Concentration and nature of ice nuclei in rim of the North Pacific Ocean

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ABSTRACT

Simultaneous collections of ice nuclei in the air were made with instruments of the same type at four sites in the rim of the North Pacific: College, Alaska; Blue Glacier, Washington; Mauna Loa, Hawaii; and Nagoya, Japan. Ice nuclei were collected on filter paper for counting of their number and also sheet meshes for examination with electron microscope.

During the period of the collection from the beginning of February through the beginning of March, 1968, three marked maxima of the concentrations of ice nuclei effective at $-15°C$ appeared at each of the sites except Mauna Loa. The peak values were the largest at Nagoya (5.3 nuclei/litre) followed by College (2.7 nuclei/litre) and Blue Glacier (1.3 nuclei/litre). At the Mauna Loa Observatory, no marked peak was observed. Neither a diurnal variation nor any other variations with a specific period have been detected. The result of identification of materials of ice nuclei collected at the four sites shows that clay and other mineral particles constitute the main part of the ice nuclei.

The results of the studies on the features of ice nucleus concentration, the trajectories of air masses and the examination of ice nuclei suggest that the ice nuclei detected originated from arid and semi-arid regions of the Asian Continent.

1. Introduction

The origins and the natures of atmospheric ice nuclei have been pursued by many workers on the basis of observations of ice nculeus concentration (e.g. Schaefer, 1954; Isono et al., 1959*a*, 1959*b*; Soulage, 1957; Bigg & Miles, 1964) and examination of center nuclei of snow crystals with electon-microscope (Kumai, 1951, 1961; Isono, 1955, 1959; Kumai & Francis, 1962). According to the results of these studies, particles of clay and other minerals have been found to constitute a great part of the atmospheric ice nuclei. However, meteoritic dust particles and some kinds of combustion products from industrial sources have also been proposed to be important constituents.

In most of the work which has been done, observations have been made of ice nucleus concentrations with the use of ice nucleus counters and their origins have been inferred. Ice nucleus counters of various types have been designed and used for this purpose. The phenomenon of ice nucleation is very complex and its occurrence on a particle of a specific material depends not only on temperature and supersaturation, but also on the environmental history of the particle. Generally speaking, therefore, ice nucleus concentrations measured with different counters differ from each other. In the case of actual clouds, even particles of the same material and the same size will behave differently from cloud to cloud.

In order to determine the origin and the nature of ice nuclei and to obtain useful information on cloud glaciation, it is necessary to collect and count ice nuclei simultaneously at several sites with instruments of the same type and to determine their sizes and their materials. The purpose of our observations was to obtain information on the nature and the origin of ice nuclei which must play an important part in the glaciation of super-cooled clouds over the North Pacific area.

This work was carried out on the basis of

Japan–United States Scientific Cooperative Program. As the instruments used by our colleagues on the United States side were NCAR Ice Nucleus Counters, which were different from those used by our group, it was not necessarily expected that values of concentrations of ice nuclei determined by both instruments would coincide with each other.

2. Locations of sites of collections

The collections were made at four sites in the rim of the North Pacific: Nagoya University, Nagoya, Japan; Minitruck Station of the University of Alaska, College, in the suburbs of Fairbanks, Alaska; the Blue Glacier Observatory of the University of Washington on Mt. Olympus to the west of Seattle, Washington; and the Mauna Loa Observatory of ESSA, Hawaii (Fig. 1). Measurements were taken during the period from the beginning of February through the first week of March, 1968.

The reason why we chose these four sites is as follows. We had made observations of ice nuclei in Tokyo and Nagoya and found that clay minerals blown up from ground in the semi-arid region of the North China constitute an important part of ice nuclei in the air over Japan (Isono et al., 1959*a*). We also found that volcanic dust ejected from active volcanoes when they erupted was active as ice nuclei. It was suspected that clay minerals from the North China may be carried by the westerlies to the Pacific coast of the continent of the North America, especially in winter. In order to collect and identify these loess or clay particles, volcanic dust and meteoritic dust particles, it was thought preferable to collect ice nuclei at sites where air is as clean as possible and free from dust from nearby ground and industrial areas. In winter the ground is covered with snow around the Blue Glacier Observatory and the station at College, and the trajectories of air masses which arrive at these sites do not pass

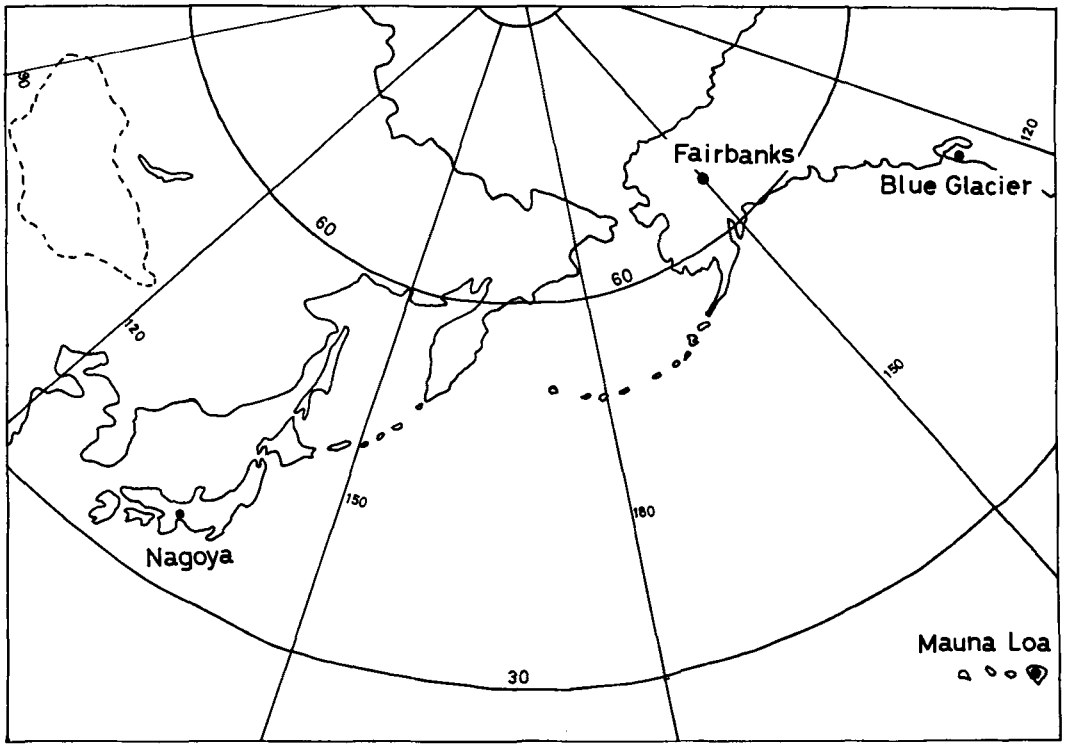

Fig. 1. Sites of collections of ice nuclei: Nagoya University, Nagoya; Minitruck Station, University of Alaska, College, in the suburbs of Fairbanks, Alaska; Blue Glacier Observatory, University of Washington, Mt. Olympus, Washington; and Mauna Loa Observatory, ESSA, Hawaii.

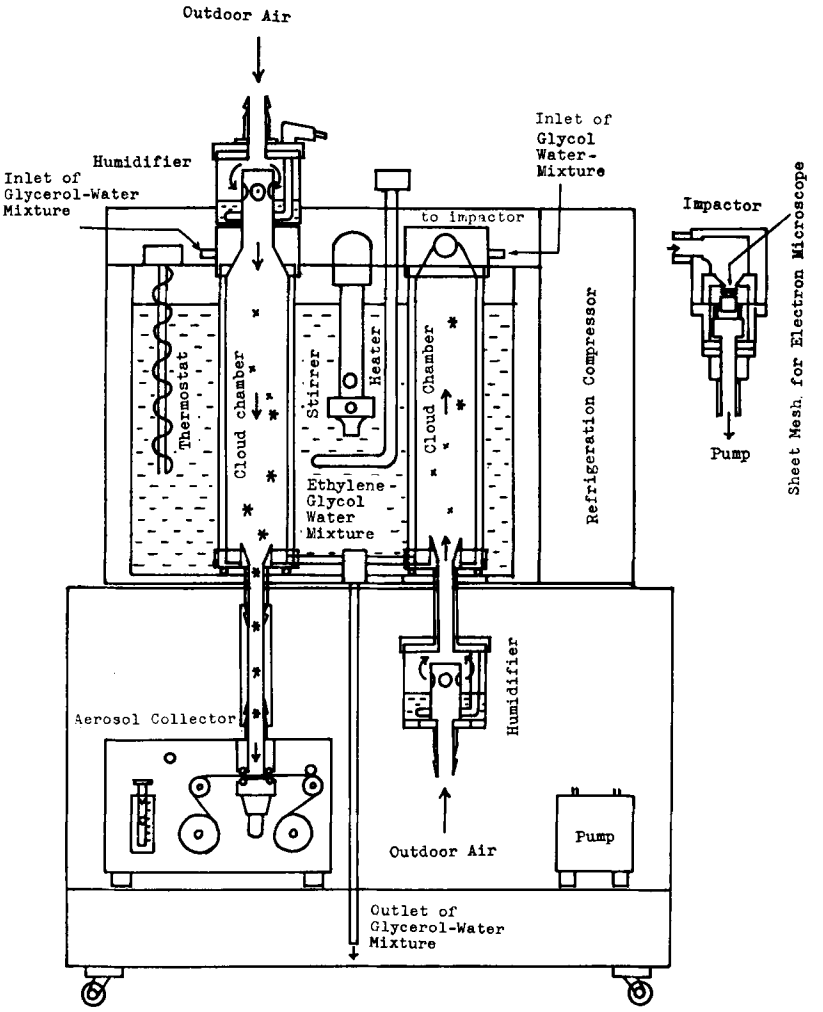

Fig. 2. Schematic illustrations of ice nucleus collector. On the right side of the figure, the impactor is illustrated in an enlarged scale.

