Study on Dry Fallout and Its Distribution of Giant Sea-Salt Nuclei in Japan

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Abstract

Weight-number distribution of giant salt nuclei which fell to the ground in Kyoto was determined every night from Jan. 4 to Feb. 3, 1962. Similar observations were also made at the same time at several stations along a transversal cross-section of Japan in the direction of the winter monsoon, at a few nights in the above period. From analysis of the data, it was concluded that giant salt nuclei in the atmosphere are primarily produced from the sea surface through an air-sea boundary process, and carried over the land by wind, as predicted by Toba (1961) based on studies of a model system utilizing a wind flume. The weight-number distribution curve, however, varied day by day and it is suggested that the accumulation of data concerning sea-salt nuclei distribution may afford some clue for further understanding of the precipitation mechanism.

Rate of the total dry fallout of sea-salt was also calculated from the above data. It was between $3 \times 10^{-12} - 2 \times 10^{-14}$ gm cm$^{-2}$ sec$^{-1}$.

1. Introduction

Sea-salt nuclei in the atmosphere are considered to be produced on the sea surface, as a result of the boundary process between air and sea. They act as condensation nuclei of water vapor, and it is known that the larger nuclei, those that contain sea-salt more than $10^{-11}$ gm in weight, can initiate rain from warm clouds, especially in tropical and subtropical oceanic areas. They are called giant sea-salt nuclei. However, it is not known how, and to what extent they participate in the phenomena of precipitation, especially in such moderate latitudes as Japan.

To clarify the role of sea-salt nuclei, it is necessary to know the actual amount of sea-salt nuclei in the air under various atmospheric and oceanic situations. One approach is to study aspects of production of the sea-salt nuclei on the sea surface. One of the authors (Hayami and Toba 1958, Toba 1959, 1961, 1962) studied the mechanism and the rate of production of sea-salt particles from wind waves by the use of a wind flume, the manner of their distribution in the atmosphere and the transportation to the land. The other way is to observe sea-salt nuclei directly in the atmosphere under various conditions. As a preliminary step, we made a series of observations of giant sea-salt nuclei fallen to the ground in Kyoto and other places along a transversal cross-section through the Kinki District of Japan, when the winter monsoon through the Sea of Japan prevailed over the district.

Fallout of sea-salt nuclei seems also to be the main source of salt in natural water. This is a problem of geochemistry. During strong winds, there is much salt fall near the coast, which becomes an important factor in ecology of coastal plants (Boyce 1954, Nobuhara et al. 1962). Furthermore, when strong typhoons strike coastal districts, power transmission lines are often affected by serious salt damage, as far as 100 km from the coast. Kita and Aya (1959) studied the distribution of the rate of fall of salt in mass to the ground within 1 km from the coast. There seems to be, however, no observation of weight and number of individual sea-salt particles falling to the ground at various distances from the coast under different meteorological situations.
This study was intended as an approach to the boundary process between air and sea, meteorological significance of sea-salt nuclei, and fundamentals of salt damage.

2. Procedure of the observation

(a) Sampling surface

A halide ion-sensitive film prepared by the method of N. H. Farlow (1954, 1957) was used as the sampling surface of sea-salt nuclei. It was made from a commercial Fuji Gravure Safety Film Normal, having a gelatin layer of 11.0 µ in thickness. The principle of the use of the film is as follows. A gelatin layer on the film contains silver dichromate gel, and when a particle of sea-salt precipitates on the film surface, it diffuses into the gelatin layer, and the chloride ions precipitate as silver chloride, producing a white circular spot on the reddish brown base of the film. We can determine the mass of chloride by measuring the size of the spot.

After the white spots developed, we prevented further attachment of salt particles on the sensitive film by dipping the film into a dilute solution of collodion and drying it. The white spots were measured by a moving microscope with 1 µ vernier.

(b) Calibration of the sampling film

To understand the relation between the size of the spot of silver chloride on the film and the mass of sea-salt nucleus, the films prepared were calibrated. The principle of the calibration technique was to measure the size of spots developed from fog droplets of NaCl solution of known concentration, the size of which could be known by their terminal velocity in a space where the vapor pressure was kept equal to the equilibrium vapor pressure on the surface of the fog droplets.

