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# Rime and Graupel: Description and Characterization as Revealed by Low-Temperature Scanning Electron Microscopy

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**Summary:** Snow crystals, which form by vapor deposition, occasionally come in contact with supercooled cloud droplets during their formation and descent. When this occurs, the droplets adhere and freeze to the snow crystals in a process known as accretion. During the early stages of accretion, discrete snow crystals exhibiting frozen cloud droplets are referred to as rime. If this process continues, the snow crystal may become completely engulfed in frozen cloud droplets. The resulting particle is known as graupel. Light microscopic investigations have studied rime and graupel for nearly 100 years. However, the limiting resolution and depth of field associated with the light microscope have prevented detailed descriptions of the microscopic cloud droplets and the three-dimensional topography of the rime and graupel particles. This study uses low-temperature scanning electron microscopy to characterize the frozen precipitates that are commonly known as rime and graupel. Rime, consisting of frozen cloud droplets, is observed on all types of snow crystals including needles, columns, plates, and dendrites. The droplets, which vary in size from 10 to 100 μm, frequently accumulate along one face of a single snow crystal, but are found more randomly distributed on aggregations consisting of two or more snow crystals (snowflakes). The early stages of riming are characterized by the presence of frozen cloud droplets that appear as a layer of flattened hemispheres on the surface of the snow crystal. As this process continues, the cloud droplets appear more sinuous and elongate as they contact and freeze to the rimed crystals. The advanced stages of this process result in graupel, a particle 1 to 3 mm across, composed of hundreds of frozen cloud droplets interspersed with considerable air spaces; the original snow crystal is no longer discernible. This study increases our knowledge about the process and characteristics of riming and suggests that the initial appearance of the flattened hemispheres may result from impact of the leading face of the snow crystal with cloud droplets. The elongated and sinuous configurations of frozen cloud droplets that are encountered on the more advanced stages suggest that aerodynamic forces propel cloud droplets to the trailing face of the descending crystal where they make contact and freeze.

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

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## Introduction

For more than 100 years, scientists have observed and photographed numerous types of precipitating snow crystals with the light microscope (LM) (Bentley and Humphreys 1931, Magono and Lee 1966, Nakaya 1954). To help characterize the basic shapes and to standardize the crystal terminology, various classification schemes were proposed (Bentley and Humphreys 1931, Hellman 1893, Magono and Lee 1966, Nakaya 1954, Nordenskiöld 1893, Schaefer 1949). However, these authors described anywhere from 6 to as many as 80 different types of snow crystals depending on which scheme was examined. To clarify this confusion, international commissions were appointed that have categorized newly precipitated snow crystals into eight basic shapes or subclasses (Colbeck *et al*. 1990, ICSI 1954).

One of the most difficult subclasses to describe accurately are atmospheric snow crystals, which initially form by vapor deposition, but continue to grow by the accretion of minute supercooled cloud droplets. Contact and freez-

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ing of these droplets onto a snow crystal result in a rimed crystal. If the riming process continues until the identity of the original crystal is no longer evident, the resulting crystal is referred to as graupel. The cloud droplets that accumulate on a snow crystal are microscopic, but their accumulation may considerably increase the size and topography of the original crystal. Therefore, the detailed structure of rime and graupel is difficult to observe and photograph with the LM, which has limited resolution and depth of field.

Recently, our laboratory has developed methods that allow us to sample, observe, and photograph snow crystals with a technique known as low-temperature scanning electron microscopy (LTSEM) (Rango *et al*. 1996a,b; 2000; Wergin and Erbe 1990; 1991;1994a,b; Wergin *et al*. 1995; 1996a,b; 1998; 1999; 2002a,b). This technique can be used to magnify snow crystals several thousand times and has a depth of field that exceeds that of the LM by at least 1,000 times (Wergin *et al*. 1998). The current study utilizes LTSEM to describe and characterize rime and graupel.

## Material and Methods

### Collection Procedure

Data illustrated in this study resulted from five different snow collections during the period of 1995 to 2002 from sites near the following locations: Beltsville, Maryland, Bearden Mountain, West Virginia, Greenwood, Wisconsin, Pinedale, Wyoming, and Fraser, Colorado. The samples that were obtained when the air temperatures ranged from –12° C to 0° C consisted of freshly fallen snowflakes. The collection procedure consisted of placing a thin layer of liquid Tissue-Tek, a commonly used cryoadhesive for biological samples, on a fabricated, flat copper plate measuring 15 × 27mm (Tissue-Tek, Albertville, Minn., USA). The Tissue-Tek and the plate were precooled to ambient outdoor temperatures before use. Next, newly fallen snowflakes were either allowed to settle on the surface of the plate, lightly dislodged and allowed to fall onto its surface, or sampled by gently pressing the plate to the surface of freshly fallen snow. The plate containing the sample 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 sample to the plate. Frozen plates containing the samples were inserted diagonally into 20 cm segments of square brass channeling and lowered into a dry shipping dewer that had been previously cooled with $LN_2$. The dewer containing the samples was conveyed from the collection sites and then either transported by van (from West Virginia) or shipped by air to the laboratory in Beltsville, Maryland. Upon reaching the laboratory, the samples were transferred under $LN_2$ to an $LN_2$ storage dewer where they remained before being further prepared for observation with LTSEM.

