Charge separation in collisions between ice crystals and a spherical simulated graupel of cm-size

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Abstract

This work reports a new laboratory study of the electric charge separated in collisions between a spherical target of 1 cm in diameter growing by riming and vapor-grown ice crystals, with the objective of studying the charging behavior of the larger ice precipitation particles in thunderstorms in terms of the non–inductive mechanism. A series of experiments was conducted for a wide range of environmental conditions; the measurements were performed for effective liquid water content between 0.5 and 5 g m^{-3}, for ambient temperatures between –5 °C and –30 °C and at air speed of 11 m s^{-1}. The magnitude and sign of the electric charge transfer on the ice sphere as a function of the ambient temperature and the effective liquid water content is presented. The results show a charge reversal temperature for the riming target which is roughly independent of liquid water concentration in the measured range. The simulated graupel charges negatively for temperatures below –15 °C, and positively at temperatures above –15 ºC.

1. Introduction

Laboratory studies with all kinds of variations of the environmental parameters have been carried out to study the non-inductive electrification mechanism. The results of these studies reveal that parameters such as ambient temperature (T_a), liquid water content (LWC) or effective liquid water content (EW), impact velocity (V), droplet and ice crystal size distributions and the difference in saturation vapor pressures over ice and liquid water may affect sign and magnitude of the charge acquired by an ice precipitation particle when colliding with ice-crystals [Takahashi, 1978; Jayaratne et al., 1983; Saunders et al., 1991, 1999, 2001, 2004, 2006; Ávila et al., 1996; Saunders and Peck, 1998; Ávila and Pereyra, 2000; Pereyra et al., 2000; Ávila et al., 2013; Luque et al., 2016, 2018].

The charging current (CC) that a simulated graupel acquires under collisions with ice crystals, has been quantified in the laboratory. Typically, the simulated graupel consists of metallic collectors over which supercooled water droplets hit and freeze, forming an ice cover that represents the surface characteristic of natural graupels (rime). Dimensions of the simulated graupel vary from study to study but they usually are around the millimeter-size.

Takahashi [1978] rotated a rimed rod of 3 mm in diameter at 9 m s^{-1} through a cloud of ice crystals and supercooled water droplets and measured the CC of the simulated graupel. He observed that the target may charge negatively or positively depending on the LWC and the ambient temperature. He found that for temperatures above -10 ºC, the CC was positive for all LWC values. For temperatures below -10 ºC, the sign of CC showed a dependence with the value of LWC, being negative for 0.1 g m^{-3} < LWC < 4 g m^{-3}, and positive for lower (< 0.1 g m^{-3}) and higher LWC values (> 4 g m^{-3}) regardless of the ambient temperature.

Jayaratne et al. [1983] rotated a rimed rod of 5 mm in diameter at 3 m s^{-1} and found significant dependencies of the CC magnitude with the simulated graupel temperature, ice crystal sizes and the impact velocity. Unlike Takahashi´s results, they found a single charge reversal temperature (T_R) as a function of LWC. The rime became positively charged at temperatures above T_R and high LWC, and negatively charged at lower LWC or at lower temperatures. The T_R value was around -20 °C at LWC = 1 g m^{-3}.

Saunders et al. [1991] measured the CC of a fixed rod of 5 mm in diameter growing by accretion while it was subjected to collisions with ice crystals in a series of experiments under controlled conditions at an impact velocity of 3 m s^{-1}. They introduced the variable effective liquid water content (EW) in the place of LWC, EW is the product of LWC and collision efficiency, therefore, is made up only of those droplets that can be captured by the rod. Thus, this parameter takes into account the part of the water droplets involved in the riming process. The EW was varied while the size of the crystals and the impact velocity were kept constant. This work extended previous measurements by Jayaratne et al. [1983] and modified the charging diagram (LWC-T) with more than one charge reversal temperature for each LWC.

Saunders et al. [1991], Takahashi [1978] and Jayaratne et al. [1983] observed that the sign of the charge acquired by the simulated graupel mainly depends on the liquid water content and T. However, their results were different. There were attempts to explain their discrepancies by recalling that the impact velocities and simulated graupel sizes used were different, but it is apparent that they could not be the only cause. The differences between the results are still under debate [Takahashi et al. 2017].

Pereyra et al. [2000] carried out measurements of the charge transfer when vapor grown ice crystals rebound from a riming target representing a graupel pellet falling in a thunderstorm. Measurements were made at temperatures between -5°C and -30°C, at EW values between 0 and 4 g m-3 and for an impact velocity of 8.5 m s^{-1}. Basically, they found positive CC at T > -17 ºC and negative CC at T < -17 ºC.