Glass cylinders of either 135 cm or 45 cm in length were held vertically, inside of which several streaks of filter paper were hung, and the lower edges were dipped into small beakers containing the same NaCl solution. A window and shutter were made at the top as well as at the bottom of each cylinder. A box was placed on the upper window and the film to be calibrated was placed just under the lower window. After fine fog droplets were produced in the box by the use of a medical atomizer, from the same NaCl solution as that in the small beakers, the upper shutter was opened for a moment and, after a measured time interval, the lower shutter was opened. Droplets that had fallen through the length of the cylinder during the time interval were selectively collected by the film. Two solution concentrations and several time intervals were used to obtain various cases of the mass of NaCl contained in the droplets. In this experiment, the space within the cylinder was presumed to have been kept at a constant vapor pressure for each NaCl solution as the experiment was carried out after a considerable time had elapsed since the filter paper and beaker containing specific NaCl solution were set, and after many fog droplets were produced in the space with the same solution. Consequently, the fog droplets are considered to have fallen with constant terminal velocities, without evaporation and condensation. Knowing the velocity of fall, we determined the diameter and volume of the fog droplets by using the formulae of C. N. Davies (1945); by knowing the diameter and volume we were able to determine the quantity of Cl contained in the fog droplets from the concentration of the solution. The relation we obtained between the size of the spot on the film and the weight of the sea-salt nucleus is shown in Fig. 1.

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(c) Procedure of sampling

The sampling film (about 20 cm²) was set at the bottom of an empty can (6 cm in diameter, and 6.5 cm in height), and exposed horizontally to the air near the ground for one night. Silica gel grains were placed under the film in the can to prevent dew or frost formation on the surface of the film. The can was set on the ground under a shelter also to prevent dew or frost formation and to protect from slight precipitation. The cap of the can was open during the night and the opening and closing times were recorded.

The effect of the can for depositic efficiency was checked once by comparing two films—one set in the can and the other set on a wire net, exposed on a warm night. The number of sea-salt nuclei that fell on the film in the can was only 10% or so smaller than that on a net.

(d) Place and date of the observation

Observations were made at the top of the Geophysical Institute of Kyoto University from Jan. 4 to Feb. 3, 1962. Similar observations were made at Maizuru Marine Observatory (Kyoto Pref.), Wachi Junior High School (Kyoto Pref.), Sonobe High School (Kyoto Pref.), Ueno Weather Station (Mie Pref.) and Matsusaka High School (Mie Pref.) on Jan. 12, 14, 16, 19, 24 and Feb. 2, and Kasumi High School (Hyogo Pref.) on Jan. 19, 24 and Feb. 2. The locations of these stations are shown in Fig. 2. An observation was tentatively made at Matsumoto Weather Station (Nagano Pref.) which is located in the most inland district of Japan.

3. Daily change of the dry fallout of sea-salt nuclei in Kyoto

(a) General trend

Rate of the total dry fallout of sea-salt nuclei measured in Kyoto is shown in Fig. 3. The lack of data for earlier days was due to damage done to the film by dew or frost (the frost-protecting procedure described before was introduced after several days). The lack of data on Jan. 19 and Jan. 23 was due to heavy snow. The figure also shows wind speed and wind direction at 1500 m level above Yonago, and daily mean precipitation in Kyoto Prefecture. The wind speed is illustrated as a running average for 24-hour periods. The wind direction was, generally, WNW throughout the period, as shown in the figure. The wind at that level was compared to that at the same level above Shionomisaki, and we felt the wind shown in the figure was an indication of the strength of the winter monsoon over the Sea of Japan. The rate of fallout corresponds to the wind speed, except during the last part of January. This indicates that the change in production rate of sea-salt nuclei in the Sea of Japan, which is governed by the intensity of the monsoon, directly reflected the dry fallout of sea-salt nuclei on land. In the last part of January, the precipitation shown in the figure represents a very severe snowfall, and we see that the snow reduced the seasalt nuclei number to about one third of the expected values during snow free periods.