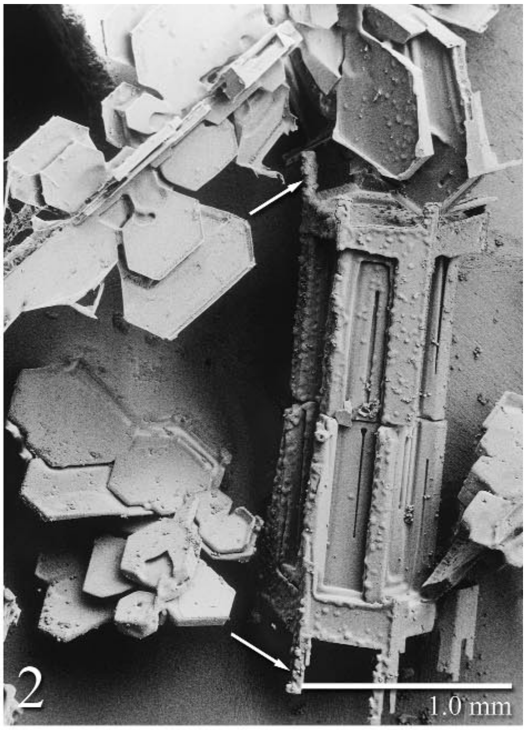
### Preparation for Low-Temperature Scanning Electron Microscopy Examination

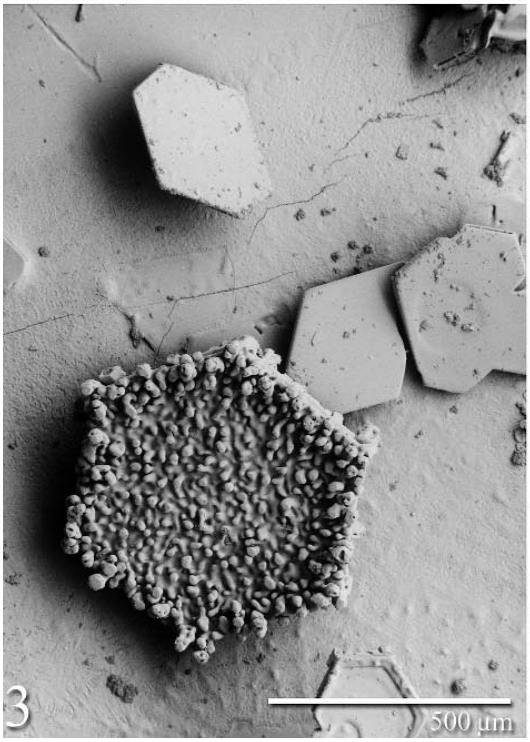
To prepare the samples for LTSEM observation, the brass channeling was removed from the storage dewer and placed in a styrofoam box filled with $LN_2$. A plate was removed from the channeling and placed in a modified Oxford specimen carrier (Oxford Instruments, Enysham, England). The carrier containing the plate was transferred to the slush chamber of an Oxford CT 1500 HF Cryotrans system that had been filled with $LN_2$. Next, the carrier was attached to the transfer rod of the Oxford cryosystem, moved under vacuum into the prechamber for etching and/or sputtercoating with Pt, and then inserted into a Hitachi S-4100 field-emission SEM that was equipped with a cold stage maintained at −176° ± 20° C (Hitachi High-Technologies Corp., Tokyo, Japan). Accelerating voltages of 500 V to 10 kV were used to observe and record images onto Polaroid Type 55 P/N film (Polaroid, Cambridge, Mass., USA). To obtain stereo pairs, a stage tilt of $6^o$ was introduced between the first and second images.

## Results

Riming, or the presence of frozen cloud droplets, is observed on the surface of all basic types of snow crystals: needles, columns, plates, and dendrites. The size of the frozen droplets varies from about 10 to 100 μm. Rime droplets consisting of the largest sizes are more frequently found on needles (Fig. 1), whereas the smallest droplets are more often associated with columns (Fig.2). On plates and dendrites, which exhibit rime deposition, the frozen droplets are generally found as a uniform layer on one side of the crystal surface. The droplets do not consist of perfect spheres, but either as irregular hemispheres, whose surfaces exhibit smooth continuity with that of the primary crystal, as illustrated in the central regions of the plate and dendrite illustrated in Figures 3 and 4, or as sinuous, elongated droplets as observed along the peripheral edges of the crystals (see Fig. 5). In the latter case, elongated droplets are attached to the primary crystals by short narrow "necks."

On most types of snow crystals, the rime appears to accumulate preferentially on one surface of the primary crystal. For example on hexagonal plates, rime tends to accumulate on one face of the crystal, whereas the opposite face remains relatively free of deposition (Fig.5). On capped bullets, which consist of short columns that taper to a point on one end and have attached hexagonal plates on the other, the rime accumulates on the upper face of the hexagonal plate, but the underside of the plate and the bullet do not show accumulations (Fig.6). On capped columns, which have hexagonal plates attached on both ends, rime deposition occurs on both outer surfaces of the plates, whereas the undersides of the plates and the columns do not exhibit rime deposition (Fig. 7). Hexagonal dendrites,