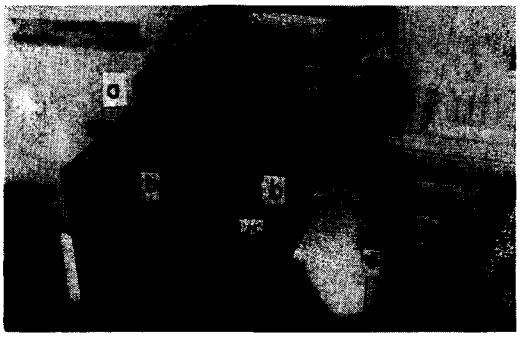

Fig. 3. Upper part of ice nucleus collector; humidifier (*a*), cloud chamber (*b*), impactor (*c*), stirer (*d*) and freezer (*e*).

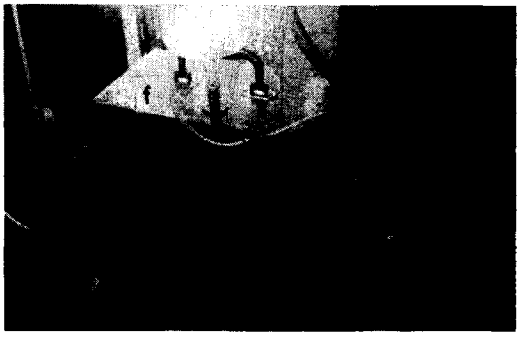

Fig. 4. Lower part of ice nucleus collector; aerosol collector (*f*) and pump for impactor (*g*).

through arid or semi-arid regions which might supply the air masses with soil particles. At the Mauna Loa Observatory the air is very clean and generally free from pollution.

At each station ice nuclei were obtained with a collector, the design and the operation of which will be described in the next section. Besides this, dust particles in the air were collected with millipore filters; these particles are now being examined with electron microscope and microfocused X-ray analyser.

3. Instrumentation

(a) *Apparatus used for the observations*

The apparatus used for the observation was designed to be portable. The schematic drawing of the unit is shown in Fig. 2 and photographs in Figs. 3 and 4. The complete assembly weights 40 kg. It consumes a maximum of 3 amperes at 100 V, 60 cycles.

The apparatus consists of two parts. The first is an ice nucleus collector for counting the number of ice nuclei and the second is a collector of ice nuclei which are to be examined under the electron microscope. Each of the collectors has a cloud chamber which is mounted in a refrigerator filled with ethylene glycol-watermixture. The upper and lower ends of one of the cloud chambers are connected to a humidifier and an aerosol collector, respectively. Those of the other cloud chamber are connected to an impactor and a humidifier, respectively. The water temperature in the humidifier is usually kept at about 30°C and the temperature of the humidified air is approximately 25°C.

Both cloud chambers consist of a brass cylinder having eight helical ridges wound on the inner wall from top to bottom in parallel. The dimensions of the cylinder are 50 mm in inside diameter and 300 mm in length. The humidified air is cooled down to −15°C after travelling about half of the way from the inlet to the outlet where the air temperature becomes −20°C. At the top of the cylinder a glycol-water mixture (two parts of glycol and one part of water) is supplied to the helical ridges and flows down along them to a drain at the bottom. This device prevents frost formation on the wall. The helical ridges increase the rate of heat exchange between the wall and air.

(b) *Method of measurement of ice nucleus concentration*

As shown in Fig. 2, outdoor air containing ice nuclei which is introduced into the left cloud chamber at the rate of 0.5 litre/min is humidified and cooled down to −20°C, by the time it reaches the outlet of the chamber. Ice crystals formed on ice nuclei grow to a size about 50 microns in diameter. The ice crystals are collected on filter paper in the aerosol collector. As the pore size of the filter paper is a few microns, the ice crystals grown in the cloud chamber are collected. As the temperature of filter paper is kept higher than 0°C, the ice crystals collected on the filter paper melt and subsequently evaporate. A roll of filter paper is rolled up intermittently and each section of the roll is exposed to the air flow for 29 min. The size of each of the sections is 25 mm in diameter.

The number of ice nuclei collected on the filter paper is counted as follows. Each of the sections is cut off from the roll of filter paper and put in a metal dish. A pad of sponge soaked with water at room temperature is used for humidification. The metal dish is put on the lid with the sponge pad and subsequently placed on a cooled metal plate kept at −15°C in a refrigerator. After about 10 min, the sponge pad is removed and supercooled sugar solution (120 g of sugar in 100 ml of water; −12°C) is poured into the dish. Ice crystals formed on the section of filter paper are counted.

Even if frost forms on some part of the cloud chamber and is collected on the filter paper, this does not produce an error in the ice nucleus count with this method. Background noises have been found to be very low.

(c) *Collection of ice nuclei for electron microscope examination*

As shown in Fig. 2, a sheet mesh covered with collodion (0.5 %) film is mounted in the impactor connected with the right cloud chambers. Air from the outlet of the cloud chamber is sucked through the impactor at the flow rate of 0.5 litre/min by means of pump.[1] The collection efficiency on the sheet mesh of this impactor is 100 % for particles larger than 25 microns in diameter at that flow rate and 50 % for particles of 17 microns in diameter. Particles smal-

[1] Each sheet mesh is exposed to the air flow for 12 hours

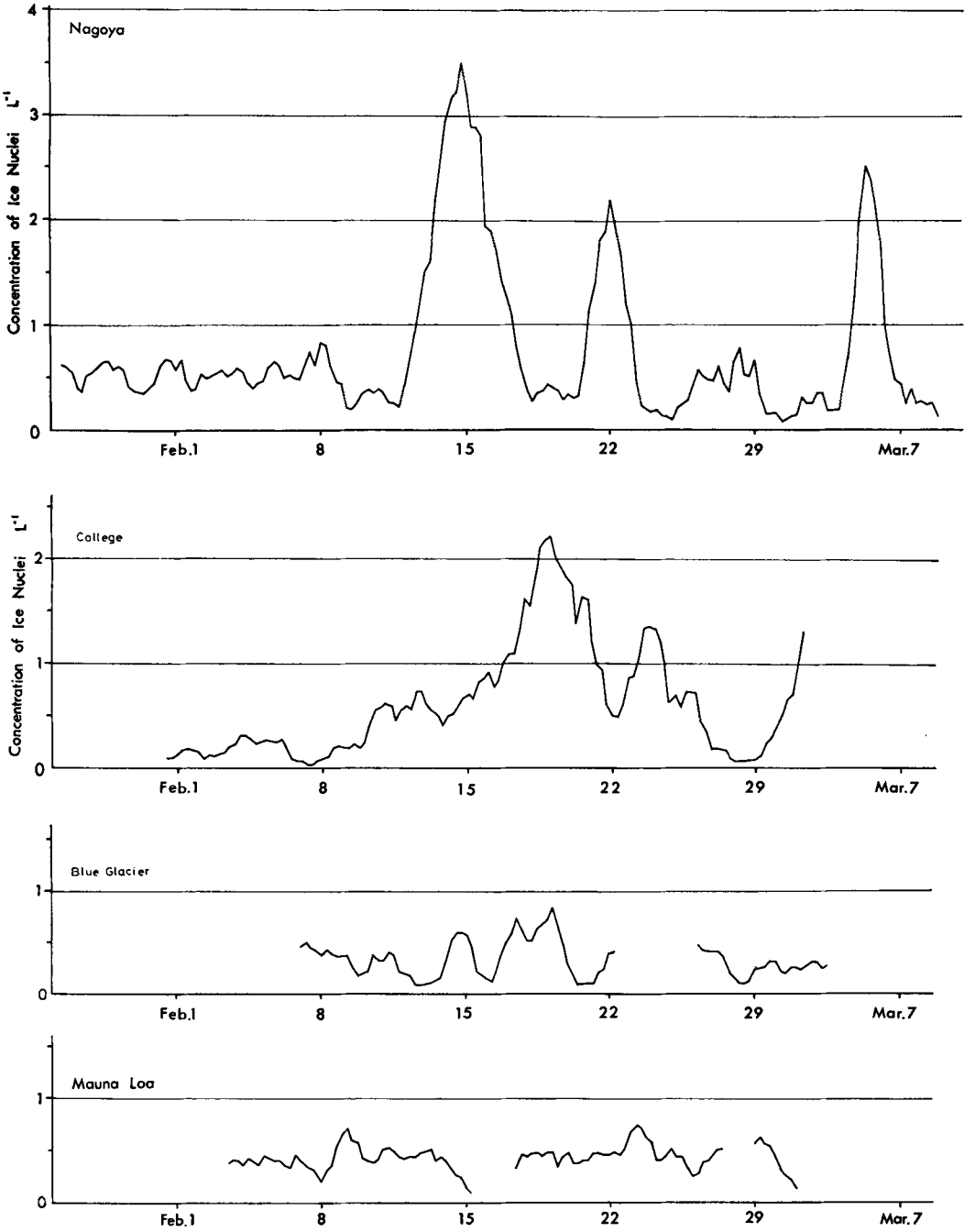

Fig. 5. Running average of ice nucleus concentration with date. Running average is taken for a period o 28 hours.

ler than 10 microns in diameter are not collected. The samples of ice nuclei collected on the sheet mesh are examined with electron microscope and their materials are identified

4. Concentrations of ice nuclei at four sites and their variations with respect to time

By means of the cold chamber–filter paper–sugar solution method described in the previous section, the number concentrations of ice nuclei effective at −15°C were counted at the four sites. At Nagoya, College and the Blue Glacier the variations in the number concentration of ice nuclei with respect to time showed similar features. However, a different feature was observed at Mauna Loa.

The common feature of the variations for Nagoya, College and the Blue Glacier was that at each site there were three groups of days with markedly high concentrations of ice nuclei. This feature can be seen in Fig. 5 where a 28 hours running average of counted values are illustrated. The durations of each group were between three days and a week. As seen from Table 1, each group of the days with markedly high concentration consists of several peaks of the concentration of ice nuclei which occurred during these days.

