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Irregular Snow Crystals: Structural Features as Revealed by Low Temperature Scanning Electron Microscopy

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Summary: For nearly 50 years, investigators using light microscopy have vaguely alluded to a unique type of snow crystal that has become known as an irregular snow crystal. However, the limited resolution and depth-of-field of the light microscope has prevented investigators from characterizing these crystals. In this study, a field-emission scanning electron microscope, equipped with a cold stage, was used to document the structural features, physical associations, and atmospheric metamorphosis of irregular snow crystals. The crystals appear as irregular hexagons, measuring 60 to 90 mm across, when viewed from the a-axis. Their length (c-axis) rarely exceeds the diameter. The irregular crystals are occasionally found as secondary particles on other larger forms of snow crystals; however, they most frequently occur in aggregates consisting of more than 100 irregular crystals. In the aggregates, the irregular crystals have their axes oriented parallel to one another and, collectively, tend to form columnar structures. Occasionally, these columnar structures exhibit rounded faces along one side, suggesting atmospheric metamorphoses during formation and descent. In extreme cases of metamorphoses, the aggregates would be difficult to distinguish from graupel. Frost, consisting of irregular crystals, has also been encountered, suggesting that atmospheric conditions that favor their growth can also occur terrestrially.

Key words: field-emission scanning electron microscopy, low-temperature scanning electron microscopy, snow crystals, irregular crystals, snowflakes, rime, graupel

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Introduction

Snow crystals have been studied with the light microscope for more than 100 years. However, because of their small size, delicate structure, and topography, this instrument has failed to elucidate many of the structural details of the crystals. In addition, snow is difficult to transport, store, image, and photograph without subjecting samples to structural changes resulting from sublimation, melting or recrystallization. Examination of snow crystals with a scanning electron microscope (SEM) would solve problems associated with resolution, topography, and their delicate nature. However, for obvious reasons, frozen samples cannot be imaged at ambient temperatures in a conventional SEM.

In 1970, Echlin *et al*. (1970) solved this problem for biologists by describing a cold stage that could be interfaced with an SEM and operated at temperatures below –130° C. At these temperatures, the vapor pressure of water is not significant and sublimation does not occur at a detectable rate. Furthermore, recrystallization of pure water-ice does not occur (Beckett and Read 1986) and frozen, fully hydrated samples remain stable for several hours while being observed (Wergin and Erbe 1991).

Recently, this procedure, which has become known as low-temperature (LT) SEM, was used to compare frozen, fully hydrated, fractured membranes of yeast in the SEM with the platinum/carbon (Pt/C) replicas of the identical cells in the transmission electron microscope (TEM) (Wergin and Erbe 1990). Results indicated that macromolecular particles, as small as 10 nm in diameter, retained their structure, were stable, and could be imaged and photographed with LTSEM. The images, which were obtained in the LTSEM, were comparable with those of the same cells in the Pt/C replicas, which were photographed in a TEM. Encouraged by these results, identical snow crystals were imaged and photographed with a light microscope, imaged and photographed in the

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LTSEM, and then rephotographed with a light microscope. The results further demonstrated that observation in the LTSEM did not alter the structure of the snow crystals (Wergin *et al.* 1998). In addition, techniques were developed to collect, ship, and store fresh and metamorphosed snow crystals for subsequent imaging with an LTSEM. These techniques have been successfully used to record detailed images of thousands of snow crystals that were collected from numerous states including Alaska, Colorado, Montana, Utah, Wyoming, Wisconsin, Maryland, and West Virginia. In previous studies, we have described the detailed structure of snow and ice grains including columns, needles, plates, stellar dendrites, depth hoar, and glacial ice that were collected from several states (Rango *et al.* 1996a, b, 2000; Wergin and Erbe 1994a, b; Wergin *et al.* 1995, 1996a, b, 1998, 1999). The current study uses LTSEM to illustrate the features and distinctive characteristics of the elusive group of crystals recognized by The International Commission on Snow and Ice (Colbeck *et al.* 1990) and simply referred to as "irregular crystals."

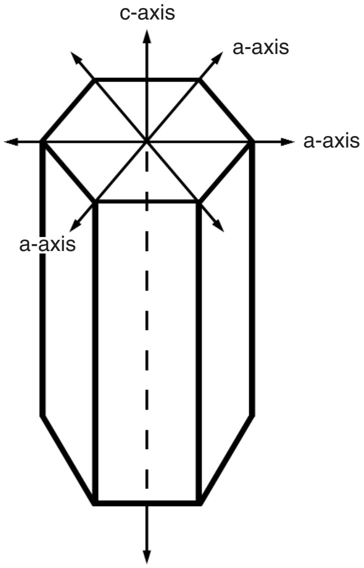


FIG. 1 Diagram illustrating the crystallographic axes of snow crystal growth. Environmental conditions that favor growth along the a-axes result in flat hexagonal plates and stellar dendrites, whereas conditions that favor c-axis growth produce elongated columns and needles. Neither type of growth predominates in the formation of irregular crystals.

