Deformations of frozen droplets formed at −40°C

María Laura López[1] and Eldo E. Ávila[1]

Received 28 October 2011; revised 2 December 2011; accepted 7 December 2011; published 10 January 2012.

[1] The optical properties of cirrus clouds are frequently computed with respect to various ice-crystal size distributions and shapes; small ice crystals are generally considered to be quasi-spherical. This approximation can lead to significant errors in the analysis of the in-situ microphysical data and also in the theoretical study of the radiative properties of cirrus clouds. Effective size and aspect ratio of the particles are the key parameters required to determine the optical parameters that are included in modeling and prediction of climate; their parameterizations need to be as accurate as possible. In this work, laboratory experiments were conducted to study the ice crystal shape formed from the freezing of water droplets at −40°C. Liquid and ice clouds were separately formed and sampled in a cloud chamber at −30 and −40°C, respectively. The effective water droplet diameter ranged from 8 to 20 μm, while the effective ice particle diameter ranged from 8 to 30 μm. The average effective diameters were 11 μm for water drops and 14 μm for frozen droplets. The deformation of the frozen droplets was evident during inspection under microscope; bulges and spikes protuberances were found in many of the observed ice particles. The results show that the aspect ratio of the frozen droplets is 1.2. **Citation:** López, M. L., and E. E. Ávila (2012), Deformations of frozen droplets formed at −40°C, *Geophys. Res. Lett.*, *39*, L01805, doi:10.1029/2011GL050185.

1. Introduction

[2] Cirrus clouds are widely spread over the globe; they cover about 30% of the Earth's surface while in the tropics this number can increase to about 60% [*Sassen et al.*, 2008]. They can absorb and scatter solar radiation, and absorb and emit infrared (IR) thermal radiation. For these reasons, cirrus clouds influence the radiative energy budget of the Earth and constitute an essential component of the Earth's climate system [*Liou*, 1986; *Lynch et al.*, 2002].

[3] Cirrus clouds frequently appear in the upper troposphere, these clouds mainly consist of non-spherical ice crystals with complicated scattering and absorption features. To understand the net radiative impact of cirrus (combined solar and thermal IR), radiative transfer simulations are required which are often based on microphysical measurements and on the optical properties (scattering and absorption) of the individual ice crystals of the cirrus cloud. These optical quantities depend on the size and shape of the ice crystals, as well as on the wavelength of the incident radiation. It is important to note that the size of ice crystals in cirrus covers a broad range from micrometers to millimeters, with a clear predominance of small particles [*Vogelmann and Ackerman*, 1995].

[4] Deep convection in the tropics generates anvil cirrus that covers vast areas [*Salby et al.*, 1991]. The initial ice crystal number and mass densities are largely determined by the evolution of the freezing drop size distribution during upward transport. The concentrations and sizes of ice crystals in these clouds affect the reflection of incoming solar radiation: for a given ice water content, a cloud with smaller, more numerous crystals will have a higher albedo. In addition, deep convective clouds are associated with intense updrafts, growth of particles in the solid phase and lightning production. Based on experimental evidence, *Ávila et al.* [2011] suggested that the charging mechanism associated with graupel–frozen droplet collision may be relevant in clouds in which internal temperatures are substantially lower than −37°C and could be the main generator or high-altitude lightning.

[5] Research aircraft have practically been unable to reach the tops of tropical anvil cirrus that can extend up to 18 km. The inaccessibility of anvil cirrus has restricted the sampling of their microphysical composition. Therefore, it is difficult to investigate ice nucleation mechanisms in cirrus, since the conditions when sampled crystals are produced may not be known and instrumentation to measure crystal shapes, particle size distributions and relative humidity may be insufficient [*Lynch et al.*, 2002]. In short, it is currently not possible to characterize these small ice particles using in situ measurements, but it is possible to do this under laboratory conditions.

