RR 245

AD 691326

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Research Report 245

MICROSPHERULES IN SNOW AND ICE-FOG CRYSTALS

Motoi Kumai

March 1969

U.S. ARMY MATERIEL COMMAND
TERRESTRIAL SCIENCES CENTER
COLD REGIONS RESEARCH & ENGINEERING LABORATORY
HANOVER, NEW HAMPSHIRE

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DA TASK 1T061102B52A02

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PREFACE

This report was prepared by Dr. Motoi Kumai, Research Physicist, of the Research Division, Cold Regions Research and Engineering Laboratory (CRREL), U.S. Army Terrestrial Sciences Center (USA TSC). It covers research on microspherules found in snow crystals and ice-fog crystals in the cold regions of the Northern Hemisphere and is published under DA Task 1T061102B52A02, *Research in Earth Physics - Cold Regions and Related Environments.*

The author wishes to thank Dr. C.C. Langway, Jr., for his discussions on microspherules.

USA TSC is a research activity of the Army Materiel Command.

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ABSTRACT

Spherules found in snow crystals, ice-fog crystals, fallout particles, and fly ash were studied with an electron microscope using the electron diffraction method. The central part of the residues of 1004 specimens of natural snow crystals from Greenland, the United States, and Japan were examined; 14 spherules 0.1 to 1.5μ in radius were found among them. The residues of 658 artificial ice-fog crystals formed from water vapor in flue gases of coal-burning electric power plants at Fairbanks, Alaska, were also examined; nine spherules were found. Spherules similar to those found in ice-fog residues were found in furnace-produced fly ash fallout at Fairbanks, Alaska. Electron and optical microscope examination of spherules found in Greenland snow reveals a size distribution of the form $dN/d(\log r) = C r^{-\beta}$ where $\beta \approx 3$. The properties of spherules and the mean mass of snow crystals from Greenland are described. The electron microscope study indicated that less than 0.7% of the 1004 snow crystals contained spherules of possible extraterrestrial origin, and that snow crystals are formed mainly on clay mineral particles by heterogeneous nucleation.

MICROSPHERULES IN SNOW AND ICE-FOG CRYSTALS

by

Motoi Kumai

INTRODUCTION

Spherules found in polar ice are thought by many workers (Wright *et al.*, 1963; Langway and Marvin, 1964) to be extraterrestrial materials. In comparing volcanic ashes with sedimentary salt samples, Mutch (1964) concluded that magnetite spherules in the Paleozoic salts were extraterrestrial in origin. Other workers (Fredriksson and Martin, 1963) have found spherules in volcanic rocks which they maintain are volcanic and have suggested that some spherules in other collections may also be volcanic.

The chemical compositions of spherules from the Greenland ice sheet, the Antarctic ice sheet, and other polar and temperate climate locations have been studied by Wright *et al.* (1963) and Hodge *et al.* (1964) with an electron-beam microanalyzer.

Experiments on cloud modification by seeding in Greenland were made by Jiusto and Rogers (1960), and experiments on snow crystal nuclei at Site 2, Greenland, have been described by Kumai and Francis (1962). Spherules larger than 15μ radius were studied by Thiel and Schmidt (1961) and spherules larger than 3μ radius by Wright *et al.* (1963) with an optical microscope and an electron-beam microanalyzer.

This paper describes electron microscope observations of spherules smaller than 5μ in radius (microspherules) collected from Greenland, Alaska, the United States, and Japan, and the shape and size distribution of snow crystals in Greenland.

SNOW CRYSTALS IN GREENLAND

Observations of the shapes of snow crystals from photomicrographs and temperature measurements of snow-producing clouds by radiosonde and a tethered blimp were made during 8 weeks in summer 1960 at an inland Greenland location, Site 2. This site is located at an altitude of 2000 m, about 320 km east of Thule. Precipitation at Site 2 is generally in the form of snow crystals and amounts to about 40 cm water equivalent per year. Almost all shapes of snow crystals were observed, and 700 photomicrographs were taken. The major forms of snow crystals include needles (simple and combination), capped columns, plates, stellars, dendrites, spatial dendrites, irregular shapes, and graupel. From these observations, a relationship between the temperature of the snow-forming clouds and the shapes of falling crystals was obtained. The results agree well with Nakaya's (1954) diagram and the findings of Hallet and Mason (1958).