On the other hand, in Hawaii, irregular small peaks continued to occur throughout the whole

Table 1. *Concentration of ice nuclei effective at −15°C at four sites (per litre)*

Date	Local Standard Time 00	06	12	18
Nagoya				
Jan. 26	0.26	0.73	1.33	0.66
27	0.05	0.20	0.49	0.58
28	0.43	0.89	0.37	0.69
29	0.81	0.49	0.52	0.50
30	0.49	0.02	0.37	0.37
31	0.44	0.77	0.25	1.20
Feb. 1	0.70	0.37	0.30	0.72
2	0.28	0.14	0.50	1.03
3	0.44	0.41	0.30	0.65
4	0.69	0.62	0.64	0.14
5	0.10	0.45	0.84	0.80
6	0.78	0.34	0.29	0.25
7	0.88	0.65	0.30	0.96
8	0.90	0.18	1.79	0.17
9	0.04	0.05	0.29	0.49
10	0.10	0 34	0.48	0.45
11	0.38	0.26	0.16	0.00
12	0.38	0.22	1.20	1.33
13	1.27	2.00	1.73	1.73
14	4.00	3.33	4.00	2.67
15	2.00	5.33	1.93	2.40
Feb. 16	2.67	1.60	1.01	1.73
17	1.47	1.33	0.76	0.17
18	0.16	2.93	0.49	0.24
19	0.49	0.29	0.61	0.34
20	0.08	0.05	0.61	0.38
21	0.46	1.53	2.67	1.73
22	2.53	0.89	3.07	1.47
23	0.32	0.18	0.11	0.30
24	0.18	0.17	0.09	0.18
25	0.02	0.04	0.05	0.77
26	0.32	0.22	0.77	0.80
27	0.49	0.10	0.24	1.33
28	0.09	0.02	1.60	0.84
29	0.02	0.04	0.69	0.00
Mar. 1	0.00	0.04	0.08	0.33
2	0.01	0.13	0.14	0.93
3	0.01	0.05	0.60	0.13
4	0.09	0.04	0.06	1.87
5	1.60	2.67	4.00	2.40
6	1.25	0.34	0.10	0.52
7	0.25	0.20	0.14	0.09
8	0.70	0.08	0.28	0.01
9	0.16	0.04	0.10	0.05
College				
Jan. 31			0.10	0.10
Feb. 1	0.13	0.20	0.40	0.05
2	0.13	0.08	0.10	0.10
3	0.20	0.06	0.20	0.21
4	0.36	0.29	0.45	0.20
5	0.05	0.14	0.38	0.52
6	0.10	0.04	0.02	0.00
7	0.00	0.00	0.04	0.08
8	0.06	0.17	0.10	0.12
9	0.50	0.13	0.05	0.10
10	0.32	0.33	0.40	1.07
11	0.66	0.40	0.53	0.26
12	0.40	1.13	0.60	0.38
13	1.13	0.41	0.46	0.29
14	0.14	0.66	0.84	0.56
15	0.73	0.50	0.84	0.60
16	1.40	0.89	0.77	0.17
17	0.93	2.40	1.16	0.70
18	1.24	2.53	2.00	2.67
19	2.00	1.64	2.67	0.93
20	1.15	2.67	1.37	0.66
21	2.27	1.01	*	0.33
22	0.46	0.56	0.44	0.57
23	0.93	1.73	0.64	1.33
24	2.00	1.07	1.47	0.26
25	0.14	0.12	1.40	0.89
26	1.07	0.06	0.08	0.05
27	0.42	0.21	0.10	0.00
28	0.00	0.01	0.12	0.10
29	0.05	0.05	0.02	0.28
Mar. 1	0.73	0.32	0.50	0.64
2	1.05	0.97	1.67	2.13

(*Table 1 cont.*)

Date	Local Standard Time 00	06	12	18
Blue Glacier				
Feb. 6			0.53	0.42
7	0.24	0.36	0.71	0.69
8	0.20	0.08	0.16	0.98
9	0.48	0.10	0.09	0.13
10	0.44	0.11	0.17	0.24
11	0.91	0.18	0.08	0.57
12	0.11	0.12	0.04	0.03
13	0.12	0.07	0.18	0.05
14	0.19	0.26	0.82	1.27
15	0.37	0.18	0.17	0.22
16	0.07	0.17	0.01	0.04
17	0.85	0.73	0.77	0.47
18	0.85	0.32	0.11	0.82
19	1.00	1.05	0.56	0.67
20	0.00	0.04	0.16	0.00
21	0.16	0.07	0.05	0.20
22	0.52	0.32	0.83	0.17
23	*	*	*	*
24	*	*	*	*
25	*	*	*	*
26	0.73	*	0.21	0.33
27	0.73	0.49	0.46	0.21
28	*	0.01	0.08	0.12
29	0.12	0.20	0.60	0.16
Mar. 1	0.18	0.42	0.16	0.12
2	0.07	0.44	0.42	0.08
3	0.29	0.32	0.44	0.05
4	0.23			
Mauna Loa				
Feb. 3	0.46	0.26	0.34	0.33
4	0.46	0.69	0.15	0.15
5	0.61	0.35	0.49	0.59
6	0.11	0.50	0.33	0.25
7	0.42	0.82	0.15	0.07
8	0.15	0.05	0.49	0.77
9	0.38	1.06	0.59	0.69
10	0.23	0.25	0.30	0.49
11	0.66	0.42	0.66	0.37
12	0.33	0.39	0.34	0.70
13	0.39	0.53	0.45	0.43
14	0.22	0.53	0.31	0.17
15	0.03	0.13	0.06	0.02
16	*	*	*	*
17	0.13	0.26	0.78	0.11
18	0.34	0.79	0.15	0.93
19	0.09	0.27	0.91	0.14
20	0.18	0.65	0.47	0.39
21	0.13	0.33	0.66	0.79
22	0.37	0.10	0.33	0.79
23	0.61	0.79	0.86	0.62
24	0.71	0.15	0.47	0.06
25	0.65	0.95	0.42	0.07
26	0.03	0.15	0.50	0.59
27	0.65	0.05	0.50	0.70
28	*	*	0.07	0.53
29	0.66	0.66	0.79	0.39
Mar. 1	0.26	0.46	0.15	0.26
2	0.06	0.05	0.15	

observation period, but there was no markedly high concentration. The average concentration of ice nuclei throughout the whole period at Mauna Loa was about 0.4 nuclei/litre which is similar to the concentrations of ice nuclei at College and the Blue Glacier except on those days with markedly high concentrations.

The highest values observed at each site were 5.3 nuclei/litre at Nagoya on 15 February, 2.7 nuclei/litre at College on 20 February, 1.3 nucleus/litre at the Blue Glacier on 14 February and 1.1 nucleus/litre at Mauna Loa on 9 February. This suggests that Nagoya was positioned nearest to a source region of ice nuclei if the main portions in the numbers of ice nuclei observed at each site were originated from a common source region in the northern hemisphere.

Inspecting the running averaged curves in Fig. 5 for Nagoya, College and the Blue Glacier and if it were permitted to postulate that the air of high concentration of ice nuclei moved from west to east over the Pacific area owing to westerlies, one could consider that the peak at Nagoya on 14 February reached College on 19 February where the value of the peak had been lowered and the duration period of the peak (the width of the peak with respect to time) had been broadened on account of a large scale atmospheric diffusion. Also one could consider that the same peak reached the Blue Glacier at about the same time.

If this theory is correct, the travelling time of the peak of ice nucleus concentration from Japan to Alaska and the West Coast of the United States would be about five days. Similarly one could refer other peaks observed at College and the Blue Glacier back to the corresponding peaks observed at Nagoya by a lag period of a few days.

It is difficult to apply the method of trajectory analysis to the very air parcel which reached College or the Blue Glacier after a long distance of travel over the Pacific Ocean. Nevertheless, it cannot be considered to be meaningless to apply the trajectory analysis to the air mass which covered College or the Blue Glacier area and trace back its general path in order to estimate its general travel velocity. The reason why trajectories on 500 mb isobaric surface were used instead of those on an isentropic surface is as follows. Dust clouds from semi-arid regions in the Asiatic Continent are often observed in the western part of Japan.