[6] Micrometer-size cloud water droplets have been detected in situ at temperatures down to about −37°C [*Sassen and Dodd*, 1988; *Rosenfeld and Woodley*, 2000; *Heymsfield et al.*, 2005], where ice nucleation evidently does not take place heterogeneously on immersion or contact of ice nuclei (IN), suggesting that such nuclei are scarce in the upper troposphere. As temperature decreases below −37°C, the droplets freeze spontaneously without the need for IN. Ice particle concentrations that exceed the number densities of ice nuclei available for heterogeneous nucleation have frequently been reported, providing evidence for the importance of homogeneous nucleation [*Kuhn et al.*, 2011, and references therein].

[7] The droplets in clouds usually form on soluble cloud condensation nuclei (CCN), which depress the freezing point of the resulting solution relative to pure water. The temperature to which liquid water can supercool has been a subject of academic interest over the past years. Several studies have demonstrated that the probability of homogeneous freezing depends on droplet size and that micron-size dilute solution droplets can supercool to about −38°C before freezing [*Schaefer*, 1962; *DeMott and Rogers*, 1990].

[1]FaMAF, Universidad Nacional de Córdoba, IFEG-CONICET, Ciudad Universitaria, Córdoba, Argentina.

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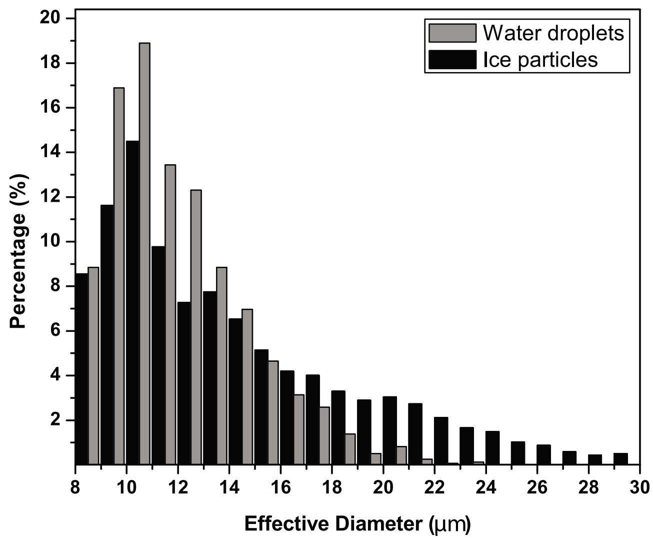

Figure 1. Water droplet and ice particle size spectra.

[8] The small ice crystals located at cloud tops are often regarded as quasi-spherical. As the optical properties of cirrus clouds are frequently computed with respect to various size distributions and ice crystal shapes [*Baran*, 2009], this approximation can lead to significant errors in the analysis of the in-situ microphysical data and also in the theoretical study of the radiative properties of cirrus clouds [*Mischenko et al.*, 1996; *Yang et al.*, 2001; *Um and McFarquhar*, 2011]. As the asymmetry factor for cirrus clouds is one of the most important optical parameters required in modeling and prediction of climate, its parameterization needs to be as accurate as possible [*Fu*, 2007]. This parameterization in terms of the effective size is very sensitive to the habit and shapes of ice crystals adopted for the light scattering calculations [*Edwards et al.*, 2007]. Reports have suggested that the shape-sensitive parameters are an important feature to be considered in order to deduce more accurate microphysical and optical properties of ice clouds. For example, *Fu* [2007] found that the asymmetry factor is very sensitive to the particle aspect ratio.

[9] As previously stated, homogeneous freezing of cloud droplets is a very important mechanism of ice formation at temperature less than −37°C [*Rosenfeld and Woodley*, 2000]. The relevance of the knowledge of the shape and realistic morphology of the small ice crystals for the calculation of parameters crucially affecting the overall radiative effects of cirrus has also been pointed out. Thus, laboratory experiments were conducted to examine the shape of the ice crystals formed when micrometer-sized water droplets freeze at −40°C, resembling the conditions that commonly occur in anvil cirrus clouds.