The size of snow crystals is not uniform but varies through periods of growth under slightly different conditions of supersaturation and temperature in the cloud. The snow crystals observed at Site 2, Greenland, were precipitated mainly from stratus clouds. The size distribution of snow crystals from Greenland(the diameter of plate crystals and stellar crystals, and the largest dimension of needle and column-shaped crystals) is plotted in Figure 1. The sizes of diamond-dust crystals formed at the ground surface and wind-blown crystal fragments are not included in Figure 1.

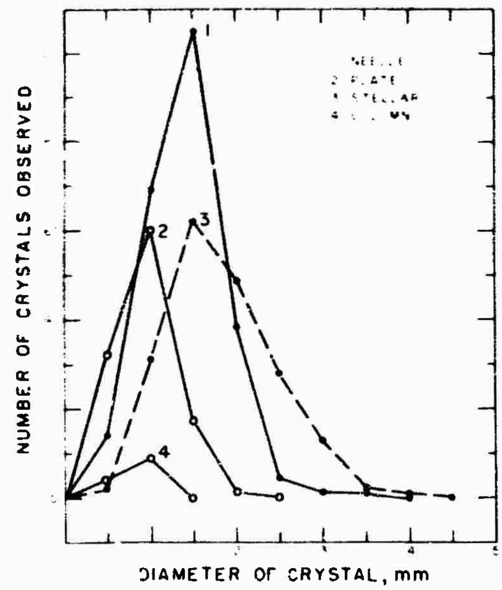

Figure 1. Size distribution of snow crystals in summer at Site 2, Greenland. The size is diameter of plate and stellar crystals and the largest dimension of needle and column-shaped crystals.

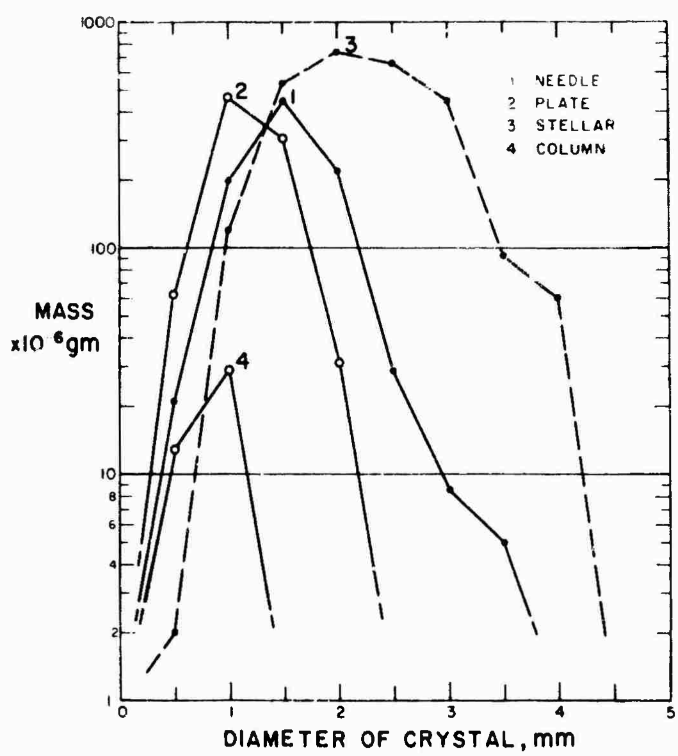

Figure 2. Mass distribution of snow crystals in summer at Site 2, Greenland.

Table I. Size and mean mass of snow crystals.

	Hokkaido, Japan	Site 2, Greenland	
Crystal shape	Most frequently occurring diameter (mm)	Most frequently occurring diameter (mm)	Mean mass (g)
Needle	1.75	1.5	4.1×10^{-6}
Plate	1.0	1.0	7.9×10^{-6}
Stellar	1.0, 2.5	1.5	14.1×10^{-6}
Column	0.5	1.0	3.15×10^{-6}

The results are quite similar to those at Mount Takachi, Hokkaido, Japan (Nakaya, 1954) with respect to the size range and the most frequently occurring size for each type (Table I). This indicates that conditions of snow crystal growth for each type of crystal are similar in different geographical locations.