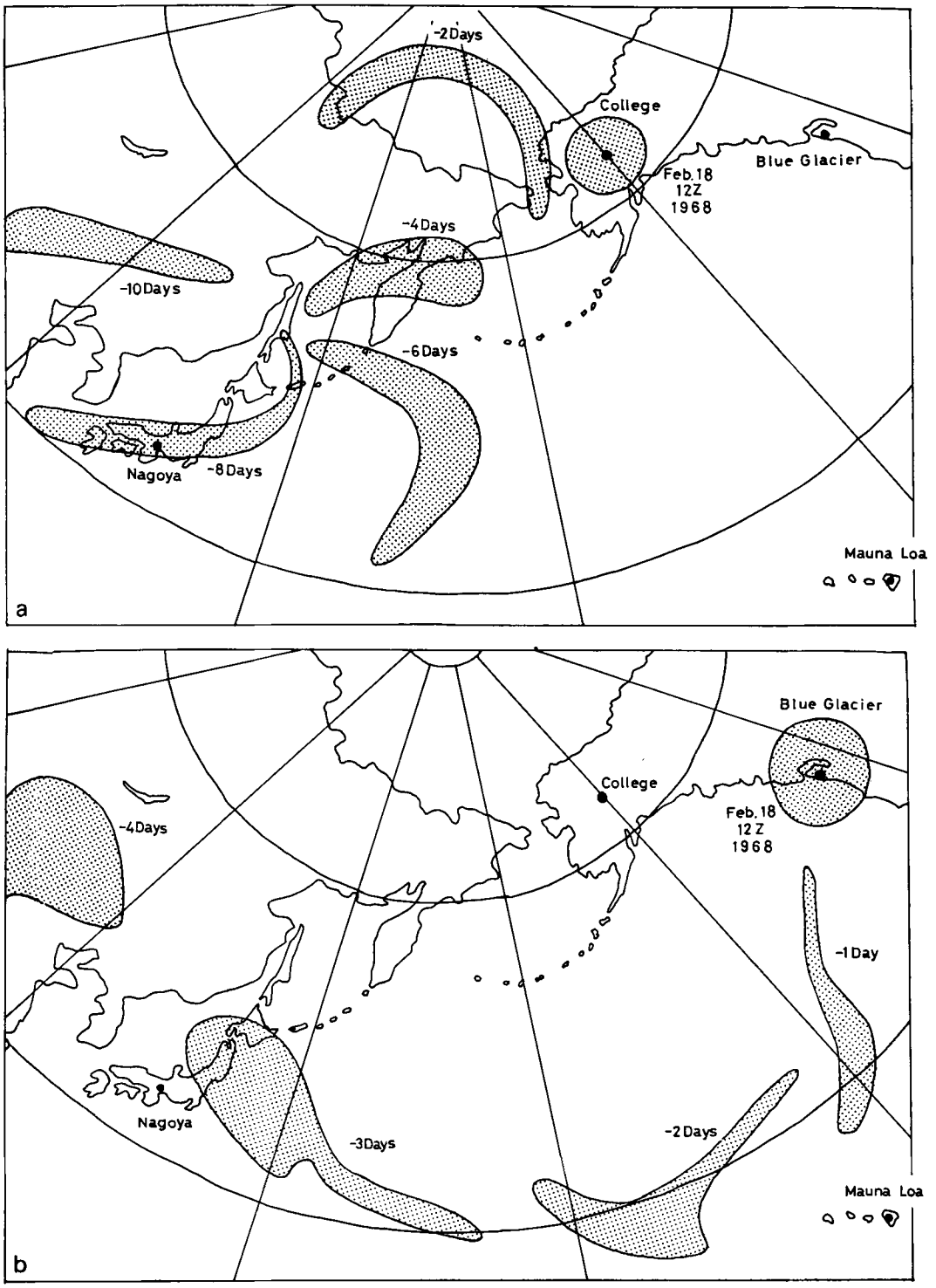

Fig. 6. Air trajectory at the level of 500 mb to College (*a*) and Blue Glacier (*b*).

An example of them was described in a previous paper (Isono et al, 1959*a*). In such cases dust particles were often observed from aircraft even at levels between 3 000 m and 5 000 m above sea level. When dust particles, especially those which were active as ice nuclei, were transported by air currents and came to cyclonic or frontal regions they would move along complicated trajectories, especially at low levels. In order to determine their sources it may be preferable to use trajectories on an isobaric surface which does not intersect frontal surfaces or mountain slopes. Fig. 6 shows the examples.

Fig. 6*a* indicates the past position and the shape of the air mass which took the shape of a circular disc with diameter 500 km around College on 18 February, 12 GCT, 1968 at the level of 500 mb. February 18 was one of the days with especially high concentration of ice nuclei observed at College. The movement of the air on 500 mb level was that the air on the Asian Continent at −10 days (10 days before) moved in a manner of cyclonic motion passing Japan at −8 days and reached Kamchatka at −4 days. This cyclonic movement was due to a long-lived upper cyclone which stayed over Sakhalin. Then, the air moved in a manner of anticyclonic motion passing the Arctic area and reached Alaska. This anticyclonic movement was due to a long-lived anticyclone which stayed near the Bering Strait.

It was considered that the air was under conditions of upward motion in the early period on account of the cyclonic motion and the air was under conditions of subsidence in the later period on account of the anticyclonic motion. The circumstances were favorable for dust particles of small size to be blown up into the atmosphere in the early period and to settle down towards the ground level in the later period.

Fig. 6*b* indicates the past position and the shape of the air which reached the Blue Glacier on 18 February, 12 GCT, 1968 in the shape of circular disc with diameter of 500 km at the level of 500 mb. February 18 was one of the days with high concentration of ice nuclei at the Blue Glacier. It can be seen that owing to the jet stream the air travelled across the Pacific Ocean in a relatively short period.

Though the trajectory analysis of such a long distance is inaccurate, if it were permitted to consider that the air from the Asia had travelled to the Blue Glacier owing to the jet stream in a shorter period than to College, the group of days with especially high concentration at College on 18–19 February should correspond to the peak at Nagoya on 8 February rather than the large peak at Nagoya on 14 February. As will be described in the following section, the electron-microscopic study has revealed that the size distribution of ice nuclei on 18 February at College was different from that at Nagoya on 14 February, while that at the Blue Glacier on 18 February coincides well with that at Nagoya on 14 February.

During the observation period the jet stream was always to the north of Hawaiian Islands and Hawaii was in a different air mass. The absence of any large peaks and the presence of irregular variation may support the idea that ice nuclei observed at Mauna Loa were of different origin from those in the westerly region in the North Pacific.

Fig. 7 shows the results of analyses of the diurnal variation in the concentration of ice nuclei. No appreciable diurnal variation was found at the four sites so far as ice nuclei effective at −15°C are concerned.

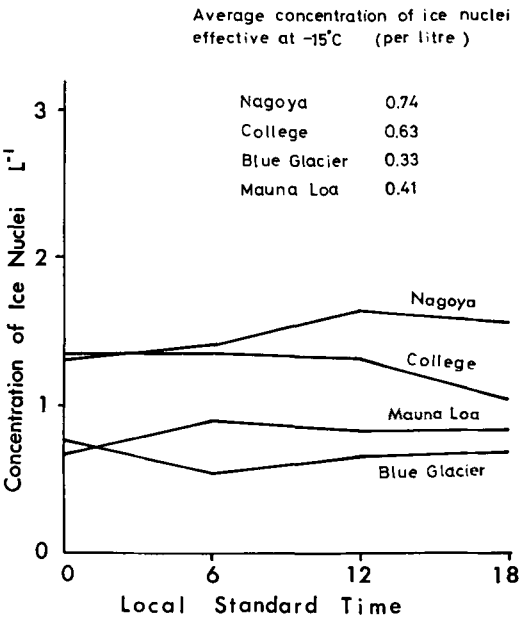

Fig. 7. Diurnal variation of ice nucleus concentration and average concentration throughout the whole period.

5. Materials of ice nuclei

Ice nuclei collected on collodion films were examined with an electron microscope.

Figs. 8 to 12 show several examples of electron-micrographs of ice nuclei collected in Nagoya together with their electron diffraction patterns. The first three micrographs show ice nuclei collected on 14 February when ice nucleus concentration was high in Nagoya. These were identified as mineral particles. They gave spot patterns as shown in Figs. 8*b* and 9*b*. Interplanar distances determined from these patterns are given in Fig. 13 together with those of kaolin minerals. Though the numbers of particles of this kind were small, they were also found on days when ice nuclei concentrations were low. Typical size distribution curves of ice nuclei in Nagoya are shown in Fig. 14*a*,

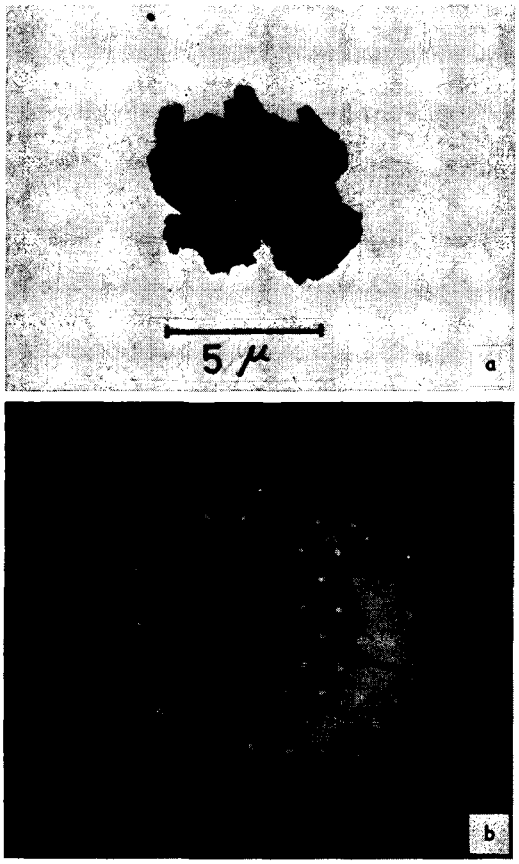

Fig. 8. (*a*) Ice nucleus (clay mineral) collected on 14 February at Nagoya and (*b*) its electron diffraction pattern (kaolin mineral).

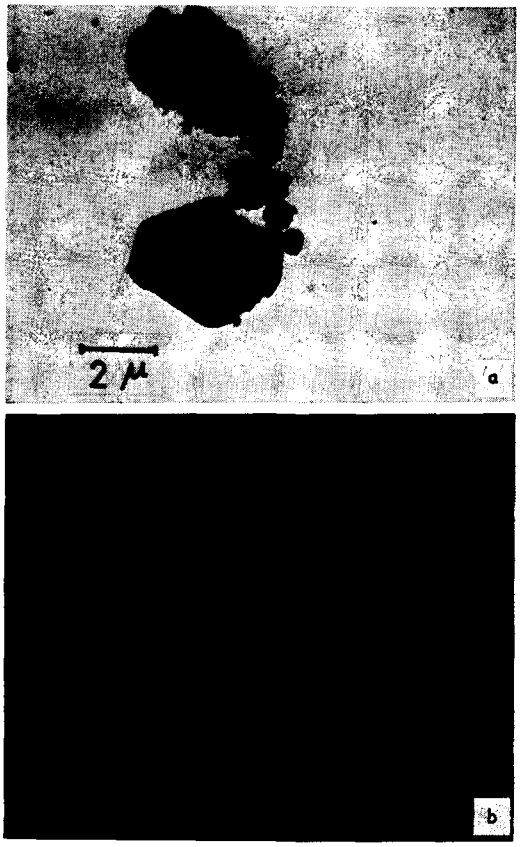

Fig. 9. (*a*) Ice nuclei (clay mineral) on 14 February at Nagoya and (*b*) the electron diffraction pattern from the particle of hexagonal shape (kaolin mineral).

