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Relation between the Center Nucleus of Snow Crystals and Aerosol Particles in Arctic Canada

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Abstract

To clarify the formation mechanisms of the snow crystals of low temperature types, observations on the shapes of snow crystals, the samplings of center nucleus of snow crystals and aerosol particles in the free atmosphere during snowfalls were carried out at Inuvik (68°22'N, 133°42'W), Northwest Territories, Canada from December 25, 1985 to January 23, 1986. As a result of the analysis, a higher frequency of the element Si as a center nucleus was recognized in all types of snow crystals as a whole. The center nucleus of snow crystals around Inuvik area, therefore, appears to consist of soil components. In the analysis classifying the snow crystals into single crystals and polycrystals, the chemical components of the center nucleus that were found in single crystals were similar to that found in polycrystals in soil components, that is to say, the elements of Si, Al and Ca. The chemical components of the center nucleus found in polycrystals, on the other hand, were higher than that of single crystals in sea salt components such as Na, Mg, Cl and K. Furthermore, the sampling of airborne aerosol particles during snowfalls was made, and the analysis of their chemical components was carried out classifying the cold and warm periods when the observation site was covered by the cold Arctic and the relatively warm North Pacific air masses, respectively. During the cold periods, the predominant chemical components were soil particles, and in the warm periods, the predominant chemical components were sea salt particles in the free atmosphere. If the chemical component ratio of the center nucleus of snow crystals was classified into the cold and warm periods, it may be concluded that the general shapes of snow crystals such as plate and column types have a tendency to consist of soil and anthropogenic components as the center nucleus. On the other hand, the peculiar shapes of snow crystals of low temperature types such as gohei twin and seagull types have a tendency to take soil and sea salt components as the center nucleus. This result showed a quite similar tendency which

was obtained by Kikuchi et al. (1972) in Hokkaido.

1. Introduction

Recently, studies on aerosol particles have been carried out from a point of view of the long range transportation of air pollution and acidity of precipitation particles. Furthermore, to determine the chemical components of aerosol particles is very important for the consideration of the origin and the role of the center nucleus of the precipitation particles (Kumai, 1951, 1961, 1976; Isono, 1955). Because, the nucleation activities of aerosol particles as condensation nuclei and freezing nuclei also largely depend on the chemical components of the nuclei (Schaefer and Cheng, 1968). In the Arctic area, however, only a limited number of investigations have been made on the chemical components of aerosol particles especially from a point of view of their roles as nuclei (Kumai and Francis, 1962; Parungo et al., 1979). Moreover, they did not measure the chemical components of aerosol particles floating free in the atmosphere when the snow crystals were collected as specimens for the analysis. On the other hand, an attention has been focused on the formation mechanisms of the snow crystals of low temperature types, for instance, "Gohei twin", "Seagull", and "Spearhead" type crystals reported by Kikuchi and his coworkers (Kikuchi, 1969, 1970; Kikuchi and Hogan, 1976; Kikuchi and Kajikawa, 1979; Kajikawa et al., 1980; Sato and Kikuchi, 1985; Kikuchi and Sato, 1988) in the Antarctica and Arctic areas. Therefore, the relation between the chemical components of aerosol particles and center nucleus of snow crystals of the general and basic types, and low temperature types was investigated in this study.

2. Sampling location and methods

Observations of snow crystals and aerosol particles were carried out at Inuvik (68°22′N, 133°42′W), Northwest Territories, Canada as shown in Figs. 1 and 2 from December 25, 1985 to January 23, 1986. At the actual site, snow crystals were photographed by a polarization microscope. For the preparation of specimens to determine the center nucleus of snow crystals, the following two methods were used; (1) A vapor replica solution method was used, that is, sampling films coated with Polyvinyl Formvar replica solution were adhered on glass slides and the snow crystals were collected on the films. The snow crystals descended on the film were replicated by using 30% Chroloform vapor. (2) Snow crystals were collected on carbon stages for a scanning electron

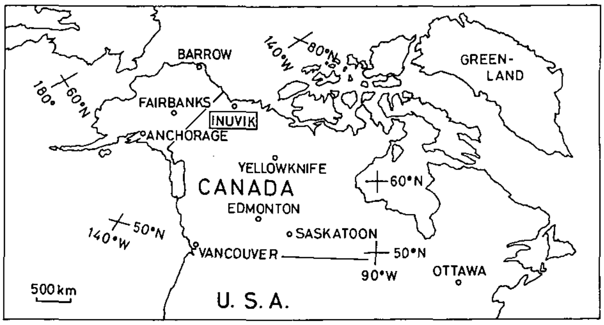


Fig. 1. Location of the observation site.

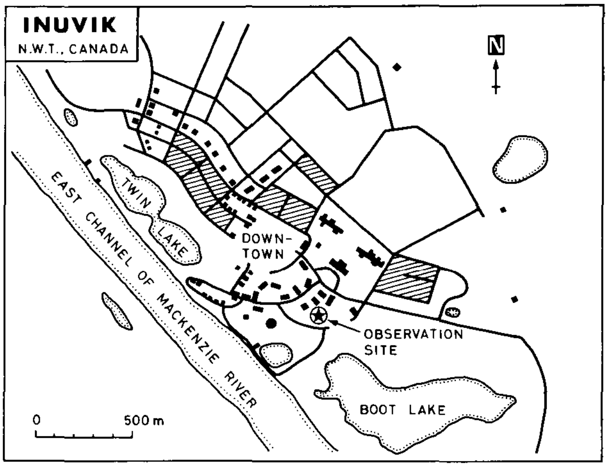


Fig. 2. Map of Inuvik. Observation site is shown by a star mark.