(b) Relation between number of sea-salt nuclei and wind speed

As will be shown in the next section, the fallout rate of smaller sea-salt nuclei does not seem to change significantly according to distance from the coast, so that the fall-out rate of smaller nuclei in Kyoto during snow free periods depends primarily on the production rate of them on the surface of the Sea of Japan, even though wind direction or wind speed changes. We selected the period of Jan. 11-21, and plotted the fallout rate of sea-salt nuclei of log $m < 1.75$ (cf.
Fig. 3. Rate of the dry fallout of sea-salt nuclei observed in Kyoto; wind speed (running average for 24-hr periods) and wind direction at 1500 m level above Yonago; and daily average precipitation in Kyoto Prefecture.

next section), where $m$ represents weight of sea-salt nuclei in $10^{-12}$ gm unit, in Fig. 4 (left) together with the wind speed above Yonago. The relation between the two curves is more obvious in the right figure. This seems to indicate that sea-salt nuclei suddenly increase in number when wind speed exceeds several meters per second. We plotted this relation in a semi-logarithmic chart in Fig. 5. It seems to show that the dependence of the production rate of sea-salt nuclei on wind speed, (predicted by Toba (1961) following a

Fig. 4. Relation between rate of the dry fallout of sea-salt nuclei of log $m < 1.75$ observed in Kyoto, and wind speed (running average for 24-hr periods) at 1500 m level above Yonago, in a generally snow free period.

Fig. 5. Extrapolation of the dependence of the production rate of sea-salt nuclei $F$ (number/sec cm$^2$) on wind speed at 10 m level above sea surface $u$ (m sec$^{-1}$) : $F \propto u^{0.40}$, predicted by Toba (1961), and replotting of circles in Fig. 4.
wind flume experiment for wind speed at 10 m level above sea surface) can be extrapolated to several meters per second in wind speed, if we take into account that the wind speed at 10 m level is somewhat lower than that at 1500 m level. Toba's conclusion, however, was that there is a critical wind speed at about 13 m sec\(^{-1}\) at 10 m level, and above this speed the production rate followed the indicated relation. The present study includes only cases of wind speed below 13 m sec\(^{-1}\) at 1500 m level, and we do not have exact information about the relation between wind speed at 1500 m level and that at 10 m level. Further studies should reach conclusive concepts on this.

4. Change of the dry fallout of sea-salt nuclei with distance from the coast

In order to know the horizontal distribution of sea-salt nuclei on land, observations were made at several stations along the direction of the winter monsoon on the same days, as were described in Section 2 (d). Among the observations, those on Jan. 16-17 and Feb. 2-3 were analyzed, since complete sets of the sample were available for these days only.

(a) Observation on Jan. 16-17, 1962

Meteorological conditions on the night of Jan. 16 may be seen in Fig. 3. The wind at 1500 m level above Yonago was about 16 knots in speed and from W to WNW in direction. There was a slight trace of precipitation in Kyoto Prefecture. The fallout rate of sea-salt nuclei for each grade of weight at 6 stations is shown in Fig. 6. The curves have a characteristic feature—a peak at about log \(m=1\) to 2. In the upper graph, it is seen that the peak moves left as distance from the coast increases. This seems to indicate that the number of smaller nuclei does not change much according to distance.
from the coast as the fall velocity is small, while the number of larger nuclei decreases. This relation may be seen in Fig. 7 where the fallout rate is illustrated for two ranges of the nuclei weight, divided by the line "a" in Fig. 6 (log $m<1.75$ and log $m>1.75$), and further for a narrow range between the line "a" and "b" (1.75 < log $m$ < 2.50), as a function of distance from the coast in a strictly WNW direction. As is seen in the figure, the fallout rate of smaller nuclei was of the same order at all stations, while that of larger nuclei gradually decreased from Maizuru to Kyoto. The conspicuous feature of Fig. 7 is, first, that the values were very high at Ueno. Inspection of the figure will remind one that Ueno is nearer to the coast than is indicated in the figure, and it is assumed that nuclei at Ueno entered from Osaka Bay. In the second place, the tendency of ups and downs of the three graphs is in the same direction. This seems to indicate that the ups and downs that were not related to the distance from the coast were caused by some local effect such as humidity, structure of air motion according to the topography, etc. Actually, the height of each station above sea level that is closely related to humidity, is entered in the top graph of Fig. 7, and the ups and downs are similar to those of the three graphs.