Ávila and Pereyra [2000] reported measurements using a rod of 4 mm in diameter and found that the cloud droplet spectrum can affect the EW-T charge diagram of the simulated graupel. They found that small droplets promote a predominance of positive CC on the simulated graupel. Bürgesser et al. [2006] used the same experimental device and studied the influence of the impact velocity on the reversal temperature. They found a shift of the reversal temperature T_R to warmer temperatures when the impact velocity was increased.

Ávila et al. [2013] modified the previous target geometry and carried out their measurements using as simulated graupel a network constituted by brass wires of 2 mm in diameter. They measured the CC for lower values of EW representing the microphysical conditions typically found in the stratiform regions of mesoscale convective systems (MCS). The results found on this study showed a dependence of the sign of the charge acquired by the simulated graupel with the impact velocity. Luque et al. [2016] also used the network but in that case, they measured CC in absence of supercooled water droplets and found a prevalence of negative charge transfer.

Recently, Jayaratne and Saunders [2016] found substantial positive CC when ice crystals impacted with a rod of 4 mm in diameter under wet growth conditions. Luque et al. [2018]

corroborated the results from Jayaratne and Saunders [2016] by working with a spherical simulated graupel of 1 cm in diameter.

Baker et al. [1987] suggested that the sign of the CC acquired by the simulated graupel, when ice crystals collide with it, is related to the diffusional growth rates of the ice particles involved in the collision. They proposed that the ice particle growing faster by vapor diffusion will acquire positive charge while the other particle acquires negative charge. This empirical observation was supported for many subsequent experimental results [Emersic and Saunders, 2010]. On the other hand, Williams et al. [1991] proposed that simulated graupel being warmed by riming will charge negatively while a simulated graupel that is not sufficiently warmed by riming will grow by diffusion and charge positively. Both hypotheses stressed the importance of the microphysics of the mixed phase clouds to influence the surface state of the interacting particles. Small changes in the atmospheric conditions or in the microphysical parameters may affect the surface states of the ice particles and consequently modify the CC sign and magnitude during the interactions among them.

As previously described, in most of the former experimental studies, mm-size graupels of cylindrical geometry were used. These sizes are surely representative of most of the ice precipitation particles found in thunderstorms. However, it is well documented that graupel pellets and hailstones of cm-size can also be present in significant concentrations within convective regions of mesoscale convective systems and within severe and ordinary thunderstorms as well; these large ice particles could play a key role in the cloud electrification processes [Williams, 2001]. There is certainly a need for more laboratory studies of the charge separation during collisions between ice precipitation particles and ice crystals. Such studies are needed, for example, to determine the charging behavior of larger ice precipitation particles in thunderstorms in terms of the non–inductive mechanism. Thus, in the present study, the sign and magnitude of the average charge separated during ice crystal-graupel collisions have been measured for a simulated graupel of 1 cm in diameter, ambient temperatures between –5 °C and –30 °C, effective liquid water content between 0.5 and 5 g m^{-3} and impact velocities of 11 m s^{-1}.

2. Experimental Setup

The measurements of the charging current were carried out inside a cold chamber using the experimental setup displayed in Figure 1. The arrangement and the experimental procedure are the same as those used by Luque et al. [2018].

Airflow velocity inside the wind tunnel was controlled with an air pump. Measurements were performed at an impact velocity V = (11 ± 1) m s^{-1}, which is representative of the fall speed of cm-size graupel pellets inside thunderstorms [Heymsfield and Kajikawa, 1987].

Three temperatures were recorded during the experiment: the ICC temperature (T_{ICC}), the wind tunnel temperature (T_{WT}) and the target temperature (T_T). These values were sensed using three previously calibrated thermistors. The location of the thermistors can be seen in Figure 1. The ICC and target temperatures were recorded only for control and were not used in the data analysis. The wind tunnel temperature was considered as the ambient temperature for each experiment, and its variation during the runs was typically less than 1°C.

A brass sphere target was connected to a current amplifier capable of detecting currents above 1 pA. The current amplifier output was recorded using an A/D converter.

Plastic replicas of ice particles generated in the ICC and cloud particles generated in the cloud chamber were taken. Clouds were swept with glass slides previously covered with a 3% Formvar solution following the sampling procedure based on Schaefer [1956]. After analyzing the samples, size distributions for ice crystals and supercooled droplets were determined.

The size distributions of the ice crystals used in the experiments at different temperatures are displayed in Figure 2. The average size of the crystals varied with T; in fact, at –10 °C the average size is 15 μm, at –13 °C the average size is 26 μm, at –17 °C the average size is 32 μm, at –21 °C the average size is 26 μm, at –26 °C the average size is 19 μm and at –30 °C the average size is 14 μm. Most of the sampled ice crystals were hexagonal plates with sizes up to 60 μm, broad branches ice crystals were observed at –13 °C and –17 °C with sizes up to 80 μm.