[10] The shapes of ice crystals and the changes in the shapes of water drops during freezing have been reported in many works for a wide range of size particles and temperatures. These studies analyzed the physical parameters which affect the freezing behavior and fragmentation of water drops. They report that droplets nucleated at equilibrium in ordinary air at atmospheric pressure formed spikes and protrusions during freezing [*Dye and Hobbs*, 1968; *Johnson and Hallett*, 1968; *Takahashi*, 1975, 1976; *Wood et al.*, 2002]. All these previous studies have examined water droplets larger than 50 μm in diameter. To our best knowledge, this is the first laboratory work studying the representation of nonspherical ice crystals smaller than 30 μm in diameter.

2. Experimental Device and Measurements

[11] Based on the fact that the freezing process in water droplets drastically increases for temperatures lower than −35°C and that theoretical and experimental results have shown that the droplets freeze spontaneously without the need for IN for temperatures below −38°C, liquid and ice clouds were separately formed and sampled in a cloud chamber in the laboratory at −30 and −40°C, respectively.

[12] The cloud chamber is a box 170 L of capacity. The temperature control was carried out with two thermocouples placed on the wall and the center of the chamber. As the difference in both measurements was less than 0.2°C throughout the whole experiment, the temperature was considered to be homogeneous.

[13] Water droplets were generated from distilled water by using an ultrasonic atomizer; these droplets were introduced into the cloud chamber. The particles produced in each experiment were collected on microscope slides previously covered with a thin layer of a 3% w/w Formvar solution in chloroform [*Schaefer*, 1956]. The collection was made by pumping out a small air volume from the cloud chamber into the coated slide; special care was taken to prevent the introduction of external air masses. After chloroform and water were evaporated, the plastic replicas of the particles were analyzed at ambient temperature. For each Formvar replica, the particles (water droplets or ice crystals) were observed under a microscope and digital images were acquired. Each image was processed with the ImageJ software. Shape identification and particle-size distribution were obtained using this software. The shape descriptors calculated in this work were the effective diameter (D), the aspect ratio (AR) and the circularity (C) defined as:

$$D = 2.\left(\sqrt{\frac{Area}{\pi}}\right)$$

$$AR = \frac{Mayor.axis}{Minor.axis}$$

$$C = \frac{4\pi Area}{(Perimeter)^2}$$

The aspect ratio and the circularity are shape-sensitive parameters that allow examining the deformation of the frozen drops. A circularity value equal to 1.0 indicates a perfect circle. As the value approaches to 0.0, it indicates an increasingly elongated shape.

[14] At both temperatures, the experiments were carried out three times to ensure the repeatability of the results. During both experiments, the total sampling allowed to quantify 5000 and 3000 particles at −40 to −30°C, respectively.

[15] The water droplets' effective diameter ranged from 8 to 20 μm, while the ice crystals' effective diameter ranged from 8 to 30 μm. The average effective diameters and the standard deviation were 11 μm and 3 μm for water drops and 14 μm and 5 μm for ice crystals, respectively. Figure 1 displays the histograms of the droplet size distribution for