Based on the above consideration, the points of the top graphs were leveled, and points of the other graphs were modified by the use of the same reduction factors as those used in the leveling of the top graphs for each station, the distances to Ueno and Matsusaka were taken from the coast of Osaka Bay. Then the circles in Fig. 8 were obtained, corresponding to the bottom graph of Fig. 7 as shown below, and they appear in good series, although the leveling of the points of log $m<1.75$ doesn’t seem to have a specific meaning.

Fallout rate $P$ of sea-salt nuclei of fall velocity $w$ can be expressed as a function of distance from the coast $x$ by the following equation (Toba, 1961),

$$P = \frac{w}{\theta_0} \cdot \frac{1}{\sqrt{\pi \gamma u x}} e^{-\frac{w^2 x}{\gamma u x}}$$

$$- \frac{w}{2\gamma u} \text{erfc} \left( \frac{w \sqrt{x}}{2 \sqrt{\gamma u}} \right) dx$$

where $\theta_0$ represents the number of sea-salt nuclei in the air of unit volume near the ground, $F$ production rate of the nuclei on the sea surface, $u$ wind speed, $\gamma$ mixing coefficient in the atmosphere which is now taken as constant. We tried to derive the distribution of fallout rate of sea-salt by the above equation from the production rate of the nuclei on the sea surface. The curve $F$ in Fig. 9, as a function of weight of the nuclei $m$, was derived as an extrapolation of the data of the wind flume experiment by Toba (1961) to wind speed of 10 m sec$^{-1}$ at 10 m level above the sea surface (cf. Section 3 (b)). The calculation curves for $x=50$ km and $x=150$ km were obtained by the above equation with values of $\gamma u=10^6$, $w$ for sea-salt particles that are in equilibrium state in the air of RH=80%, and $F$ indicated by the curve of $F$. The black circles and crosses are entered as the dry fallout rate at Maizuru and Kyoto, respectively, that are values modified from Fig. 6 in the same way as described several passages before.
The curve "a" in Fig. 8 shows the same calculation with values of $\eta u=10^6$, RH=80% and $F$ for $1.75<\log m<2.5$. Black circles were entered by calculating $\theta_0=P/w$ from the bottom graph of Fig. 7, by assuming RH =80%.

The calculation and the observation appear consistent, and it seems that the study of production of sea-salt nuclei by Toba (1961) approximated the actual phenomena, and the transportation of the nuclei may be expressed, to a considerable degree, by the above equation. However, it must be noted that, aside from the problem noted in the last part of Section 3 (b), there is some question as to the values used as the constants. For example, if we use $w$ for $\log m=2.05$, and RH <75% where sea-salt nuclei are dry particles, and $\eta u=10^5$ and $10^6$, respectively, we will obtain the curves "b" and "c", respectively, provided that we consider $F$ as unknown and draw curves through nearly middle points of white circles in Fig. 8, where white circles also show the condition for RH<75%. It should be noted that if we use the same value of $w$ with "b", and $\eta u=10^6$, we will obtain the curve "d". It may be said, however, that the mixing coefficient $\eta$ was of the order of $10^2*10^3$ cm$^2$ sec$^{-1}$, because wind speed $u$ was about $10^3$ cm sec$^{-1}$.

(b) Observation on Feb. 2-3, 1962

Fig. 10 shows the observation at 7 stations from Kasumi to Matsusaka on Feb. 2-3. In this case, the circumstances were more complicated, and it seems that there are different types of weight distribution curves. This may have been caused by the condition where wind speed was steeply decreasing on those days as seen in Fig. 3, and by some local precipitation. These points will be expanded upon in the following section.

5. Shape of the weight distribution curves of dry fallout of sea-salt nuclei and precipitation mechanism

Weight distributions of the rate of dry fallout of sea-salt nuclei in Kyoto from Jan. 11 to Feb. 3, 1962, and one example taken at Matsumoto, the most inland spot in Japan, are shown in Fig. 11. In Figs. 10 and 11, we see that the curves show very interesting behavior from day to day, and that there are several types in the curves. For example, type 1 — this can be seen on Jan. 11 in Kyoto, and this is tentatively considered as the normal type, type 2 — a case where smaller nuclei of $\log m<2$ are cut off as on Jan. 18 and 29 in Kyoto, type 3 — a part between $\log m=2$ and 3 is much reduced as on Jan. 17, 30, 31 in Kyoto, and Feb. 2 at Wachi (Fig. 10), it seems that curves of type 2 and 3 arose from cutting off of the sharp peak at around $\log m=1.5$ or the middle part of $\log m=2-3$ by snow forming mechanism or some other factor, type 4 — a very sharp peak appears at around $\log m=1.25-1.5$ as in the case of Kasumi and Maizuru on Feb. 2 (Fig. 10), this seems to have arisen from the rapid falling of larger nuclei because of the sudden weakening of the wind.