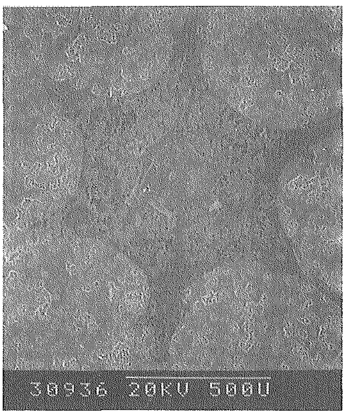
microscope (SEM) and they were replicated on the stages by using a Methyl 2-Cyanoacrylate (MCA) vapor (Odencrantz and Hildebrand, 1971).

For the preparation of specimens of aerosol particles, the following two methods were used ; (1) A newly developed aerosol sampler was used to collect the atmospheric aerosol particles on the filter papers continuously for every one hour (Taniguchi and Kikuchi, 1989). (2) Atmospheric aerosol particles during snowfalls were collected occasionally on membrane millipore filters using a suction pump. In the analyses of the chemical components of these particles, a scanning electron microscope (HITACHI SEM-S430) and an energy dispersive X-ray microanalyzer (HORIBA EMAX-1800E) were used in our laboratory.

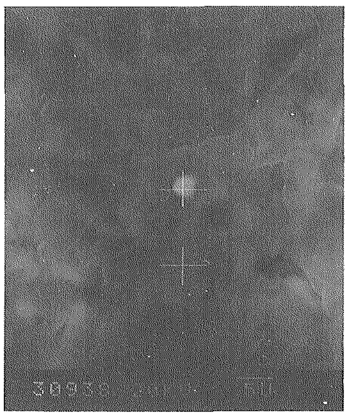
3. Analysis of center nucleus of snow crystals

3.1 Analysis of individual shapes of snow crystals

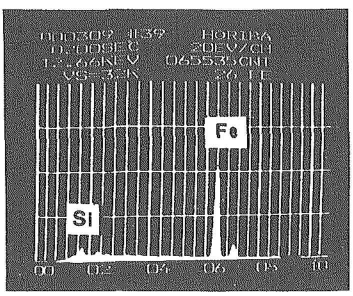
Figure 3 shows an example of snow crystals of plate type. In a strict sense, the crystal was a plate with dendritic extensions coded by (P2g) (Magono and Lee, 1965) as seen in Fig. 3(a). Figure 3(b) shows a close up photograph of Fig. 3(a). As seen the photograph, a relatively large nucleus of 2.5 μm in diameter which resembles the center nucleus of this crystal was recognized around the central part of this crystal. Two white cross marks indicate points which were radiated by an electron beam (target point) in the center nucleus (upper position) and background around the center nucleus (lower position). As shown in Fig. 3(c), chemical components of the nucleus consisted of components of Si and Fe.



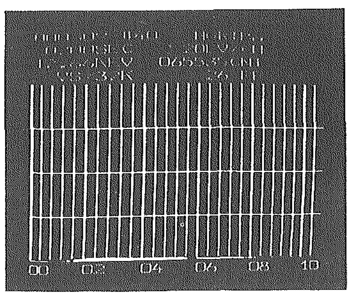
(a)



(b)

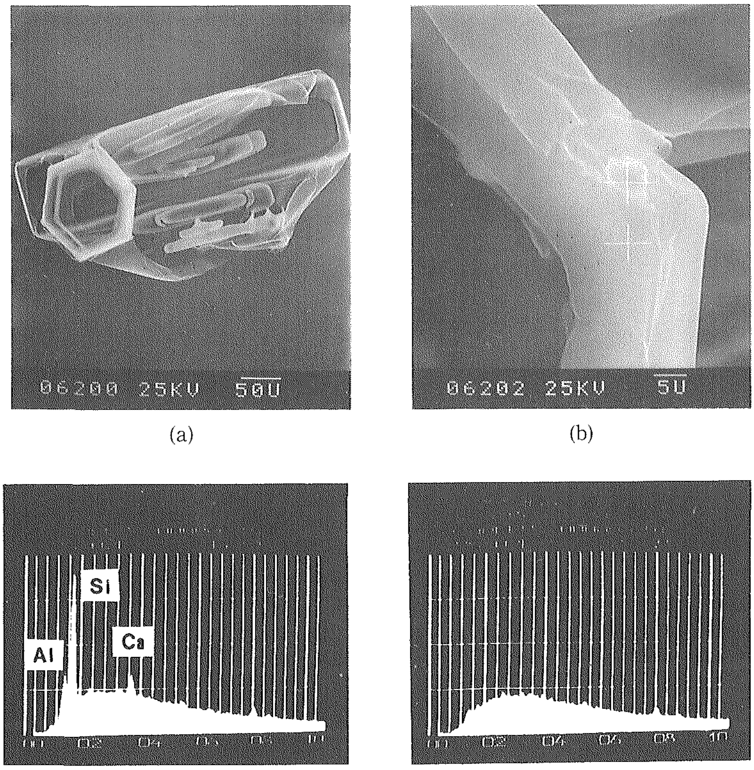


(c)



(d)

Fig. 3. Analysis of a plate with dendritic extensions. (a) and (b) SEM photographs, (c) and (d) EMAX spectra of the particle and background.



(a) (b)

(c) (d)

Fig. 4. Same as Fig. 3 but for column.