The diameter distributions of the cloud droplets used in the experiments for temperatures between –8 °C and –26 °C are displayed in Figure 3. Unlike the ice crystal sizes, droplet sizes do not show significant variations with temperature. The median diameters of the droplets ranges between 14 μm and 17 μm. Ávila and Pereyra [2000] showed that the size distribution of the droplets used for riming could affect the sign of the charge transfer to the target. They used two size distributions A (larger droplets) and B (smaller droplets) to make their point; and found that larger droplets promote a predominance of negative CC on the target. In the current work, the laboratory setup used to generate the cloud droplets was the same as those used by Ávila and Pereyra [2000] to generate the larger size distribution (spectrum A), as well as the droplet size distribution subsequently used by Pereyra et al. [2000], Bürgerser et al. [2006], Ávila et al. [2013], Luque et al. [2016] and Luque et al. [2018].

The measurements were performed following the next steps:

1. Cold chamber temperature was fixed.
2. Reservoir water temperature was heated up to the desired temperature.
3. Distilled water droplets were introduced in the ICC for 3 minutes.
4. Ice crystals were nucleated and they grew by vapor deposition for 60s.
5. Water reservoir tap was opened to initiate supercooled droplets cloud formation. After 10 seconds, the pump was turned on initiating the airflow inside the wind tunnel. This airflow drags both clouds from their respective chambers and while the target is growing by riming, it also collides with the ice crystals.

Measurements lasted between 50 and 200 seconds. Complementary experiments were performed to corroborate that ice crystal concentration remained constant during measurement time.

3. Results and Discussions

The measurements were performed for ambient temperatures (T) between –5 ^{o}C and –30 ^{o}C and for an impact velocity (V) of 11 m s^{-1}. The effective liquid water content value (EW) was determined weighing the accreted mass (ΔM) on the target after each experiment. The accreted mass relates to the effective liquid water content through the equation:

$$\Delta M = EW\ V\ A\ \Delta t \quad (1)$$

where A is cross sectional area of a sphere and Δt is experiment time. The values of EW in this study ranged between 0.5 and 5 g m^{-3} and the uncertainty in the measured EW was typically about 30%.

Figure 4 displays the sign of the charging current (CC) of the target as a function of the effective liquid water content and the ambient temperature. Open circles indicate T-EW combinations for which the simulated graupel is charged positively, and filled circles indicate negative charging of the simulated graupel. The solid line in the lower region of the figure represents the transition between sublimation and deposition of the simulated graupel surface set up by the difference in saturation vapor pressures over ice and liquid water. The transition between sublimation and deposition zones is determined from Macklin and Payne [1967] and from the Clausius-Clapeyron equation taking into account that the simulated graupel sublimates if its surface temperature becomes higher than the ambient temperature and if the equilibrium vapor pressure of ice at the simulated graupel surface temperature falls below the equilibrium vapor pressure of the environment at ambient temperature. The dot-dashed line belongs to the transition between wet and dry growth of the simulated graupel. The wet growth regimes were calculated from Macklin and Payne [1967], assuming that wet growth is reached when the surface temperature is 0 ºC. It is possible to observe that the measurements were mainly performed under the dry growth regime of the simulated graupel, which was corroborated from direct observations of the surface of the simulated graupel after each measurement.

Figure 4 shows almost entirely negative CC at temperatures lower than –15 ºC and positive CC at warmer temperatures. It suggests that under the studied conditions there is a sort of charge reversal temperature for the simulated graupel (T_R= –15 ºC) which is roughly independent of the liquid water content. The results indicate that precipitation ice particles of cm-sizes, under impacts with ice crystals at 11 m s^{-1}, charge positively at temperatures above –15 ºC, and negatively at temperatures below –15 ºC. These results are valid for 0.5 g m^{-3}< EW < 5 g m^{-3} and under all these conditions the simulated graupel was mainly sublimating and in the dry growth regime. These results could be no longer valid for EW < 0.2 g m^{-3} since some researchers reported that the graupel charges positively for EW < 0.2 and T < -15C [Takahashi, 1978; Saunders et al., 1991; Ávila and Pereyra, 2000; Bürgeser et al., 2006]. The range EW < 0.2 was unexplored in the current study.

Williams et al. [1991] proposed that a simulated graupel growing by riming and sublimating simultaneously, as in the case of the current experiments, charges negatively. This hypothesis is consistent with the results at T < –15 ºC, but inconsistent with the results at T > –15 ºC. This inconsistency was already observed in most of the previous results [Takahashi, 1978; Saunders et al., 1991; Pereyra et al., 2000; Saunders et al., 2006]. On the other hand, Baker et al. [1987] proposed that the ice-particle growing faster by vapor deposition will charge positively while the other will remain negative. In the sublimation regime, the ice-particle which sublimates slower will acquire positive charge. In the case of the present work, the ice crystals were growing by vapor deposition during the experiments because they were surrounded by supercooled water droplets (Bergeron´s mechanism); while the simulated graupel was losing mass by sublimation but gaining mass due to its growth by riming. At cold temperatures, sublimation prevails over accretion so, according to the Baker's hypothesis, the simulated graupel acquires negative charge. However, as ambient temperature increases, the freezing time of the accreted water droplets increases, generating extra vapor sources onto the simulated graupel surface which enhances its diffusional growth rate. At some point, the simulated graupel stops sublimating and its diffusion growth rate exceeds the ice crystal growth rate causing a change in the CC sign from negative to positive. Although this hypothesis is plausible and consistent with the current results, it still needs to be checked with more experimental evidence.