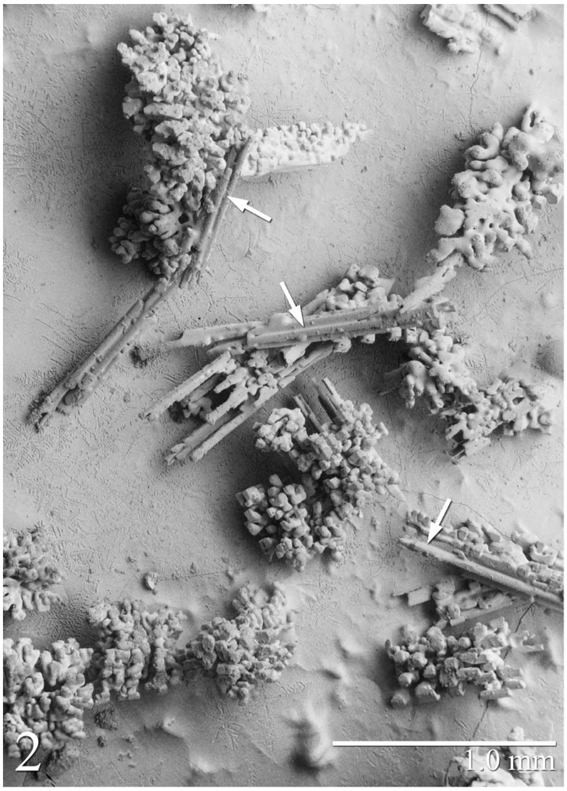
Materials and Methods

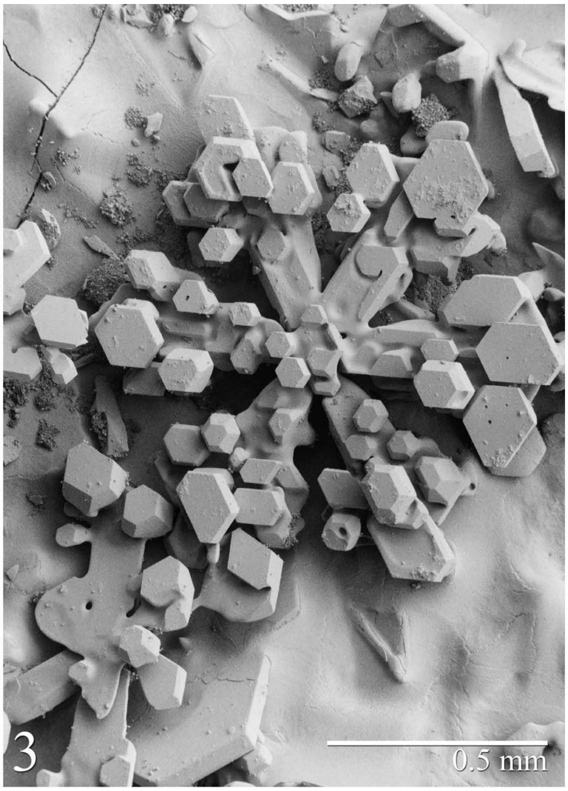
Collection Procedure

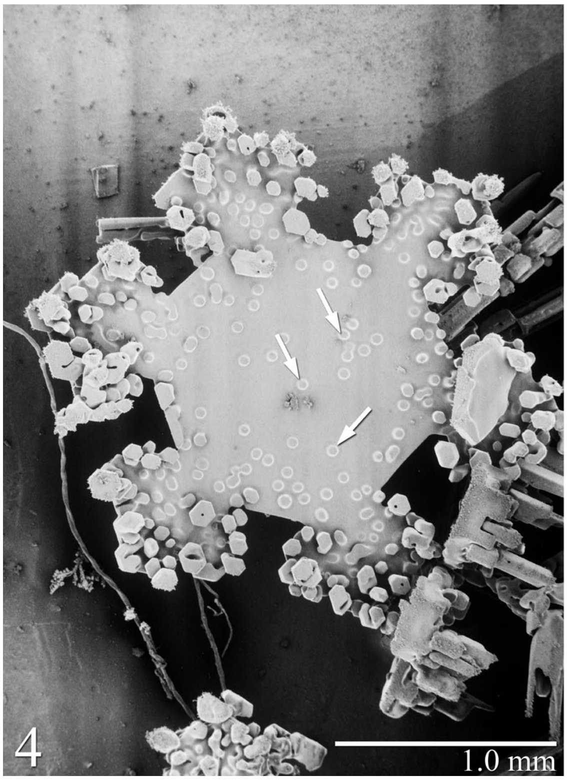
Data illustrated in this study resulted from six different snow collections during 1993–1999 from sites near the following locations: Beltsville, Maryland; Bearden Mt., West Virginia and Greenwood, Wisconsin. The samples, which were obtained when the air temperatures ranged from –5°C to 0°C, consisted of freshly fallen snowflakes. To collect samples, a thin layer of liquid Tissue-Tek, a commonly used cryoadhesive for biological samples, was spread on a flat copper plate (15 × 27 mm). The Tissue-Tek and the plates were precooled to ambient outdoor temperatures (below freezing) before use. Newly fallen snowflakes were either permitted to settle on the surface of the Tissue-Tek or were lightly dislodged from the snow surface and allowed to fall onto the surface of the cryoadhesive. Next, the plate was either rapidly plunged into a Styrofoam vessel containing liquid nitrogen (LN_2) or placed on a brass block that had been precooled with LN_2 to –196°C. This process, which solidified the Tissue-Tek, resulted in firmly attaching the snow crystals to the plate. The frozen plates were inserted diagonally into prefabricated 20 cm segments of square, brass channel tubing, containing an end cap, and lowered into dry shipping dewars that had been previously cooled with LN_2. The dewars containing the samples were conveyed from the collection sites and then either transported by van (from West Virginia) or shipped by air (Wisconsin) to the laboratory in Beltsville, Maryland. Upon reaching the laboratory, the samples were transferred under LN_2 to a LN_2 storage dewar where they remained at –196°C for as long as 9 months before being further prepared for observation with LTSEM.

Preparation for Low-Temperature Scanning Electron Microscopy Examination

To prepare the samples for LTSEM observation, a numbered segment of brass channel tubing was extracted from the storage dewar and placed in a styrofoam work chamber that was filled with LN_2. A plate was removed from the channel tubing and inserted into a modified Oxford specimen holder. The holder, which contained the plate, was transferred to the slush chamber of an Oxford CT 1500 HF Cryotrans system that had been filled with LN_2 (Oxford Instruments, Enysham, England). Next, the holder was attached to the transfer rod of the Oxford cryosystem, moved under vacuum into the prechamber for etching and/or sputter coating with Pt or Au/Pd, and then inserted into a Hitachi S-4100 field emission SEM equipped with a cold stage that was maintained at –185°C (Hitachi High-Technologies Corp., Tokyo, Japan). Accelerating voltages of 500 V to 10 kV were used to observe samples. However, 2 kV was most commonly used because it minimized charging and provided adequate resolution. The samples were imaged for as long as 2 h without observing any changes in the morphologic features or the coating integrity of the snow crystals. Selected images were recorded onto Polaroid Type 55 P/N film (Polaroid, Cambridge, Mass., USA).