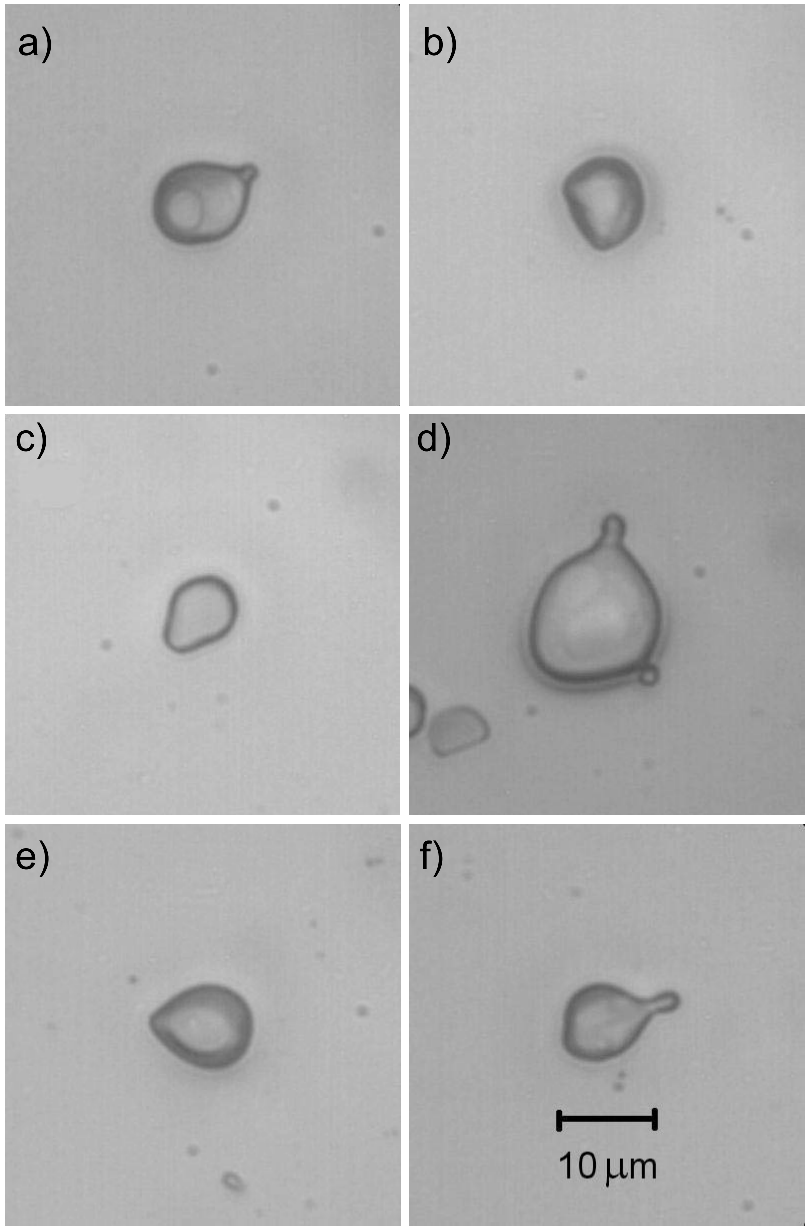

Figure 2. (a–f) Images of collected ice particles.

both temperatures. Using aircraft measurements, *Rosenfeld et al.* [2006] reported the microphysical evolution of severe convective storms from the cloud base to the −45°C isotherm level. The authors measured significant values of liquid water content up to just below the level of the −40°C and then an abrupt decrease indicating that most of the condensed water has frozen at higher levels. The droplet size distribution showed particles with sizes around 20 μm, supporting the idea that the particles used in this study are representative of those found in real clouds.

[16] During both experiments (−40 and −30°C), the evolution of the number of particles was recorded with a video camera. A lamp was used to enable the visualization of the particles. The videos obtained were processed with the ImageJ software. The analyses of the corresponding frames allowed the quantification of particle concentration in the cloud chamber. The measured concentrations were $(6 \pm 1)10^3$ particles L^{-1}, which corresponds to a liquid water content ~0.01 g m^{-3}.These auxiliary experiments also showed that the ice particles completely disappear from the cloud chamber (sublimation process) after 4 minutes of introducing the water droplets into the chamber, indicating that the ice crystals were not growing by vapor deposition after being formed. This result allows us to infer that the shapes of the ice crystals (originated by the freezing process of the droplets) have not been substantially modified after freezing.

3. Results and Discussion

[17] The deformation of the frozen droplets at −40°C was evident during inspection under microscope. As an example, Figure 2 shows the photographs of typical shapes of the small ice particles; most of these particles exhibit protuberances. *Takahashi* [1975] classified these types of deformations as bulge and spike. The former is defined as a protuberance shorter than one fourth of the drop diameter, while the latter

