Review of experimental studies on secondary ice production

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Abstract

10   Secondary ice production (SIP) plays a key role in the formation of ice particles in tropospheric clouds. Future improvement of the accuracy of the weather predictions and climate models relies on a proper description of SIP in numerical simulations. For now, laboratory studies remain a primary tool for developing physically based parameterizations for cloud modeling. Over the past seven decades, six different SIP-identifying mechanisms have emerged: (1) shattering during droplet freezing; (2) the rime splintering (Hallett-

15   Mossop) process; (3) fragmentation due to ice-ice collision; (4) ice particle fragmentation due to thermal shock; (5) fragmentation of sublimating ice; (6) activation of ice nucleating particles in transient supersaturation around freezing drops. This work presents a critical review of the laboratory studies related to secondary ice production. While some of the six mechanisms have received little research attention, others consist of contradictory results obtained by different research groups. Unfortunately, despite past investigative

20   efforts, the lack of consistency and the gaps in the accumulated knowledge hinder the development of quantitative descriptions of any of the six SIP mechanisms. The present work is aimed at identifying gaps in our knowledge on SIP and on stimulating further laboratory studies in obtaining a quantitative description of efficiencies for each of SIP mechanism.

25   **1.   Introduction**

Secondary ice production (SIP) is defined as the formation of atmospheric ice as a result of processes involving pre-existing ice particles, in contrast to primary ice production, which commences by the nucleation of ice either homogeneously in strongly supercooled droplets or heterogeneously on the surface of ice nucleating particles (INP) (e.g. Kanji et al, 2017). SIP is one of the fundamental cloud microphysical

30   processes, recognized as a major contributor to the observed concentration of ice particles at temperatures warmer than the homogeneous freezing temperature.

Even though SIP was observed in early laboratory experiments (e.g. Dudetski and Sidorov, 1911, Findeisen, 1940; Findeisen and Findeisen, 1943; Brewer and Palmer, 1949; Malkina and Zak, 1952; Puzanov and Accuratov, 1952; Schafaer, 1952; Bigg, 1957), the geophysical significance of SIP was recognized only

35   after the beginning of regular airborne studies of cloud microstructure in different geographical regions (e.g. Koenig 1963, 1965; Hobbs, 1969; Mossop, 1970, 1985; Mossop et al. 1972; Ono, 1972; Hallett et al. 1978; Hobbs and Rangno 1985, 1990; Beard 1992; and many others). A systematically observed enhancement of the

number concentration of cloud ice particles over the concentration of INP in the same air mass, urged for the

provision of an explanation of the physical processes underlying this discrepancy. One of the explanations

40   linked the enhancement of the concentration of ice particles to a mechanism unrelated to heterogeneous ice

nucleation.

From the late 1950s to early 1970s, six possible mechanisms were proposed explaining the secondary

production of ice crystals. However, since then, limited progress has been made in understanding of how each

of those mechanisms contribute to the ice particle concentrations and what the necessary and sufficient

45   conditions are for initiating each of these mechanisms. This situation is complicated by the fact that numerical

cloud models tend to focus on only one of the six possible mechanisms, namely the rime-splintering (Hallett-

Mossop) process, whereas other mechanisms have been disregarded.

Beyond recent reviews on in-situ studies of ice multiplication (e.g. Cantrell and Heymsfield, 2005; Field et

al., 2017), little attention has been devoted to exploring the details of laboratory studies on SIP mechanisms.

50   To bridge this gap, this paper provides an extended review of experimental works on SIP. Laboratory studies

are the basic means of examining physical processes underlying each SIP mechanism, as well as quantifying

the rates of secondary ice production, and identifying necessary and sufficient conditions required for initiation

of these mechanisms. Without this knowledge, a development of the physically based parameterisations of SIP

in weather prediction and climate simulations is not feasible. Due to their coarse spatial and temporal

55   resolution, in-situ airborne (by nature Eulerian) observations should be used for validation and feedback of

laboratory and theoretical SIP studies, rather than serve as a primary tool for developing parameterizations for

numerical simulations.

This work is an overview of the current knowledge on SIP obtained from laboratory studies. For the sake of

thoroughness, experimental studies of the effects of ice particle shattering on SIP observation were included

60   here as well. In-situ observations and theoretical studies of SIP were mentioned occasionally, though many of

them remained outside the frame of this review.

This review aims to provide navigation for future experimental works that seek to enhance our

understanding of SIP mechanisms.

The present paper describes laboratory studies of the following SIP mechanisms: the fragmentation of

65   droplets during their freezing (section 2), rime splintering (section 3), fragmentation due to collision of ice

particles with each other (section 4), ice particle fragmentation due to thermal shock caused by freezing

droplets on their surface (section 5), fragmentation of sublimating ice particles (section 6), activation of ice

nucleating particles in transient supersaturation around freezing drops (section 7). Section 8 describes

experimental studies that look at spurious enhancement of ice concentration during in-situ measurements,

70   which can be confused with SIP. The concluding remarks are presented in section 9.

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Discussions

2. Fragmentation of freezing drops

Historically, the first mechanism proposed to explain SIP was the fragmentation of freezing droplets (e.g.

75 Langham and Mason, 1958; Mason and Maybank, 1960; Kachurin and Bekryaev, 1960; Muchnik and Rudko, 1961). During the freezing process of a cloud droplet, liquid water may be trapped inside a growing ice shell formed around the droplet. The expansion of ice during subsequent freezing results in an increase of pressure inside the ice shell. If the pressure exceeds a critical point, then the ice shell may crack or shatter to relieve the internal pressure. The ice fragments that result from droplet cracking or shattering will serve as secondary ice.