If we accumulate much data of this type, the shape of weight distribution curves of dry fallout of sea-salt nuclei may afford some clue in understanding the precipitation mechanism.
6. Rate of total dry fallout of sea-salt

Total mass of sea-salt nuclei falling to the ground is contributed to by larger nuclei because of their larger mass and greater fall velocities, although their number is small compared to smaller nuclei. So, for the study of dry fallout of sea-salt, it is more convenient to observe dry fallout itself of individual sea-salt nuclei than to observe the concentration of them in air.

The rate of total dry fallout of sea-salt was calculated from the above data on the rate of fallout of sea-salt nuclei for each grade of weight. The values are listed in Table 1. They were of the order of $10^{-12} - 10^{-14}$ gm cm$^{-2}$ sec$^{-1}$.

Table 1. Observed rate of total dry fallout of sea-salt.

(a) Daily change observed in Kyoto (Jan. 11-Feb. 3, 1962)

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<tr>
<th>Date</th>
<th>Jan. 11</th>
<th>12</th>
<th>13</th>
<th>14</th>
<th>15</th>
<th>16</th>
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<th>20</th>
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<td>Fallout rate (10$^{-12}$ gm cm$^{-2}$ sec$^{-1}$)</td>
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<td>0.15</td>
<td>2.2</td>
<td>0.087</td>
<td>0.13</td>
<td>0.28</td>
<td>0.67</td>
<td>0.61</td>
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<td>0.099</td>
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<th>28</th>
<th>29</th>
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<th>31</th>
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<th>2</th>
<th>3</th>
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<tr>
<td>Fallout rate (10$^{-12}$ gm cm$^{-2}$ sec$^{-1}$)</td>
<td>0.36</td>
<td>0.20</td>
<td>0.28</td>
<td>0.11</td>
<td>0.14</td>
<td>0.76</td>
<td>3.1</td>
<td>2.4</td>
<td>0.19</td>
<td>0.033</td>
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(b) An example of the horizontal distribution (Jan. 16, 1962)

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<th>Wachi</th>
<th>Sonobe</th>
<th>Kyoto</th>
<th>Ueno</th>
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<td>0.34</td>
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<td>0.28</td>
<td>0.60</td>
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(c) An example of the horizontal distribution (Feb. 2, 1962)

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<tbody>
<tr>
<td>Fallout rate (10$^{-12}$ gm cm$^{-2}$ sec$^{-1}$)</td>
<td>1.3</td>
<td>0.91</td>
<td>2.0</td>
<td>0.85</td>
<td>0.19</td>
<td>0.34</td>
<td>0.14</td>
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Fig. 11. Weight distributions of the rate of dry fallout of sea-salt nuclei in Kyoto from Jan. 11 to Feb. 3, 1962, and that at Matsumoto on Jan. 4, 1962.

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References


本邦における巨大海塩核のドライフォールアウトとその分布に関する研究

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1962年1月4日から2月3日まで、京都において毎夜巨大海塩核のドライフォールアウトの観測がなされた。また、期間中数回、季節風の方向に本州を横断して分布する数か所の観測点で同時観測がなされた。その結果、海塩核は鳥羽（1961）が風洞実験による研究から推定したように、大気海洋の境界過程として海面で生成され、風にとって陸上に運ばれることが明らかとなった。しかし、その降雨率の重量分布曲線の形は日々かなり面白く変化し、海塩核分布の資料の集積は、降水量機構解明に関する一つの手がかりを与えるであろうことが示唆される。

またこの資料から、ドライフォールアウトによる降塩率を計算された。それは$3\times10^{-12}$～$2\times10^{-14}$gm cm$^{-2}$ sec$^{-1}$の範囲にあった。