Regarding that the CC sign of the simulated graupel may depend on the impact velocity of the ice crystals, we compare the charge diagram obtained in the current work with those obtained by Takahashi [1978], Pereyra et al. [2000] and Bürgesser et al. [2006] who worked with velocities > 8 m s^{-1}. Takahashi [1978] rotated a simulated graupel of 3 mm in diameter at 9 m s^{-1} through an environment of supercooled water droplets and ice crystals and measured the CC of the target. He found that for T > –10 ºC, the CC was positive independently of LWC values. For T < –15 ºC and 0.1 g m^{-3}< LWC <4 g m^{-3}, the sign of CC was negative. For –10 ºC< T < –15 ºC, the sign of the CC depended on LWC being positive for lower and higher LWC values and negative for intermediate LWC. For instance, at T = –12 ºC the CC was positive at LWC < 0.7 g m^{-3} and LWC > 3 g m^{-3}, and negative for 0.7 g m^{-3}< LWC < 3 g m^{-3}. Beyond the behavior of the charge reversal temperature in the range –10 ºC< T_R < –15 ºC, the results from Takahashi [1978] looks qualitatively similar to the present results with a region of positive charging of the simulated graupel at warmer temperature (T > –10 ºC) and a region of negative charging of the simulated graupel at colder temperatures (T < –15 ºC). Pereyra et al. [2000] measured the CC for a target of 4 mm in diameter. The measurements were made for temperatures between -5°C and -30°C, at EW values between 0 and 4 g m^{-3} and for an impact velocity of 8.5 m s^{-1}. Their results show positive CC at T > –17 ºC and negative CC at T < –17 ºC. Bürgesser et al. [2006] measured the sign of the CC for an artificial graupel of 4 mm in diameter during collisions with ice crystals at 11 m s^{-1}. The ambient temperature was varied in the range -5 ºC< T < -30 ºC and EW between

0 and 2 g m^{-3}. They found a charge reversal temperature T_R = –15 ºC, independent of EW; the same reversal temperature occurs on both a 4 mm riming rod and a 1 cm sphere (current results).

Considering the similarity between the charge diagrams found by Takahashi [1978], Pereyra et al. [2000] and Bürgesser et al. [2006] and the current results, it is plausible to assume that ice precipitation particles of mm-sizes and cm-sizes are charged in similar ways during collisions with ice crystals inside thunderstorms. Most investigators currently think that the sign of the charge transfer is dependent upon the nature of the colliding surfaces [e.g. Baker et al., 1987; Williams et al., 1991; Saunders et al., 2006 and others]. The charge separation during collisions depends on the local surface state of the interacting particles. The results seem to suggest there is no apparent size effect on the local surface state for graupel of mm or cm sizes. The fact that there is no apparent size effect makes modeling simpler and narrows the parameter space for future researchers.

The magnitude of the average charge transferred to the target per individual ice crystal collision (q) can be estimated by using the equation [Ávila et al, 2013]:

$$q = \frac{CC}{pE_C NVA}$$

Here, V is the impact velocity, A is the target cross-sectional area, and p is the probability that an ice crystal collides and bounces off the target. N is the ice crystal concentration and Ec is the collision efficiency of the spherical target for the ice crystals. Assuming that Ec of the target is the same as the glass strips used as sampler, then the factor $Ec \times N$ can be calculated from the formvar samples. Regarding the substantial dispersion of the measurements, $Ec \times N$ was estimated as (3±2) crystals per cm^3. Furthermore, in order to calculate the average charge transferred to the target, for simplicity and due to the lack of more detailed information, it was assumed p=1.

It was observed that the magnitude of the CC was different for the different charging regimes (positive and negative), for this reason q was estimated separately in each regime. The positive CC magnitude ranged between 1 and 14 pA. With the above assumed values for parameters in the equation, the corresponding values of positive charge transferred per rebounding collision varied between 1 and 6 fC. The negative CC magnitude varied between 1 and 7 pA; therefore, q can be estimated between –1 and –3 fC. These values are smaller than those from Takahashi [1978], where q values were reported between 15 and 30 fC in the positive region and between –3 and –15 fC in the negative region. It is important to note that the ice crystals can stick the target after collisions, at least at some temperatures; which suggests that actually $p < 1$. Smaller values for p would increase the estimate of charge per rebounding collision and might make the values much closer to the values cited for Takahashi [1978]. However, due to the lack of information about this coefficient, it is not possible to make a better estimation of q.

