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## Observation of Aerosol Particles Attached to Falling Snow Crystals, Part II, Utilizing an Electron Microscope

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### Abstract

Aerosol particles attached to falling snow crystals were observed in Sapporo, March 1974, utilizing an electron microscope; the size distribution was also observed.

As a result, it was found that the mode of size distribution coincided with that of Junge's distribution. And the collection efficiency of aerosol particles by falling snow crystals was estimated to be as large as several tens in percentage. These facts show that the snow crystals captured aerosol particles of submicron size at a much higher rate than generally believed, during their fall.

At present, we have no possible mechanism to explain such a high collection efficiency. It is our hope that all mechanisms related to aerosol entrapment may be re-examined.

### 1. Introduction

As previously reported by Magono, Ueno and Kubota<sup>1)</sup>, the size distribution of aerosols attached to falling snow crystals was measured under an optical microscope mainly in a range greater than  $1 \mu$  diameter. As well known however, the resolution power of the optical microscope was insufficient to measure the size distribution of aerosols of submicron size.

Accordingly the authors observed aerosols using an electron microscope. In the measurement, attention was mainly paid to following three points.

- a) Avoidance of coagulation of aerosols in sampling and photographing processes.
- b) Distinction of aerosols attached to snow crystals from background dust particles.
- c) Identification of observed location over the entire area of an original snow crystal.

Because this work was done for the purpose of measuring the washout effect of aerosols by falling snow crystals, the condensation nuclei of rimed cloud droplets were excluded, in other words, only non-rimed snow crystals

were observed.

## 2. Sampling method of aerosols attached to falling snow crystals

In order to concentrate on the three points described above, a polyvinyl formvar solution was considered to be suitable, at first for the fixation of the surface of snow crystals. In this case however, it was required that the solution is both extremely thin and dilute, otherwise the electron beam can not penetrate the film in which aerosols are embedded. Therefore the actual sampling of snow crystals was made in the following manner, as schematically shown in a series of Fig. 1.

- a) A clean glass plate is coated as thin as possible with a dilute polyvinyl formvar solution of 0.1%.
- b) A falling snow crystal is sampled about 30 sec after coating the plate,

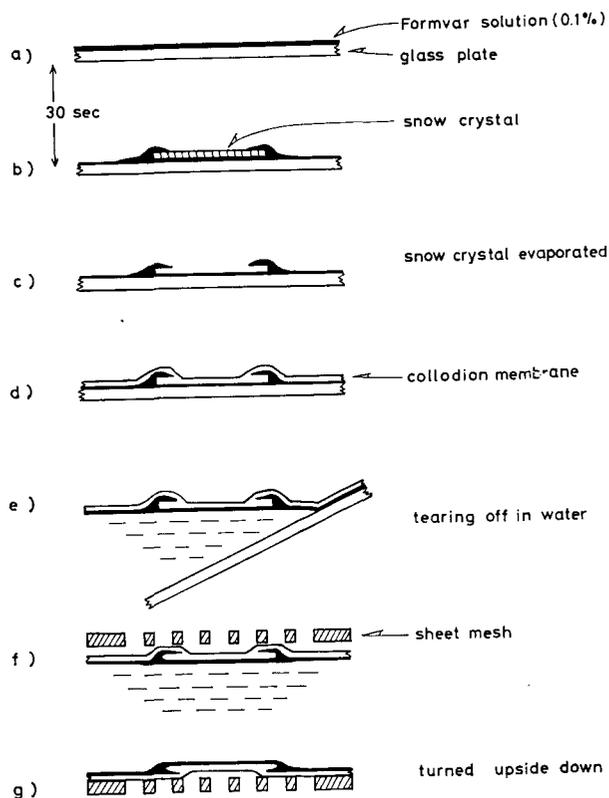


Fig. 1 Replication process by sandwich technique.

before the coated film dries up. Because the film was so thin, it could not cover the entire upper surface of the snow crystal.

- c), d) After the evaporation of the snow crystal, the replica is coated again from above by a collodion membrane of 2% solution in amyle acetate used in usual electron-microscopic observations.
- e) The replication film is gradually torn off from the glass plate with the aid of collodion membrane in clean water.
- f) A sheet mesh for the electron-microscopic observation is placed on the replicated film floating on clean water.
- g) The replication film is taken off from the water by quickly inverting it.

This fixing technique by sandwiching with two films of polyvinyl formvar and collodion is quite tedious. In addition, later it was found that aerosols were oozed out from the replication area to the neighbouring area in the later half of the fixing process. Therefore the process was changed to another resin technique found by Smith-Johannsen<sup>2)</sup>, as described schematically in the following series of Fig. 2.