On the other hand, no elements were found in the background as seen in Fig. 3(d). Figure 4(a) shows one of typical column type crystals (Cle). As seen in Fig. 4(b), the center nucleus was not recognized around the center but was found in the edge corner. The chemical components of this crystal consisted of Al, Si and Ca estimated from the compositions of soil particles. However, no elements were found out around background area. Figure 5(a) shows an example of a combination of bullets (C2a), and a close up picture is shown in Fig. 5(b). A smaller particle compared with other examples of 0.5 μm in diameter was found. The particle contained elements of Na, Cl and K. Therefore, it was considered that the particle was sea salt particle. Figure 6 is one of the microphotographs of snow crystals of low temperature types. Although the outer shape of this crystal has a square form with crossed plates at the lower right, the square form of this crystal is one of the prism planes. As well known, the snow crystals of

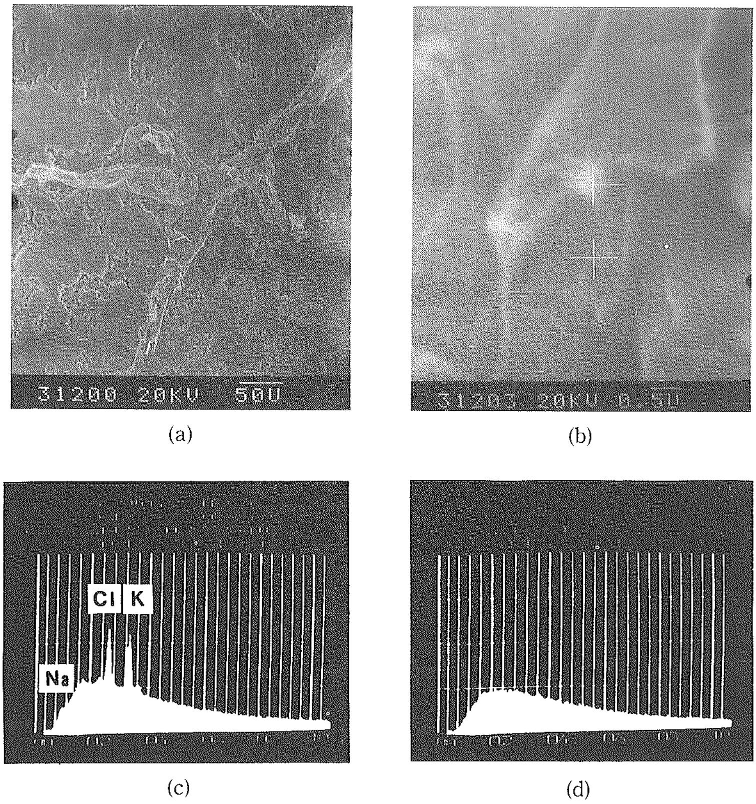


Fig. 5. Same as Fig. 3 but for combination of bullets.

low temperature types are observed with other polycrystalline snow crystals such as crossed plates and a combination of bullets. Therefore, it is very difficult to determine the center for their complicated shapes. As shown in Fig. 7(a) and (b), however, a nucleus of 3 μm in diameter was found near the center of this crystal. The chemical components of this crystal consisted of numerous kinds of elements in the nucleus such as Na, Al, Si, S, Cl and K as shown in (c). Therefore, the nucleus can be considered to be a mixed nucleus containing soil, sea salt, and combustion product components. Figure 8 shows one of the typical examples of "Seagull" type crystals. Figure 9(a) shows the crystal replicated and Fig. 9(b) shows a particle of 3 μm in diameter which was discovered at the wing joint of the crystal. The chemical components contained in the particle were Al and Pb as shown in (c), however, no components were found in the background near the particle as shown in Fig. 9(d).

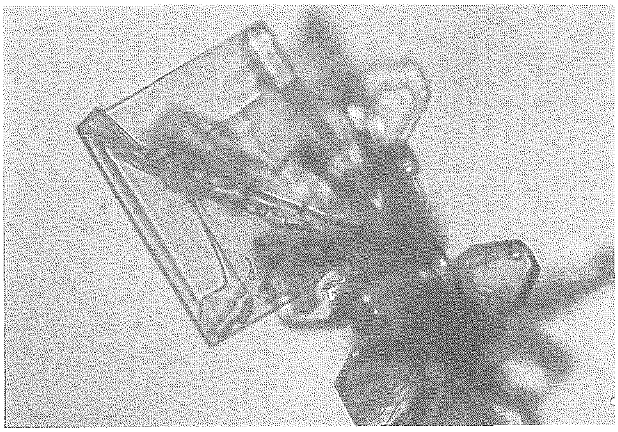


Fig. 6. A microphotograph of snow crystals of low temperature types (×130).

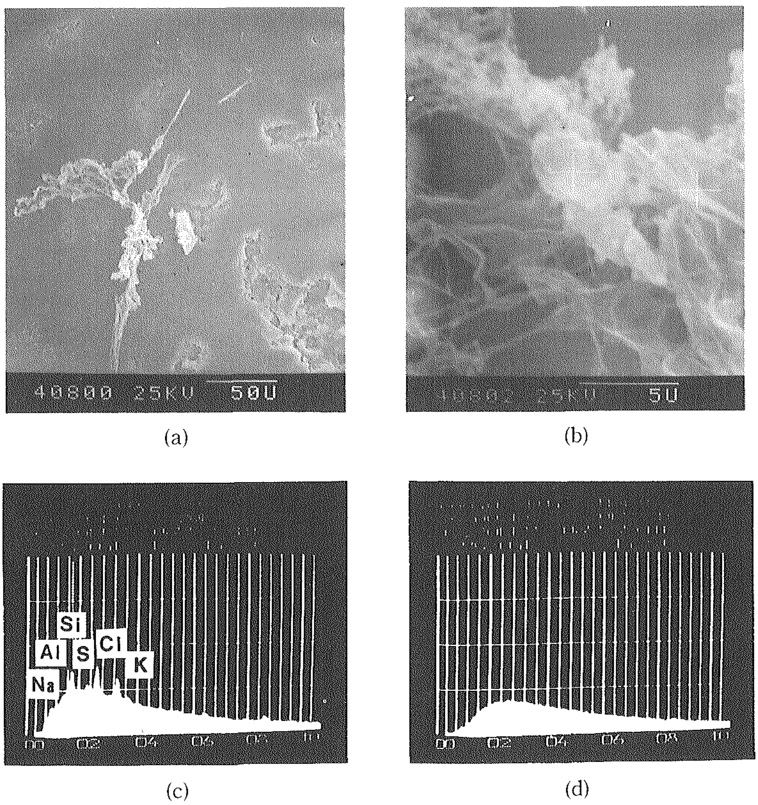


Fig. 7. Same as Fig. 3 but for the snow crystal of low temperature type.