4. Summary and Conclusion

New experimental measurements of the charge transfer during collisions between a spherical target of 1 cm in diameter growing by riming and ice crystals are reported in this work. The measurements were performed for ambient temperatures between –5 °C and –30 °C, for an impact velocity of 11 m s^{-1} and for effective liquid water content between 0.5 and 5 g m^{-3}, with the goal of studying the charging behavior of ice precipitation particles of cm-sizes under microphysical conditions like those found in updrafts of deep convective clouds.

A charge diagram of the sign of the electric charge transfer on the ice sphere as a function of the effective liquid water content and the ambient temperature was obtained. Under the studied conditions the target was mainly sublimating and under the dry growth regime. The results indicate that precipitation ice particles of cm-sizes, under impacts with ice crystals at 11 m s^{-1}, are charged positively for temperatures above -15 ºC, and negatively for temperatures below –15 ºC. It suggests that under the studied conditions there is a sort of charge reversal temperature for the simulated graupel (T_R= –15 ºC) which is roughly independent of the effective liquid water content in the range of 0.5 and 5 g m^{-3}. Under some simplifying assumptions, a rough estimate of the average charge transferred per collision is between 1 and 6 fC in the positive charging regime and between –1 and –3 fC in the negative charging regime.

Finally, we can conclude this study extends our understanding to the larger size extreme of hydrometeors and it is useful to know how trends that have been found at smaller sizes translate to the larger sizes.

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Figure captions

Figure 1. Sketch of the experimental chamber used for laboratory studies. Droplets are produced in the cloud chamber and drawn into the target receptacle by the flow from the pump. Ice crystal produced in the adjoining chamber are inserted in the airflow just above the target.

Figure 2. Ice crystal size distributions for different ambient temperatures. The total number of particles (N) and the mean diameter (dm) are included in each histogram.

Figure 3. Droplet diameter distributions for different ambient temperatures. The total number of particles (N), the mean diameter (dm), the mean volume diameter (dv) and me median diameter are included in each histogram.

Figure 4. Experimental points of the rimer charge sign as a function of EW and T at 11 m s^{-1}. Filled circles indicate negative charging of the rimer and open circles belong to positive results.

Figure 1.

