NOTES AND CORRESPONDENCE

**Relationship between the Size of Cloud Droplets
and Their Residual Particles****By Masataka Murakami and Choji Magono***

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1. Introduction

The observational study of aerosol scavenging showed that the collection efficiency of the rimed snow crystals was about one order of magnitude higher than that of the unrimed snow crystals (Murakami *et al.*, 1981). One of the reasons might be an apparent increase in the collection efficiency of rimed snow crystals; such an apparent increase might be caused by counting not only the particles scavenged by snow crystals but also the particulate residues of cloud droplets on the snow crystals. Since Murakami *et al.* (1981) treated the aerosol particles which were detectable by using an optical microscope, the question arises as to whether the cloud droplets left the substantial number of such large particles after their evaporation.

Many workers (Dessens, 1949; Kuroiwa 1951, 1957; Ogiwara and Okita, 1952; Yamamoto and Ohtake, 1953, 1955; Naruse and Maruyama, 1971) studied the particulate residues of cloud droplets from the viewpoint of cloud physics; they regarded these residues as cloud nuclei. According to these field measurements and theoretical studies which have been made hitherto, the size of cloud nuclei varies widely with the location and ranges from 0.01 μm to several microns.

The size of particulate residues of cloud droplets over Sapporo City, where Murakami *et al.* (1981) had observed the collection effi-

ciency of snow crystals, was investigated in order to clarify whether these particulate residues cause an apparent increase in collection efficiency for aerosol particles which were detectable by an optical microscope. This paper reports the size and number of particulate residues and their relation to the size distribution of cloud droplets.

2. Observation

The observation of cloud droplets were carried out at the Cloud Physics Observatory, Hokkaido University in October and November 1978; this observatory is located at the top of Mt. Teine (1,024 m), in Hokkaido, Japan. The cloud droplets were collected on a collodion coated electron microscope grid using an impactor of rectangular-shaped orifice (1.45 mm width). The diameter of droplets which are collected with 50% efficiency is estimated to be 4 μm . The cloud droplets on the grid were immediately examined by using an optical microscope with a heat insulating filter under the conditions of outdoor humidity ($\sim 100\%$). The change of droplet size due to evaporation during replacement of specimen and examination (~ 10 s) was negligible. After the evaporation of cloud droplets, their particulate residues were examined by using an optical microscope at the relative humidity of 40% to 50%. Further they were examined by using an electron microscope in order to supply the lack of resolution power of optical microscope for submicron particles.

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3. Results and Discussion

3-1. Meteorological conditions

Meteorological conditions during the observation periods and the sampling time of cloud droplets are shown in Fig. 1. Seven out of ten samplings were carried out in the precipitating clouds. During the observation periods, the temperature at the top of Mt. Teine was around 0°C so that precipitation particles were in the form of snow (including sleet and graupel). The samplings of cloud droplets were also carried out under the conditions of various wind directions as shown in Fig. 1. Sample numbers with underlines indicate that the size distribution of cloud droplets was bimodal; this bimodal distribution was similar to that obtained by Warner (1969). As far as the present investigation is concerned, there seems to be no systematic difference in the size distribution of cloud droplets according to the atmospheric conditions; wind directions or whether the cloud is precipitating. Therefore, all the samples are treated together in this paper.

3-2. Size and number of residual particles

The particulate residues, which were left after the cloud droplets had evaporated, are

called residual particles in this paper. In the case where a cloud droplet left two or more residual particles, the largest particle and the others are called "primary residual particle" and "secondary residual particle", respectively.

Figure 2 shows the optical micrographs of the cloud droplets on a collodion coated electron microscope grid (upper part) and their residual particles (lower part). As shown in Fig. 2, the residual particles were mostly identified in the position where the cloud droplets had existed before their evaporation.

Since an optical microscope, in general, does not have sufficient resolution power to examine exactly the residual particles smaller than $1\text{ }\mu\text{m}$ in diameter, an electron microscope was used in conjunction with an optical microscope. The relationship between the size of cloud droplets and that of residual particles obtained by the use of electron microscope is shown in Fig. 3. Cross marks at the bottom of the figure represent the cloud droplets which did not leave any detectable residual particles (larger than $0.3\text{ }\mu\text{m}$ in diameter). As for the cloud droplets larger than $10\text{ }\mu\text{m}$ in diameter, the size of primary residual particles was proportional to that of cloud droplets although the data was scattered to some extent. This result is similar to that obtained by Naruse and Maruyama (1971); they collected the cloud

Fig. 1 Meteorological conditions during the observation periods.

Fig. 2 Optical micrographs of the cloud droplets (upper part) and their residual particles (lower part).