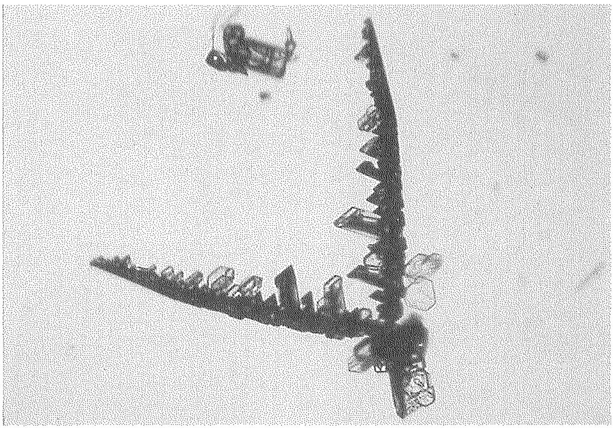
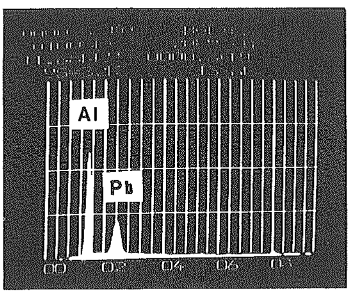


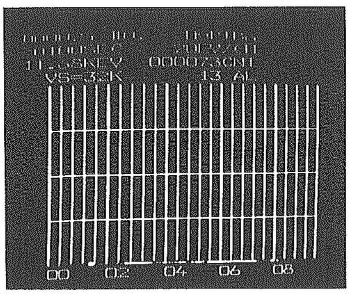
Fig. 8. Same as Fig. 6 but for the sea gull type crystal (×35).

(a)

(b)



(c)



(d)

Fig. 9. Same as Fig. 3 but for the seagull type as shown in Fig. 8.

Table 1. Elements contained in the center nucleus of snow crystal.

Types \ Element	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	Cr	Fe	Ni	Cu	Zn	Pb	No.
Plate • Column	13	9	57	78	4	43	43	30	52	4	13	35	4	13	17	0	23
Combination of bullets	71	0	14	71	0	29	71	43	14	0	0	0	0	0	0	0	7
Seagull	31	31	77	77	8	38	38	31	77	15	8	54	0	0	0	0	13
Gohei twin	38	0	38	63	0	38	50	50	13	0	0	25	0	0	63	13	8
Single crystals	13	9	57	78	4	43	43	30	52	4	13	35	4	13	17	0	23
Polycrystals	43	18	50	75	4	36	50	39	42	7	4	32	0	0	18	4	28

3.2 Percentage frequency of the chemical components of center nucleus of snow crystals

In totality, 42 snow crystals and 51 nuclei of snow crystals were analyzed using SEM and EMAX systems. The difference between the numbers of snow crystals and nuclei depends on whether a snow crystal has several nuclei around the center of the crystals occasionally. Table 1 shows the percentage frequency of the number of the chemical elements contained in the center nucleus of snow crystals. In the percentage, the peak heights of individual spectra of the particles were not considered but only the numbers were counted. Therefore, 50% of Cl, for instance, as seen in the row of the snow crystals of gohei twin means that 4 particles among 8 particles analyzed showed peaks of the element of Cl. The numbers written in the right hand side indicates the number of crystals analyzed. As seen in the table, the element of Si which is one of typical chemical components of soil particles was predominant in all kinds of snow crystals analyzed. This result was in good agreement with that obtained in the previous studies using an electron diffraction pattern (Kumai, 1951, 1961 ; Isono, 1955 ; Kumai and Francis, 1962) and in a similar study using the same system as in our experiments by Parungo et al. (1979). Regarding the element Zn, there was a possibility that the element was included during the operation of the system. Next, we examined the tendency of the chemical components by dividing crystal forms, namely, single or polycrystals. Plate and column type crystals were classified into single crystals and a combination of bullets, gohei twin, and seagull type crystals were classified as polycrystals, respectively. The results are shown in the last two lines in the Table 1. As seen in Table 1, the element Si showed a similar percentage in both crystals, namely 78% and 75%, respectively. The element Na which formed sea salt particles was three

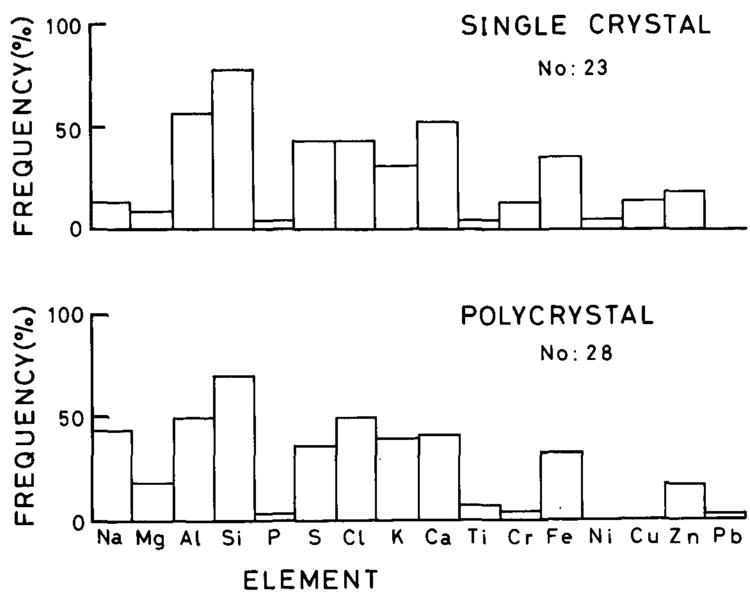


Fig. 10. Frequency distributions of the elements of single and polycrystals.