- a,) A sheet mesh on a glass plate is covered with a collodion membrane by the usual method.
- b), c,) After sampling a snow crystal on the sheet mesh, the mesh is placed in a capped dish. The inner side of the cap is previously coated by a resin of methyl 2-cyanoacrylate monomer (commercially Eastman 910

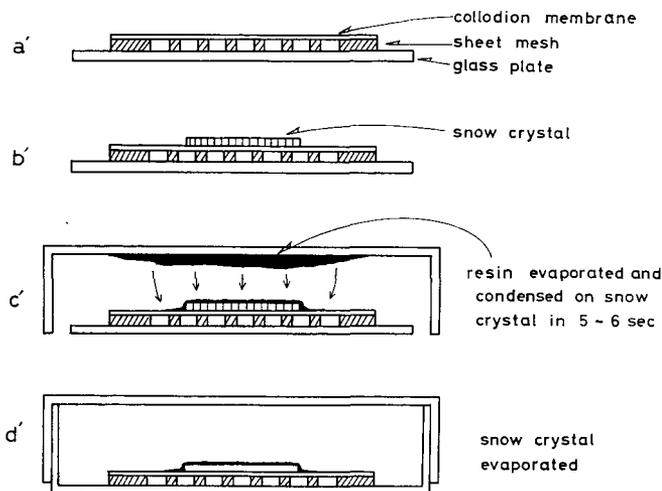


Fig. 2 Replication process by resin adhesive technique.

Adhesive).

d.) In several seconds, the resin is evaporated from the dish, and then condensed on to the surface of the snow crystal. After the evaporation of the snow crystal, a replication is obtained in which aerosol particles are fixed.

This technique was easier than the previous sandwich technique, but frequently the collodion film was broken during the observation under the electron microscope. Furthermore the resin vapor condensed not only on the sampled snow crystal but also on frozen water droplets on the surface of the sampling glass plate. Therefore the trace of the droplets become ghost particles in electron-microscopic photographs, if the vaporizing time for the resin is prolonged.

The resolution power of aerosol particles by the electron microscope was  $0.05 \mu$ .

### 3. Result

#### 3.1 *Size distribution of aerosols*

Falling snow crystals were sampled for the purpose of measuring the size distribution of aerosol particles attached to the snow crystals using an electron microscope, however we succeeded in only a few cases owing to the difficulty in replication and fixing process.

One of replicas of a snow crystal of plane dendritic type obtained by the sandwich technique is shown in Photo. 1 in Pl. I. It may be seen that the snow crystal was not rimed. Hereinafter this snow crystal will be called Crystal *A* in this paper. A portion of a branch of the crystal *A* indicated by an arrow *a* in the photograph was photographed under an electron microscope, as shown in Photo. 2. To check the background dust particles, another portion outside of the branch indicated by an arrow *a'*, was also photographed under the electron microscope, however no dust particles were detected.

The size distribution of aerosols attached to the snow crystal *A* in Photo. 2 is shown in a form of percentage at intervals of  $0.05 \mu$  in Fig. 3. The total number of observed aerosols *N* and the area of surface used for the measurement *S* are given in the upper portion of the figure. It may be seen that the frequency of aerosols increases as their size decreases down to  $0.1 \mu$  in diameter. The maximum seems to be near  $0.1 \mu$  diameter. The decrease in the range smaller than  $0.05 \mu$  is only apparent, because this range is beyond the resolution power of the microscope used.

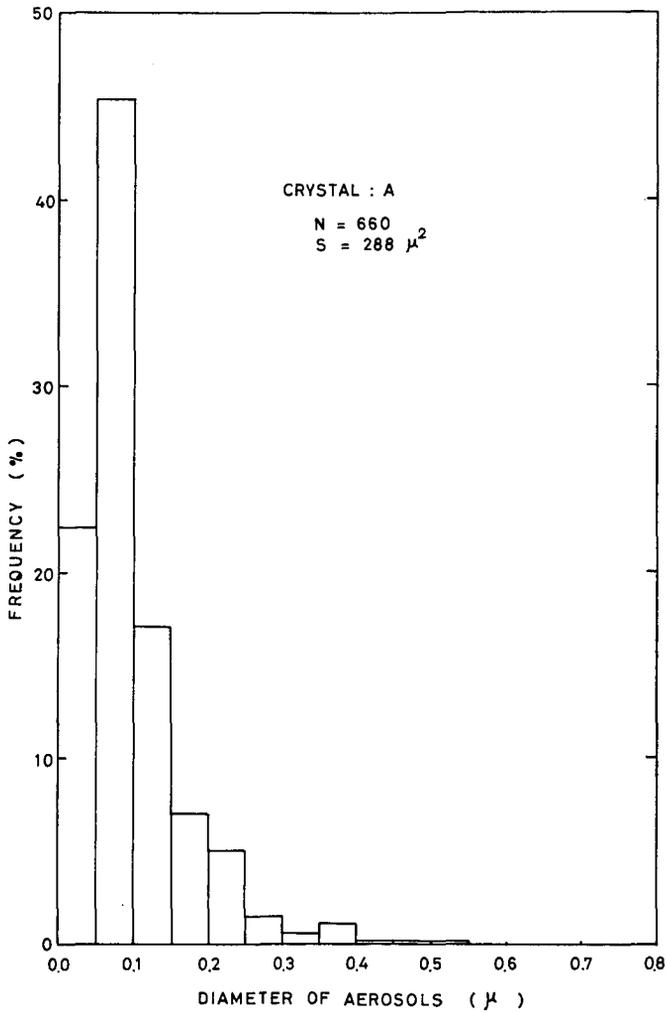


Fig. 3 Size distribution of aerosols attached to Crystal A.