80 In addition, gases dissolved in the droplet might be released during the pressure drop events. Gas bubbles may burst upon freezing at the colder droplet surface, resulting in a second source of fresh small ice fragments.

One of the necessary conditions for SIP during droplet freezing is creating a shell and freezing inward. Therefore, depending on the way in which the droplet freezes, it may or may not generate secondary ice. Hence, our consideration begins with a review of studies on the process of droplet freezing.

2.1 Freezing stages of a supercooled drop

The process of freezing of a supercooled droplet can be divided into two main stages. The first stage is a process that involves negligible heat exchange with the surrounding air. During that period, a dendritic ice network (slushy ice) forms through the liquid phase, releasing the latent heat, and heating up the liquid toward the melting point. This stage is usually referred to as the "fast" or "recalescence" stage. The second stage is

90 quasi-isothermal and determined by the freezing of the remaining liquid water. The heat transfer during this stage is directed to the air-droplet interface. The second stage is usually called the "slow" or "freezing" stage. After freezing is complete, the temperature of the frozen droplet gradually decreases towards the ambient temperature to attain a thermal equilibrium.
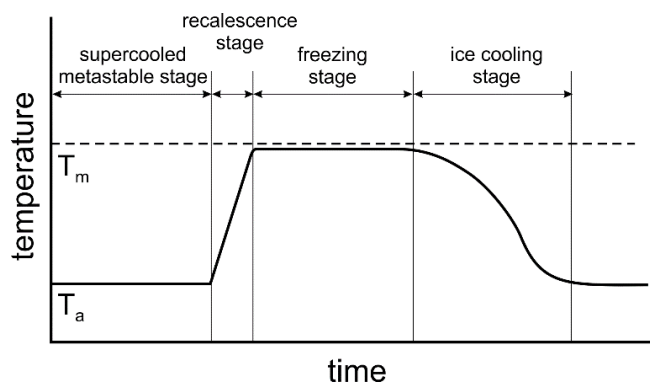
Figure 1 A conceptual diagram of temperature changes during the freezing of a supercooled droplet. Here T_m, T_0 are the melting and environmental temperatures, respectively.

A conceptual diagram of the temperature changes during the freezing of a supercooled droplet is shown in

100 Fig.1a. Documented temperature changes during the freezing of supercooled liquid drops can be found in e.g.

Mason and Maybank (1960), Muchnik and Rudko (1961), Pena et al. (1969), Bauerecker et al. (2008), Tavakoli et al. (2015).

2.2 Freezing fraction

105   The amount of frozen liquid water Δm during the recalescence stage can be estimated from a simplified equation of heat balance:

$$\Delta m L_m = \Delta m c_i \Delta T + (m - \Delta m) c_w \Delta T + \Delta Q \tag{1}$$

where m is the droplet mass, $\Delta T = T_m - T_a$ is the droplet supercooling; T_m, T_a are the melting point and initial droplet temperatures, respectively; L_m is the latent heat of freezing, c_i, and c_w are the specific heat of ice and

110   liquid water, ΔQ is the heat loss due to thermal exchange with the environment. After neglecting ΔQ and $(c_i - c_w)\Delta m \Delta T$, Eq.1 yields an approximation of the fraction of water $\mu = \Delta m / m$ frozen during the recalescence stage as

$$\mu = \frac{c_w \Delta T}{L_m} \tag{2}$$

Down to a temperature of -30°C, Eq. 2 is in very good agreement with an exact solution of (1) with T-

115   dependent material properties. Using a nuclear magnetic resonance technique, Hindmarsh et al. (2005), measured a fraction of frozen water formed in a supercooled 2mm diameter drop during the recalescence stage of freezing. They found the experimentally measured μ is in good agreement with that predicted by Eq.2 (Fig. 2).

Equation 2 yields that only a relatively small fraction of water freezes during the first stage. Thus, at -4°C and -20°C, the frozen fraction of water will be approximately 5% and 23%, respectively.
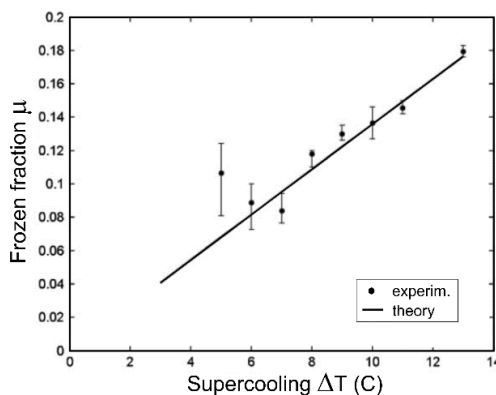
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Figure 2. A frozen fraction of water μ formed in a 2mm diameter drops during the recalescence stage versus temperature. The experimentally measured μ is in good agreement with that theoretically predicted by Eq.2 (adapted from Hindmarsh et al. 2005)

2.3 Droplet freezing time

125   The time scale of the recalescence stage can be assessed as (Macklin and Payne, 1967, 1968)

$$\tau_1 = D/u_i(\Delta T) \tag{3}$$

where D is the droplet diameter, and $u_i(\Delta T)$ is the velocity of ice crystal tips growth at water supercooling ΔT.

The growth rate $u_i(\Delta T)$ was studied by many research groups (e.g. Hallett, 1964; Pruppacher, 1967;

130 Feuillebois, et al. 1995; Lindenmeyer et al. 1959; Shibkov et al. 2003, 2005 and others). The summary of

studies of the velocity of freely growing ice as a function of ΔT is shown in Fig.3

Following Fig.3 and Eq.3 at T_0 =-4°C and -20°C the recalescence time t_1 for droplets with D =20μm will

be approximately 5ms, and 5μs, respectively; and for droplets with D=2mm, 0.5s and 5ms, respectively.