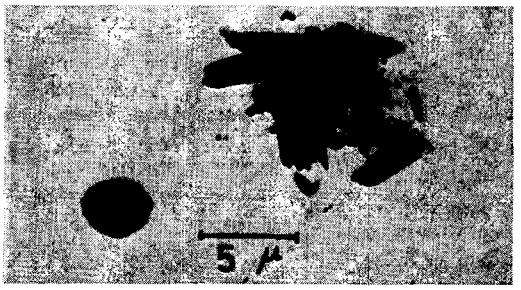

Fig. 10. Ice nucleus (clay mineral) on 14 February at Nagoya and condensation nuclei around it.

one is for a day with high ice nucleus concentration (February 14) and the other is for a day with low concentration (February 9). In the former case, particles larger than 4 microns constitute 50% of the total, whereas in the latter case 50% are particles smaller than 0.4 micron.

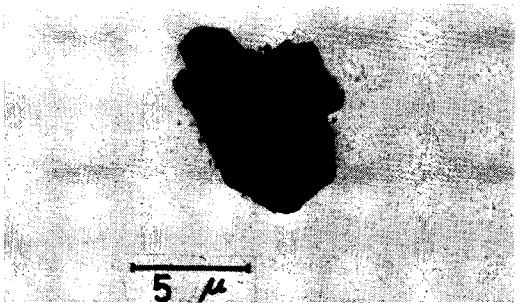

Fig. 11. Ice nucleus on 14 February at Nagoya (kaolin mineral).

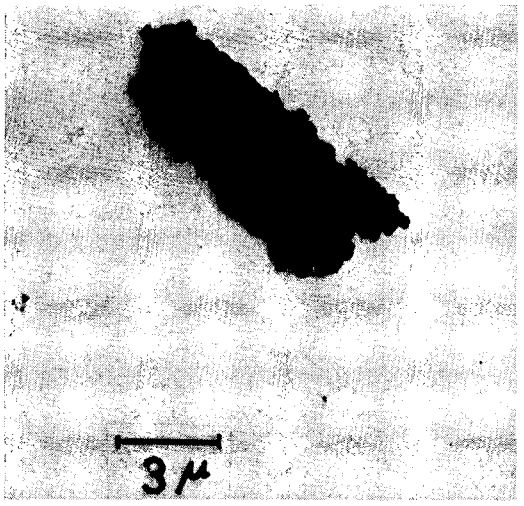

Fig. 12. Ice nucleus (clay mineral) on 25 January at Nagoya.

On days with low ice nucleus concentrations, particles which are considered to have originated from sea sprays are found. Figs. 15 and 16 show such particles. The shapes of the particles and the round traces or residuals of evaporated water droplets around them suggest that they acted as giant condensation nuclei and that water droplets which formed on them grew to sizes large enough to be collected by the impactor. The particles in Fig. 15*a* gave rings of NaCl as shown in Fig. 15*b*, whereas particles in Fig. 16*a* gave those of $CaSO_4$ and $CaCl_2$. One of the authors (Isono, 1959) found by the electron diffraction method that particles of different compositions formed when sea water was sprayed. Particles of sea salt constituents were commonly found in maritime air from the Pacific Ocean in Nagoya.

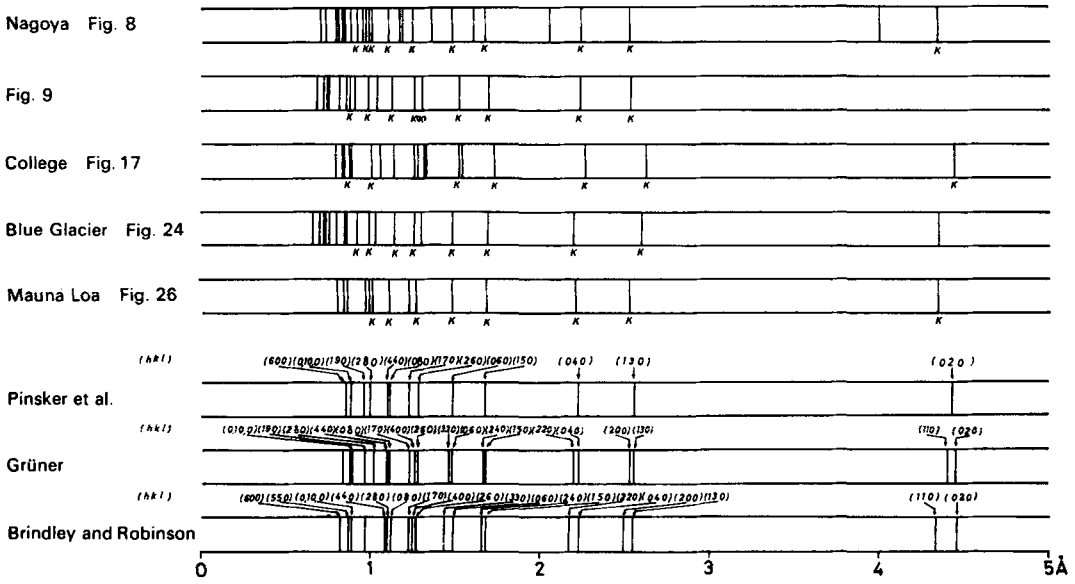

Fig. 13. Interplanar distances of crystal lattices obtained from electron diffraction patterns for particles shown in Figs. 8 and 9 (Nagoya), Fig. 17 (College), Fig. 24 (Blue Glacier) and Fig. 26 (Mauna Loa). Those of kaolinite obtained by Pinsker et al. (1948), Grüner (1932) and Brindley & Robinson (1945) are also shown in the figure.

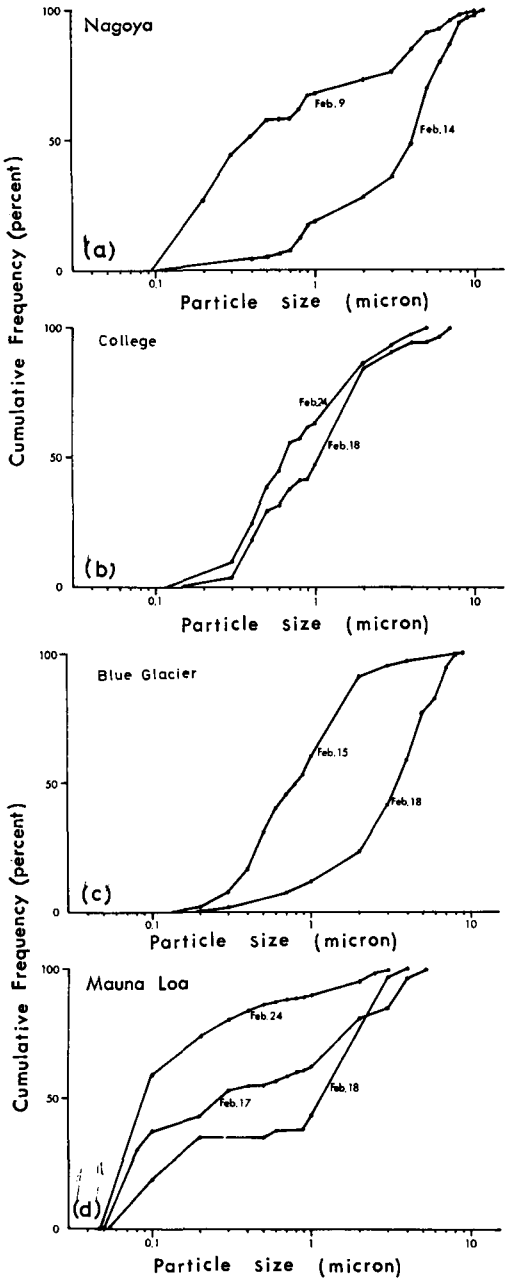

Fig. 14. Size distribution of ice nuclei of four sites. Cumulative frequency in percent is indicated.

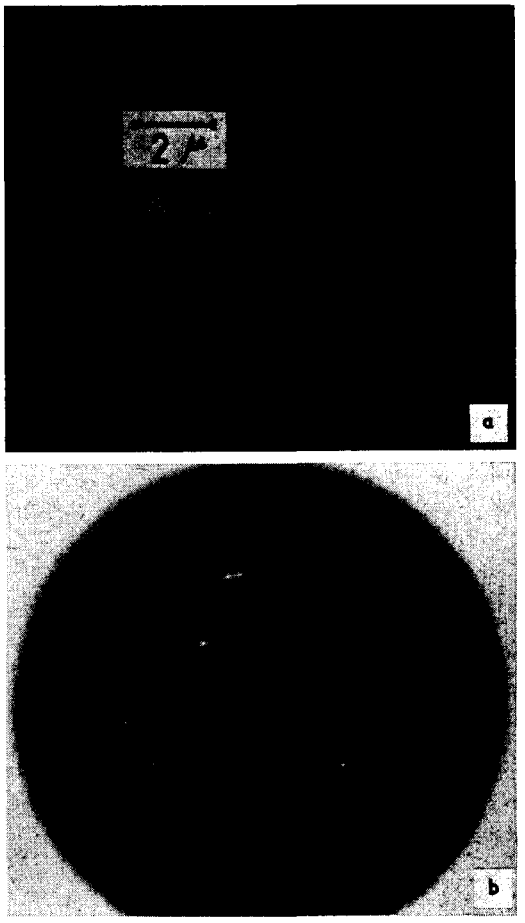

Fig. 15. (*a*) Giant condensation nucleus (sea salt) on 4 February at Nagoya and (*b*) its electron diffraction pattern (NaCl).

Figs. 17 to 19 show particles collected at College. Most of ice nuclei were clay minerals. Around these particles many tiny carbon particles were found. These are considered to have been discharged from the smokestacks of an electric power plant, other factories and houses. Carbon particles collected in Nagoya are shown in Fig. 20. It is likely that these tiny carbon particles were collected by the impactor after they had become attached to ice crystals formed on clay particles in the cloud chamber. The number density of the carbon particles on the surface of an ice nucleus was smaller than that in annulus area around the nucleus. Particles of $CaSO_4$ were also found at College as shown in Figs. 21 and 22.