Another example of a snow crystal replicated by the sandwich technique is shown in Photo. 3, Pl. II. This snow crystal will be referred to as Crystal B in this paper. It seems that this crystal was somewhat shrunken in the replication process, however the shrinkage may be ignored. The electron microscopic photograph of a portion indicated by arrow *b* in the photograph is shown in Photo. 4, and the size distribution counted from this and other

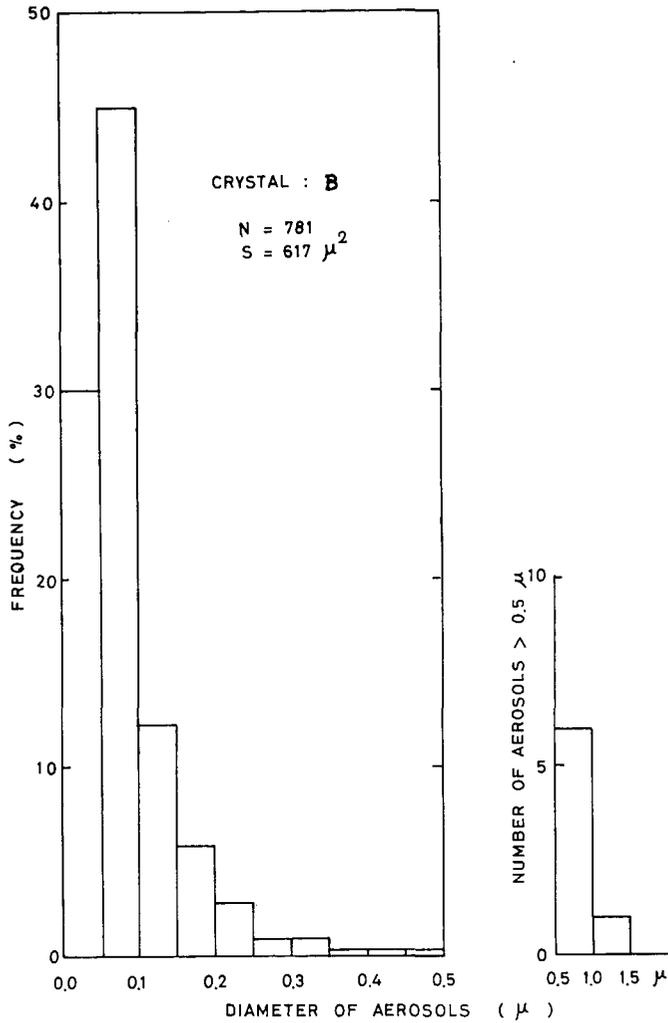


Fig. 4 Size distribution of aerosols attached to Crystal B.

neighbouring photographs are shown in Fig. 4. The distribution of aerosols with a size greater than  $0.5 \mu$  is given at the right end of the figure in their original numbers. It may be seen that the mode of the distribution is entirely similar to that in Fig. 3, except for the small number of aerosols larger than  $0.5 \mu$  in diameter.

Snow crystal C was replicated by the Eastman adhesive technique.

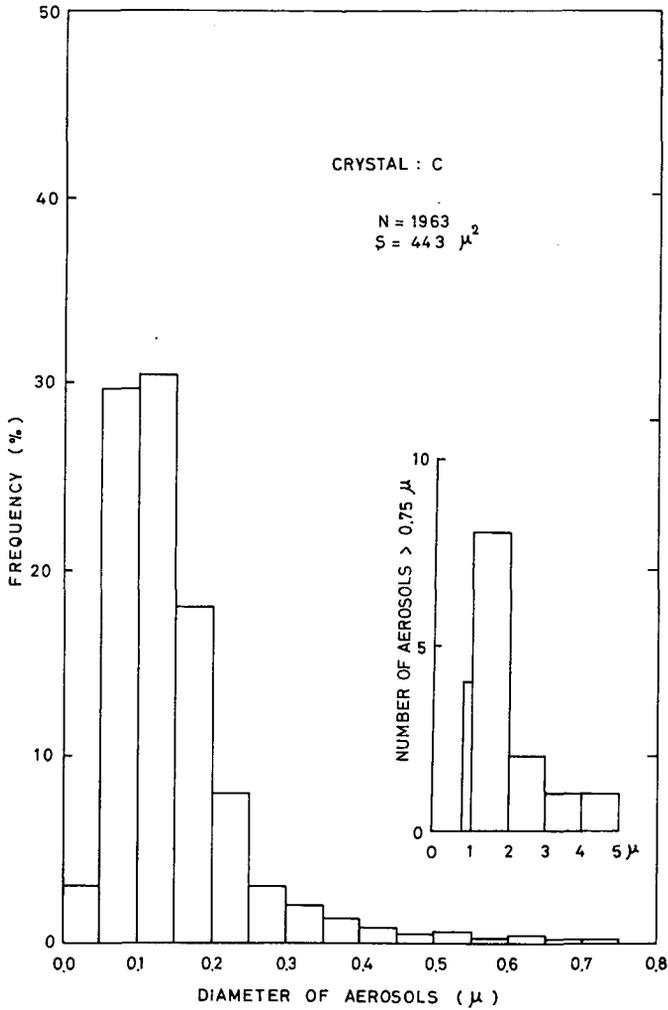


Fig. 5 Size distribution of aerosols attached to Crystal C.