The size and mass of individual snow crystals were calculated from photomicrographs of the crystals and their melt droplets with an experimental error of less than 2%. The mass distributions of snow crystals, needles, plates, stellars, and columns are shown in Figure 2. From the frequency of each snow crystal shape and its mean mass, the mean mass for all snow crystals at Site 2, Greenland, is calculated to be 8.22×10^{-6} g. One gram of snow therefore contains approximately 1.22×10^{5} crystals.

MICROSPHERULES

The size distribution in snow and ice of spherules larger than 15μ in diameter has been studied by Thiel and Schmidt (1961) (spherules from Antarctica) and by Wright *et al.* (1963), Hodge *et al.* (1964) and Langway (1965) (spherules from Greenland). Wright *et al.* (1963) and Hodge *et al.* (1964) have also investigated with an electron-beam microanalyzer the chemical composition of spherules collected from the Greenland ice sheet, the Antarctic ice sheet, and other localities. These spherules were collected carefully, avoiding contamination, from melt water samples of snow and ice by use of a microfilter, and their radii as measured with an optical microscope ranged from 2 to 50μ. In a total of 55 spherules from Greenland there were 24 iron-rich spherules, 5 silicon-rich spherules, and 26 other spherules. Of the 55 spherules from the South Pole station, 22 were iron-rich and 7 were silicon-rich. Although most of these particles were considered to be of extraterrestrial origin, from an analysis of the shape and chemical composition of weld spatter (Langway and Marvin, 1964) and volcanic particles (Wright *et al.*, 1963; Hodge *et al.*, 1964) it was considered likely that some were of volcanic or artificial origin.

EXPERIMENTAL METHOD

Three methods may be used to detect microspherules for electron microscope examination: 1) the volume of ice melt water can be reduced by evaporation and the droplet then placed on a collodion-filmed grid and evaporated; 2) a piece of ice can be placed on a collodion-filmed grid and sublimed below the freezing point; 3) a single snow crystal can be collected directly on a collodion-filmed grid and sublimed below the freezing point.

Solid particles in the ice become coagulated when methods 1 and 2 are used. Method 3 was found to be the best method of collection and was used in this research because contamination of the sample is thereby kept to a minimum and loss of nuclei (spherules) during the preparation stage is prevented.

The spherule specimens were shadowed with chromium vapor in a vacuum chamber. An oval shadow is cast by a spherical particle (Fig. 3a) and is the means by which its form can be identified. Electron photomicrographs were taken, with a magnification of × 10,000, and electron diffraction patterns obtained.

MICROSPHERULES IN SNOW AND ICE-FOG CRYSTALS

Microspherules varying in radius from 0.1 to 1.5μ were found in snow crystals. Electron microscope observations made since 1948 by Kumai and O'Brien (1965), Kuroiwa (1957), Ogiwara and Okita (1952), Yamamoto and Ohtake (1955), and other researchers are shown in Table II. In a total of 1004 snow crystals collected at Site 2, Greenland; Michigan; and Hokkaido, Japan, 14 microspherules were found (1.4%). Minute spherules such as carbon particles from 0.01 to 0.03μ in radius are not described in this paper.

Table II. Electron microscope observations of microspherules found in snow crystals, ice-fog crystals, and fog droplets.