Fig. 3 Size of primary residual particles versus size of cloud droplets; particle size was obtained by the use of an electron microscope.

droplets at Mt. Fuji. As for the cloud droplets smaller than $10\ \mu\text{m}$ in diameter, however, it seems that the size of primary residual particles have no relation to that of the cloud

droplets. Although the relationship between them based on the measurement by the use of optical microscope (not shown) was similar to that shown in Fig. 3, the number of cloud droplets which did not leave any detectable residual particles increased markedly. As for the size and number of the residual particles, hereafter, the data obtained by the use of an electron microscope are adopted.

Although Figure 3 shows all the data obtained from ten samples, the individual samples could be classified into two types. Figure 4 shows two typical size distributions of cloud droplets and relationships between the size of cloud droplets and that of primary residual particles. In the case of "Type A", the size distribution of cloud droplets showed one mode at around $20\ \mu\text{m}$ in diameter and the size of cloud droplets was proportional to that of primary residual particles. On the other hand, in the case of "Type B", the size distribution of cloud droplets showed two modes at around $5\ \mu\text{m}$ and $20\ \mu\text{m}$ in diameter. As for the cloud droplets larger than $10\ \mu\text{m}$ in diameter, their size was proportional to that of primary residual particles. For the cloud droplets smaller than $10\ \mu\text{m}$, however, such a relationship was not recognized. As seen in Fig. 1, both types of cloud droplet distribution were sometimes observed in the same cloud system.

Assuming that primary residual particles functioned as cloud nuclei, the bimodal distribution may suggest that the larger and smaller cloud droplets had different experiences in the cloud or formed on the cloud nuclei of different materials. In order to clarify the cause of the bimodal distribution of cloud droplets, it is desired that further investigation which includes the chemical composition of residual particles besides the size and number of them should be carried out.

The size distribution of the primary residual particles is shown in Fig. 5. Most of primary residual particles were smaller than $2\ \mu\text{m}$ in diameter, and the size distribution showed a marked mode at around $0.8\ \mu\text{m}$ in diameter. Figure 5 as well as Figure 3 shows that the substantial percentage of cloud droplets leave residual particles larger than $1\ \mu\text{m}$ in diameter; these particles are easily detec-

Fig. 4 Typical size distributions of cloud droplets and relationship between size of cloud droplets and size of primary residual particles.

Fig. 5 Size distribution of primary residual particles.

table by an optical microscope. In this way, it is concluded that the residual particles of cloud droplets cause an apparent increase in collection efficiency for micron-size aerosol particles.

The examination by using an electron

Fig. 6 An electron micrograph of residual particles in the positions where cloud droplets existed before the evaporation.

microscope showed that the individual cloud droplets often left two or more residual particles. An example of electron micrographs is shown in Fig. 6. Circles in the figure indicate the positions where the cloud droplets existed before their evaporation. In the two circles at the lower left of the figure, there were two or three residual particles, respec-

Fig. 7 Histogram of number of residual particles per a cloud droplet.

tively.

Most of secondary residual particles were smaller than $1\text{ }\mu\text{m}$ in diameter, and no remarkable relationship was recognized between the sizes of cloud droplets and secondary residual particles.

The frequency percentage of the number of residual particles per one cloud droplet is shown in Fig. 7; residual particles include both primary and secondary residual particles larger than $0.3\text{ }\mu\text{m}$ in diameter. About 30% of cloud droplets left two or more residual particles after evaporation.

The attachment of non-activated aerosol particles to the cloud droplets is considered to be one of probable mechanisms which account for the origin of secondary residual particles although the coagulation and dispersion of aerosol particles in the cloud droplet may have an influence on secondary residual particles to some extent.

5. Conclusion

The size of cloud droplets and their residual particles were examined by using an optical microscope and an electron microscope. The following results were obtained.

The majority of residual particles of cloud

droplets were smaller than $2\text{ }\mu\text{m}$ in diameter, and their size distribution showed a marked mode around $0.8\text{ }\mu\text{m}$ in diameter. Even when only the particles larger than $1\text{ }\mu\text{m}$ in diameter are assumed to be detected by an optical microscope, a substantial fraction of cloud droplets leave the detectable residual particles after their evaporation. This result confirms the possibility that the residual particles of cloud droplet deposited on snow crystals may apparently increase the collection efficiency of micron-size aerosol particles by rimed snow crystals.

Four out of ten samples of cloud droplets showed the bimodal distribution. In the case of bimodal distribution, the size of cloud droplets larger than $10\text{ }\mu\text{m}$ in diameter was proportional to that of primary residual particles, whereas the cloud droplets smaller than $10\text{ }\mu\text{m}$ in diameter did not show such a relationship. On the other hand, in the case of monomodal distribution, the size of cloud droplets was proportional to that of the primary residual particles independently of their size.

About 30% of the cloud droplets left two or more residual particles ($d_p > 0.3\text{ }\mu\text{m}$).

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雲粒とその残渣粒子の大きさの関係について

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