It was of non-rimed plane dendritic type similar to Crystal A, although its entire shape was not photographed. Aerosols inside and outside of replicated area of crystal C are shown in Photos. 5 and 6 in Pl. III, respectively. A small number of minute particles are seen in Photo. 6 in spite of being outside of the snow crystal area. However these particles are not considered as aerosols, but ghost particles which were produced in the Eastman adhesive

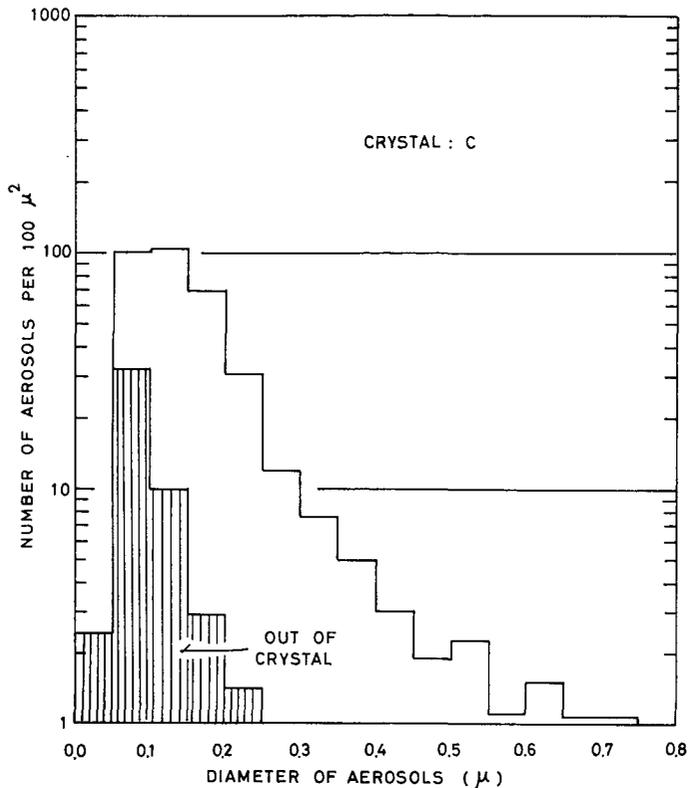


Fig. 6 Size distributions of aerosols and ghost dust particles.

replication process, as described previously. The size distribution of aerosols counted from Photo. 5 and the neighbouring photographs are shown in Fig. 5. The mode of the distribution was substantially the same as those in Figs. 3 and 4.

The size distribution of ghost particles counted from Photo. 6 is shown by a shaded histogram in Fig. 6, together with that of real aerosols. Because the size of ghost particles are limited to a range below  $0.25 \mu$  and their numbers are one order smaller than those of aerosols, the effect of ghost particles on the size distribution of aerosols may be neglected, even if ghost particles are mistakenly counted into the number of aerosols.

The final size distribution counted from all snow crystals *A*, *B* and *C* is shown in the form of a semi-logarithmic diagram in Fig. 7. It may be seen that the number of aerosols smoothly increases as their size decreases down

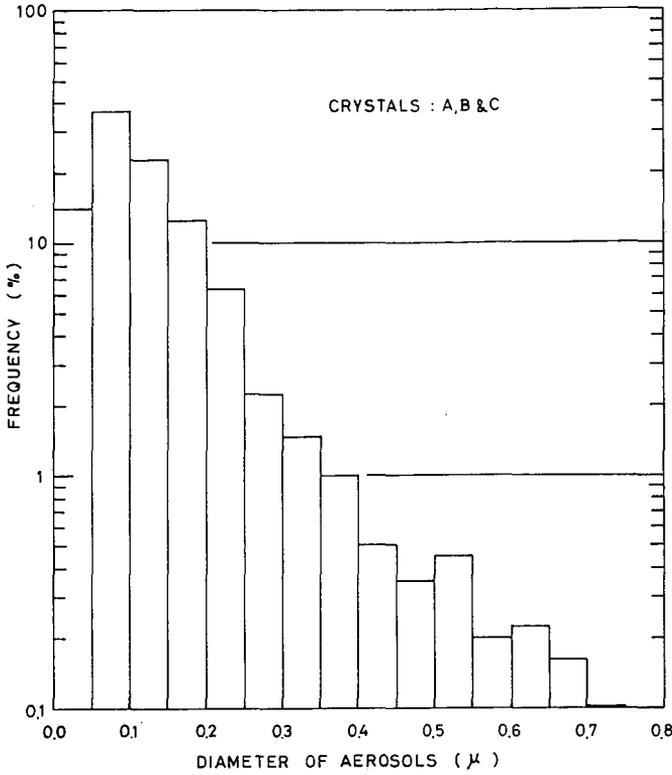


Fig. 7. Size distribution of aerosols attached to Crystals *A*, *B* and *C*.

to 0.1  $\mu$ .

Junge<sup>3)</sup> measured aerosols in various size ranges in air by various methods, and pointed out that in a range from 0.1 to 10  $\mu$ , the size distribution can be described by a straight line in the logarithmic diagram, more exactly  $dn/d\log D$  is proportional to  $D^3$ , as shown by a chain line to the right hand in Fig. 8 where  $n$  and  $D$  mean the space number density and the diameter of aerosols. In order to compare the present result with his distribution, the size distribution described in Fig. 7 was transformed to a surface density, utilizing the area of observed surface of snow crystals *A*, *B* and *C*, then plotted by black dots in logarithmic scale on the left hand in Fig. 8. The surface number density corresponds to the space number density in this case, because the observed aerosols attached to the snow crystals were swept off by the snow crystals when the crystals were falling through space.