times higher in polycrystals than in single crystals. Figure 10 shows the same results drawn by histograms. As clearly seen in the figure, the frequency percentage was higher in polycrystals than in single crystals as a whole. The reason why the percentage of single crystals was lower than polycrystals is because the chemical components of center nucleus of single crystals were made of relatively pure elements. On the contrary, it was considered that the chemical components of the center nucleus of polycrystals were made from mixed and complex elements. The elements of Al, Si and Ca which were made of soil particles were relatively higher in the single crystals, and the element S which was an anthropogenic particle. However, the elements of Na, Mg, Cl and K which consisted of sea salt particles were higher in polycrystals than in single crystals. The elements in which the atomic number was larger than 22, Ti, were not considered here. This result showed a quite similar tendency which was obtained by Kikuchi et al. (1982) in Hokkaido.

3.3 Component ratios of typical elements contained in the center nucleus of snow crystals

As discussed in the above section, the percentage frequency of chemical components of the center nucleus does not show the mass ratio of individual

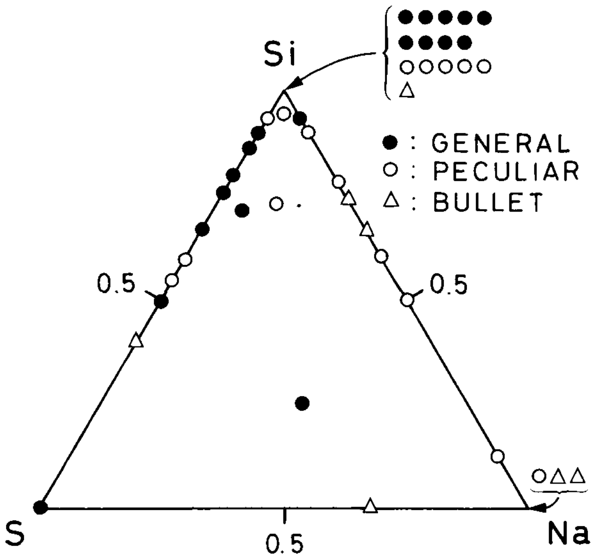


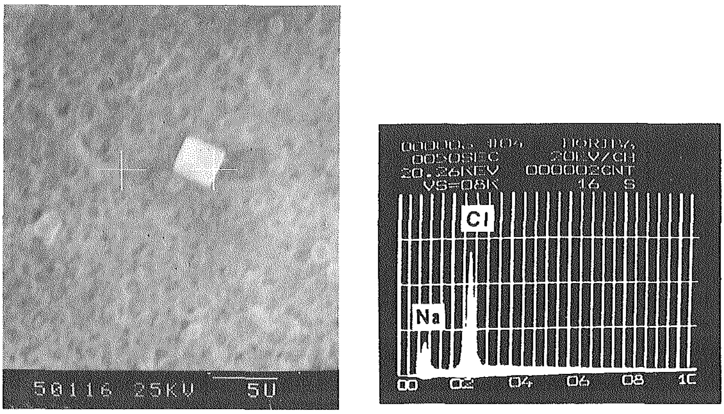
Fig. 11. Triangle diagram of the typical three elements of Si, Na and S of the center nucleus of snow crystals.

elements. Then, we examined a relative mass ratio of the typical three elements, namely, Si for soil particles, Na for sea salt particles, and S for combustion products as criteria for human activity for the center nucleus of snow crystals by Murakami and Kikuchi (1982). Although the center nucleus of 51 snow crystals in totality as shown in the Table 1 were analyzed, 42 snow crystals contained some of the elements of Si, Na and S. Figure 11 shows a ratio of Si : Na : S in a triangle diagram. In this figure, solid circles, open circles, and open triangles show the general shapes of snow crystals such as plate and column type crystals in this study, peculiar shapes such as gohei twin and seagull type crystals, and a combination of bullets, respectively. It is understood in this figure that the frequency of center nucleus of snow crystals in Arctic Canada was larger in the element of Si, because of individual marks concentrate at an apex of Si of the triangle diagram. However, solid circles are scattered on the line between apexes of Si and S rather than otherwise. In contrast, open circles and open triangles are seen scattered on the line between apexes of Si and Na. Therefore, it was understood that snow crystals of low temperature types such as polycrystals, gohei twin, seagull and a combination of bullets in this study have a tendency to contain sea salt particles rather than combustion products as the center nucleus.

4. Analyses of aerosol particles

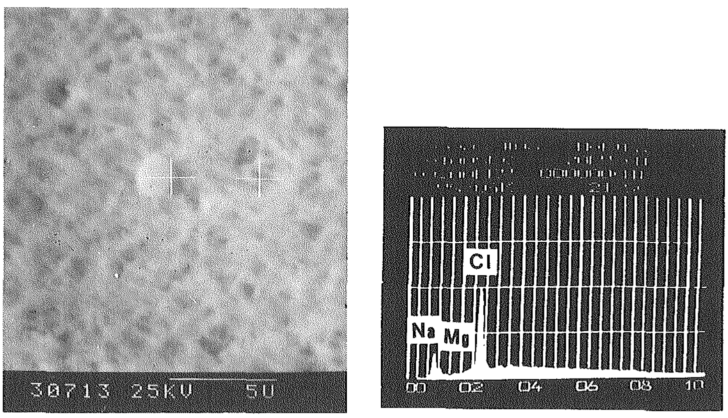
4.1 Individual analyses of aerosol particles

On January 14, 1986, a combination of bullets was mostly observed. During the period, a sampling of airborne aerosol particles was carried out at the same observation site. Figure 12 shows an example of sea salt particles. As seen in this figure, the sea salt particle was of a cubic shape and the chemical elements were Na and Cl alone. In contrast, the sea salt particle in Fig. 13 was of a



(a) (b)

Fig. 12. Analysis of aerosol particles. (a) SEM photograph, (b) EMAX spectrum.