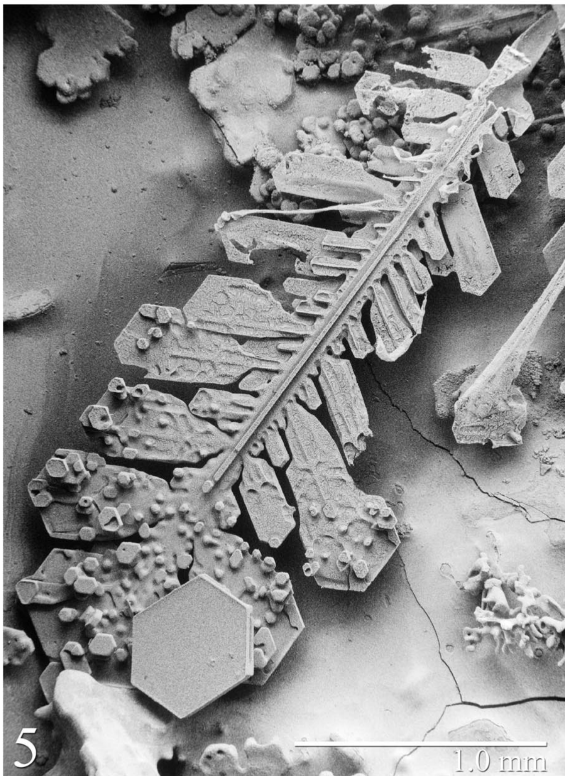


FIG. 2 Several snowflakes consisting of aggregates that are composed of numerous irregular crystals. In many cases the aggregates of irregular crystals are associated with needles (arrows). FIG. 3 Irregular crystals attached to the surface of a stellar dendrite. In general, these irregular crystals vary from 0.1 to 0.2 mm across and consist of short, nonsymmetric hexagonal columns. FIG. 4 Hexagonal plate with plate-like extensions that have numerous irregular crystals on their surfaces. Small spherical particles (arrows), possibly rime, can be seen on the surface of the hexagonal plate. FIG. 5 Dendritic arm from a stellar dendrite containing irregular crystals along the outer most edges and small droplets, which are believed to be frozen cloud droplets (rime), more central toward the median axis.

Results

General Morphology and Characteristics

Aggregates and secondary crystals: Snow crystal growth by vapor deposition generally occurs along one of two planes: the basal plane along one of three a-axes or the perpendicular plane or c-axis (Fig. 1). Basal plane growth results in flat hexagonal plates or stellar dendrites, whereas growth along the c-axis results in elongated columns or needles. In particles believed to correspond to the class "irregular crystals," growth appears to occur along both planes with neither type predominating.

The irregular crystals generally occur in aggregates, containing numerous distinct crystals (Fig. 2), but also occur as solitary crystals that are attached to the surfaces of other forms of snow crystals such as stellar dendrites (Fig. 3) and plates (Fig. 4). When found on other forms of snow crystals, they frequently occur along with small spherical droplets thought to represent rime (Figs. 4–6). The spherical shape of the droplets easily distinguishes these particles from the larger faceted irregular crystals, which generally consist of short nonsymmetric hexagonal (a-axis view) columns (c-axis view).

When both the spherical droplets and the irregular crystals occur as secondary particles on other types of snow crystals, the crystals are generally found on the peripheral edges, whereas the droplets are more median. For example, on the plate with extensions illustrated in Figure 4, the irregular crystals are found predominately on the extensions, whereas the spherical droplets occur on the primary central hexagonal plate. Similarly, on the dendritic arms illustrated in Figures 5 and 6, the crystals are found along the outer edges, whereas the droplets are more common along the median axis. In fact, the sizes of the irregular crystals increase with distance from the median axis of the primary crystal so that the largest ones are found on the most peripheral regions.

An aggregate, consisting of hundreds of irregular crystals, frequently results in a single snowflake that has a "columnar" shape (Fig. 7). Within the column, the c-axes of the irregular crystals are parallel to one another, but each individual crystal is offset from its neighbor. As a result, the files of offset crystals do not form long continuous needles or true columns.

When the columnar aggregates are viewed from their a-axes, 10 to 40 irregular crystals may form the diameters, which frequently measure 0.2 to 0.4 mm (Fig.8). When the aggregates are viewed along their c-axes, groups of 25 to 75 irregular crystals, oriented in the same plane, collectively form the lengths of the columns, which may be 1 to 2 mm long (Fig. 9).

Characteristics of the irregular crystals: An individual irregular crystal is typically about 60 to 90 μm across, but may range from 50 to 200 μm (Figs. 10 to 12). Views of their a-axes, reveal asymmetrical hexagonal faces (Fig. 11, arrows), occasionally with slight ridges around the perimeter (Fig. 12). Each crystal has the shape of a short column that is only 50 to 200 μm in length along the c-axis, which seldom exceeds the diameter of the a-axis. More often than not, the length of the crystal along the c-axis is only one-half its diameter along the a-axis. The c-axes faces occasionally contain openings or holes (Fig. 10) but are generally uninterrupted, resulting in the appearance of solid crystals (Figs. 11, 12).

Sintering of irregular crystals: The irregular crystals appear to be sintered in two different ways. Irregular crystals, which occur as single secondary crystals on other forms of (primary) snow crystals, generally have the entire faces of their a-axes flat against the a-axes of the larger forms (Figs. 3–5). Alternatively, in the aggregates, sintering of adjacent crystals occurs between partial faces of the a-axes of adjacent crystals, as well as between partial faces of the c-axes of adjacent crystals (arrows, Figs. 13, 14). Because of the varying sizes and the irregular hexagonal faces of the individual crystals, this type of sintering results in snowflakes composed of heterogeneous aggregates.