Year	Location	No. of microspherules	%	Observer
		Snow crystals		
1948-1950	Hokkaido, Japan	0 out of 103	0	Kumai
1952-1953	Honshu, Japan	0 out of 20	0	Isono
1956	Hokkaido, Japan	2 out of 202	1	Kumai
1955-1958	Honshu, Japan	0 out of 52	0	Isono
1959	Houghton, Michigan	5 out of 271	1.8	Kumai
1960	Site 2, Greenland	7 out of 356	2.0	Kumai
	Total	14 out of 1004	1.4	
		Ice-fog crystals		
1962	Fairbanks, Alaska	3 out of 340	0.9	Kumai
1963	Fairbanks, Alaska	1 out of 202	0.5	Kumai
1964	Eielson AFB, Alaska	5 out of 116	4.5	Kumai
	Total	9 out of 658	1.4	
		Fog droplets		
1944-1945	Hokkaido, Japan	0 out of 225	0	Kuroiwa
1951	Honshu, Japan	0 out of 75	0	Ogiwara & Okita
1952-1955	Honshu, Japan	0 out of 217	0	Yamamoto & Ohtake
1952-1955	Honshu, Japan	0 out of 123 (mist)	0	Yamamoto & Ohtake
1960	Thule, Greenland	0 out of 40	0	Kumai
1962	Fairbanks, Alaska	0 out of 21	0	Kumai
1964	Barrow, Alaska	0 out of 78	0	Kumai
	Total	0 out of 809	0	

Site 2, Greenland

Aerosol concentration at Site 2 is one of the lowest on the earth. The concentration of Aitken nuclei is less than 50 particles cm^{-3} in the summer. Of the 356 snow crystals collected at Site 2 in the summer of 1960 (Kumai and Francis, 1962), 85% had nuclei of clay minerals, 2% contained spherules, and 9% contained other substances; 4% had no nuclei. The spherules (Fig. 3a and 3b) ranged in radius from 0.1 to 1.3μ (Table II) and were found in the centers of hexagonal snow crystals. The spherules are generally polycrystalline, and they produce electron diffraction patterns for a suitable mass of specimen. The suitable mass depends on the intensity of the electron beam and the accelerating voltage of the electron microscope. The spherules 0.1μ in radius were too small to produce an electron diffraction pattern; spherules of radius 1μ and larger showed diffraction patterns. Possibly all spherules from Site 2, Greenland, are of extraterrestrial origin. They have no minute particle contamination, such as carbon particles, on their surfaces.

Houghton, Michigan

Snow crystals were collected in the winter of 1959 at Houghton, Michigan (Kumai, 1961), which is situated on a small peninsula along the southern shore of Lake Superior. In the 271 snow crystals examined, 5 spherule nuclei were found. Silicate mineral nuclei were found in 87% of the crystals, spherules in 2%, hygroscopic nuclei in 1%, solid combustion products in 2%, and other substances in 7%; 1% of the crystals had no nuclei. The spherules varied in radius from 0.5 to 1.2μ (Fig. 3c). Spherules of radius 1μ and larger gave electron diffraction patterns but the indexing of the diffraction patterns was not sufficient for positive identification. All spherules had minute particle contamination on their surfaces, as shown in Figure 3c, and appeared to be different from those found in snow crystals at Site 2, Greenland.

Hokkaido and Honshu, Japan

Snow crystals were collected in mid-winter on the slopes of Mount Taisetsu, Hokkaido, at an elevation of 1050 m (Kumai, 1951), at approximately 1000 m near Mount Asama, Honshu (Isono, 1955), and at Mount Tokachi, Hokkaido, at an elevation of 1030 m (Kumai, 1957). A total of 377 crystals were collected from 1948 to 1958 (Table II); 62% had silicate mineral nuclei, 0.6% spherules, 16% hygroscopic nuclei, 8% combustion by-products, and 13.4% other substances. Only two spherules were found, with radii of 3μ and 5μ, and they were similar in shape to the spherules found in snow crystals collected at Houghton, Michigan (Fig 3c). They may be fly ash particles.

Fairbanks, Alaska

During winter, the air around Fairbanks is contaminated with flue gases derived from coal-burning power and heating plants and other industrial sources, producing what is known as fly ash. The concentration of Aitken nuclei in Fairbanks is about 3×10^4 particles cm^{-3} in the winter. Spherules that were collected in fallout during the winter of 1962 at Fairbanks, Alaska (Kumai, 1964), are shown in Figure 3d. These spherules of 0.05μ to 0.1μ radius are coagulated and are covered or associated with many carbon particles of about 300 Å radius, unlike the spherules found in Greenland. Spherules, also associated with many carbon particles, were found in furnace ash (Fig. 3e). During severe cold, dense ice fog forms at around -40C from water vapor in these flue gases. Nine spherules with radii varying from 1μ to 5μ were found in the residues of 658 ice-fog crystals from Alaska (Fig. 3f). They were similar in shape and diffraction pattern to the spherules found in the furnace ash and are believed to be fly ash.