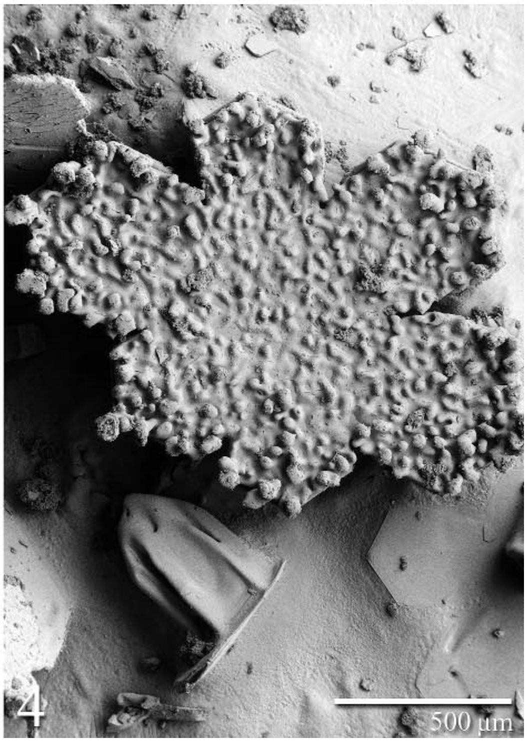
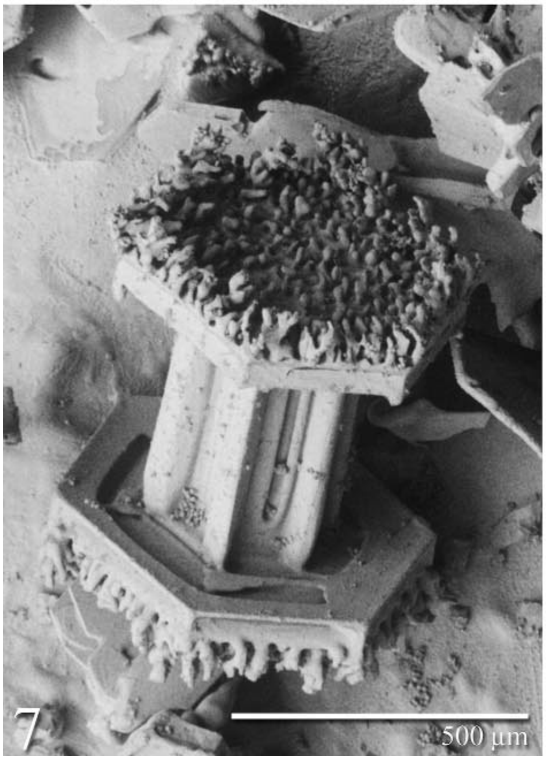


FIG. 1 Aggregation of needles with deposition of frozen cloud droplets or rime. Rime associated with needles tends to be the largest of the droplets, frequently measuring up to 100 µm in diameter. Sample collected from Bearden Mt., West Virginia, when surface air temperature was –2° C.

FIG. 2 Capped column having needle-like extensions that emanate from the plates (arrows). Rime droplets on the column appear as small hemispheres whose surfaces are continuous with that of the column. These frozen droplets, which are about 10 µm in diameter, represent the smallest forms of rime that are encountered on snow crystals. Sample collected from Bearden Mt., West Virginia, when temperature was –3° C.

FIG. 3 Hexagonal plate containing a uniform deposition of frozen rime droplets. The droplets near the center of the plate are smaller and less spherical than those that have accumulated on the peripheral regions of the crystal. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

FIG. 4 Hexagonal plate with broad branches. The pattern of rime deposition of this crystal is similar to that illustrated in Figure 3. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.



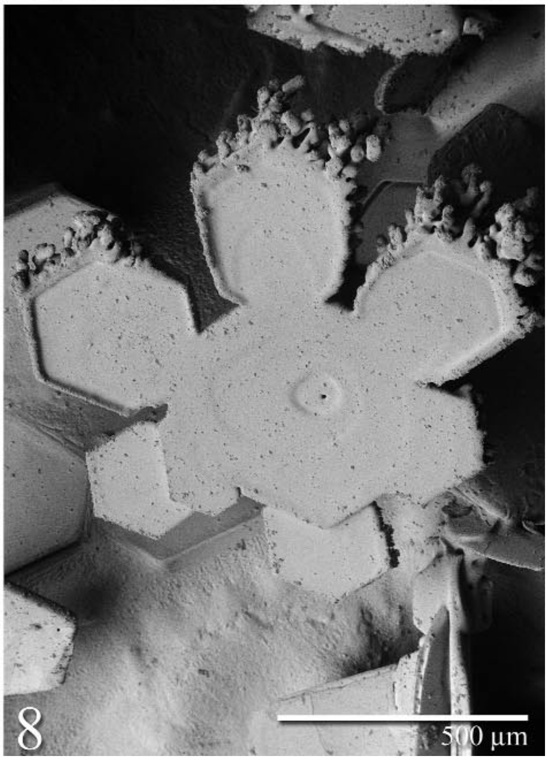


FIG. 5 Hexagonal plate illustrating an accumulation of elongated frozen rime droplets emanating from one surface of the crystal. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

FIG. 6 Capped bullet. The rime accumulation is restricted to the outer surface of the hexagonal plate, whereas the underside of the plate and the lateral surfaces of the bullet are free from the deposition. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

FIG. 7 Capped column. Rime deposition is concentrated on the outer surfaces of the two hexagonal plates that formed on the ends of the column. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

FIG. 8 Hexagonal dendrite with broad branches. Although the surface of the crystal illustrated in the figure is free of deposition, rime accumulation can be observed on the opposite surface of this crystal. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

whose extensions or branches are either broad (Fig. 8) or more stellar (Fig. 9), exhibit rime deposition on only one of the flat surfaces, while the opposite surface remains free of the frozen droplets.

Some aggregations of crystals, such as the radiating assemblage of plates shown in Figure 10, also show preferential accumulation of rime along one surface of the aggregate. Even needles tend to exhibit preferential accumulation of rime. Apparently, once rime deposition begins to occur on the crystal, this process continues on the same surface area and results in the elongated accumulation of frozen droplets evident in Figure 11. As this process continues, layers of rime as thick as 0.5 mm can be observed on the surfaces of crystals (Fig. 12). These layers appear as accumulations of elongated droplets that are interconnected to the underlying droplets by narrowed necks. The accumulation does not exhibit any crystalline characteristics. Alternatively, the surfaces of the rime droplets are sinuous, continuous, and appear to flow smoothly from one droplet to the next all the way back to the primary crystal.