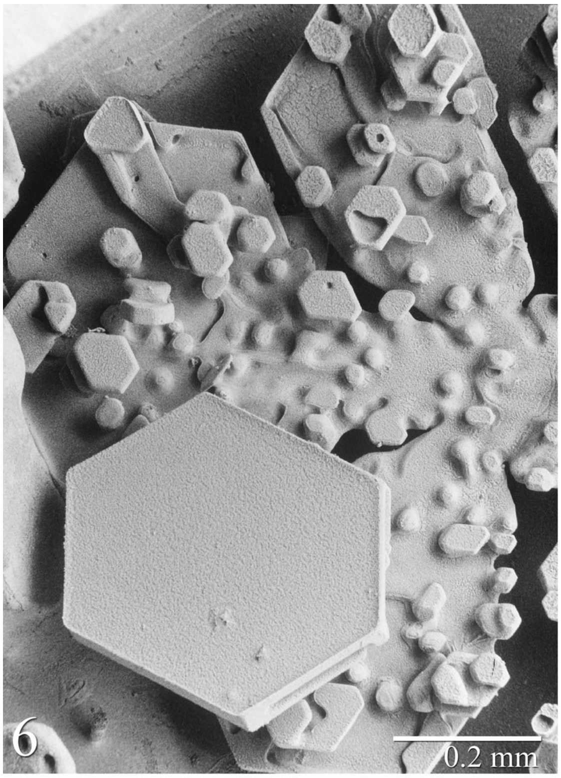
Atmospheric Metamorphosis of Irregular Crystals

In addition to aggregates in which nearly all the irregular crystals are distinctly faceted, other aggregates are encountered in which the faces of the individual crystals are more rounded. These examples appear to have undergone sublimation or metamorphosis (Figs. 15–20). At low magnifications, the irregular crystals in the aggregate appear faceted (Fig. 15). Higher magnifications reveal that the aggregates consist of a mixture of sharply faceted crystals, rounded or metamorphosed crystals, and spherical droplets (Figs. 16, 17). In these heterogeneous aggregates, the metamorphosed crystals more commonly occur along one side of the columnar aggregate (Figs. 17, 18). Consequently, one side of the columnar aggregate may consist primarily of metamorphosed crystals without sharply defined faces, whereas the opposing side of the column may exhibit irregular crystals with sharply defined faces.

In more advanced stages of metamorphosis, sharply defined faces of the individual crystals are less frequently encountered in the aggregate (Fig. 19). These aggregates appear to be largely enveloped by a continuous, amorphous layer of ice, which fills the crevices between adjacent crystals. Finally, in the most advanced stages of metamorphosis, the aggregate appears as a mass of elongated, interconnected, spherical droplets; only occasional remnants of faces from the original irregular crystals are apparent (Fig. 20, arrows). These later stages of metamorphosed aggregates of irregular crystals would be very difficult to distinguish from rime or graupel when viewed in a light microscope.

Terrestrial Frost

Aggregates of irregular crystals have also been observed as frost. These crystals are very similar in size and shape



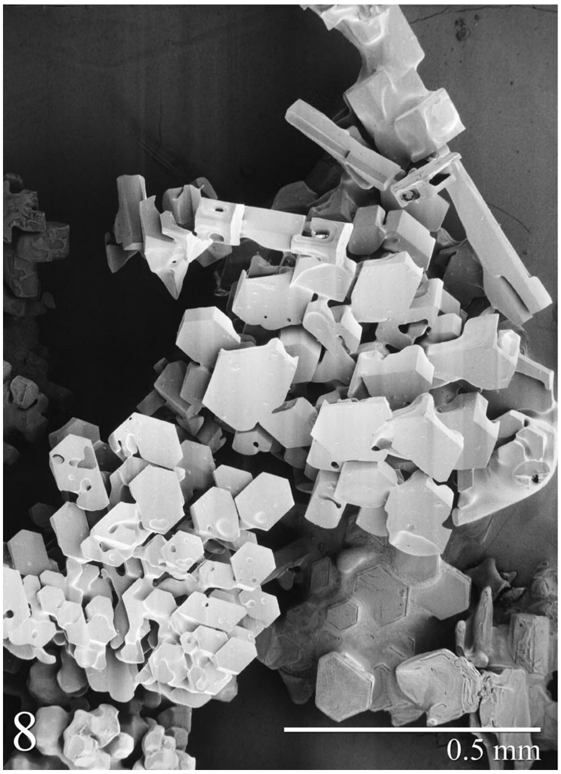
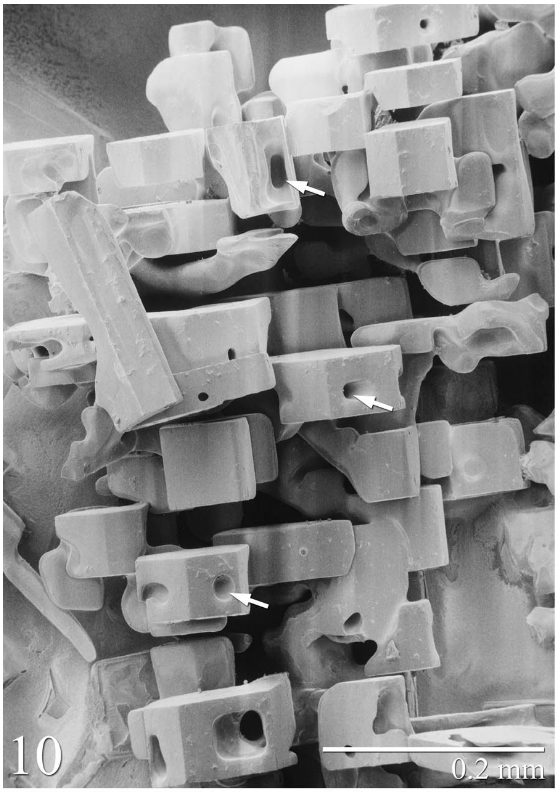
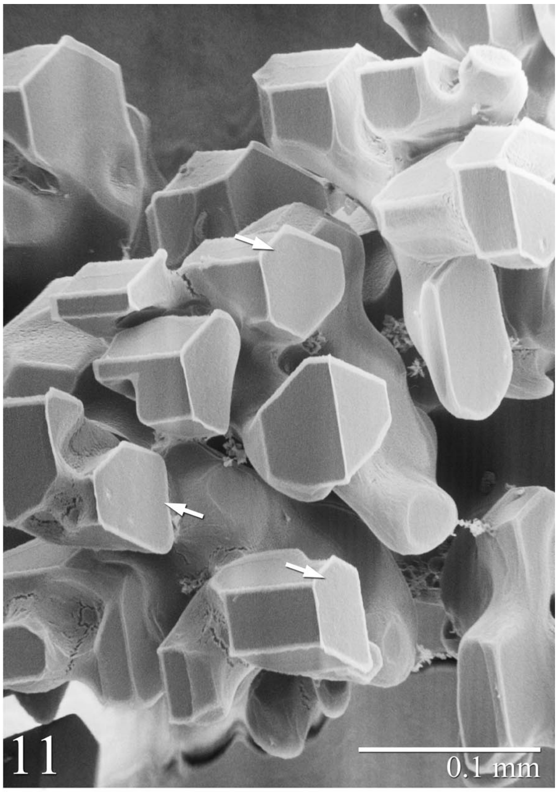
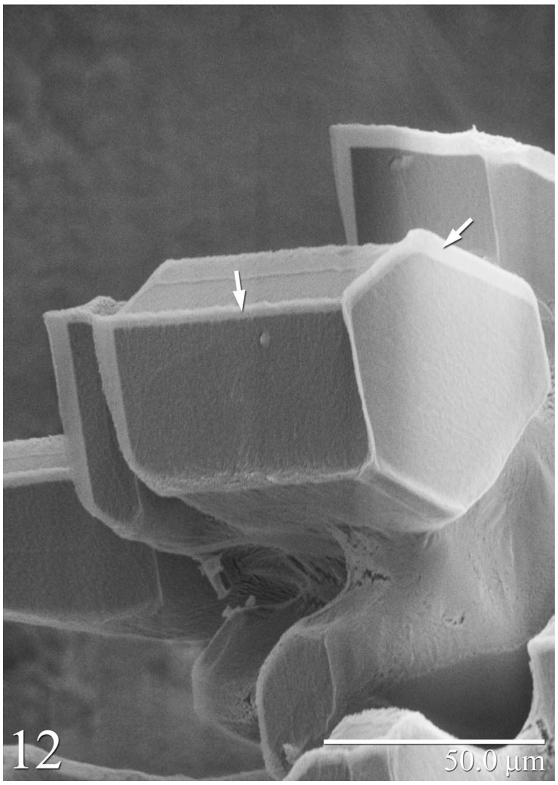