Figs. 23–25 show particles collected at the Blue Glacier. They are again clay particles. Many tiny carbon particles are also seen around them. These are considered to have come from

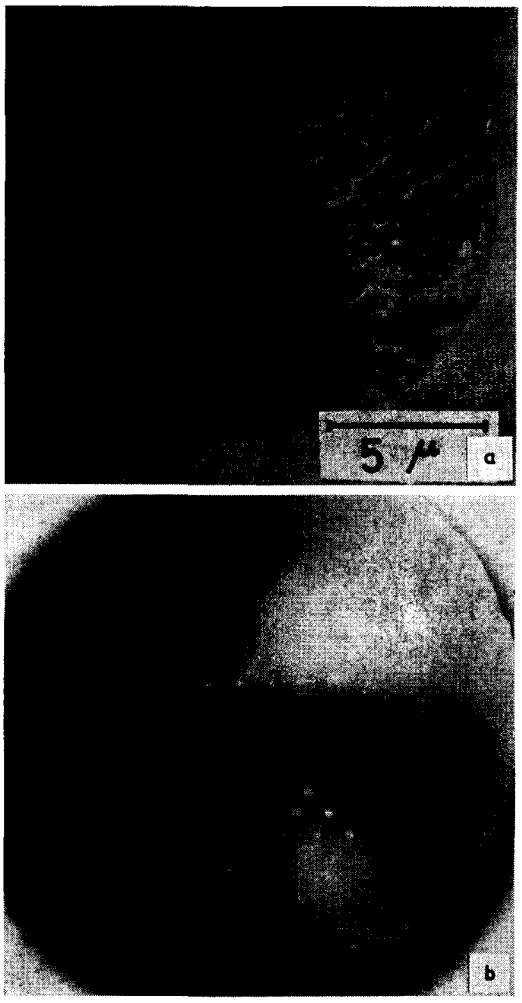

Fig. 16. (*a*) Giant condensation nuclei (sea salt) and (*b*) their electron diffraction pattern ($CaSO_4$ and $CaCl_2$).

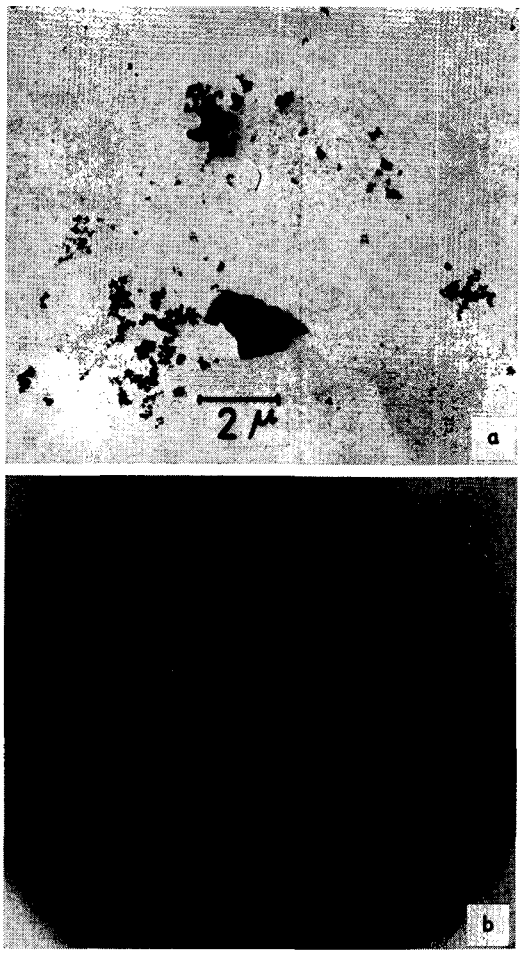

Fig. 17. Ice nucleus (clay mineral) and tiny carbon particles on 18 February at College and (*b*) its electron diffraction pattern (kaolin mineral).

engines used for electric power generator at the observatory. Clay particles collected at the Blue Glacier were similar to those collected in Nagoya and College. The fact that most of the clay particles collected at the Blue Glacier and College were not contaminated with sea salt seems to indicate that they came from a continent, having been carried by air, not from the sea surface near the station as suggested by some workers.

Particles in Figs. 26 to 28 were collected at Mauna Loa Observatory. As is seen in each photograph, contaminating particles such as carbon particles are completely absent around each ice nucleus. Ice nuclei collected there also consisted of clay and other mineral particles, but in comparison with those collected at the other three sites they were more massive and smaller in size on the average. Some of them may be of volcanic origin. Spherical particles shown in Figs. 29 and 30 were found at Mauna Loa. As we could not obtain electron diffraction pattern from them, we cannot identify their materials, exactly. It is likely that they are particles ejected from volcanoes on the island of Hawaii, though there is the possibility that they are of meteoritic origin. Examples of size distribution of ice nuclei collected at Mauna Loa are shown in Fig. 14*d*. They indicate that

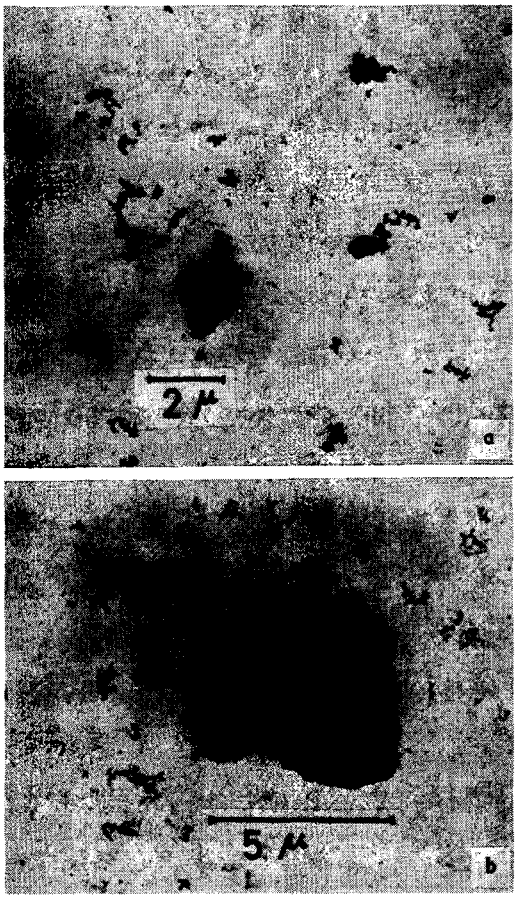

Fig. 18. (*a*) and (*b*) Ice nuclei (clay mineral) and tiny carbon particles around them on 18 February at College.

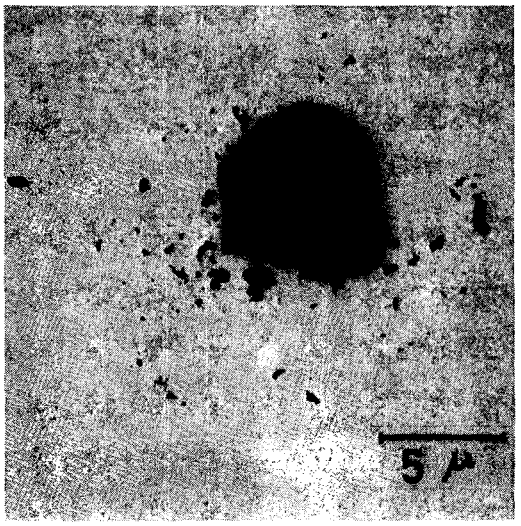

Fig. 19. Ice nucleus (clay mineral) and tiny carbon particles around it on 23 February at College.

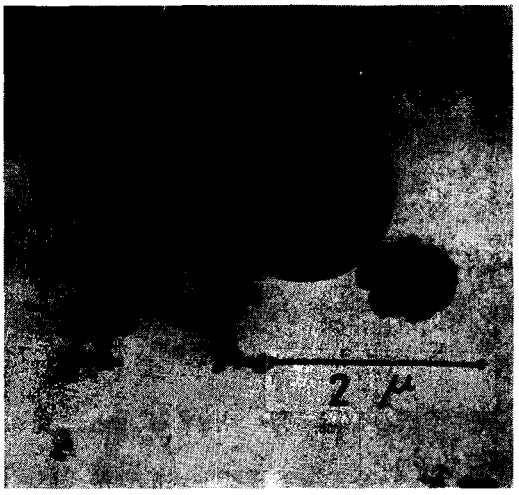

Fig. 20. Spherical carbon particles collected at Nagoya.

ice nuclei collected there were much smaller on the average than those collected at the other three sites.

Fig. 13 shows the interplanar distances of crystal lattices obtained from electron diffraction patterns for particles shown in Figs. 8, 9, 17, 24 and 26. Fig. 13 also shows those of kaolinite obtained from an oriented polycrystalline film of kaolinite in a position normal to the beam (Pinsker et al., 1948) and corresponding interplanar distances determined by Grüner (1932) and Brindley & Robinson (1945) (which are referred to by Pinsker (1953)). Remarkable coincidence can be seen between interplanar distances of ice nuclei collected at different sites and those of kaolinite obtained by Pinsker, taking into account that clay minerals which belong to a species (for instance, kaolinite) have lattice spacings which are different in a limited range according to specimens. As clay particles were collected on collodion film with their basal plane parallel to the surface of the film as shown in the photographs, only (*h k O*) reflections were obtained. Therefore, these particles collected at the four sites can be identified as kaolin minerals including kaolinite, nacrite and

Fig. 21. (*a*) Particle of $CaSO_4$ and tiny carbon particles on 22 February at College. (*b*) A part of the particle of $CaSO_4$ in higher magnifcation which gives spot pattern (*c*) of $CaSO_4$.

dickite. This result coincides with that of center nuclei of snow crystals collected in the middle

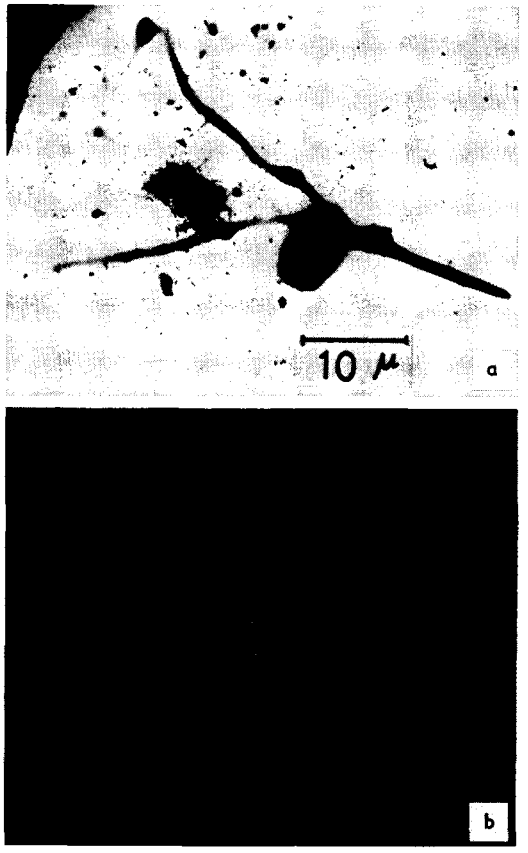

Fig. 22. (*a*) Particle of $CaSO_4$ and tiny carbon particles on 22 February at College. (*b*) Ring pattern of the particle of $CaSO_4$.

mountainous district of Japan which were identified by one of the authors (Isono, 1955).