Although the types of snow crystals described above have preferential deposition of rime droplets, some crystalline aggregations do not exhibit this characteristic. For example, the single capped bullet described in Figure 6 exhibits preferential deposition; however, aggregates consisting of four capped bullets, which are attached at their tapered apices, have rime droplets on the surfaces of the plates, as well as along the lateral surfaces of the bullets (Fig. 13). Likewise, rime accumulates on one surface of a single dendrite (Figs. 4, 8, and 12), whereas a radiating assemblage of dendrites, as illustrated in Figure 14, contains a heavy accumulation of rime in the center of the aggregate; the outer arms of the dendrites are free of deposition.

Rime accumulation can completely obscure the identity of the original or primary crystal. This process results in a crystal type known as graupel. In Figure 15, the primary crystal appears to have been a needle; however, the entire surface that is exposed is encumbered with frozen cloud droplets. In Figure 16, the original crystal may have consisted of a hexagonally branched stellar dendrite, but the extensive accumulation of rime makes identification difficult; consequently this snow crystal is more properly referred to as graupel. Continuation of riming may result in the formation of lump graupel, which may measure 1 to 3 mm (Fig. 17). Some advanced stages of graupel have a cone-like shape, which may result from the preferential deposition of frozen cloud droplets on one surface of a hexagonal plate (Fig. 18).

The droplets that accumulate and result in the formation of graupel exhibit no crystalline characteristics and no flat surfaces or facets (Fig. 19). Consequently, the graupel particle consists of an aggregation of sinuous droplets whose smooth surfaces are continuous. Although the frozen droplets are well sintered, considerable air space exists within the graupel particle (Fig. 20).

## Discussion

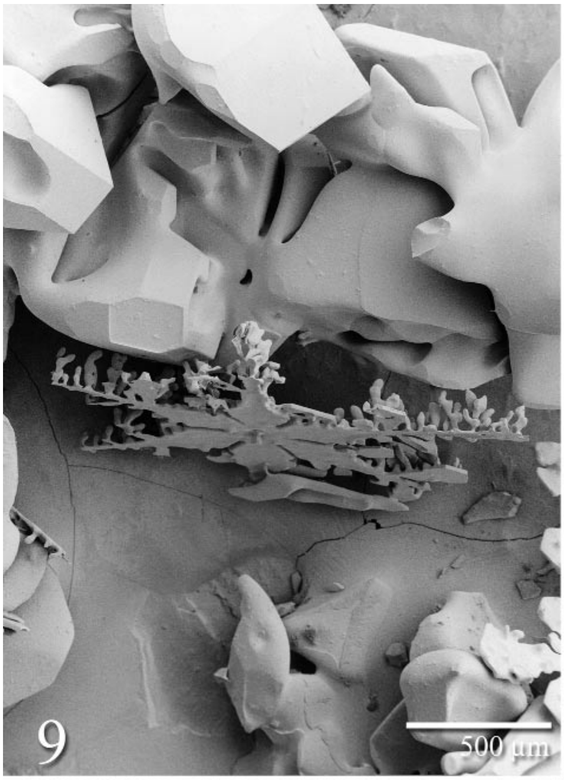
The formation of ice crystals in the atmosphere can result from vapor deposition, accretion, or aggregation (see reviews Hobbs 1974, Lamb 1999). Vapor deposition gives rise to discretely shaped crystals such as needles, columns, plates, and dendrites. Accretion of supercooled cloud droplets results in rime and graupel, whereas aggregation, or the association of two or more crystals, produces what is normally referred to as a snowflake. Rime and graupel have been observed with the LM for nearly 100 years. However, comparing LM images with those obtained in an SEM indicate that many structural details such as crystal thickness, topography, and microstructure are difficult to determine with the LM (Domine *et al.* 2001, Wergin *et al.* 1998). The current study utilizes LTSEM to describe and characterize the details of rimed snow crystals and the microstructure of graupel for the first time.

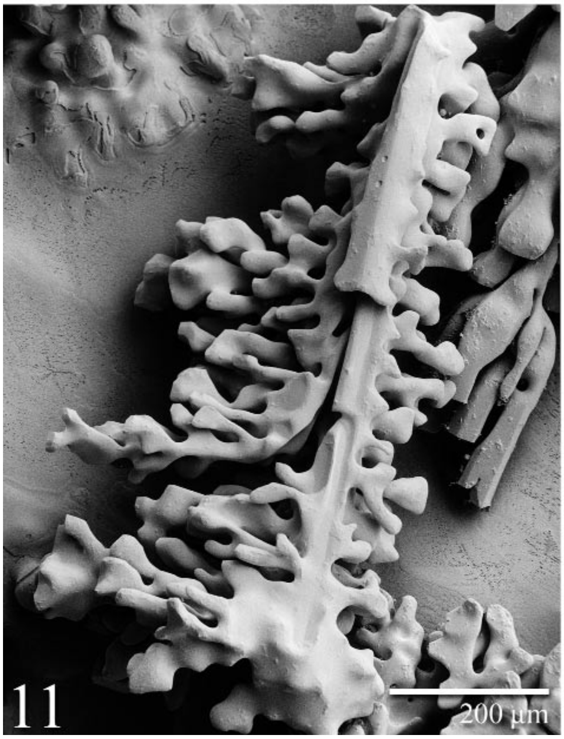
Rime and graupel occur in clouds that contain forming snow crystals and supercooled cloud droplets (Pruppacher and Klett 1997). The unfrozen cloud droplets can occur over a wide range of temperatures that extend from 0° C to –45° C (Hallet 1984, 1988; Pruppacher and Klett 1997). Likewise, snow crystals can form under a similar range of temperatures; however, their shape or growth habit, that is, whether they become needles, columns, plates, or dendrites, seems to be primarily influenced by specific temperatures (Lamb 1999). For example, atmospheric temperatures around –5° C favor needle growth, whereas temperatures colder than –30° C favor dendrite formation. Because unfrozen cloud droplets and the various forms of snow crystals exist under the same temperatures, one would expect to encounter riming on all types or shapes of snow crystals. In our study, riming was indeed encountered on needles, columns, plates, and dendrites.