(a) (b)

Fig. 13. Same as Fig. 12. (a) SEM photograph, (b) EMAX spectrum.

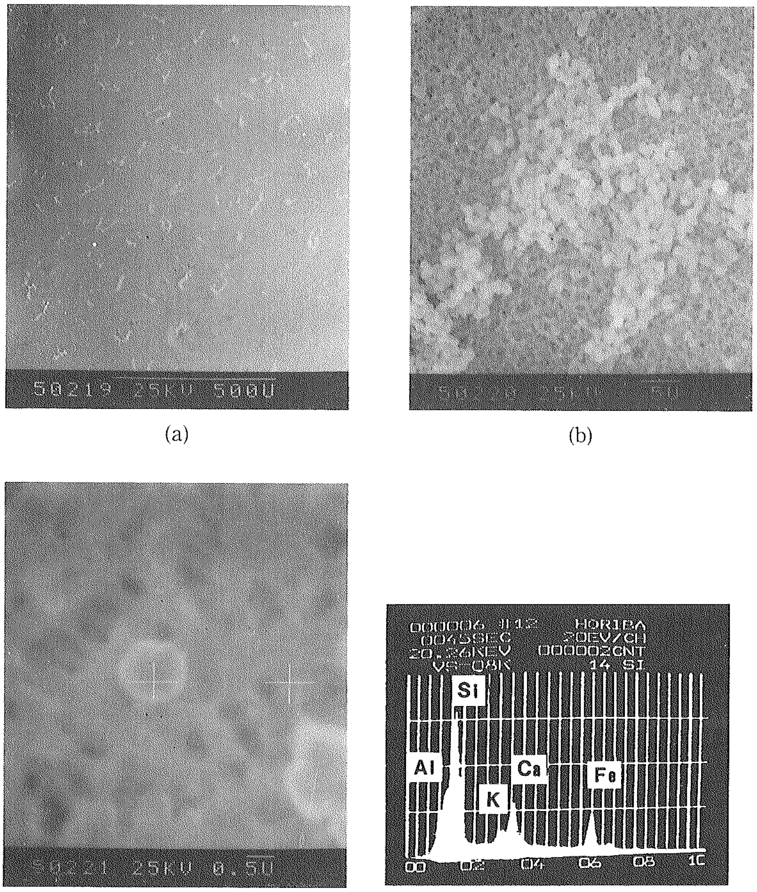
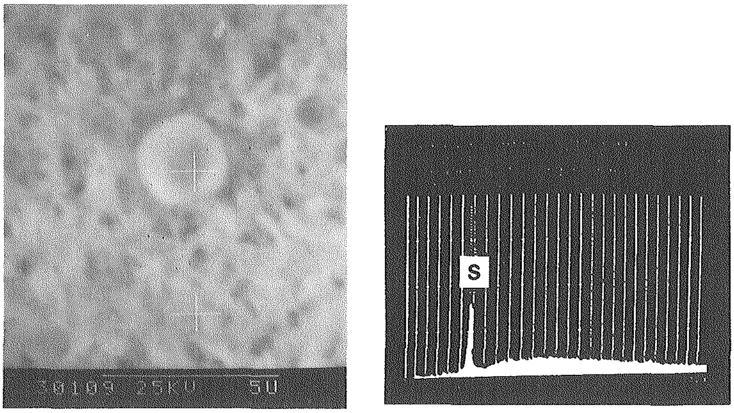


Fig. 14. Same as Fig. 12. (a), (b) and (c) SEM photograps, (d) EMAX spectrum.

spherical shape and the chemical elements were Na, Mg and Cl. Diameters of each particle were 3 μm and 1 μm, respectively. As shown in these examples, the size was large when the particle had a cubic shape and was small when the particle had a spherical shape. It was considered that the particles having a cubic shape were relatively short lasting ones in the atmosphere and other ones relatively long lasting. On the other hand, on January 16, 1986 when seagull type crystals were observed relatively abundantly, aerosol particles consisted of small spheres coagulated as shown in Fig. 14(a) and (b) were observed. The chemical components of these particles were almost soil particles as shown in



(a) (b)

Fig. 15. Same as Fig. 12. (a) SEM photograph, (b) EMAX spectrum.

Fig. 14(c) and (d). The particles closely resembled coal flyash reported by Mamane et al. (1986). When such particles were observed, the filter papers for sampling showed charcoal to blackish colors. Therefore, it was presumed that the particles were some kinds of combustion products as a result of human activity. Figure 15(a) shows an example of aerosol particles when plate type crystals were falling predominantly. As seen in Fig. 15(b), the EMAX spectrum shows the element S alone. A spectrum such as this was reported by Mamane et al. (1986) as fuel flyash.

4.2 Percentage frequency of the chemical components of aerosol particles

Table 2 shows percentage frequency of the number of chemical components of aerosol particles when the snow crystals were falling, namely, on January 1, the gohei twin type crystals were prevalent, on January 5, the plate, on January 16, 1986, the seagull type crystals were prevalent, respectively. 30 particles on millipore filters sampled aerosol particles were selected for analysis at random.

Table 2. Elements contained in the aerosol particles.