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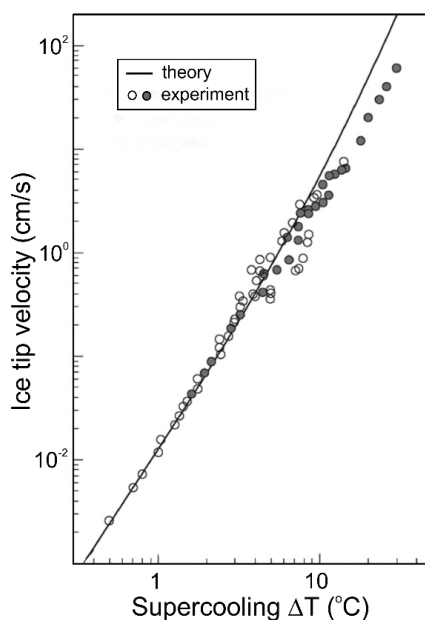
Figure 3. The measured velocity of freely growing ice as a function of supercooling measured by Hallett (1964), Pruppacher (1967), Kullinghall and Barduhn (1977), Furukawa and Shimada (1993), Feuillebois, et al. (1995; Ohsaka and Trinh (1998), Lindenmeyer et al. (1959) (open circles), Shibkov et al. (2003) (solid circles). The theoretical curve is based on Langer Muller-Krumbhaar results. Adapted from Shibkov et al. (2003)

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During the freezing stage droplets are cooling due to the thermal exchange with the ambient environment,

and thus, the remaining liquid water gradually freezes. The second stage is quasi-isothermal and it is

approximately 100-1000 times slower than the first stage. According to Pruppacher and Klett (1998), the time

of the second stage of the droplet freezing inward can be estimated as:

145 $$t_2 = \frac{\rho_w L_m D^2 (1 - \frac{\Delta T c_w}{L_m})}{12 f \Delta T \left(k_a + L_s D_v \left(\frac{\overline{d\rho_v}}{dT}\right)_{sat,i}\right)} \tag{4}$$

where ρ_w liquid water density; f ventilation coefficient; D_v is the water vapor diffusion coefficient; k_a is the

thermal conductivity of the air; L_s latent heat of ice sublimation; $\left(\frac{\overline{d\rho_v}}{dT}\right)_{sat,i}$ is the mean slope of the ice

saturation vapor density curve over the interval from T_0 to T_m.

Following Eq.4 at T_0 =-4°C and -20°C, the freezing time t_2 for droplets with D =20μm will be

150   approximately 70ms, and 11ms, and for droplets with D=2mm, 80s and 13s, respectively.

Since $t_2 \gg t_1$ the droplet freezing time is determined by the freezing during the second stage. Experimentally, the freezing time was studied by Muchnik and Rudko (1962), Murray and List (1972), Hindmarsh et al. (2003).

It should be noted that there is a good wealth of theoretical studies on the freezing time t_2 (e.g. Macklin and

155   Payne, 1967; King, 1975; Gupta and Arora, 1992; Feuillebois et al., 1995; Tabakova et al., 2010). However, Eq.4 (Pruppacher and Klett, 1998) provides a reasonably accurate assessment of t_2, which is in good agreement with experimental measurements.
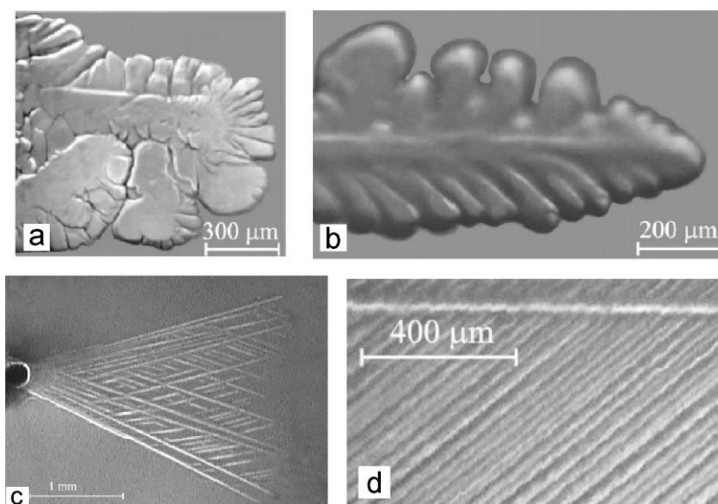
Figure 4. The morphology of ice crystal habits freely growing in pure water, supercooling at (a) ΔT =0.3°C dense brunching structure; (b) ΔT =1.5°C, developed dendrite; (c) ΔT =4.1°C, needle-like crystals; (d) ΔT =-14.5°C compact needle mesh. (adapted from Shibkov et al. 2003)

2.4 Crystalline structure of ice

165   The way in which ice crystals grow through the freezing droplet during the recalescence stage is of great importance for two reasons. First it affects the formation of the ice shell and it also impacts the way in which the liquid water freezes inside the droplet. The morphology of ice formation during water freezing was explored by Kumai and Itagaki, (1953), Hallett (1960, 1964), Macklin and Ryan (1965, 1966), Pruppacher (1967a,b), Furukawa and Shimada (1993), Ohsaka and Trinh (1998), Shibkov et al. (2003, 2005). It was found

170   that the shape of the ice crystals depends on the water supercooling ΔT. At low supercooling (1°C< ΔT <3°C), ice crystals appear as stellar dendrites or dendritic sheets growing parallel to the basal plane. With the increase of supercooling, ice crystals start splitting, causing a formation of three-dimensional complex structures (e.g. Pruppacher 1966b; 1998; Shibkov et al, 2003). Splitting leads to so-called "non-rational"

growth, i.e. growth that cannot be explained by rational crystallographic indices. Hallett (1964) and Macklin

175   and Ryan (1965, 1966) suggested that this non-rational growth is explained by the hopper structure of ice

crystal growth. One of the important findings of studies on water freezing is that the density of the ice mesh

increases with the decrease of temperature, whereas the spatial scale of the ice crystals parallel to the basal

plane becomes smaller. These features can be clearly seen in Fig.4. The shape of the ice crystals and the

density of their network has direct impact on the size and the number of isolated water pockets formed during

180   freezing as well as the tensile stress that is required to rupture the droplet.