The residues of fog and mist droplets from Thule, Greenland; Barrow and Fairbanks, Alaska; and Honshu and Hokkaido, Japan, have been examined with the electron microscope by many workers; however, no spherules have been found in these droplets (Table II).

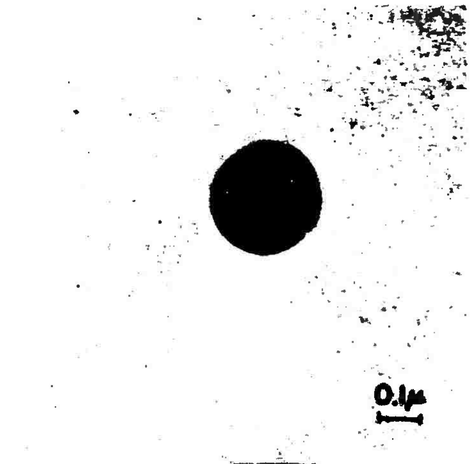

Figure 3a. Spherule (possibly extraterrestrial) in snow crystal at Site 2, Greenland.

Figure 3b. Spherule (possibly extraterrestrial) in snow crystal at Site 2, Greenland.

Figure 3c. Spherule (possibly fly ash) in snow crystal at Houghton, Michigan.

Figure 3d. Spherules (possibly fly ash) in fallout particles at Fairbanks, Alaska.

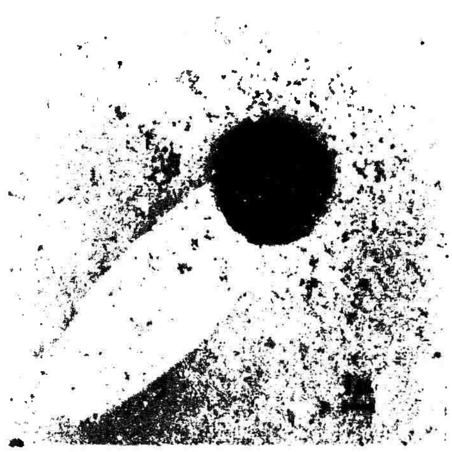

Figure 3e. Spherule found in ashes from coal furnace at Fairbanks, Alaska.

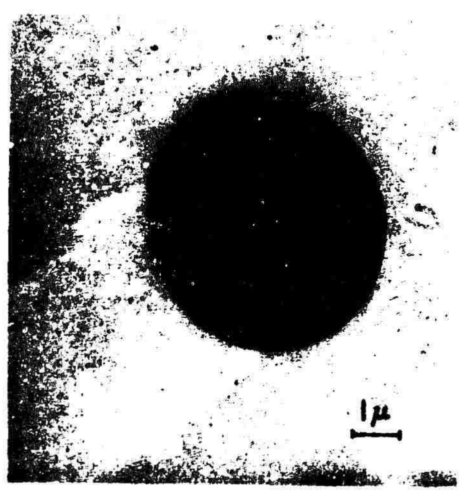

Figure 3f. Spherule (possibly fly ash) in ice fog crystal at Fairbanks, Alaska.

Concentration and Radii of Spherules

Most of the spherules from Greenland are believed to be extraterrestrial material. The concentration and radii of the spherules have been calculated from observations with both electron and optical microscopes. Spherules smaller than 2μ in radius were observed by electron microscopy in the residues of 1.96% of the snow crystals examined (Table II). Since the mean mass of a snow crystal was calculated to be 8.22×10^{-6} g, 1 g of snow contains 1.22×10^{5} crystals and the number of spherules (from 0.1 to 1.5μ in radius) in 1 g of snow is calculated to be 2.4×10^{3}. From observations of melted glacier samples from Greenland with an optical microscope (Langway, 1963, 1965) the number of larger spherules (2 to 120μ in radius) in 1 g of snow is calculated to be 4.4×10^{-2}.