	Elements (%)																No.
Date	Na	Mg	Al	Si	P	S	Cl	K	Ca	Ti	Cr	Fe	Ni	Cu	Zn	Pb	
Jan. 1, 1986	43	30	53	47	7	63	57	63	53	7	0	37	0	3	3	10	30
Jan. 5, 1986	53	47	33	40	0	40	80	40	47	3	13	27	3	0	7	3	30
Jan. 16, 1986	37	53	63	60	0	63	40	63	57	13	0	53	0	7	7	7	30

Regarding the aerosol particles in the free atmosphere as seen in the table, several kinds of elements were prevalent differing from the cases of center nucleus. That is to say, in the case of January 1, the elements of Al, Si, S, Cl, K and Ca were contained abundantly. In the case of January 5, on the other hand, the elements of Na, Mg, Cl and Ca were abundant. The tendency of the case of January 16 was a quite similar to the first one. Comparing the cases of January 1 and 16 with the case of January 5, the difference between both cases was clear, that is to say, the former prevailed in soil particles and combustion products and the latter prevailed in sea salt particles mainly.

4.3 Component ratios of typical elements contained in the aerosol particles

Figure 16 shows ratios of Si : Na : S in triangle diagrams described above for three snowfall days. In this figure, solid and open circles show large (0.2 μm $\leq$d$<$2 μm) and giant (d$\geq$2 μm) particles, respectively. However, we have no discussion about the difference between the particle sizes. As seen in the figure, it is understood that the frequency of chemical components of aerosol particles was larger in the elements of Si for all days. It was recognized, however, that the chemical components of aerosol particles when the snow crystals of low temperature types were observed, have a tendency to contain soil particles and

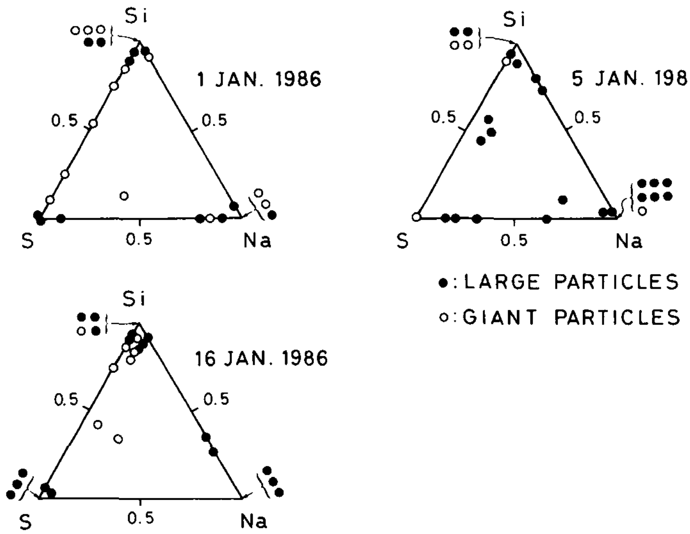


Fig. 16. Triangle diagrams of the typical three elements of Si, Na and S of the aerosol particles on each date.

combusition products, that is to say, the data are scattered on the line between the apexes of Si and S. On the other hand, the chemical components of aerosol particles when the plate type crystals were observed, have a tendency to contain sea salt particles, that is to say, the data are scattered on the line between apexes of Si and Na.

5. Relation between chemical components of the center nucleus of snow crystals and aerosol particles in the free atmosphere

In the previous sections, the chemical components of the center nucleus of snow crystals and aerosol particles in the free atmosphere were discussed separately. Then, in this section, we have examined the relation between chemical components of the center nucleus and aerosol particles. Figure 17 shows sounding curves when the polycrystals were falling mostly on January 1, 14 and 16, 1986. In this figure, the ordinate and abscissa shows the height and temperature, respectively, and solid large circles, open circles, and solid small

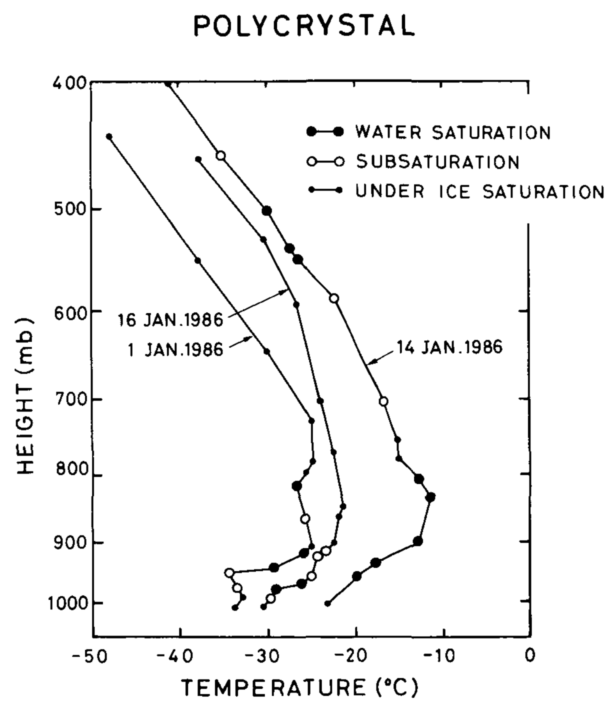


Fig. 17. Sounding curves when the polycrystalline snow crystals were falling mostly.