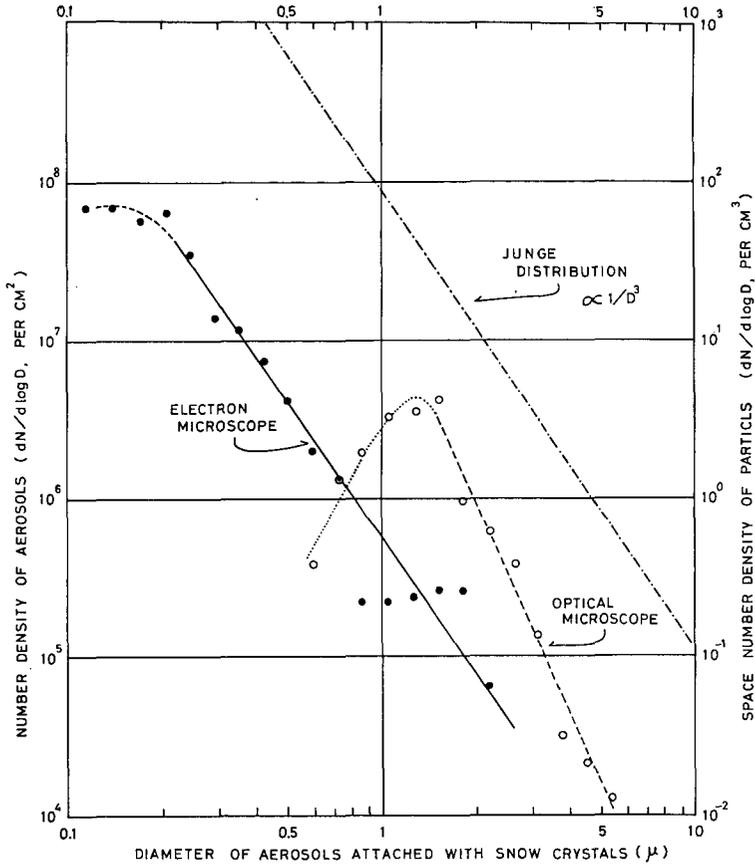


Fig. 8 Size distributions of aerosols observed by electron microscopic and optical microscopic observations, and Junge distribution.

In Fig. 8, it will be seen that the black dots are distributed on a straight line (solid line in the figure) parallel to Junge's distribution, although they are fairly scattered in a range greater than  $1\mu$ . This scattering in the greater range was caused by the insufficient number of data. In case of the optical microscopic observation<sup>1)</sup>, the distribution was described by a straight line in a range up to  $5\mu$  at least, as seen in the broken line in the figure.

Both in the cases of the electron microscope and the optical microscope observations, the number of aerosols seems to be smaller than those expected by the respective straight lines in the smaller range ( $<0.2\mu$  in electron microscope,  $<1.5\mu$  in optical microscope). These differences originated

from the transformation process from linear scale description to logarithmic scale description, because the number of counted aerosols in a smaller range was extremely small in the logarithmic scale, although the respective number was comparable with other ranges when it was counted on the linear scale. If sufficient numbers of aerosols were counted in extremely smaller and greater ranges, the range of distribution described by a straight line would be much wider, i.e. from 0.1 to  $5 \mu$ . Finally it may be concluded that the mode of the size distribution of aerosols attached to falling snow crystals was nearly the same as the typical distribution of aerosols in air known as Junge's distribution in a range from 0.1 to  $5 \mu$ . This means that there was no particular range in which the collection efficiency of aerosols by falling snow crystals is extremely low, in the range 0.1 to  $5 \mu$  in diameter. In other words, aerosols are captured by the snow crystal with nearly a homogeneous collection efficiency in the range during their falls, although the collection mechanisms are different.

### 3.2 Surface density of aerosols

The observed area of surface of a snow crystal by electron-microscope was only a small portion of the entire snow crystal, however it was selected randomly as far as the photographing was possible. If the result of measurement in the limited area is uniformly applicable to other areas, it would be possible to calculate the surface number density of aerosols on a snow crystal. Based on this assumption, the surface number density of aerosols was calculated as shown in the left portion of Table 1.

Table 1. Surface number density of aerosols on snow crystal

Snow crystals	<i>A</i>	<i>B</i>	<i>C</i>	Estimation
Number of aerosols, observed	660	781	1963	$500m \times 1.0 \times 10^4 \text{cm}^{-3}$
Area, observed( $\mu^2$ )	288	617	443	
Surface density ( $\times 10^8 \text{cm}^{-2}$ )	2.3	1.3	4.4	$5.0 \times 10^8 \text{cm}^{-2}$
(Mean)	(2.7)			

Aerosols both on the lower and upper surfaces of snow crystals are included in the number of aerosols, because it was impossible to distinguish these two surfaces from each other in the case of the electron microscope of a see-through type.

## 4. Consideration

### 4.1 *Rain out or wash out*

It is obvious that the minute particles attached to snow crystals are carried by the crystals, however some consideration will be required for determining whether these particles were washed out by the crystals under a cloud base or rained out as condensation nuclei of cloud droplets from which the snow crystals were formed.