FIG. 6 Portion of a dendritic arm, from the crystal shown in Figure 5, illustrating the peripheral irregular crystals, which vary from about 50 to 100 µm, and the more central, spherical rime particles, which vary from 30 to 50 µm across. FIG. 7 Several snowflakes, each consisting of an aggregate of numerous irregular crystals. In each snowflake, the irregular crystals, which may vary from a few dozen to several hundred, tend to align along the same axis resulting in a "columnar" type of formation. FIG. 8 An aggregate of irregular crystals viewed from the a-axis. The cross-sectional areas of the aggregates, which frequently measure 0.2 to 0.4 mm across, can be composed of 10 to 40 individual irregular crystals. FIG. 9 An aggregate of irregular crystals viewed along the c-axis. Twenty-five to 75 irregular crystals, having their axes oriented in the same direction, form the length of a "column" that may be 1 to 2 mm long.







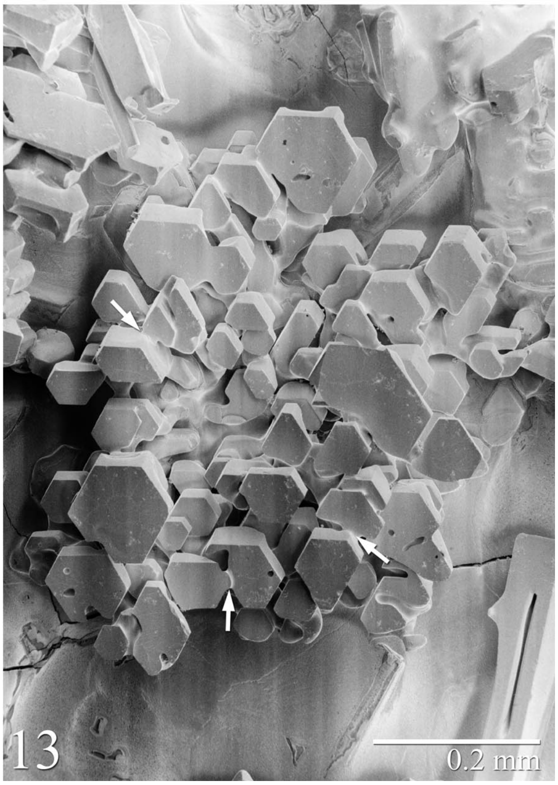
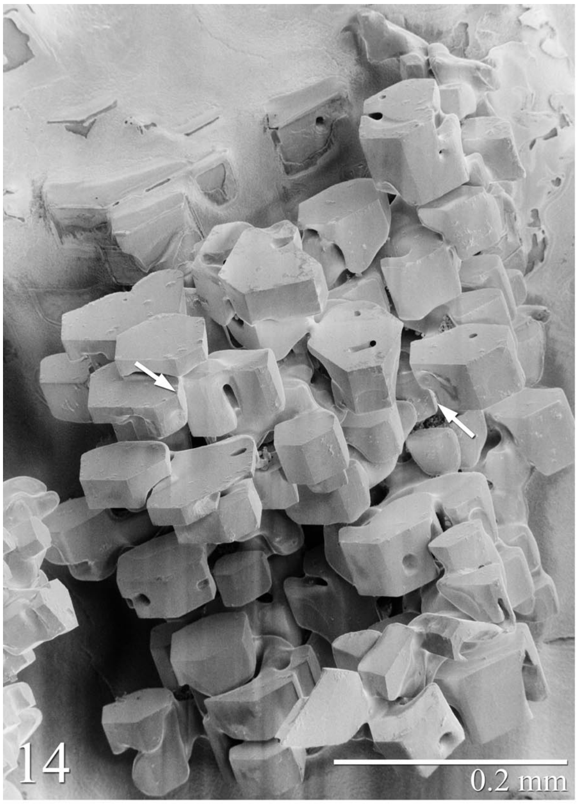