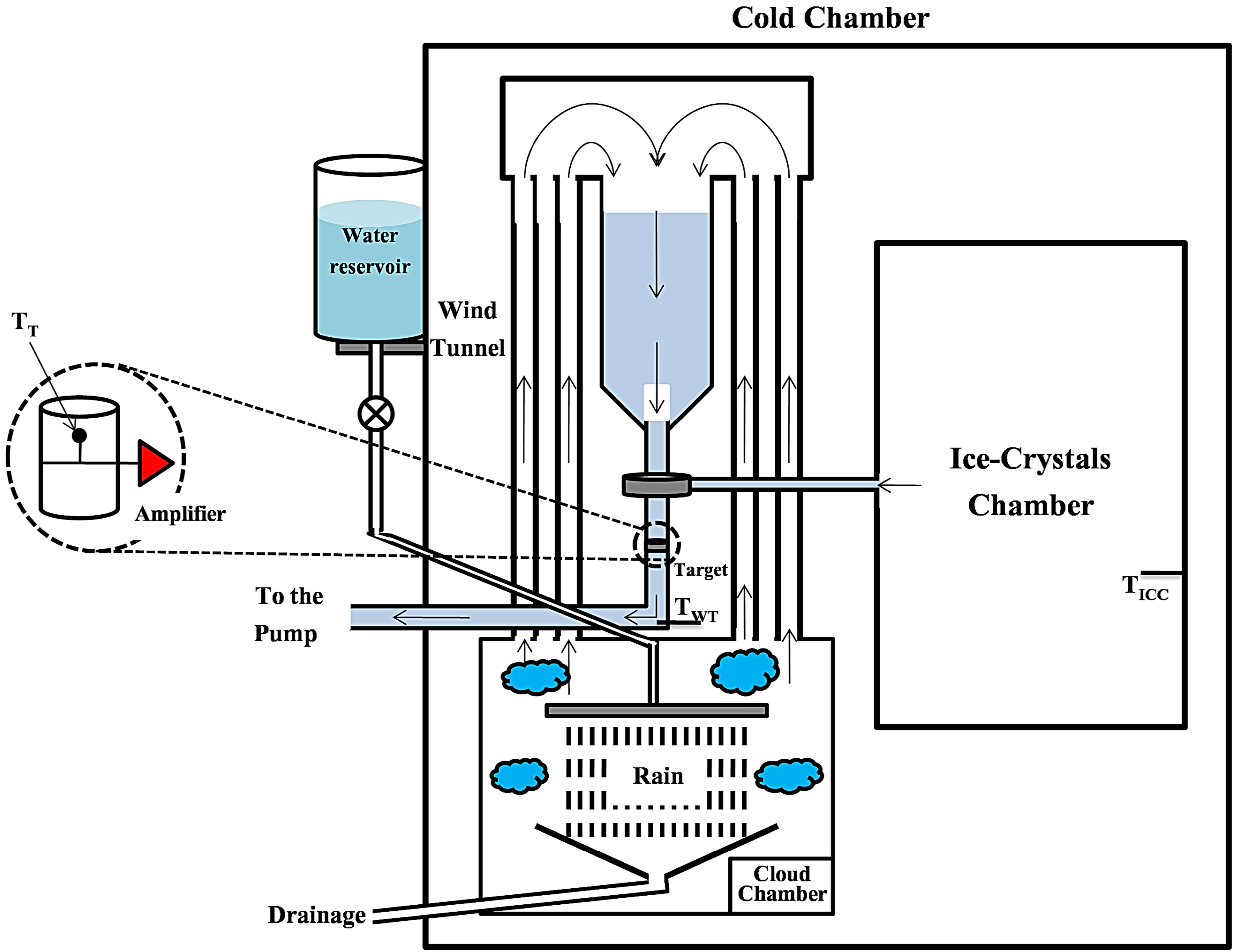
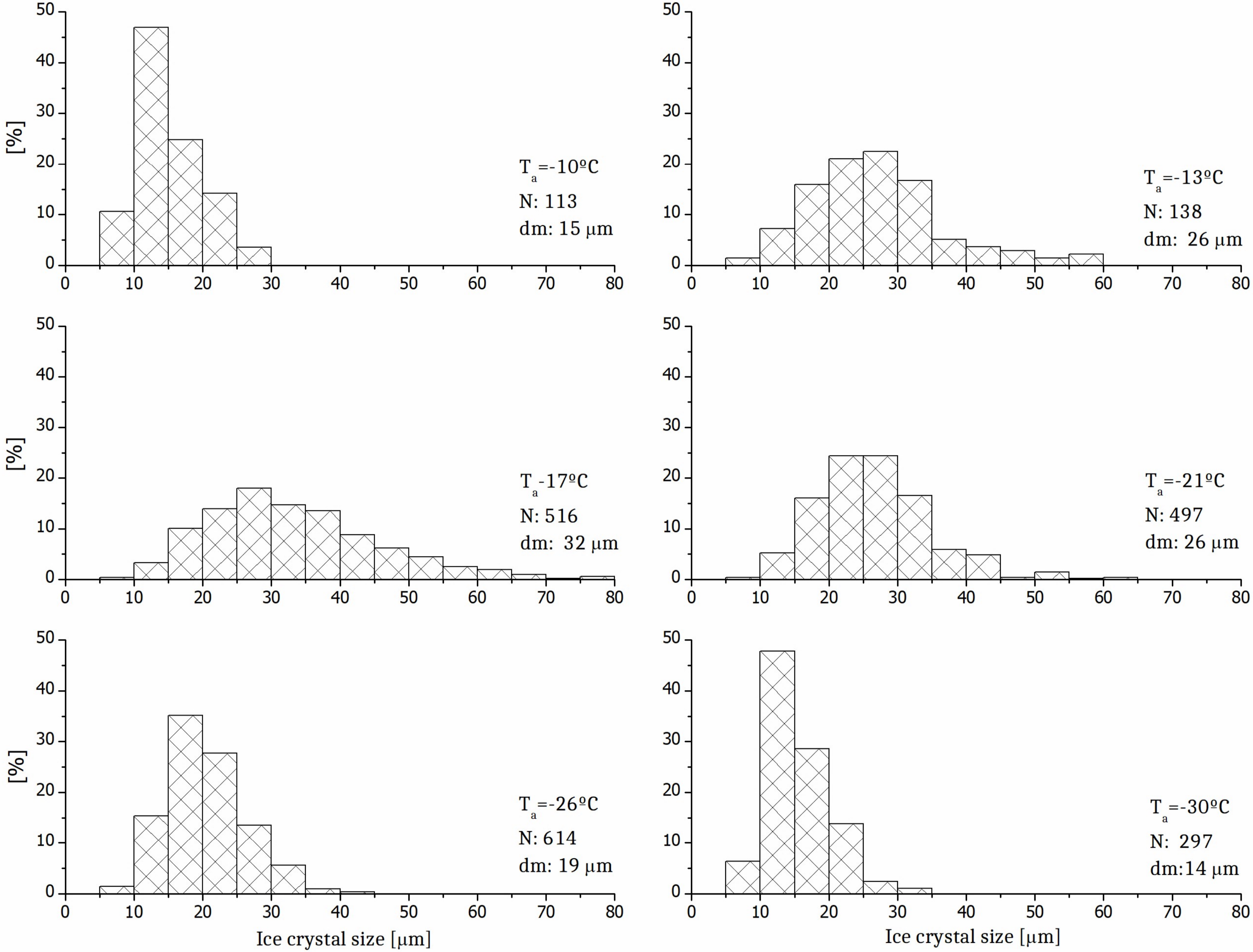