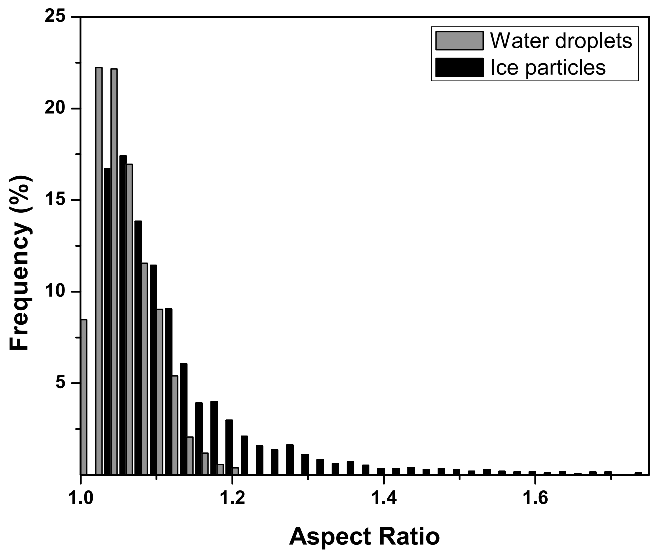

Figure 3. Histograms for the aspect ratio calculated for water droplets and ice particles.

is defined as a protuberance longer than one fourth of the drop diameter.

[18] Spikes and bulges were not observed in the cloud droplets sampled at −30°C, but they were the main kinds of deformation observed in the experiments performed at −40°C. Examples of bulges are shown in Figures 2a and 2b, the aspect ratios being 1.25 and 1.16, respectively. Examples of spikes are shown in Figures 2c–2f. The aspect ratios of these ice crystals are 1.32, 1.32, 1.33 and 1.58, respectively. Note that this last value indicates that the length of the protrusion can be the same (or even larger) than the effective radius of the particle. *Takahashi* [1975] explained the formation of bulges because of the expansion of the ice shell. The air bubbles which are found in a bulge indicate that unfrozen water is left in a bulge until the last stage of freezing as a result of the expansion of the ice shell. On the other hand, he explained that a spike is formed by the subsequent freezing of water pushed out of the ice shell through a small crack. This justification was made by *Takahashi* [1975] to explain the protrusions observed on freely falling water drops of diameters between 45 and 765 μm at temperatures ranging from −5 to −25°C. The same explanation seems plausible to understand the deformations found in the current work.

[19] In order to quantify the deformations of the frozen droplets, the aspect ratio of the particles was analyzed. Figure 3 shows the normalized frequency distribution of the aspect ratio for the experiments at −30 and −40°C. The differences between the aspect ratio of water droplets and ice crystals can be clearly observed.

[20] The aspect ratios of the droplets are mostly close to one and no higher than 1.2. The average and standard deviation values of the aspect ratio were 1.06 and 0.04. Considerations regarding surface tension allow assuming that, these small water droplets are spherical particles. It is plausible to assume that the deviation of the sampled droplets from the spherical shape observed in the current experiments can be a consequence of the freezing mechanism during the sampling process. Moreover, it is important to point out that the Formvar-coated slide was at the same temperature as the cloud chamber during the sampling process. Thus, we can assume that the aspect ratio of the water droplets is one.

[21] Higher aspect ratio values can be observed for ice particles than for water droplets. The average and standard deviation values of the aspect ratio for the ice particles were 1.2 and 0.2, respectively. A statistical analysis shows that there are significant differences in the aspect ratio between both experiments (significance level = 0.05).

[22] Figure 3 shows a general analysis of the aspect ratio regardless of effective diameter. To study the effective diameter dependence, all the aspect ratio values were grouped in effective diameter intervals of 1 μm. This procedure was applied to all data from the two experiments. Figure 4 shows the aspect ratio average and standard deviation as a function of the effective diameter for water and frozen droplets. The results confirm that the aspect ratios of the frozen droplets are larger than the aspect ratios of the unfrozen droplets for all the effective diameters measured.

[23] The results suggest that average aspect ratio of the frozen droplets is independent from the effective diameter, since it does not show a significant trend when the effective diameter is increased. This result is the same for both temperatures. While the statistical analysis showed that there is a significant difference between the two experiments, no significant differences were detected between the different effective diameters in each experiment (significance level = 0.05). For the effective diameter range studied in this work, deformations remain approximately constant as the droplet size increases. This result seems to contradict the results reported by *Takahashi* [1975] who showed that the deformation of frozen droplets in free fall depends on the droplet size for droplet diameters between 45 and 765 μm. However, it is important to note that the measurements in both experiments were performed in a different range of droplet sizes; thus, these discrepancies could be explained because of differences in the range of the diameter of the frozen droplets. Considering that surface tension is inversely proportional to the radius, then it is reasonable to expect that smaller droplets suffer less distortion than larger ones.