     Regardless of the visual randomness of crystals growing through supercooled water, the non-rational

structures may compose single crystals after the droplet freezing is completed (Macklin and Ryan 1965, 1966).

Hallett (1963, 1964) Magono and Aburakawa (1968) Hallett (1963, 1964), Pitter and Pruppacher (1977)

studied the formation of monocrystalline and polycrystalline droplets during droplet freezing. They

185   found that the diameter of the monocrystalline frozen drops decreases with the increase of supercooling ΔT

(Fig.5). As it will be discussed below, shattering and secondary ice production depend on whether droplets

freeze as mono or polycrystal.
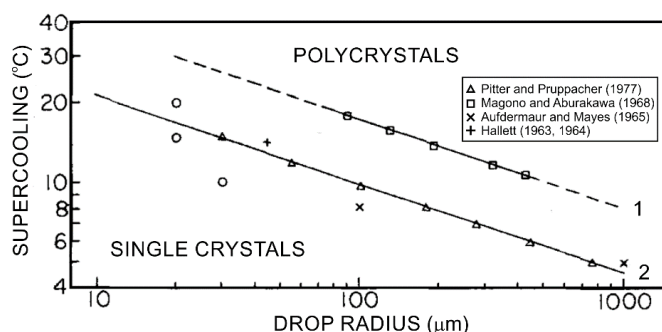
Figure 5. Dependence of the polycrystallinity on the diameter of the frozen drop and freezing temperature. (1) drops
frozen on the surface of large single crystals; (2) drops freely suspended in vertical airflow and nucleated by contact with
clay particles. Adapted from Pitter and Pruppacher (1977)

2.5 Pressure inside freezing droplets

195   The pressure inside freezing drops was measured by Visagie (1969) and King and Fletcher (1973). Water

drops with immersed pressure sensors were suspended in between paraffin oil and a carbon tetrachloride bath

inside a temperature-controlled chamber. The size of the drops varied from 7mm to 12mm. It was found that,

during freezing, the pressure inside a drop gradually built up as the shell became thicker. The pressure increase

was repeatedly interrupted due to the complete or partial pressure relief brought on by cracking (Fig.6). In this

200   period, water extruded through a crack and froze on the surface of the drop. After the crack was sealed by

frozen water, the pressure would climb back to the previous value and continue to grow. Both studies showed

that the pressure increased until reaching its maximum value $P_{\max}$ near the point of complete freezing. The

highest pressure, P_{max}= 89 bar in an 11mm diameter drop at -5°C, was observed by King and Fletcher (1973)

and 79 bar in 7mm drop at -12.8°C by Visagie (1969). However, no appreciable pressure growth was observed

205   inside drops freezing at temperatures warmer than -3°C. King and Fletcher (1973) noted that about 20% of

droplets contained a residual pressure of 10-20 bar at the completion of freezing.

The formation of cracks during droplet freezing was accompanied by an audible noise detected by

microphone in Visagie (1969) experiments. Loud sounds during droplet freezing and fragmentation were also

reported by Dudetski and Sidorov (1911).

210   Visagie (1969) pointed out that besides the shell wall thickness, the cracking pressure is also a function of

the temperature gradient across the ice shell (see Fig.6 in Visagie 1969).

King and Fletcher (1973) concluded that large droplet freezing at warm temperatures will exhibit

substantial viscous flow, and the smaller droplets freezing at colder temperatures will exhibit more elastic

behaviour and crack more often. Between these two extremes, there is probably a size-temperature domain, in

215   which sufficient elastic energy is stored in the shell to shatter it violently.

Both studies found that the cracking pressure increases with the increase of the thickness of the ice shell

during the droplet freezing. However, the dependence of the cracking pressure versus droplet size and

temperature remains unknown.
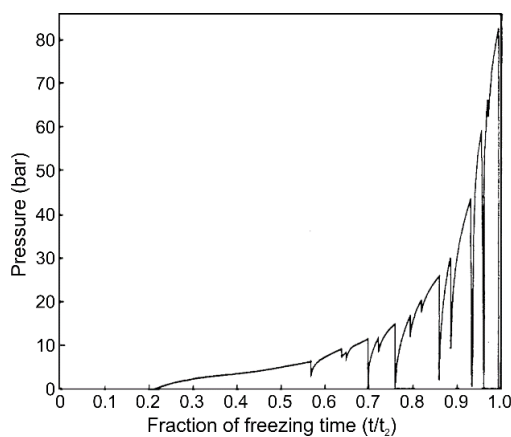
220   **Figure 6.** A time series of pressure changes inside an 11mm diameter drop freezing in a bath at -5C (adapted from King
and Fletcher, 1973).