Kumai<sup>4)</sup> found numerous particles of submicron size near the center of an electron-microscopic photograph of a snow crystal, and he considered that those particles were condensation nuclei of cloud droplets from which vapor was deposited on the crystal. However his consideration is not applicable to the present case for the following reasons.

- 1) Condensation nuclei in riming growth: Because the snow crystals in the present observation were not rimed as previously described, condensation nuclei of rimed cloud droplets are not included.
- 2) Condensation nuclei in deposition growth: It is, therefore considered that the snow crystals were formed purely by deposition growth. However, it is not yet generally determined whether condensation nuclei of cloud droplets remain on a snow crystal or not, when the snow crystal is formed purely by deposition growth.

Here, based on the assumption that those condensation nuclei remain, an estimation of surface number density of nuclei in the snow crystal was made as follows.

Because the thickness of non rimed snow crystals of plane dendritic type is estimated to be about  $10 \mu$  according to Nakaya<sup>5)</sup>, the surface number density of the condensation nuclei is estimated as one per  $10 \times 10 \mu^2$  in electron microscopic photographs. This value is one or two orders smaller than those in the present observation, as seen in Photos. 2 and 4. It is, therefore considered that the majority of minute particles observed, are not condensation nuclei of cloud droplets from which snow crystals were formed, but are rather airborne aerosols which were washed out under the cloud base by the falling snow crystals.

### 4.2 *Collection efficiency of aerosols*

It is noted in Fig. 8 that the mode of size distribution of aerosols attached to snow crystals was almost in agreement with the Junge distribution. If we assume that the airborne aerosols (mostly man-made) over Sapporo

City originally had a mode of size distribution the same as Junge distribution, in other words, if the mode of distribution may be described by a straight line parallel to  $1/D^3$ , then this agreement shows that regardless of the aerosol size, the scavenging efficiency of aerosols by falling snow crystals was uniform, at least in the range 0.2 to 5  $\mu$  in diameter, although the scavenging mechanism is not yet clear.

Besides the mode of size distribution, it may be interesting to compare the number of aerosols attached to the falling snow crystals with that of aerosols suspended in air during a snowfall. According to the results of measurement made by an aerosol counter of Gardner type, the space number density of aerosols near the ground surface was  $8 \times 10^3$  to  $2 \times 10^4$   $\text{cm}^{-3}$  during the sampling time. The height of cloud base was estimated as 500 m. According to the results of measurement at 400 m height by Endoh and Magono<sup>6)</sup>, the aerosol density ranged from  $1 \times 10^3$  to  $1 \times 10^4$   $\text{cm}^{-3}$  in this season. Considering those data, it can be estimated that the mean space density of aerosols was  $1 \times 10^4$   $\text{cm}^{-3}$  in the falling path of snow crystals through polluted air layers below the cloud base over Sapporo City.

The snow crystal of plane type falls, maintaining its plane horizontal and in case of needle type, it falls, keeping its axis horizontal. If these snow crystals capture all the aerosols suspended in air in their falling path, the surface density of aerosols attached to the snow crystals would be estimated as  $1.0 \times 10^4$   $\text{cm}^{-3} \times 500$  m, namely  $5 \times 10^8$   $\text{cm}^{-2}$  as shown in Table 1. This value is somewhat greater than the observed values of snow crystals, 2.3, 1.3 and  $4.4 \times 10^8$   $\text{cm}^{-2}$ , as shown in the left bottom of Table 1, however they are in the same order with each other. This means that the apparent collection efficiency of aerosols by snow crystals is about 0.2 to 0.9, although these values are only the apparent ones. These values also agree with those which were obtained by an entirely different method by Magono et al<sup>7)</sup>. It is noted that these values are much greater than those expected from the usual aerodynamical consideration.

Owing to the limitation due to the resolution power of the electron microscope used, aerosols of diameters smaller than 0.05  $\mu$  were mostly missed in the present counting. While the aerosol counter of Gardner type is considered to count all aerosols greater than 0.001  $\mu$  in radius. Therefore it is considered that the surface density of aerosols calculated here, are underestimated. Taking into account of this possible underestimation, it is considered that falling snow crystals washed out almost all aerosols suspended

in space in their falling path, in other words, the apparent collection efficiency is near unity.

### 4.3 Attachment of aerosols by diffusion

An analysis was made, considering the diffusion phenomenon. Because the wash-out distance by falling snow crystals was about 500 m and their speed is estimated as about 0.5 m sec<sup>-1</sup>, the snow crystals are considered to remain 1,000 sec in air. On the other hand, the mean value of surface density of aerosols was  $2.7 \times 10^8$  cm<sup>-2</sup>. Accordingly the collection rate of aerosols by the snow crystals is calculated as  $2.7 \times 10^5$  cm<sup>-2</sup>sec<sup>-1</sup>.

Let us estimate the rate of capture theoretically. In a steady diffusion field, the collection rate of aerosols of a snow crystal of spherical shape at rest is given by the following formula

$$dN/dt = 4\pi rK(n_0 - n_r) \quad (1)$$

Here  $N$ ,  $r$ ,  $K$ ,  $n_0$  and  $n_r$  mean the number of aerosols collected by the snow crystal, the radius of the crystal, the diffusion coefficient and the space number densities of aerosols near the surface of the crystal and in air. In this case the radius  $r$  is considered to be constant as far as time is concerned. And for the unit surface of the spherical snow crystal,

$$dN_s/dt = K(n_0 - n_r)/r \quad (2)$$

Here  $N_s$  is the surface number density of aerosols of the snow crystal.