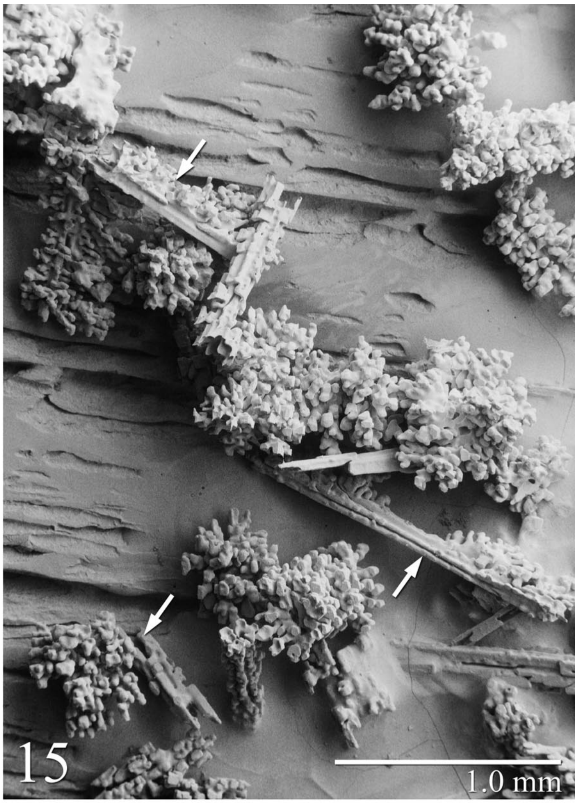
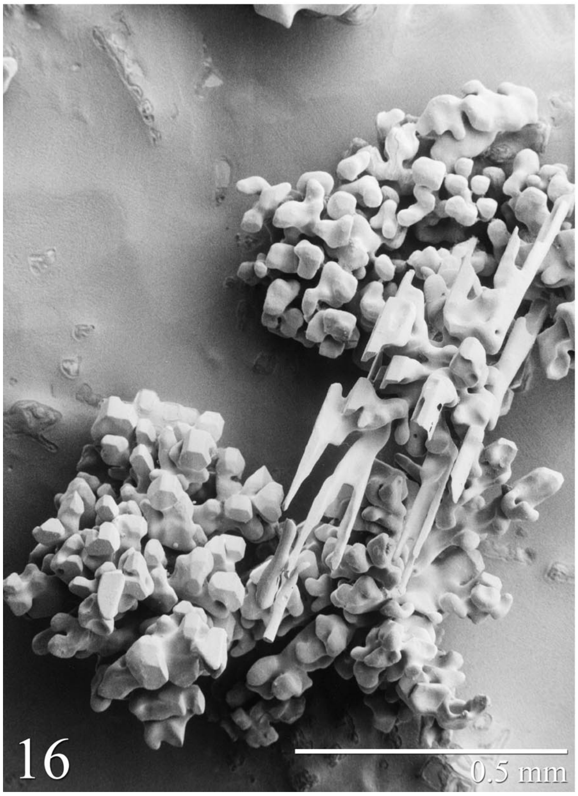


Fig. 10 Portion of an aggregate of irregular crystals viewed along the c-axis. The individual crystals are typically 60 to 90 µm across (a-axis) and 50 to 100 µm in length (c-axis). Several of these crystals exhibit holes or openings along their c-axes faces (arrows). Fig. 11 A-axis view of an aggregate reveals that the irregular crystals generally have asymmetrical hexagonal faces (arrows). Fig. 12 Single irregular crystal, having an irregular hexagonal face (a-axis), measuring about 60 µm across, and flat face or side (c-axis), approximately 50 µm in length. Occasionally slight ridges (arrows) can be observed around the perimeter of the crystals. Fig. 13 A-axis view of an columnar aggregate of irregular crystals illustrating the partial sintering (arrows) that occurs between the a-axes faces of adjacent crystals.







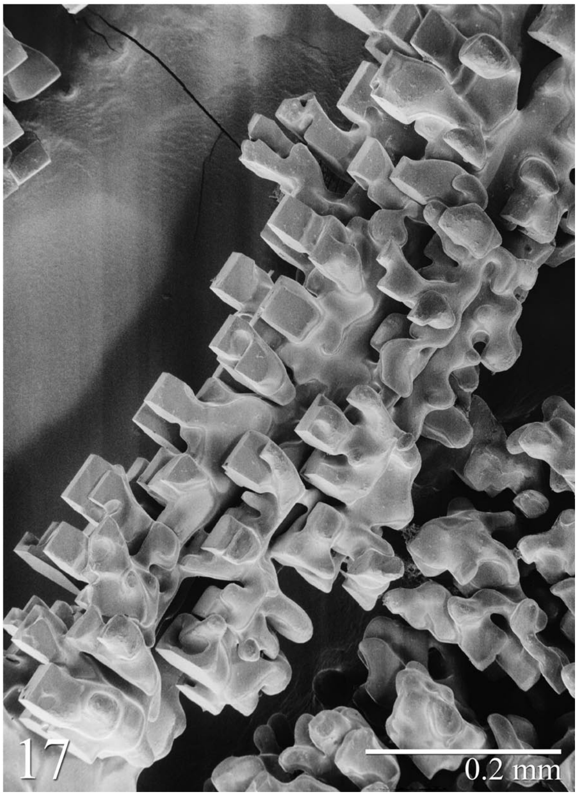
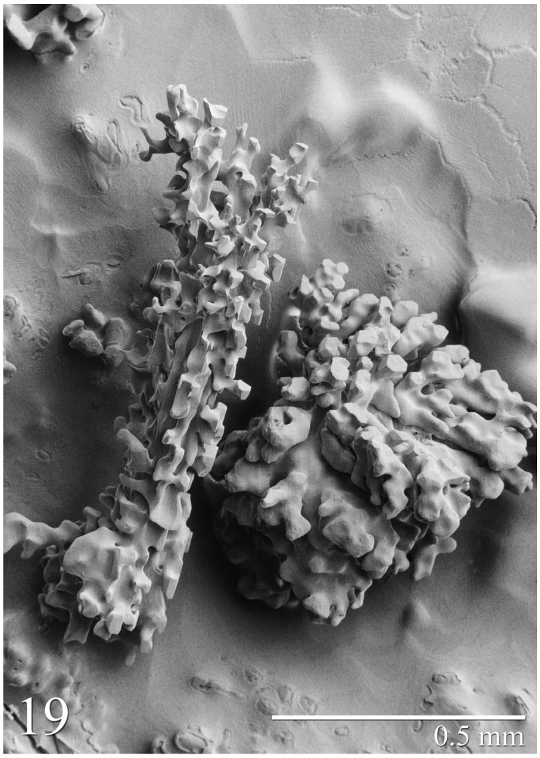