Light microscopic studies indicate that the frozen cloud droplets that form rime have diameters of 10 μm (Hallet 1984), 10 to 20 μm (Black and Hallet 1998), 15 to 45 μm (Nakaya and Terada 1934), and 10 to 80 μm (Pruppacher and Klett 1997). The technique we have used in this study also indicates that the dimensions and shapes of rime are variable and diverse. Indeed, the shapes may vary from the small, 10 μm hemispheric particles that are initially observed on the surface of snow crystals to the large 100 μm elongated droplets that constitute the graupel particles.

In a previous study, Wergin *et al.* (2002a) indicated that, in LM studies, rime particles might have been confused with irregular crystals, whose sizes overlap with those of rime. In fact, even with the SEM, the distinction between rime particles and irregular crystals that may have been exposed to partial atmospheric melting is not always clear (see Fig. 15).

Black and Hallet (1998) indicated that rime initially freezes as “near hemispheres” on the surface of a snow crystal. The images recorded with LTSEM also support this observation (Figs. 1–4). Perhaps the initial impact of the cloud droplet on the surface of the falling snow crystal





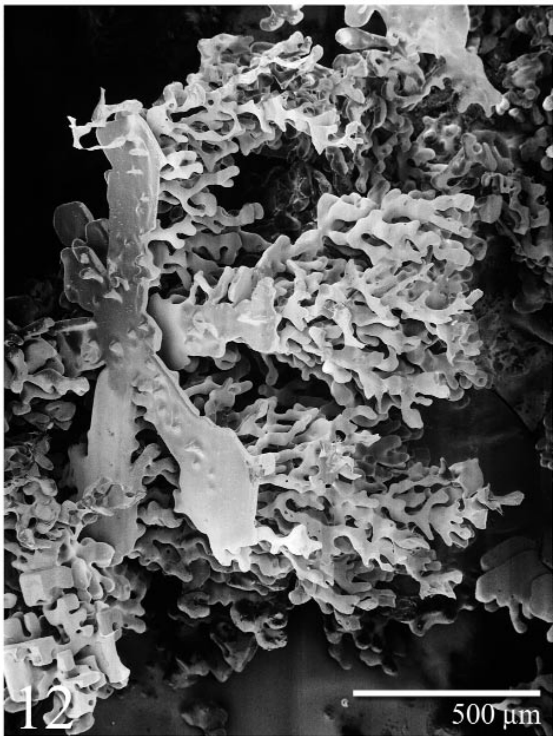
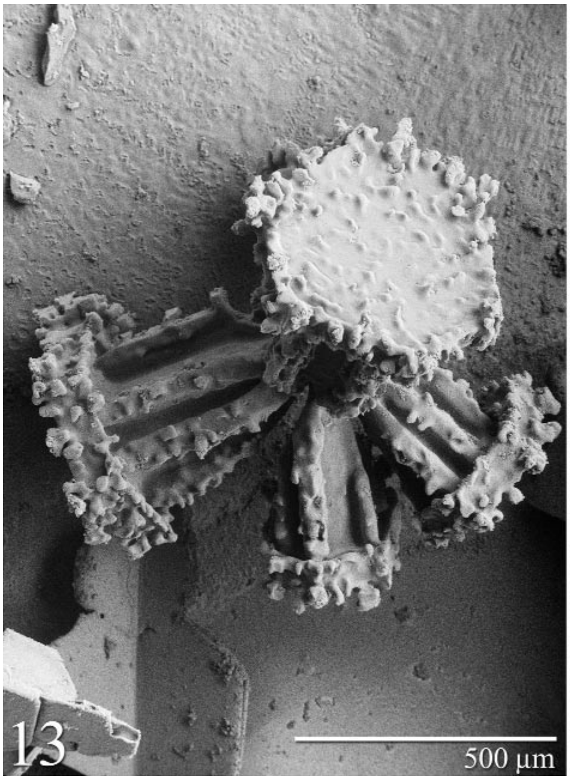


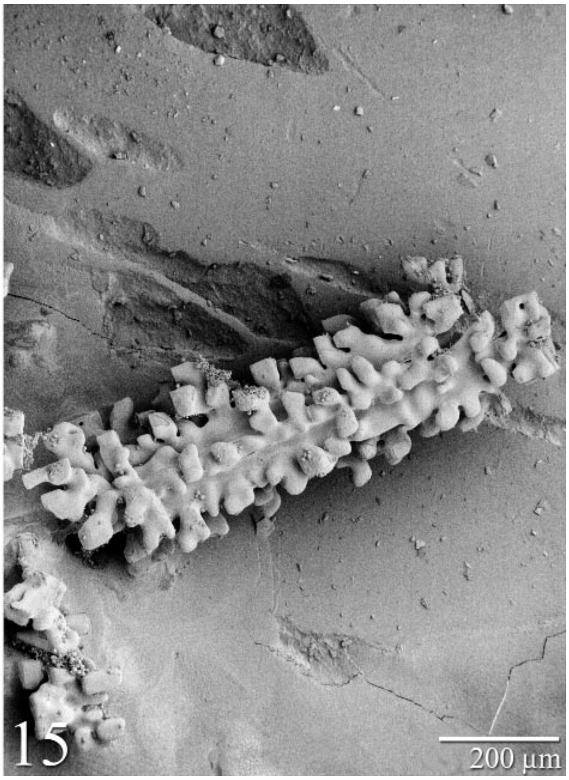
FIG. 9 Stellar hexagonal dendrite in vertical view. One side of the crystal does not contain rime, while the opposing side shows significant accumulation of the frozen droplets. Sample collected near Greenwood, Wisconsin, when the temperature was –4° C.