Discussion

Figs. 14*a–d* show some examples of cummulative size distribution curves of ice nuclei collected at the four sites. It is seen in Fig. 14*a* that particles larger than 2 microns in diameter constitute about 75% of the total particles in the case of ice nuclei collected on 14 February at Nagoya, whereas only about 25% in the case of 9 February. On 14 February ice nucleus concentration at Nagoya was as high as 4 nuclei/litre, whereas on 9 February it was as low as 0.1 nuclei/litre. A similar feature is seen in Fig. 14*c* for the Blue Glacier, where a marked peak in ice nucleus concentration appeared on 18 February.

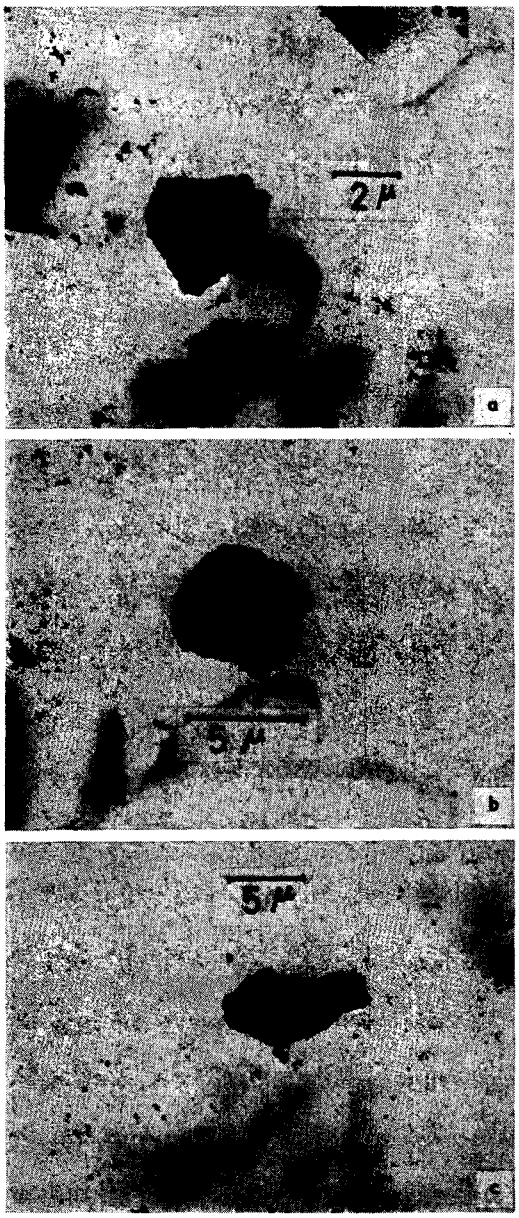

Fig. 23. (*a*), (*b*) and (*c*) Ice nuclei and tiny carbon particles on 18 February at Blue Glacier.

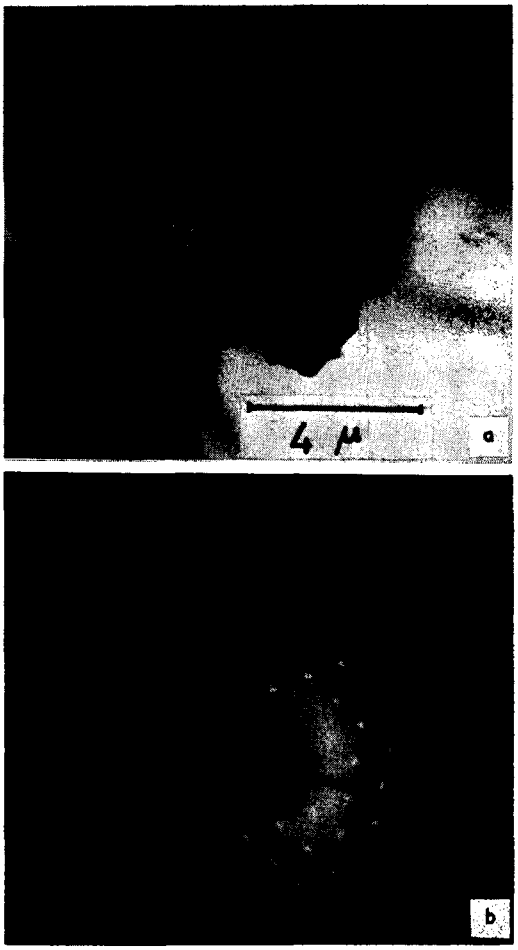

Fig. 24. (*a*) Ice nucleus (clay mineral) on 18 February at Blue Glacier and (*b*) its electron diffraction pattern (kaolin mineral).

In the case of the peak found on 18–19 February at College, the feature of size distribution was quite different from this as is seen in Fig. 14*b*.

The result of trajectory analysis described in Section 4 suggests that the large peak on 14 February at Nagoya corresponds to that on 18 February at the Blue Glacier. However, the air mass which arrived at College on 18–19 February was not over Japan on 14 February. It was over the Asian Continent about 10 days and over Japan about 8 days before the day of arrival at College.

In the case of an ice nucleus storm which was observed at Tokyo, Japan (Isono et al., 1959*a*), dense clouds of loess from the North China were observed at levels higher than 2 km. It is likely that clouds of particles which originated from the North China gave rise to a peak of ice nucleus concentration in Japan. Settling velocities of particles of 3 g/cm^3 in density are about 130 m/day for particles of 4 microns in diameter and 1.8 m/day for particles of 0.4

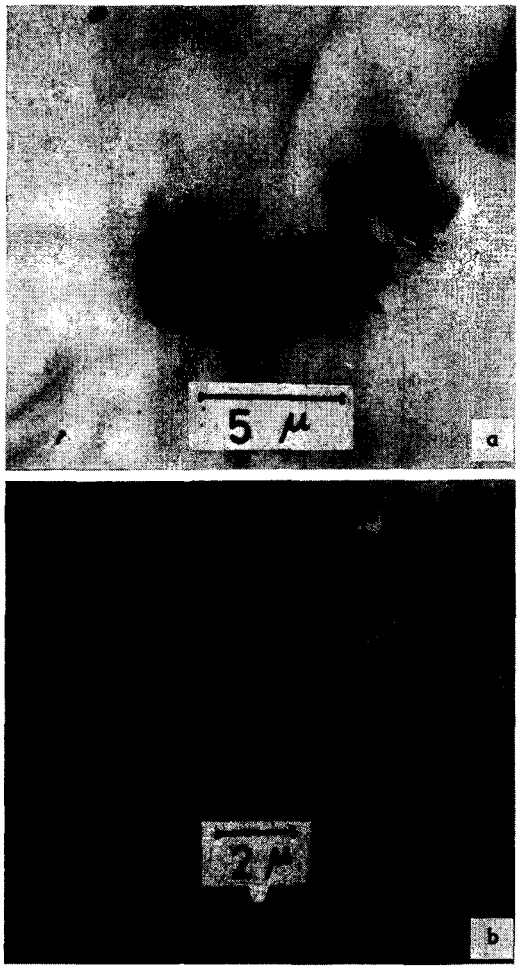

Fig. 25. (*a*) and (*b*) Ice nuclei (clay minerals) on 15 February at Blue Glacier.

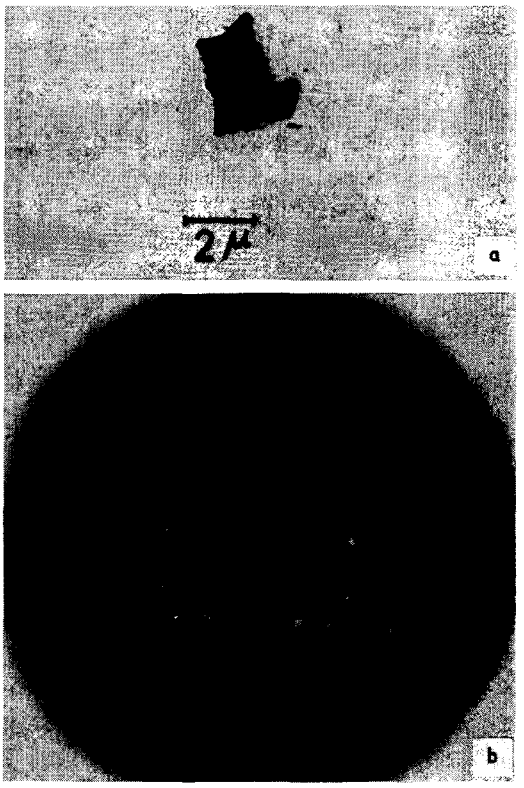

Fig. 26. (*a*) Ice nucleus (clay mineral) without any contaminant on 9 February at Mauna Loa and (*b*) its electron diffraction pattern.

micron. It is, therefore, possible that these particles were transported by air current from the North China to the west coast of the North America without significant change in their size distribution, when the travelling time from Japan to the North America was as short as three to four days as in the case of the peaks on 14 February at Nagoya and 18 February at the Blue Glacier.