Because the majority of aerosols were distributed in a range around 0.1  $\mu$  in diameter, the diffusion coefficient can be estimated as  $6.8 \times 10^{-6}$  cm<sup>2</sup> sec<sup>-1</sup> on the average. Applying the value to formula (2), we obtain

$$dN_s/dt = 0.68 \text{ cm}^{-2} \cdot \text{sec}^{-1}$$

for the condition  $r=0.1$  cm and  $n_0=1.0 \times 10^4$  cm<sup>-3</sup>. This value of collection rate is the greatest value considered, because  $n_r$  was neglected compared with  $n_0$ . Integrating this rate for 1,000 sec in which the snow crystal was falling through a polluted air layer, we obtain  $N_s=680$  cm<sup>-2</sup> at most, as a total number of aerosols captured by the snow crystal with a radius of 1 mm. This value is extremely small, compared with the observed value of  $2.7 \times 10^8$  cm<sup>-2</sup>. Accordingly it appears hopeless to explain the observed number of aerosols only by the diffusion phenomenon.

According to the results of Greenfield's calculation<sup>8)</sup>, the effect of turbulent motion on the aerosol scavenging is almost the same as that of the Brownian motion in the size range of aerosols of 0.1  $\mu$ . Therefore even

if we take into account the turbulent motion, it is impossible to explain the great number of aerosols attached to snow crystals, by means of diffusion phenomenon alone.

#### 4.4 *Attachment of aerosols by impaction collision and Facy effect*

The collection efficiency of submicron aerosols by snow crystals due to the difference in falling speeds is so low that the estimation by extrapolating the calculated results of collision efficiency of cloud droplets is difficult. The result of laboratory experiments by Kerker and Hampl<sup>9)</sup> shows that the collection efficiency of submicron aerosols is in the order of  $10^{-2}$ . Even if the wake effect is taken into account, the observed collection efficiency as high as almost unity can not be explained. And even if we consider the complex shape of snow crystals on the collection efficiency, it is difficult to explain the high collection efficiency.

As described previously, the attachment of aerosols mostly occurred under the cloud base. Accordingly the Facy effect due to the growth of snow crystals is out of the question.

#### 4.5 *Effect of electrostatic force on the attachment of aerosols*

It is generally expected that the electrostatic force is effective on the attachment of aerosols with raindrops or snow crystals. However only a few calculations were made in a range of submicron size for such a purpose. Endoh et al<sup>10)</sup> calculated the force due to the electric charge between an ice sphere of 1 mm diameter with a charge of  $1 \times 10^{-3}$  esu and an aerosol of  $0.1 \mu$  with charge of  $1 \times 10^{-8}$  esu. According to the results of their calculations, the image force is more effective than the Coulomb force when the distance between the ice sphere and the aerosol is smaller than 1 mm. Here 'distance' means the distance between their centers.

About the closed distance, the author calculated the image force in the case of distance of 0.6 mm, in other words the distance between the surface of ice and aerosols was about 0.1 mm. If the attractive force due to the image force is balanced with viscosity force,

$$Q^2d/2l^3 = 3\pi\eta v \quad (3)$$

Here  $Q$ ,  $d$ ,  $l$ ,  $\eta$  and  $v$  mean the charge on the ice sphere, the diameter of aerosol, the distance between the ice sphere and the aerosol, the viscosity of air and the speed in which the aerosol is attracted, respectively. Substituting the respective numerical values, we obtain

$$v = 0.13 \text{ cm} \cdot \text{sec}^{-1}$$

If we estimate the fall speed of the ice sphere as  $50 \text{ cm} \cdot \text{sec}^{-1}$ , the effective time for the image force is estimated as only about  $2/500 \text{ sec}$ . Accordingly it is estimated that the aerosol is attracted only by  $5 \mu$  towards the surface of ice sphere under the effect of electrostatic force in  $2/500 \text{ sec}$ . This means that the electrostatic attractive force has no practical effect on the movement of the trajectory of the aerosol, in other words, no effect to increase the collection efficiency, although the force has an effect to increase the coagulation efficiency by completely capturing aerosols which approached very closely.

### 5. Conclusion

It was observed under an electron microscope that numerous aerosols of submicron size were attached to falling snow crystals. It was ascertained that these aerosols were captured under the cloud base, not originated from condensation nuclei of cloud droplets. It was estimated that falling snow crystals captured aerosols at a high collection efficiency, namely, they scavenged almost all aerosols suspended in a volume which were swept out by the falling snow crystals.

In order to explain such a high collection efficiency, the effect of impact collision, Facy effect, diffusion (Brownian and turbulent motions), and electrostatic force were theoretically estimated, however all the effects were insufficient to explain the high efficiency.

Accordingly it is expected that all of these effects will be theoretically examined again, particularly the collision efficiency due to the turbulence in submicron size, and the electrostatic force will be scrutinized.

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Plate I Snow crystal *A*.