FIG. 10 Radiating assemblage of plates. On this aggregation of hexagonal plates, rime accumulation appears to be preferentially localized along one surface of the assemblage; the opposing surfaces remain unrimed. Sample collected near Beltsville, Maryland, when temperature was –5° C.

FIG. 11 Needle illustrating preferential accumulation of rime along one surface. The layer of rime consists of elongated droplets interconnected by more narrow, sinuous necks. Sample collected from Bearden Mt., West Virginia, when temperature was –4.5° C.

FIG. 12 Dendritic crystal containing a significant accumulation of rime preferentially accumulated on one surface. The rime deposit is nearly 0.5 mm thick. Sample collected from Bearden Mt., West Virginia when temperature was 0° C.





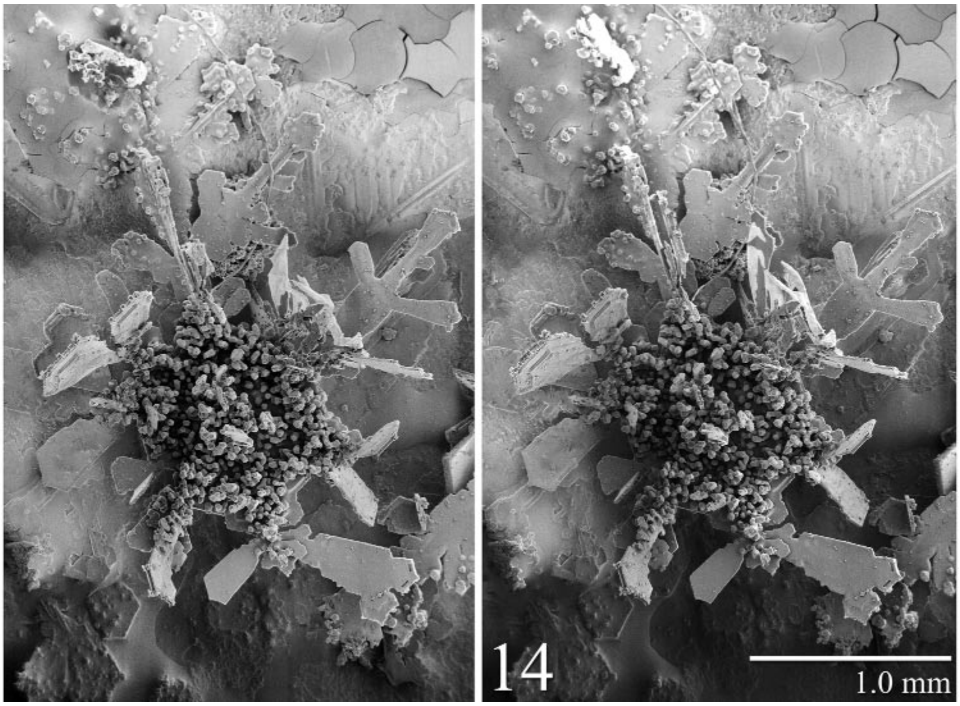
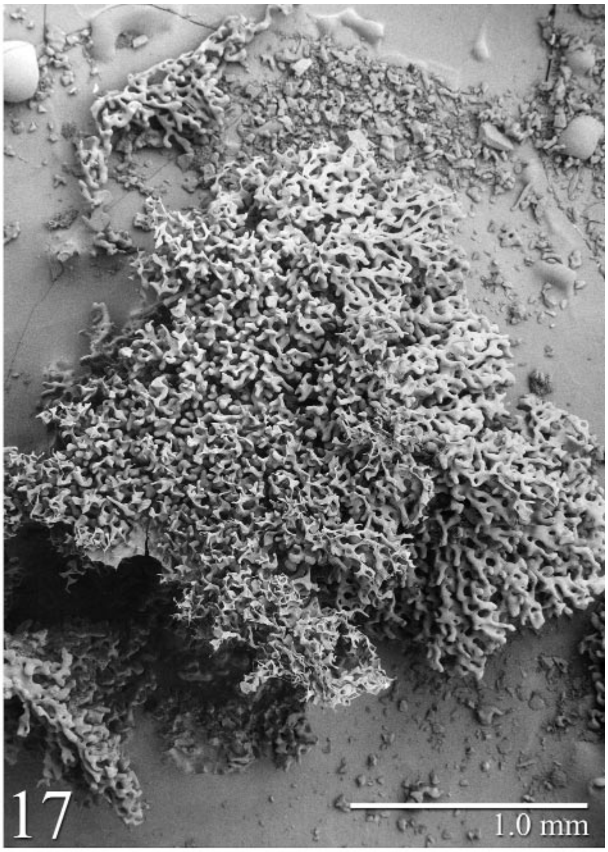


FIG. 13 Four bullets, which are attached at their tapered apices, each capped with a hexagonal plate. This aggregation of snow crystals does not exhibit preferential deposition of rime but alternatively has frozen droplets on the flat surfaces and sides of the plates, as well as on the lateral sides of the bullets. Sample collected from Bearden Mt., West Virginia, when temperature was –12° C.