As for the peak on 18–19 February at College, the travelling time of air mass from Japan to Alaska was as long as eight days, that is, more than twice that from Japan to Washington. It is suspected that particles of larger sizes were efficient as relatively rapid ice nuclei and ice crystals which formed on them precipitated out, when the air mass was kept cold and humid in passing through cyclonic region over Sakhalin and arctic region. As particles contained in the air mass are considered to have been kept at temperatures below $-10°C$ for a considerably long time before they reached College, even slow nuclei were activated in the cloud chamber of the collector used for the observation and collected by filter paper. This is an explanation why ice nuclei collected on 18 and 24 February at College were smaller in their sizes as compared with nuclei collected on the days with peaks at Nagoya and the Blue Glacier and that the peak values were, nevertheless, large. Rau & Schmidt (1961) found that ice nucleus concentration at Weissenau increased, when polar air masses flowed into Central Europe in cold season. This might be also due to a similar cause.

As for ice nuclei at Mauna Loa Observatory, the feature of the variation of their concentra-

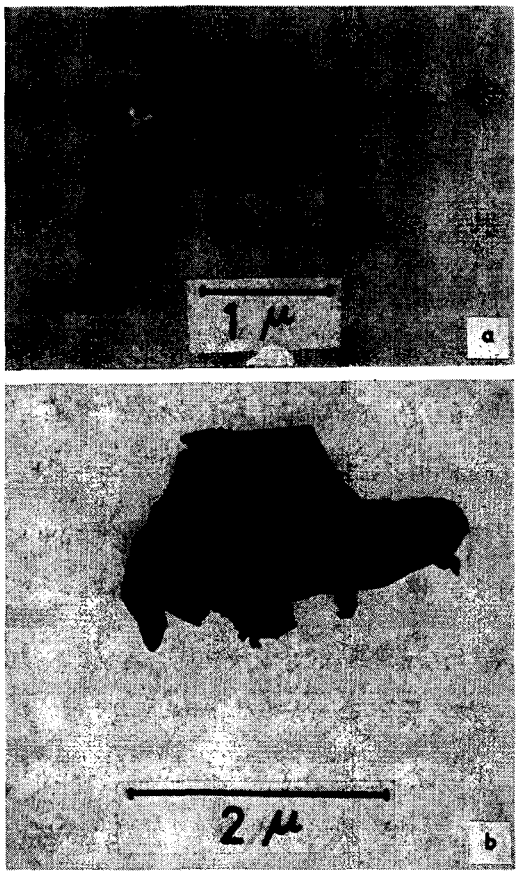

Fig. 27. (*a*) and (*b*) Ice nuclei (clay mineral) on 9 February at Mauna Loa. No other tiny particles are seen.

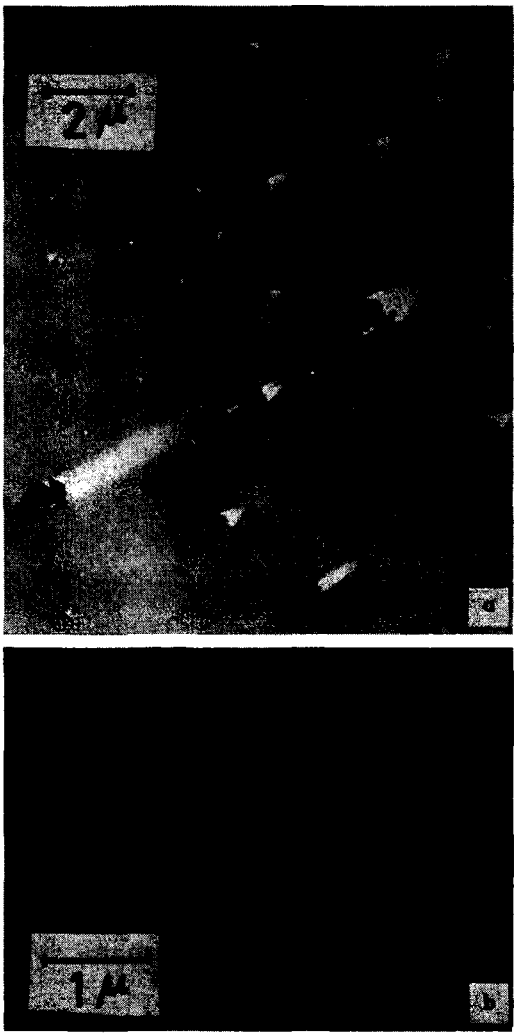

Fig. 28. (*a*) and (*b*) Ice nuclei (chrome shadowed) on 19 February. These are small and massive mineral particles.

tion and their size distribution were different from those at other three sites. Particles smaller than 0.2 micron in diameter consitute 30 to 60% of the total ice nuclei; they were smaller than those collected at other three sites. This may be primarily because their origin and travelling path in the atmosphere were different from those of nuclei which reached other three sites. This is inferred from trajectory analysis. The fact that they were not contaminated with substances of industrial origin and sea salt indicates that they had not been carried by upslope winds from the sea surface or from lower levels near Hawaiian Islands. They would have been carried by upper wind from a semi-arid region on an Asian Continent, perhaps, in the southern part of the Continent.

Summary and conclusion

Simultaneous collections of ice nuclei in the atmosphere were made with instruments of the same type at four sites in the rim of the North Pacific from the beginning of February to the first week of March, 1968. The result of the identification of their materials shows that most of ice nuclei effective at -15°C collected were mineral particles of continental origin, especially kaolin minerals. The result of the analysis of air mass trajectories and the features of variation of ice nuclei concentrations suggest that

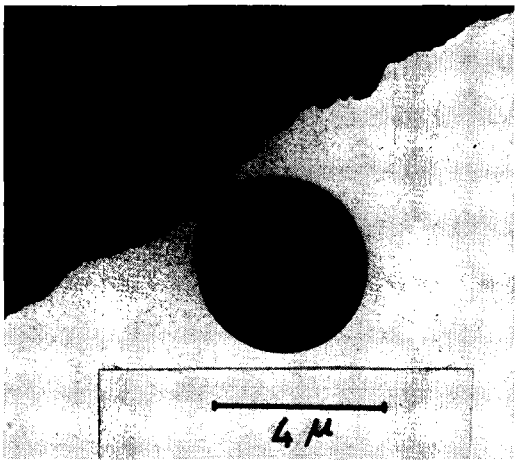

Fig. 29. Spherical ice nucleus (its material is unknown) on 13 February at Mauna Loa, presumed to be of volcanic origin.

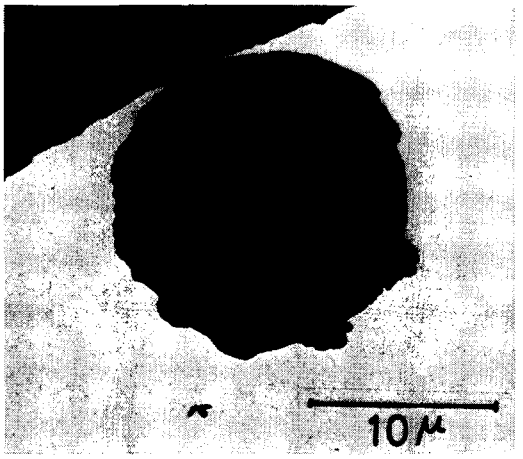

Fig. 30. Spherical ice nucleus (its material is unknown) on 9 February at Mauna Loa.

ice nuclei which reached College, Alaska and the Blue Glacier, Washington originated from the Northern China or Mongolia and those which reached Mauna Loa Observatory, Hawaii originated from a semi-arid region in the southern part of the Asian Continent. In the case of a marked peak in ice nucleus concentration at the Blue Glacier on 18 February, it can be inferred that the corresponding peak at Nagoya occurred about four days before and the size distribution of ice nuclei at both sites were similar to each other.

The result described above shows that ice nuclei are transported by air current over the Pacific Ocean. This suggests that simultaneous observations and collections of ice nuclei in a large area which are made with instruments of the same type at sites as free as possible from local sources of ice nuclei will give valuable information on their origin.

Acknowledgements

Thanks are due to Dr E. J. Workman and Dr C. M. Fullerton, University of Hawaii and Dr P. V. Hobbs, University of Washington and Dr T. Ohtake, University of Alaska for giving facilities for the measurements made at Mauna Loa Observatory, Blue Glacier Observatory and the Minitruck Station.

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КОНЦЕНТРАЦИИ И ПРИРОДА ЛЕДЯНЫХ ЯДЕР НА ГРАНИЦАХ СЕВЕРНОЙ ЧАСТИ ТИХОГО ОКЕАНА

В четырех местах на границах северной части Тихого океана: Колледж, Аляска; Блю Гласьер, Вашингтон; Мауна Лоа, Гавайи; Нагойя, Япония по одной и той же методике были произведены одновременные заборы проб с ледяными ядрами. Они собирались на бумажные фильтры для подсчета их числа и на сеточки для исследования под электронным микроскопом. В течение периода сбора с начала февраля до начала марта 1968 г. в каждом месте, за исключением Мауна Лоа, появились три заметных максимума концентрации ледяных ядер, эффективных при $-15°C$. Величины максимумов были наибольшими в Нагойя (5,3 ядер на литр), затем в Колледже (2,7 ядер на литр) и Блю Гласьер (1,3 ядер на литр). В обсерватории Мауна Лоа какого-либо заметного пика не наблюдалось. Не наблюдались также ни суточные, ни другие вариации с каким-либо другим периодом. Результаты определений вещества ледяных ядер, собранных в четырех местах, показывают, что глина и другие минеральные частицы составляют главную часть этих ядер. Результаты изучения особенностей концентрации ледяных ядер, траекторий воздушных масс и исследование самих ядер дают основание предложить, что собранные ядра возникли в засушливых и полузасушливых частях азиатского континента.