FIG. 14 Longitudinal view of an aggregate of irregular crystals illustrating the partial sintering (arrows) that occurs between the c-axes faces of adjacent crystals. FIG. 15 Low magnification of several aggregates of irregular crystals in which the individual crystals do not exhibit sharply defined faces. Needles are associated with several of these aggregates (arrows). FIG. 16 Aggregate consists of a mixture of partially faceted crystals and rounded or metamorphosed crystals. The aggregate is also associated with a group of needles. FIG. 17 Aggregate of partially faceted crystals (left) and metamorphosed crystals (right). Sintering of the crystals on the right appears as an amorphous layer of ice.



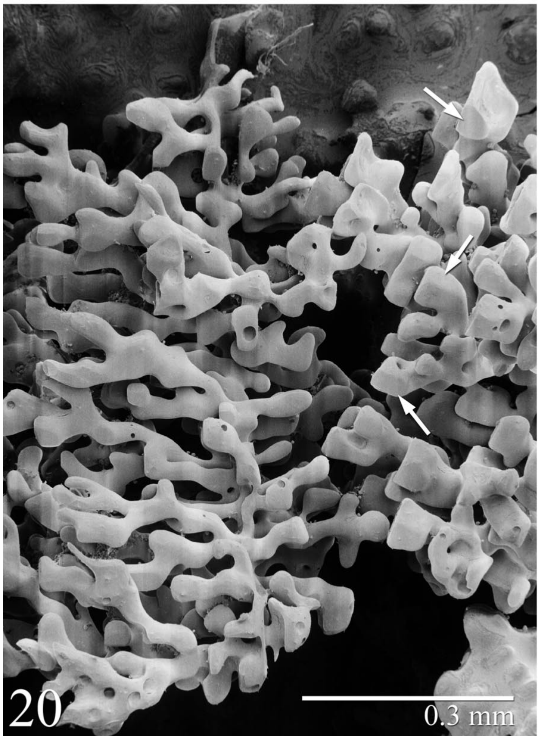


FIG. 18 High magnification of a heterogeneous aggregate of partially metamorphosed irregular crystals. In this aggregate, the crystals in the lower portion of the micrograph are more metamorphosed than those in the upper portion of the micrograph where the irregular crystals exhibit distinct faces along their a- and c-axes. FIG. 19 Two snowflakes illustrating advanced stages of metamorphosis of the irregular crystal aggregates. Distinct flat faces of the original irregular crystals are less frequently encountered. The aggregates appear to be largely enveloped by a continuous, amorphous layer of ice, which is more developed in crevices between the individual crystals. FIG. 20 An advanced stage of metamorphosis. The aggregate appears as a mass of elongated, interconnected, spherical droplets; only occasional remnants of sharply defined faces from the original irregular crystals are apparent (arrows). FIG. 21 Frost that consists of irregular crystals. These crystals are similar in size and shape to the aggregates of irregular crystals that form in the atmosphere. However, in this sample, which formed on the surface of a leaf, the crystals are more randomly arranged.

to the aggregates that form in the atmosphere (Fig. 21). However, their orientation within the frost aggregate is more random than that found in the atmospheric aggregate. The occurrence of frost, which is composed of irregular crystals, on the surface of a leaf blade suggests that the atmospheric conditions that favor this type of crystal growth also occur at ground level. This sample was collected at 8:00 AM when the air temperature was –2°C, suggesting that conditions just below freezing may favor this type of crystal growth.

Discussion

For more than 100 years, investigators who observed the microscopic features of snow crystals proposed classification systems to categorize, characterize, and name the numerous forms that occur in nature (Bentley and Humphreys 1931, Hellman 1893, Magono and Lee 1966, Nakaya 1954, Nordenskiöld 1893, Schaefer 1949). Both Hellman and Nordenskiöld (1893) described two basic forms, columns and plates, along with their variations and combinations. Bentley and Humphreys (1931), who admitted that their "scheme … is in no sense immutable," described five major groups and lavishly illustrated variations of the dendritic form. Schaefer (1949) expanded the classification to nine groups by adding such basic structures as needles, graupel, and sleet. Nakaya (1954) reduced the basic groups to seven, but Magano and Lee (1966) added a group called "germ of snow crystals" and illustrated eighty variations of the eight major groups.

To standardize the terminology and to seek general agreement on a classification system, international committees, which were convened in 1948 (see ICSI 1954) and again in 1985, published "The International Classification for Seasonal Snow on the Ground" in 1990 (Colbeck *et al.* 1990). This system, which is widely used today, divides precipitation particles into eight subclasses which include columns, needles, plates, stellar dendrites, irregular crystals, graupel, hail, and ice pellets. The "irregular crystals" that are included in this scheme were initially characterized by Nakaya (1954) who described them as a snow particle "… with so many cloud particles that it appears as a lump of frozen droplets." The ICSI (1954) commission retained the term "irregular crystal" for this class of solid precipitation and described the form as "… a snow particle made up of a number of small crystals grown together in random fashion. Generally the component crystals are so small that the crystalline form of the particle can only be seen with the aid of a magnifying glass or microscope." The most recent revised guide from this commission (Colbeck *et al.* 1990) merely described irregular crystals as "clusters of very small crystals." In our study, the resolution and depth of field that is characteristic of the field-emission SEM (FESEM) provided distinct images of this class of snow crystals. This instrument allowed us to document the atmospheric and terrestrial occurrence of irregular crystals and to characterize their features.