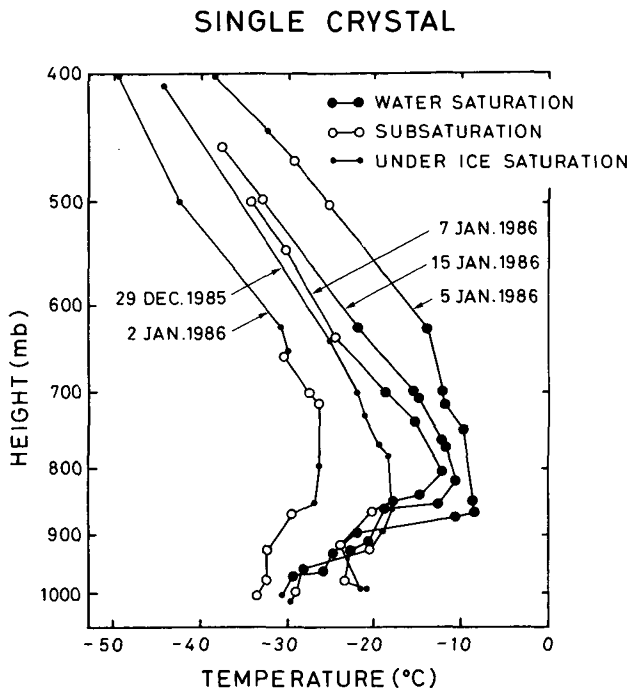


Fig. 18. Same as Fig. 17 but for single crystals were falling mostly.

dots show the water saturation, subsaturation (ice saturation and under water saturation) and under ice saturation, respectively. According to the observations of the Polar Experiment-North (POLEX-North) by Takeda et al. (1982) at the same observation site, sounding curves of January 1 and 16 except for January 14 were classified as the cold periods. Under these conditions, the observation site was covered by the arctic air masses and the snow crystals of low temperature types were frequently observed with a combination of bullets and crossed plates. On the other hand, the sounding curve of January 5 as shown in Fig. 18 was classified as warm periods (Takeda et al., 1982) except for January 2. Under these conditions, the observation site was covered by warm air from the Northern Pacific air masses. It was considered therefore that during cold periods when the snow crystals of low temperature types fell, soil particles and combustion products were predominant as aerosol particles in the free atmosphere. On the other hand, during warm periods when the general shapes of snow crystals fell, soil particles and sea salt particles were predominant. Then, we examined the relation between the chemical components of

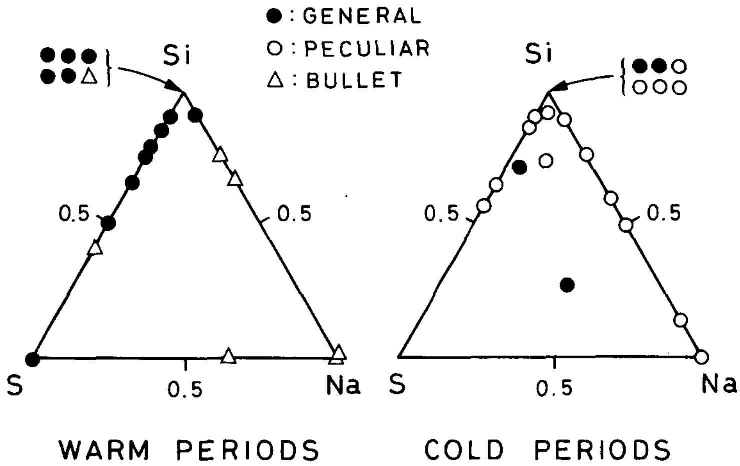


Fig. 19. Triangle diagrams of the typical three elements of Si, Na and S of the center nucleus of snow crystals in the warm and cold periods.

center nucleus of snow crystals and warm and cold periods. Figure 19 displays results obtained. In this figure, solid circles, open circles, and open triangles show general shapes, peculiar shapes of low temperature types including gohei twin, seagull and so on, and a combination of bullets, respectively. As seen in this figure, in the warm periods, the general shapes of snow crystals have soil particles and combustion products as their nucleus and the combination of bullets have soil particles and sea salt particles as their nucleus. On the other hand, in the cold periods, the snow crystals of low temperature types (peculiar shapes) have soil particles and sea salt particles as their nucleus.

6. Concluding remarks

Regarding the analyses of chemical components of center nucleus of snow crystals, the shapes of snow crystals were divided into four groups; plate and column, combination of bullets, gohei twin, and seagull type crystals. A higher frequency of Si as a center nucleus was recognized in all types of snow crystals. Therefore, the center nucleus of snow crystals around Inuvik area appears to predominantly consist of soil particles. Next, we classified the snow crystals into single, namely, plate and column types, and polycrystals, namely, gohei twin, seagull, and combination of bullets. As a result, the chemical components of single crystals were found to be nearly equal to polycrystals in soil particles, that is to say, Si, Al and Ca. On the other hand, the components of polycrystals were higher than that of single crystals in sea salt particles, that is, Na, Mg, Cl and K. Furthermore, the analysis of chemical components of airborne aerosol particles during snowfalls was carried out classifying them into the cold periods and warm periods when the observation site was covered by the Arctic air masses and the Northern Pacific air masses, respectively. During the cold periods, the predominant chemical components were soil particles and combustion products. And in the warm periods, the predominant components were soil particles and sea salt particles in the free atmosphere. If the chemical component ratio of the center nucleus of snow crystals was classified into the cold and warm periods, it may be concluded that plate and column type crystals have soil particles and combustion products as a center nucleus. On the other hand, polycrystals including the snow crystals of low temperature (peculiar) types have soil particles and sea salt particles as the center nucleus. This result showed the quite similar tendency which was obtained by Kikuchi et al. (1982) in Hokkaido, the northern part of Japan. It is considered therefore that the sea salt particles are more suitable materials as condensation nuclei, however, they can not freeze easily in relative warmer temperature ranges below the freezing point by their salinity, that is to say, they are unsuitable materials as ice nuclei in relative warmer temperature ranges below the freezing point. Therefore, the sea salt particles which condensed water vapor on their surface would be act as ice nuclei in relative lower temperature ranges. It is well known that the water droplets are frozen in a polycrystalline structure in relative lower temperature ranges below $-20^{\circ}C$ in the laboratory experiments (Murray and List, 1972; Uyeda and Kikuchi, 1978). It stands to reason therefore that the snow crystals of low temperature types such as gohei twin, seagull, and other square form type crystals which have a polycrystalline structure are observed frequently below $-25^{\circ}C$ in the free atmosphere.

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