FIG. 14 Stereo pair of micrographs illustrating a radiating assemblage of dendrites that contains a heavy accumulation of rime in the center of the aggregate. The arms of the dendrites remain free of the frozen droplets. Sample collected from Bearden Mt., West Virginia, when temperature was –1° C.

FIG. 15 Needle that is encumbered with rime. In some cases, these larger "droplets" of rime are difficult to distinguish from secondary crystals, known as irregular crystals that may form on a primary crystal and then partially melt when exposed to higher atmospheric temperatures during descent. Sample collected from Bearden Mt., West Virginia, when temperature was –4.5° C.



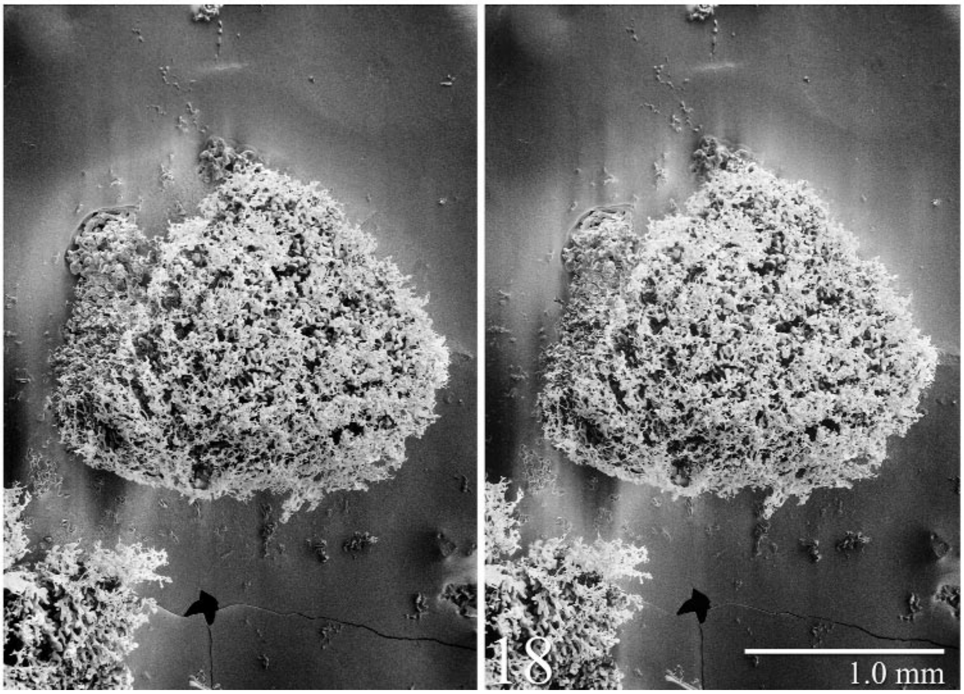
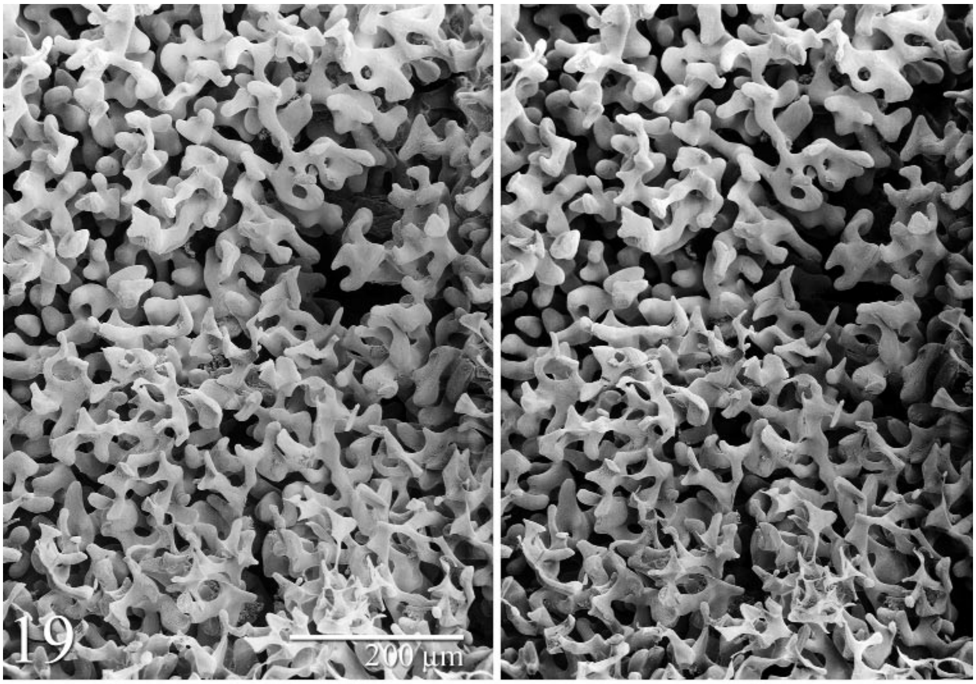


FIG. 16 Graupel particle whose primary crystal may have been a hexagonal dendrite. Sample collected from Bearden Mt., West Virginia when temperature was –0° C.

FIG. 17 Large graupel particle measuring over 2 mm in diameter. Primary crystal is no longer identifiable because of the accumulation of frozen droplets. Sample collected near Fraser, Colorado.