[24] The increment in the aspect ratio value of the frozen droplets is correlated to the appearance of protuberances.

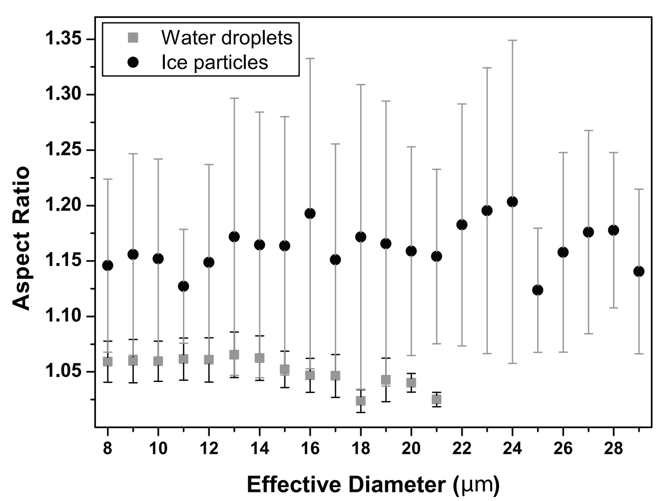

Figure 4. Average aspect ratio and error bars as a function of effective diameter, for water droplets and ice particles.

However, the increment in aspect ratio does not imply the loss of circularity of the particles. For example, circularity values of the particles in Figures 2b, 2c, and 2e are 0.96, 0.95 and 0.96, respectively; even though they have a considerable elongation (aspect ratio values higher than 1.6). In early studies of drop shattering during freezing, *Dye and Hobbs* [1968] and *Takahashi* [1975] observed the ejection of splinters from droplets of radii 50–1000 μm as they froze in free-fall over a range of temperatures from −5 to −30°C. In the current work we found no evidence of shattered particles.

[25] *Zhang et al.* [1999] examined the effect of the morphology of the ice crystals on the cirrus radiative forcing. They found that the solar radiative forcing of cirrus cloud is reduced by non-spherical ice crystals, due to larger albedo effects of non-spherical crystals compared to those of equivalent spherical crystals. Furthermore, the cloud radiative forcing (CRF), solar albedo and IR emittance are changed significantly as the mean crystal size approaches the smaller size end (<30 μm). The net CRF is positive for cirrus clouds containing relatively large ice particles. However, for thin clouds containing small particles, the values of CRF would be negative, which means that this kind of cirrus can produce a cooling effect in the Earth-atmosphere.

4. Conclusions

[26] Laboratory experiments were performed in order to characterize the shape of the particles formed when micrometer-sized water droplets freeze at −40°C. These results will contribute to improve the estimation of the scattering properties of cirrus clouds. Considering that some of the new algorithms use the aspect ratio as a variable to estimate the optical parameters of clouds, the quantifying of this shape-parameter is important for the understanding of the processes in real atmospheric conditions. Accurate determination of ice crystal shape is very important for the calculation of parameters crucially affecting the overall radiative effects of cirrus.

[27] In clouds consisting of supercooled droplets, the non-spherical frozen droplets are potentially important because they will grow by vapor deposition and eventually collect supercooled water droplets to form rime ice. The capture and subsequent freezing of supercooled cloud droplets is a very efficient mechanism of ice growth which leads to the production of precipitation in the form of rainfall or snowfall.

[28] **Acknowledgments.** This work was supported by Secretaria de Ciencia y Tecnología de la Universidad Nacional de Córdoba, Consejo Nacional de Investigaciones Científicas y Tecnológicas (CONICET), and Agencia Nacional de Promoción Científica (FONCYT). We want to thank Jose Barcelona for his technical assistance.

[29] The Editor thanks the two anonymous reviewers for their assistance in evaluating this paper.

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E. E. Ávila and M. L. López, FaMAF, Universidad Nacional de Córdoba, IFEG-CONICET, Ciudad Universitaria, 5000 Córdoba, Argentina. (avila@famaf.unc.edu.ar)