Visagie (1969) and King and Fletcher (1973) conducted their experiments with overly large drops (7mm to

11mm) placed in a paraffin oil and carbon tetrachloride bath. This experimental setup affects the temperature

225   gradients in the ice shell around the freezing drops and the rate of heat exchange between the inner part of the

drops and their surrounding environment. These are the critical components for the cracking behavior and the

inner pressure changes. This brings up the issue of whether the obtained results are applicable to drops of

smaller sizes, which typically form in natural clouds.

2.6 Metamorphosis of droplet shape during freezing

230   Visagie (1969) and King and Fletcher (1973) also documented that in addition to cracking, the release of internal pressure inside freezing drops also occurred through deformation of the shape of the ice shell. Deformation of freezing drops was reported in early observations of freezing rain and ice pellets (e.g. Bentley, 1907). However, the physical explanations of freezing drop deformation were provided almost half a century later by Dorsey, (1948) and Blanchard (1951). Deformation of freezing droplets was observed by many

235   authors in their laboratory studies (e.g. Mason and Maybank, 1960; Jonson and Hallett, 1968; Takahashi and Yamashita, 1969; Pitter and Pruppacher, 1973; Takahashi, 1975, 1976; Iwabuchi and Magono, 1975; Pruppacher and Schlamp, 1975; Uyeda and Kikuchi, 1978; Lauber 2018 and many others). Furthermore, Takahashi (1975) identified four main categories of drop deformation: (a) spike (Figs.7a and 8); (b) bulge (Fig.7bc); (c) split (Figs.7c), (d) crack (Figs.7d and 8). During freezing, droplets may simultaneously develop a

240   combination different types of deformations depending on the droplet diameter and temperature, e.g. spikes and cracks (Fig.8). Sketches of a variety of different forms of bulges, cracks and spikes are available from Takahashi (1975).

   Takahashi (1976) found that deformation and shattering are closely related to crystalline structure formed during freezing. Thus, 90-100% of spikes are formed if droplets are polycrystalline. The spikes usually

245   protrude from the crystal boundary whose mechanical connection is weaker compared to monocrystalline locations. Spikes are also formed if, at the moment of nucleation, the droplet temperature is higher than the ambient temperature. However, spikes scarcely formed when the droplet was in thermal equilibrium with the environment. Takahashi (1975) found that the probability of spike formation increases with the increase of droplet size. This can be explained by the increase of the occurrence of polycrystalline frozen drops with the

250   increase of their sizes as in Fig. 5. Takahashi (1976) and Uyeda and Kikuchi (1978) studies also showed c-axis of a frozen monocrystalline droplet coincide with the c-axis of the seed crystal and that bulges are usually aligned with the c-axis.

   Most experiments on observation of droplet deformation were performed with relatively large drops $D >50\mu m$ and at temperatures $T>-25°C$. However, López and Ávila (2012) observed the formation of spikes

255   and bulges on small droplets with $8\mu m< D_{eff} <30\mu m$ freezing at temperatures $-40°C$. Microphotographs of small frozen drops obtained in their experiments did not reveal cracks and splitting. The authors also did not find any evidence of shattering. However, no deformation of small droplets was observed by López and Ávila at $T=-30°C$. It is worth noting that the interpretation of López and Ávila is hindered by an absence of information about the nucleating temperature of droplets. Since the droplets were introduced in the cloud

260   chamber at positive temperatures, there is good reason to consider that they froze at temperatures higher than that of the environment. This kind of condition is favorable for spike formation (Takahashi, 1975). Deformed small droplets frozen at $T_a<-40°C$ were also observed by Schaefer (1962).

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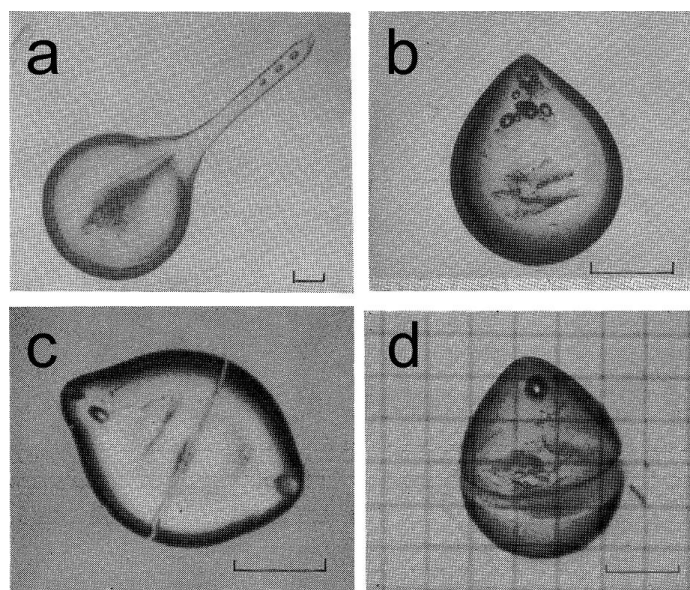
Figure 7. The main types of droplet deformations during freezing. (a) spike, long thin protrusion usually longer than one-fourth of the drop diameter; (b) bulge, protrusion shorter than one-fourth of the droplet diameter; (c) crack; (d) split. Scales are 100μm. Adapted from Takahashi (1975).

2.7 Fragmentation during freezing

The following discussion will consider works focused on laboratory studies looking at the processes behind splintering and fragmentation of freezing droplets.

Mason and Maybank (1960) studied the fragmentation of freezing droplets 30μm$< D <$1mm in the temperature range of -15°C$< T <$-1°C. Droplets were suspended on a fiber in a small (~40cm^3) cloud chamber. It turned out that, on average, the occurrence of droplet shattering decreased with the decrease of air temperature and droplet size. The occurrence of shattering for a 1mm diameter drop reached up to 47% with a maximum number of 200 splinters per drop. Such a high rate of splinter production is an important factor in the INP economy during precipitation formation.