The irregular crystals that are observed on most other crystalline forms, such as dendrites or plates, generally occur as single crystals. In our experience, when the aggregates of irregular crystals are encountered, they are most frequently found associated with the needles. This association would suggest that the atmospheric conditions that promote growth of the needles would be close to those that also promote the formation of the irregular crystals. Under laboratory conditions, Nakaya (1954) indicated that the c-axis growth, which promotes needle crystal growth, becomes extremely rapid at –6°C. Furthermore, when the temperature is increased to –4°C to –1°C and the supply of water vapor is abundant, crystals no longer grow into complete needles but rather form irregular combinations of needle-like crystals. Nakaya further states that the "details about its structure are still unknown." Perhaps these represent the irregular crystals described in our study. This suggestion is supported by the fact that we most frequently observed the aggregates in association with the needles and that the irregular crystals were found as frost when the air temperature was –2°C.

Conclusion

This study uses LTSEM to characterize small discrete crystals believed to correspond to the irregular crystals that previous investigators have attempted to describe with the light microscope. These crystals, which appear as irregular hexagons when viewed along their a-axes, generally measure 60 to 90 µm across. Their lengths (c-axis) rarely exceed their diameters. Although the irregular crystals are occasionally found as individual, secondary particles on other larger forms of snow crystals, such as plates and dendrites, they are most frequently encountered in aggregates consisting of more than 100 crystals that tend to form a column. The aggregates are often associated with needles, suggesting that the atmospheric conditions that favor needle growth may be close to those that favor growth of irregular crystals. Furthermore, examples believed to represent atmospheric metamorphoses of irregular crystals are commonly encountered. Metamorphosis can result in aggregates that consist of partially sublimated crystals and droplets that collectively would be difficult to distinguish from graupel when using a light microscope.

References

Beckett A, Read ND: Low-temperature scanning electron microscopy. In *Ultrastructural Techniques for Microorganisms* (Eds. Aldrich HC, Todd WJ). Plenum Publ Corp., New York (1986) 45–86

Bentley WA, Humphreys WJ: *Snow Crystals*. McGraw-Hill Book Co., Inc. New York (1931)

Colbeck S, Akitaya E, Armstrong R, Gubler H, Lafeuille J, Lied K, McClung D, Morris E: *The International Classification for Seasonal Snow on the Ground. International Commission on Snow* *and Ice (IAHS).* World Data Center for Glaciology, University of Colorado, Boulder, Colorado (1990)

Echlin P, Paden R, Dronzek B, Wayte R: Scanning electron microscopy of labile biological material maintained under controlled conditions. *Scan Electr Microsc* 1970, 49–56 (1970)

Hellman G: *Schneekrystalle*. J. Mückenberger, Berlin, Germany (1893)

ICSI: *The International Classification for Snow. IAHS International Commission on Snow and Ice. Technical Memorandum No. 31, Associate Committee on Soil and Snow Mechanics*. National Research Council, Ottawa, Canada (1954)

Magono C, Lee C: Meteorological classification of natural snow crystals. *J Fac Sci, Hokkaido Univ, Ser VII* 2, 321–335 (1966)

Nakaya U: *Snow Crystals: Natural and Artificial*. Harvard University Press, Cambridge, Massachusetts (1954)

Nordenskiöld G: The inner structure of snow crystals. *Nature Land* 48, 592–594 (1893)

Rango A, Wergin WP, Erbe EF: Snow crystal imaging using scanning electron microscopy: Part I, Precipitated snow. *Hydrological Sci J* 41, 219–233 (1996a)

Rango A, Wergin WP, Erbe EF: Snow crystal imaging using scanning electron microscopy: Part II, Metamorphosed snow. *Hydrological Sci J* 41, 235–250 (1996b)

Rango A, Wergin WP, Erbe EF, Josberger EG: Snow crystal imaging using scanning electron microscopy, III—Glacier ice, snow and biota. *Hydrological Sci J* 45, 357–375 (2000)

Schaefer VJ: The formation of ice crystals in the laboratory and the atmosphere. *Chem Rev* 44, 291–320 (1949)

Wergin WP, Erbe EF: Comparison of identical areas of freeze-fractured membranes from yeast using low temperature SEM and conventional TEM. *Proc Royal Microscopical Soc* 25, S21 (1990)

Wergin WP, Erbe EF: Increasing resolution and versatility in low temperature conventional and field emission scanning electron microscopy. *Scan Microsc* 5, 927–936 (1991)

Wergin WP, Erbe EF: Can you image a snowflake with an SEM? Certainly! *Proc Royal Microsc Soc* 29, 138–140 (1994a)

Wergin WP, Erbe EF: Use of low temperature scanning electron microscopy to examine snow crystals. *Proc 14th Int Cong Electron Microsc* 3B, 993–994 (1994b)

Wergin WP, Rango A, Erbe EF: Observations of snow crystals using low-temperature scanning electron microscopy. *Scanning* 17, 41–49 (1995)

Wergin WP, Rango A, Erbe EF, Murphy CA: Low temperature SEM of precipitated and metamorphosed snow crystals collected and transported from remote sites. *J Microsc Soc Amer* 2, 99–112 (1996a)

Wergin WP, Rango A, Erbe EF: The structure and metamorphism of snow crystals as revealed by low temperature scanning electron microscopy. *Proc Eastern Snow Conf* 53, 195–204 (1996b)

Wergin WP, Rango A, Erbe EF: Image comparisons of snow and ice crystals photographed by light (video) microscopy and low temperature scanning electron microscopy. *Scanning* 20, 285–96 (1998)

Wergin WP, Rango A, Erbe EF: Observations of rime and graupel using low temperature SEM. *Scanning* 21, 87–88 (1999)