From these combined measurements (2021 spherules) the concentration and size of spherules in glacier material in the range $10^{-1}\mu$ to $10^{3}\mu$ radius can be expressed by a straight line (Fig. 4), using the form

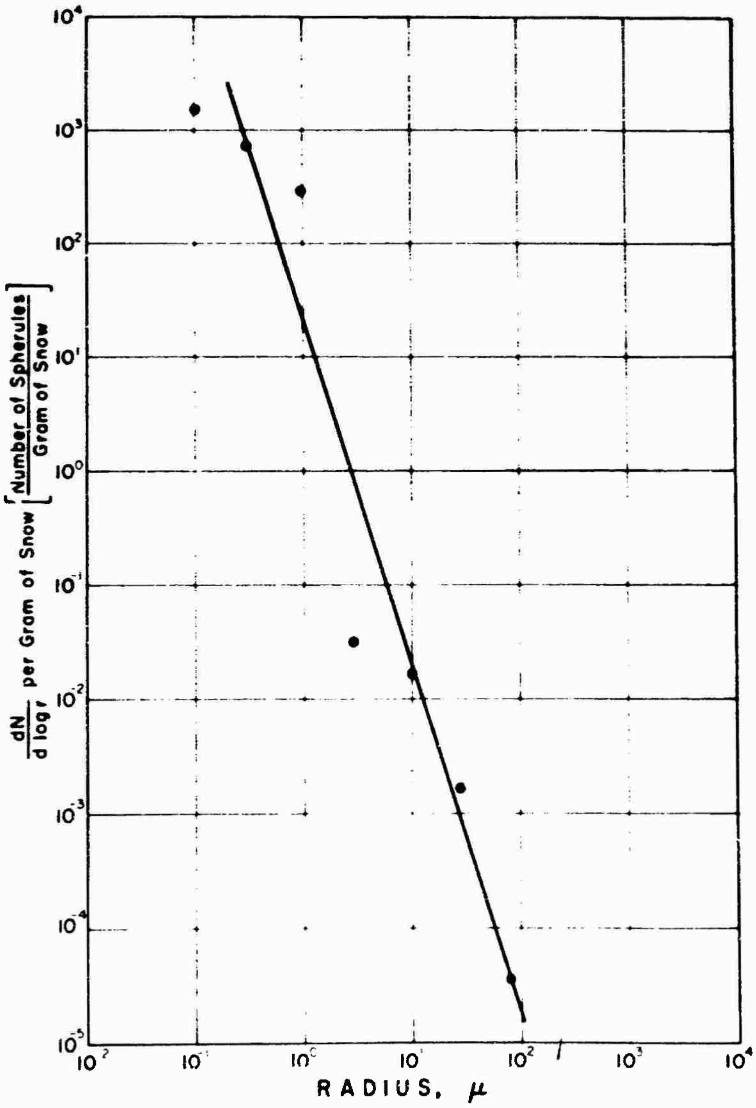

Figure 4. Size distribution of spherules per gram of snow at Site 2, Greenland.

$$\frac{dN}{d(\log r)} = C\, r^{-\beta}$$

or

$$\frac{dN}{dr} = \frac{C}{2.30} \cdot r^{-(\beta+1)}$$

where N is the total concentration of spherules of radius smaller than r and C is constant. This form is similar to the one found in aerosol studies by Junge (1963). The exponent β is 3 (see Fig. 4), which corresponds to $\beta = 2$ as published by Soberman and Hemenway (1965) for a meteoric dust size distribution with radii 10^{-2} to 1μ obtained by rocket sampling at an altitude of 75 to 98 km.

There is a gap of the order of 10^4 in distribution values between observations with the electron microscope and the optical microscope. It is possible that some of the spherules smaller than 1μ were lost during the melting and filtering of the bulk snow samples.

DISCUSSION AND CONCLUSIONS

Bowen (1953) proposed the theory that precipitation may be influenced by meteor showers. Observing the residue in the center of a snow crystal with the electron microscope provides a means for testing Bowen's hypothesis. Since 1948, 1004 snow crystals collected at various locations in Greenland, the United States, and Japan have been examined. Only 14 spherules of radius 0.1 to 1.5μ were found. Seven of these are probably terrestrial materials so that less than 0.7% of the snow crystals observed contained possible extraterrestrial material. The center nucleus of about 85% of all snow crystals from Site 2, Michigan, and Honshu consisted of clay minerals. Our own observations in Greenland, the United States, and Japan, and the studies of Schaefer (1955), Mason and Maybank (1958), Kline (1960), Battan and Riley (1960), Isono (1955), and Byers (1965) show that the ice-forming nuclei are mainly of terrestrial origin.