However, Pruppacher (1967) pointed out that when Mason and Maybank (1960) performed their experiments, droplets did not reach thermal equilibrium at the moment of nucleation, and their temperature was warmer than that of the air by 1°C to 12°C. He argued that these conditions are favorable for the formation of an ice shell and for droplets freezing inward, which are critical for droplet shattering. Pruppacher questioned the relevance of the conditions used in the Mason and Maybank experiment to those in natural clouds.

Dye and Hobbs (1968) and Johnson and Hallett (1968) attempted to reproduce the Mason and Maybank (1960) experiments. They found that a 1mm diameter water drop suspended on a fiber did not shatter when nucleated after attaining thermal equilibrium. Dye and Hobbs (1968) also demonstrated that enhanced concentration of dissolved CO_2 resulted in increasing the occurrence of droplet shattering. They argued that the

Mason and Maybank (1960) experiments were affected by increased concentrations of CO_2, which was used as a coolant. Johnson and Hallett (1968, Fig.2) also demonstrated that for the drops where the nucleating temperature was higher than that of the air ($T_n > T_a$), the ice shell forms around a pure liquid core and ice mesh does not penetrate their centre. Such drops may create a stronger ice shell with a higher internal pressure, and therefore, be more susceptible to shattering.

290   Later, Hobbs and Alkezweeney (1968), Takahashi and Yamashita (1968, 1970), Pruppacher and Schlamp (1975) found that during freefall, droplets shatter after reaching a temperature quasi-equilibrium with the environment. It is important to note, that these results are in disagreement with those obtained by Dye and Hobbs (1968) and Johnson and Hallett (1968).

295   Despite the differences in experimental setups, most laboratory studies showed a general trend that large droplets are more susceptible to shattering during freezing than small ones (summarized in Lauber et al. 2018). However, Takahashi (1975) found that the relationship between the occurrence of shattering, droplet diameter and air temperature is more complex. He showed that in the air temperature range -20C<T_a<-7C, free falling drops have the highest occurrence of shattering in the size range 75µm<D<135µm. Whereas at T_a=-25C, the

300   probability of droplet shattering nearly monotonically increases from 50µm to 500 µm. Takahashi (1975) also found that at T_a=-4C, droplets with 50µm <D<200µm do not shatter. In this regard, it is worth mentioning that Brownscombe and Thorndike (1968) observed a 9% occurrence of shattering in droplets with 50µm <D<90µm at -5C, which is in agreement with Keinert et al. (2020) reporting a 15% occurrence of droplet breakup at   -5°C, which occurred only under free fall ventilation but not in stagnant air.

305   Laboratory studies also did not show a consistency for the lower threshold diameter for droplet fragmentation. Adkins (1960) found no splintering for droplets D<10µm. Hobbs and Alkezweeney (1968) observed no fragmentation of droplets 20µm <D<50µm. Johnson and Hallett (1968) reported no shattering observed for droplets 5µm <D<38µm. However, Mason and Maybank (1960, Table 1) observed droplet shattering in the size range of 30µm<D<80 µm when the droplets were at thermal equilibrium. The

310   inconsistency of the latter result may be related to the enhanced concentration of CO_2 in the laboratory setup. It is worth noting, that based on the theoretical analysis of the energy balance, Wildeman et al. (2017) concluded that symmetrically freezing droplets smaller than 50µm in diameter cannot shatter.

Ambient air temperature has a significant effect on the occurrence of freezing drop shattering. Both Takahashi and Yamashita (1970) and Lauber et al. (2018) found that the maximum rate of shattering is

315   observed between -10°C and -20°C for droplets ranging in size 85µm <D<350µm. This is generally consistent with the results found by Brownscombe and Thorndike (1968) for droplets with 80µm <D<120µm, although their temperature range was limited by -15°C<T_a<-5°C. However, for large drops with D >500µm the maximum occurrence of shattering was observed at T_a<-25°C (Takahashi, 1975). Hobbs and Alkezweeney (1968) found that the rate of shattering of droplets 50µm <D<150µm does not depend on the temperature over

320   the range -32°C<T_a<-20°C. Whereas, in his experiments, Takahashi (1975, Fig.7) found a strong temperature

dependence of droplets shattering in this size range.

A review of the laboratory studies showed that the reported rate of shattering during droplet freezing varied

significantly. For example, Takahashi (1976) found that the maximum rate of shattering for free fall droplets

(200µm< D <350µm) at -20ºC< T <-10ºC was close to 40%. However, Lauber et al. (2018) showed that, for

325   droplets suspended in electro-dynamic balance, the maximum shattering rate for the same temperature size

range is close to 12%. This, however, increased notably to about 25% when the experiments were conducted

under terminal velocity ventilation (Keinert et al., 2020). Brownscombe and Thorndike (1968) observed a 14%

rate of shattering for free fall droplets with 80µm <D<120µm freezing at -15C.

A significant inconsistency of the efficiencies of ice splintering and their dependency on temperature and

330   droplet size obtained by different research groups is quite evident. This poses a key question about the

differences in experimental setups and the potential effects of other parameters. Already, Johnson and Hallett

(1968) have pointed out the importance of the effect of ventilation on droplet shattering. When a droplet with

D=500µm was suspended on a thread and ventilated at an equivalent to the free fall speed, no shattering was

observed. However, when the droplet was rotated around an axis perpendicular to the airflow, shattering and

335   cracking invariably occurred. This finding raised questions about the realism of the experiments that had a

droplet is suspended with a fixed orientation on a fiber or other mount. Under these conditions, the thermal

exchange between the droplet and the ambient air is different compared to the free fall condition.

