evolution of raindrops in the shower rains of marine air

masses. The techniques used here should also prove useful in exploring the role of sea-salt nuclei in the formation of raindrops in continental rains.

It should be pointed out that in the analysis presented here, the quantities of salt falling in the rains at stations 1 and 4 amount to only about 20 to 40 per cent of the total salt in the air (see summary Table 5, column J). Since, of the remaining 60 to 80 per cent of the atmospheric salt, only about 6 per cent falls at stations 3 and 5 (see summary Table 6, column G), it is supposed that the remaining salt rained-out in the drops falling between these stations, as the rain showers were carried by the winds up the mountain from the lower to the upper stations. In a later publication (of "Project Shower" observations) the authors plan to include an analysis of the effect of wind and wind shear upon the distribution of raindrops at various locations on the mountainside.

In the Hawaiian shower rains, rapid changes in intensity with time are very common, as shown on Tables 1 and 2, and by BLANCHARD (1953; see fig. 2). Most of this change is thought to be due to winds transporting the showers over the sampling stations. For instance the average clouds which were producing the showers in the present study were probably moving at about 7 meters per second up the mountain slope. During a sixty second interval between rain samples a shower-producing cloud could thus move almost one-half kilometer. The showers are seldom large in diameter, and in aircraft a flight of one-half kilometer is often sufficient to carry the observer from the misty peripheral regions to the relatively intense rains of the center.

The interpretation of the measurements presented here, in terms of the details of the raindrop-forming processes operating in these Hawaiian rains, is uncertain. They seem, however, to be clearly useful in narrowing the areas of search for these "details". One can say from the present measurements that a drop-forming process seems to be required which will, (a), involve most of the larger salt nuclei in a simultaneous growth, (b) cause this growth to occur, presumably very rapidly, in a manner which prevents marked changes in the distribution of the nuclei (or drops), and (c) add water to the nuclei without at the same time greatly adding to their salt content.

**Table 7.** Weights of sea salt added to a 0.9 mm diameter raindrop due to coalescence with 10 and 20  $\mu$  radius cloud droplets containing sea-salt nuclei of various sub-micron sizes

| Cloud droplet radius $\mu$ | Cloud droplet no. required to form 0.9 mm diameter raindrop | Weight sea salt ( $10^{-12}$ g) in 0.9 dia. raindrops<br>(cloud droplet no $\times$ nucleus wt.) |                              |                              |                               |                               |
|----------------------------|-------------------------------------------------------------|--------------------------------------------------------------------------------------------------|------------------------------|------------------------------|-------------------------------|-------------------------------|
|                            |                                                             | (0.5 $\mu$ )<br>$10^{-12}$ g                                                                     | (0.2 $\mu$ )<br>$10^{-13}$ g | (0.1 $\mu$ )<br>$10^{-14}$ g | (0.05 $\mu$ )<br>$10^{-15}$ g | (0.02 $\mu$ )<br>$10^{-16}$ g |
| 10                         | 91,000                                                      | 91,000                                                                                           | 9,100                        | 910                          | 91                            | 9.1                           |
| 20                         | 11,400                                                      | 11,400                                                                                           | 1,140                        | 114                          | 11.4                          | 1.1                           |

In the present study the quantities of salt in the individual large salt nuclei are adequate to account for all of the salt found in the rains. This suggests that the drops have grown entirely by condensation processes, or through coalescence with relatively chloride-free cloud droplets<sup>1</sup>. If one assumes, for instance, that the 0.9 mm diameter raindrops (which, on rain curve 2, fig. 2, are supposed to have formed on 2,100  $\mu\mu$ g salt nuclei) have grown through coalescence with 10  $\mu$  radius cloud droplets, about 91,000 of these droplets would be required. Table 7 shows amounts of salt which would be added to the initial giant (2,100  $\mu\mu$ g) nucleus due to coalescence with cloud droplets which have formed on sea-salt nuclei of various sub-micron sizes. On this table it is evident that the quantities of salt added through accretional growth might in some instances equal or greatly exceed the weight of the giant nucleus upon which the raindrop initially formed. The very small nuclei would, of course, require rather large water vapor supersaturations in the clouds to become activated.

Junge's work at Round Hill (JUNGE, 1954) indicates that most of the numerous nuclei which are less than about 0.8  $\mu$  in radius (or  $< 5 \times 10^{-12}$  g, as shown on fig. 2) are composed largely of ammonium sulfate. However, in Hawaii Junge recently sampled aerosols which were less than 0.8  $\mu$  radius, and he found relatively very little ammonium sulfate present.

<sup>1</sup> Hawaiian clouds, which are not producing rain, have been found to contain from 30 to 140 cloud droplets per cubic centimeter, though the air at sea level usually contains several hundred Aitken nuclei in an equal volume.

From the results of this paper, we cannot say that the raindrops in Hawaii do or do not grow by accretion. If the nuclei smaller than about  $10^{-12}$  g are sea salt, then accretion would seem to be ruled out unless the nuclei are very small (see Table 7). Thus we feel that one of the important steps in future physical-chemical studies of raindrop formation in shower clouds is to determine the chemical nature and the number of the majority of the individual cloud-droplet nuclei which are smaller than about 0.8 microns and to relate these particles to the solutes found in the cloud and rain waters.

A conservation of the number of the larger nuclei per unit volume of air, during their growth to raindrops, is indicated by the results given here. This number conservation is not consistent with the model proposed by BOWEN (1950), in which accretional growth involves marked alterations in drop distributions. If these Hawaiian raindrops grow largely by accretion, or by condensation, the authors feel that it is necessary to suppose that this growth occurs very rapidly in turbulent volumes of air which are ascending within clouds in a way similar to the "bubble parcels" proposed by SCORER and LUDLAM (1953).

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