The spherules found in snow crystals where aerosols are scarce, at Site 2, Greenland, are believed to be extraterrestrial materials. Spherules found in ice-fog crystals which were formed with water vapor from flue gases are considered to be fly ash. Some similar spherules found in snow crystals from Michigan and Hokkaido are considered to be a material similar to fly ash.

Concentration and radii of spherules found in Greenland in the range $10^{-1}\mu$ to $10^{3}\mu$ may be expressed in the form $dN/d(\log r) = C\, r^{-\beta}$, where the exponent $\beta \approx 3$.

Precipitation at Site 2, Greenland, is generally in the form of snow crystals. The major forms of snow crystals include dendrites, plates, spatial dendrites, needles, columns, irregular crystals, and graupel. The most frequently occurring diameters were 1.0 mm for plates and columns and 1.5 mm for dendrites (stellar) and needles. From the frequency of occurrence of each snow crystal shape and its mean mass, the total mean mass of snow crystals at Site 2, Greenland, is calculated to be 8.22×10^{-6} g. One gram of snow therefore consists of 1.22×10^{5} crystals. Diamond-dust crystals which were formed near the ground surface and wind-blown crystal fragments are not included in this calculation.

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Unclassified

Security Classification

DOCUMENT CONTROL DATA - R & D

(Security classification of title, body of abstract and indexing annotation must be entered when the overall report is classified)

1. ORIGINATING ACTIVITY (Corporate author) Cold Regions Research & Engineering Laboratory U.S. Army Terrestrial Sciences Center Hanover, New Hampshire	2a. REPORT SECURITY CLASSIFICATION Unclassified 2b. GROUP

3. REPORT TITLE

MICROSPHERULES IN SNOW AND ICE-FOG CRYSTALS

4. DESCRIPTIVE NOTES (Type of report and inclusive dates)

Research Report

5. AUTHOR(S) (First name, middle initial, last name)

Motoi Kumai

6. REPORT DATE	7a. TOTAL NO. OF PAGES	7b. NO. OF REFS
March 1969	14	29

8a. CONTRACT OR GRANT NO. b. PROJECT NO. c. DA Task 1T061102B52A02 d.	9a. ORIGINATOR'S REPORT NUMBER(S) Research Report 245 9b. OTHER REPORT NO(S) (Any other numbers that may be assigned this report)

10. DISTRIBUTION STATEMENT

This document has been approved for public release and sale; its distribution is unlimited.

11. SUPPLEMENTARY NOTES	12. SPONSORING MILITARY ACTIVITY Cold Regions Research & Engineering Laboratory U.S. Army Terrestrial Sciences Center Hanover, New Hampshire

13. ABSTRACT

Spherules found in snow crystals, ice-fog crystals, fallout particles, and fly ash were studied with an electron microscope using the electron diffraction method. The central part of the residues of 1004 specimens of natural snow crystals from Greenland, the United States, and Japan were examined; 14 spherules 0.1 to 1.5μ in radius were found among them. The residues of 658 artificial ice-fog crystals formed from water vapor in flue gases of coal-burning electric power plants at Fairbanks, Alaska, were also examined; nine spherules were found. Spherules similar to those found in ice-fog residues were found in furnace-produced fly ash fallout at Fairbanks, Alaska. Electron and optical microscope examination of spherules found in Greenland snow reveals a size distribution of the form $dN/d(\log r) = Cr^{-\beta}$ where $\beta \approx 3$. The properties of spherules and the mean mass of snow crystals from Greenland are described. The electron microscope study indicated that less than 0.7% of the 1004 snow crystals contained spherules of possible extraterrestrial origin, and that snow crystals are formed mainly on clay mineral particles by heterogeneous nucleation.

14. Key Words:

Snow crystals	Ice fog
Ice crystals	Electron microscopy
Microspherules	Electron diffraction

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