Pitter and Pruppacher (1973) demonstrated that a droplet suspended in the airflow begins to tumble and

spin immediately after nucleation, thus providing a radially more symmetric heat loss. Drop spinning after

340   nucleation was also reported by Dye and Hobbs (1968), Kolomeychuk et al. (1975) and Keinert et al., (2020).

Initiation of tumbling and spinning after droplet nucleation can likely be explained by the asymmetrical shape

and heterogeneous surface roughness that builds up quickly after freezing, thereby leading to a fluctuating

torque being exerted by the terminal airflow.

Takahashi (1976) also revealed the importance of the crystalline nature of ice that forms inside freezing

345   drops to their subsequent shattering. He found that 90-100% of shattering occurs when drops freeze as single

crystals. Takahashi also showed that splitting occurs perpendicular to the *c*-axis dividing the drop in two equal

parts (e.g. Fig.9c,d). The equatorial cracking and splitting of freezing drops was also reported by Wildeman et

al. (2017, Fig.3a,b) and Lauber et al. (2018, Figs.5,6). Takahashi (1976) systematized how drops may shatter

with respect to their crystalline boundaries. In most cases of polycrystalline drops, their fragmentation occurs

350   along the crystal boundaries, where mechanical connectivity is weaker.

One of the first classifications of 'types of fragmentation' during drop freezing goes back to the work of

Stott and Hutchinson (1965). They nucleated 0.9mm to 1.9mm diameter drops that were suspended on a fiber

at -1°C and then froze them at the air temperature of -15°C. Even though this particular arrangement is not

fully relevant to conditions in natural clouds, it helped identify the most common patterns of the drop

355   fragmentation. The droplet fragmentation was classified as follows: (a) violent shattering with multiple pieces;

(b) central breaks or splitting; (c) spicule breaks with liquid; (d) spicule breaks after solidification; (e) spicule

bubble breaks; (f) cracks.

Wildeman et al. (2017) conducted experiments with millimeter sized drops freezing on a super-

hydrophobic substrate. The high-speed videos documented explosive shattering of freezing drops, which

360   generated a cascade of ice fragment sizes. One of the videos (V2) documented secondary shattering of one of

the fragments formed after primary shattering. This suggests that during droplet freezing, liquid water may

form several pockets across the droplet volume, rather than one big unfrozen volume in the central part. As

discussed above, the connectivity of unfrozen pockets of water inside the ice shell is likely to be controlled by

the type of the ice network formed inside the droplet and temperature exchange between the droplet and

365   environment.

One of the caveats of the Wildemann et al. laboratory setup is that the experiments were performed at very

low pressure (3.4 10^{-3}atm), and the droplets cooled much faster than they would cool in the atmosphere.

Johnson and Hallett (1968) showed that below 0.13atm, every drop in their experiments shattered violently. In

this way, the results are not directly applicable to environmental conditions.
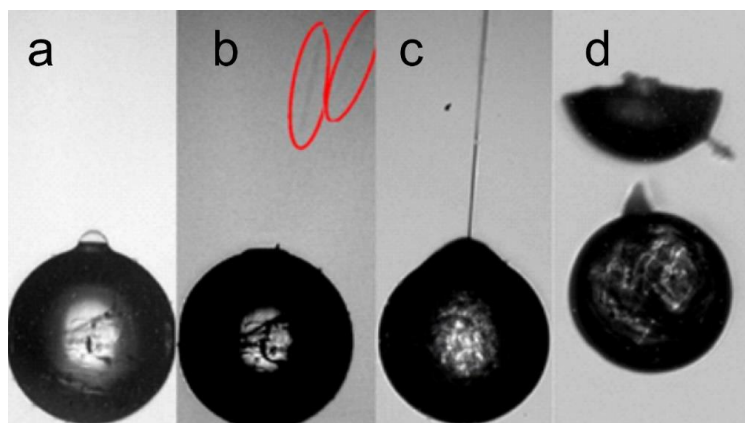
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Figure 8: Secondary ice processes, as observed by high speed microscopy (a) a bubble has formed on the surface of a
freezing droplet. Cracks are visible in the surface. (b) the droplet from (a) 12 ms later: the bubble has burst, two fragments
are highlighted. (c) jetting: a jet of liquid water is expelled violently through a hole in the ice shell. (d) breakup: a freezing
droplet splits in two halves, a few small fragments are sometimes observed (adapted from Lauber et al. 2015).

375

In a series of experiments conducted with electrically charged droplets levitated in an electrodynamic

balance, Kiselev and colleagues observed droplet freezing with a high speed video microscope and categorized

secondary ice processes as breakup, cracking, bubble bursting and jetting, cf. Fig. 8. Opposite to previous

studies, they did not observe violent shattering of freezing droplets into many fragments. The relative and

380   absolute frequency of the secondary processes did not only depend on droplet size and temperature, but also on

droplet ventilation and the presence of solid inclusions or dissolved salts. The effect of solid inclusions

(polystyrene latex particles) was twofold. While they suppressed droplet shattering upon freezing of large

(d=300µm) drizzle droplets (Lauber et al. 2018), they strongly enhanced droplet shattering in small (d=80µm) drizzle droplets (Pander et al. 2015). Large droplets were found to shatter at warmer temperatures and much

385 more frequently when suspended at terminal air velocity compared to being suspended in stagnant air under otherwise identical conditions (Keinert et al. 2020). Dissolved sea salt hindered droplet shattering at all sizes at concentrations above about 100 mg/L. It is reasoned that dissolved substances and solid inclusions are expelled from the growing ice phase and concentrate in the liquid phase during freezing. Here, they hinder the formation of a monocrystalline ice shell. So, on one hand, this reduces the pressure needed for breakup, but on