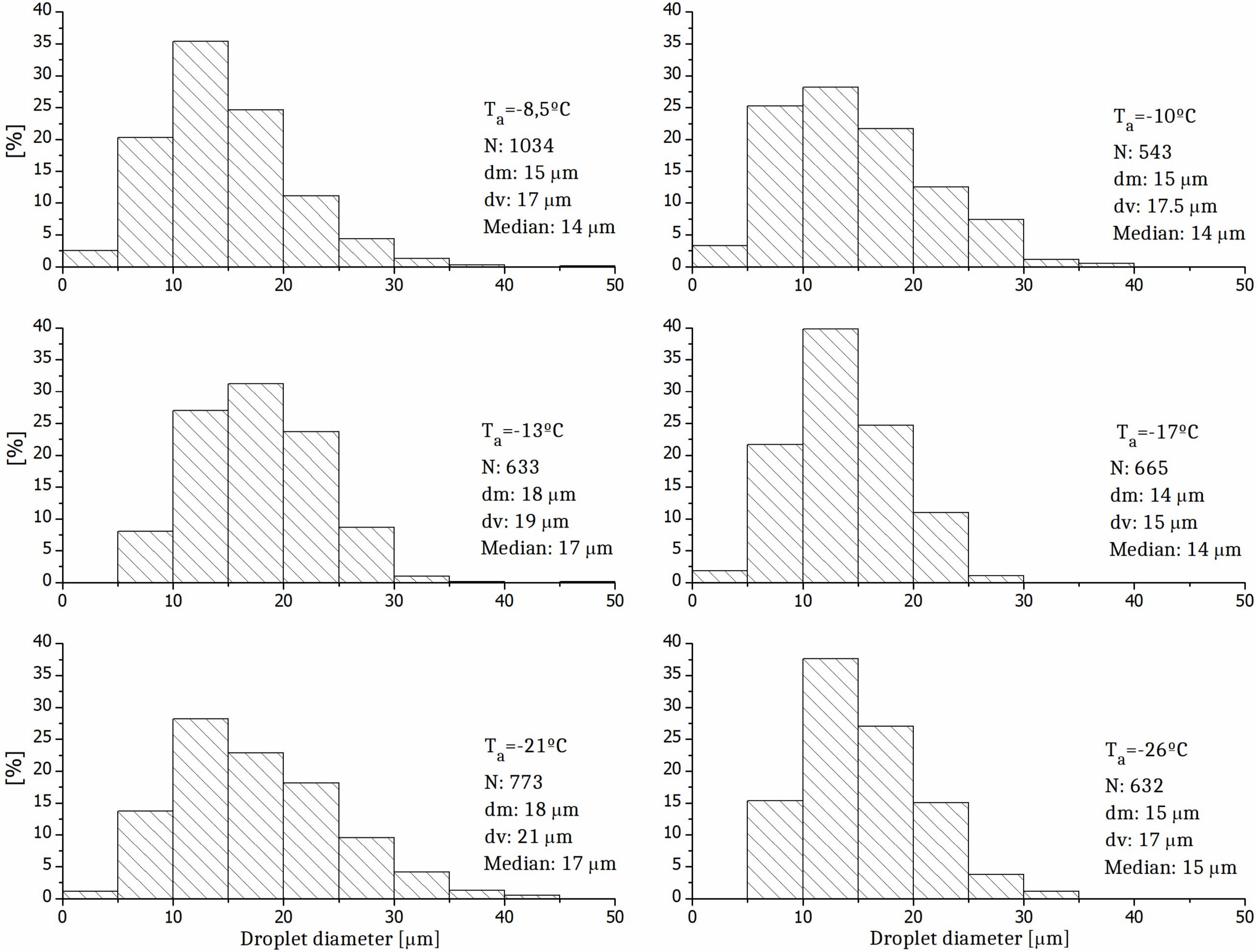