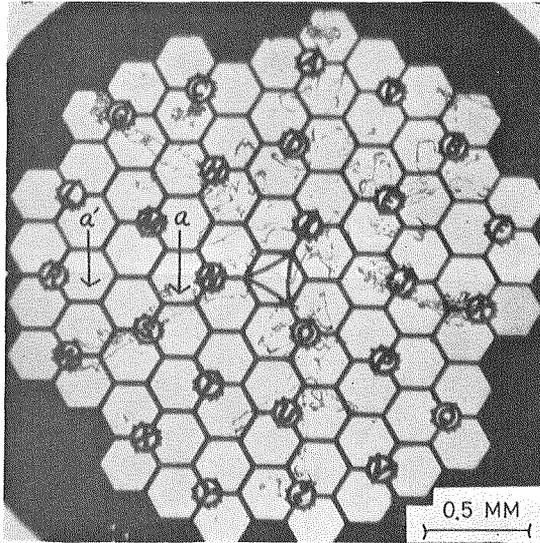


Photo. 1 Optical microscopic photograph of Crystal *A*.

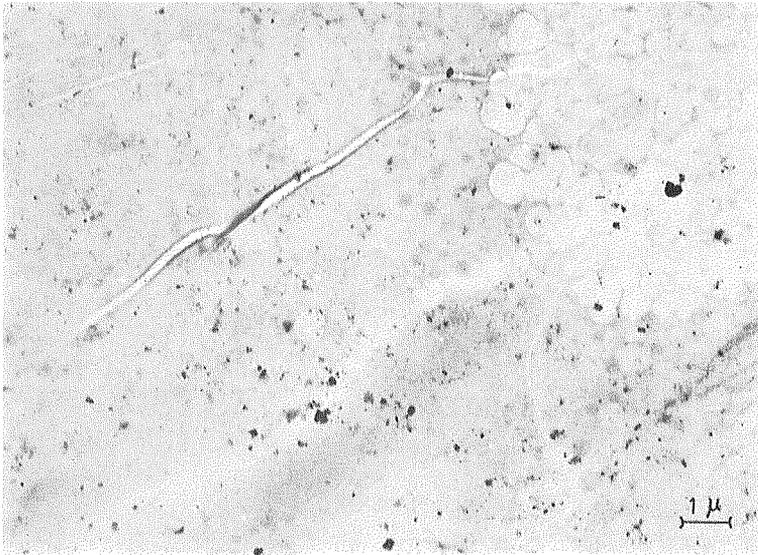


Photo. 2 Aerosols attached to the portion indicated by arrows in Photo. 1.

Plate II Snow crystal *B*.

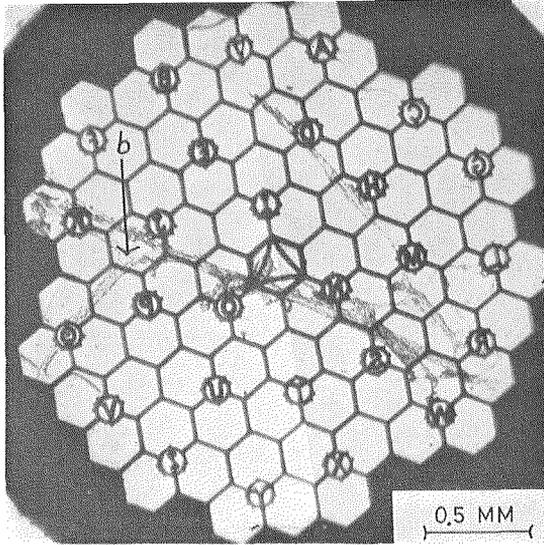


Photo. 3 Optical microscopic photograph of Crystal *B*.

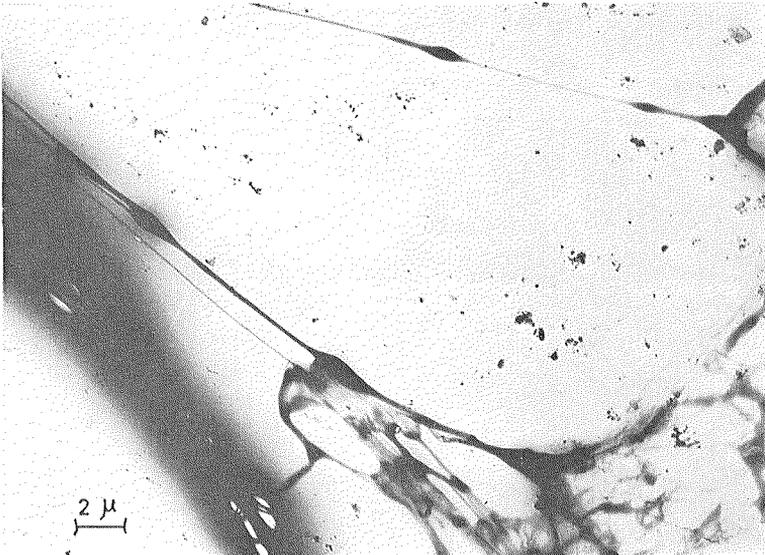


Photo. 4 Aerosols attached to the portion indicated by an arrow in Photo. 3.

Plate III Snow crystal *C*.



Photo. 5 Aerosols attached to a portion of Crystal *C*.



Photo. 6 Ghost dust particles found in a portion outside of a replicated area of Crystal *C*.