FIG. 18 Stereo pair of micrographs showing graupel with a cone-like shape that may have resulted from preferential accumulation of droplets along one side of a primary crystal such as a hexagonal plate. Sample collected near Pinedale, Wyoming.



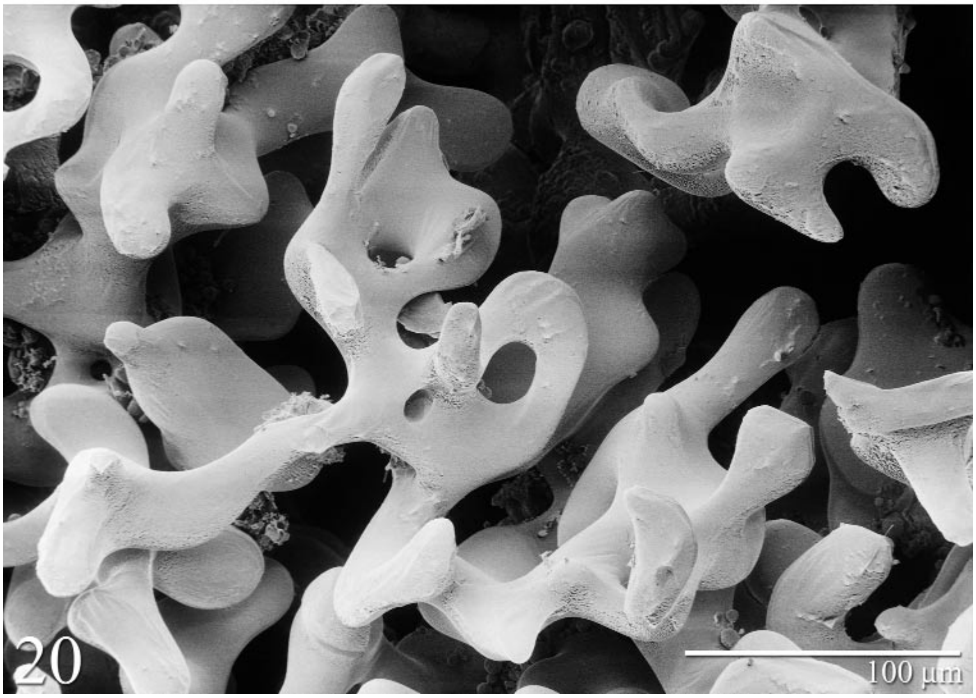


FIG. 19 Stereo pair of micrographs illustrating a large accumulation of frozen cloud droplets that formed a graupel particle. The droplets, which exhibit no crystalline characteristics, are indistinguishable from the early accumulations that are found on heavily rimed crystals. Sample collected near Fraser, Colorado.

FIG. 20 High magnification of droplets from a graupel particle. The "droplets" are well sintered, but considerable air space exists within the graupel particle. Sample collected near Fraser, Colorado.

could result in this configuration. However, when riming continues on many types of snow crystals, the accumulation of the frozen droplets is frequently more pronounced on one surface. This observation was by Nakaya (1954), who described "thick plane crystals" as having frozen cloud droplets on only one surface. Ono (1969) also noted that, for plate crystals, droplets accumulated on one side of the basal plate and suggested that this accretion must have been the front or leading face. We are not able to discern the leading or front face from the trailing face. However, on many of the ice crystals that are more heavily rimed on one side, the accumulated droplets appear to be elongated. We suggest that this appearance could result from the freezing of the droplets on the trailing face brought about by air currents, rather than from droplet impact on the leading face which would tend to result in a more flattened appearance, such as that found on the initial or earliest stages of rime formation. Perhaps the initial droplets impact on the leading face of a crystal and consequently have a flattened appearance. However, their presence could change the aerodynamics and cause the crystal to "flip" $180^0$. As a result, further accumulation of the rime particles would continue on the trailing face and have the elongated appearance that is typical of the heavily rimed crystals observed in this study.

When accretion of the frozen cloud droplets continues, the encumbered mass of frozen droplets obscures the identity of the original snow crystal. These types of particles are referred to as graupel. Barkow (1908) was the first person to recognize this type of crystal. Nakaya (1954) suggested that graupel formed in the lower atmosphere where supercooled water droplets were most abundant. Although the limited resolution and depth of field of the LM did not reveal the true nature of graupel particles, investigators (Hobbs 1974, Nakaya 1957) stated that the density of graupel particles was about 0.125 Mg $m^{-3}$ and suggested that the particles had a very open network. Our study, which allows visualization of the three-dimensional nature of the graupel particle, clearly illustrates the internal structure of graupel that indeed has internal air spaces resulting from the manner in which the frozen droplets freeze to the original snow crystal and subsequently to one another.

## Conclusion

Low-temperature scanning electron microscopy can be used to illustrate and characterize the frozen cloud droplets, commonly known as rime, which adhere and freeze to snow crystals during formation and descent. This technique shows that rime is found on all types of snow crystals including needles, columns, plates, and dendrites. The droplets vary in size from 10 to 100 μm and frequently accumulate along one side of a single snow crystal, but are found more randomly distributed on crystal aggregations, which are commonly known as snowflakes. Continued accumulation of the frozen cloud droplets leads to the formation of graupel, a particle that frequently measures 1 to 3 mm across and consists of hundreds of frozen cloud droplets that completely engulf the original snow crystal. The frozen cloud droplets are initially encountered as flattened hemispheres on the face of a snow crystal. However, as accumulation continues, they appear more sinuous and elongated, suggesting that aerodynamic forces may influence whether the droplets impact and freeze to the leading face of the snow crystal or are attracted to the trailing face of the crystal as it descends through the atmosphere.

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