Cold Chamber
Water reservoir
Wind Tunnel
T_T
Amplifier
Ice-Crystals Chamber
Target
T_{WT}
T_{ICC}
To the Pump
Rain
Cloud Chamber
Drainage

Figure 2.



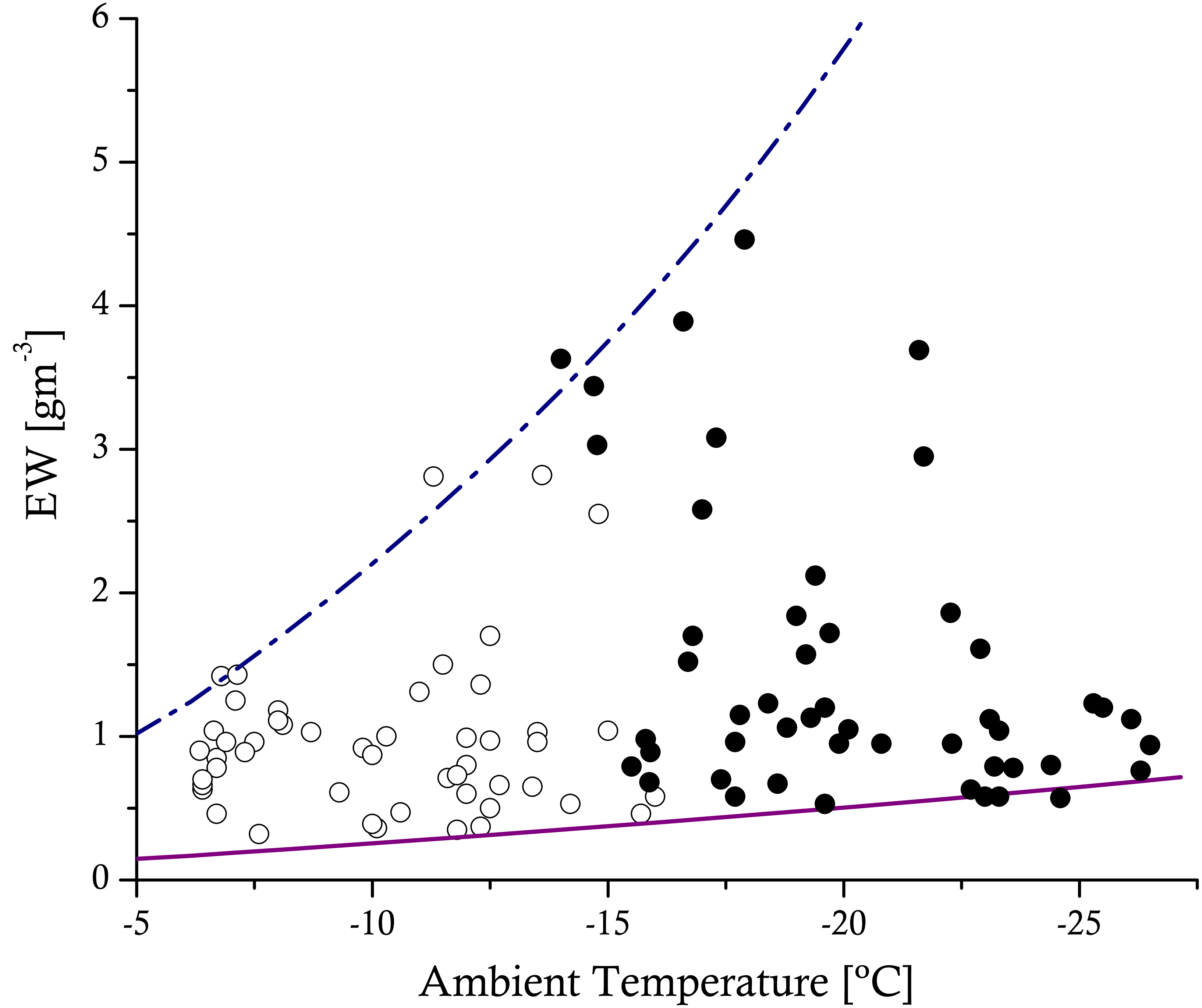
[%]
T_a=-10ºC
N: 113
dm: 15 μm
T_a=-13ºC
N: 138
dm: 26 μm
T_a-17ºC
N: 516
dm: 32 μm
T_a=-21ºC
N: 497
dm: 26 μm
T_a=-26ºC
N: 614
dm: 19 μm
T_a=-30ºC
N: 297
dm:14 μm
Ice crystal size [μm]

Figure 3.



[%]
Droplet diameter [μm]
T_a=-8,5ºC
N: 1034
dm: 15 μm
dv: 17 μm
Median: 14 μm
T_a=-10ºC
N: 543
dm: 15 μm
dv: 17.5 μm
Median: 14 μm
T_a=-13ºC
N: 633
dm: 18 μm
dv: 19 μm
Median: 17 μm
T_a=-17ºC
N: 665
dm: 14 μm
dv: 15 μm
Median: 14 μm
T_a=-21ºC
N: 773
dm: 18 μm
dv: 21 μm
Median: 17 μm
T_a=-26ºC
N: 632
dm: 15 μm
dv: 17 μm
Median: 15 μm

Figure 4.



EW [gm-3]
Ambient Temperature [ºC]
0
1
2
3
4
5
6
-5
-10
-15
-20
-25