390 the other hand may open pathways for pressure release prior to breakup. Pressure release events such as jetting or spiking have been found to occur. Once high concentrations of dissolved gases build up in the liquid phase of the droplet interior, pressure release induces gas bubble formation in the droplet interior. These bubbles may escape through spikes or cracks in the ice shell giving rise to bubbles. Upon freezing of the bubble skin, the skin breaks and may form a source of additional tiny ice particles. Even though bubble bursting has been found

395 to be a frequent secondary ice process (Pander et al. 2015; Lauber et al. 2018), the number of emitted ice particles has not been quantified up to date. Droplet ventilation had a major influence on secondary ice process frequency and type (Keinert et al 2020). Droplets moving at terminal velocity with respect to the surrounding air generally showed more frequent secondary ice processes when compared to droplets levitated in stagnant air. The dominant process observed shifted from cracking at stagnant conditions to breakup under free fall

400 conditions. The latter could be observed even at temperatures warmer than -5°C.

King and Fletcher (1973) hypothesized that the numerous discontinuities in the pressure changes inside the freezing drops are indicative of the large-scale movements of the ice shell, and therefore, it may be a source of particles, even if the droplet does not shatter. This hypothesis was confirmed in experimental studies of droplet freezing by Wildeman et al. (2017). The production of ice splinters during cracking of 2mm freezing drop was

405 documented in the supplementary high-speed video V2. Visual analysis of this video allowed for the identification of several ice splintering events during cracking prior to final droplet shattering. Four of those events are shown in Fig.8. In general, the number of secondary ice particles due to droplet cracking during freezing could be formulated as the product of the number of cracking events per freezing event and the average number of secondary particles per cracking event. However, the actual number of splintering during

410 cracking events may be higher in comparison to those observed visually. This is because microphotography allows for the detection of only those splinters that occurred within the depth-of-field of the microscope or whose sizes were larger than the detecting threshold of the optical system, and because not all cracking events are detectable by optical microscopy.

Splintering during cracking is an important finding, since it shows that freezing droplets may be a source of

415 secondary ice even though they do not shatter by the end of freezing.
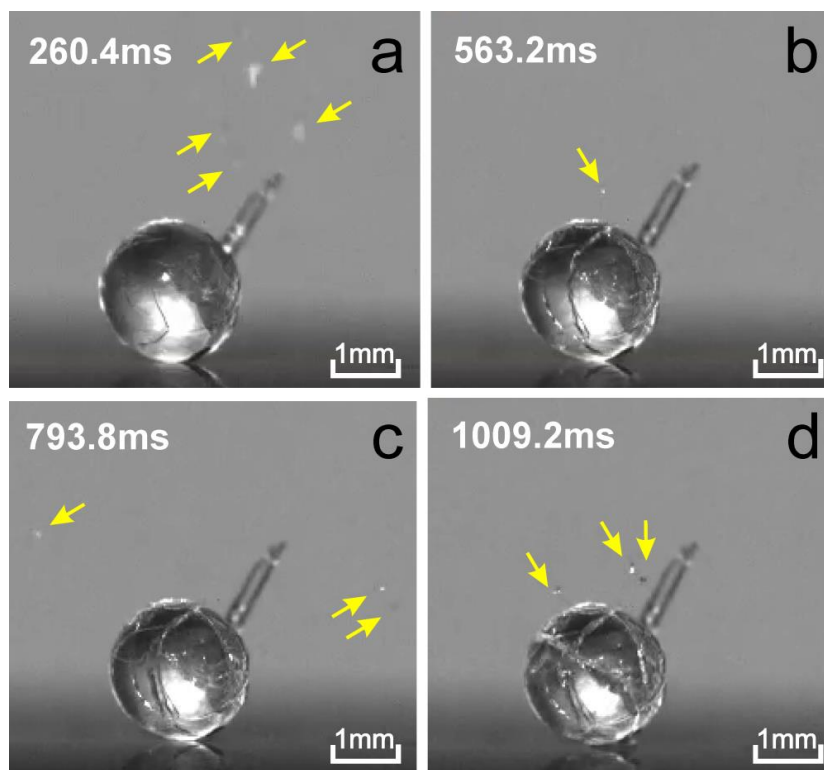
Figure 9. High-speed video snapshots of a 2mm drop at different stages of freezing. The pictures show progressive increase of the number of cracks covering the drop during its freezing. The yellow arrows indicate the locations of ice splinters ejected during cracking. The ambient temperature T=-7°C. The numbers in the top left corners indicate time since nucleation. Adapted from video V2 from the supplementary material to Wildeman et al (2017).

2.8 Summary

The review of the laboratory studies showed that the fragmentation of freezing drops is sensitive to a large number parameters such as: (a) droplet size D, (b) environmental temperature T_a, (c) droplet nucleating temperature T_n, (d) air pressure P, (e) type of ice mesh formed during the recalescence stage (dependant on T_n), (f) crystalline nature of freezing droplet (i.e. monocrystalline or polycrystalline), (g) ventilation (e.g. static air, free fall, drop rotation during freezing), (h) fall velocity (dependant on D and P), (i) dissolved gases (specifically CO_2, dependant on T_a and P). Several types of ice fragmentation during droplet freezing were documented: (a) splitting with few fragments, (b) explosive shattering with multiple fragments, (c) cracking-splintering, (d) bubble bursting, (e) jetting. Unfortunately, the dependency of ice fragmentation during droplet freezing on the above parameters remains poorly understood or unknown.

A review of the laboratory studies on droplet freezing showed a large diversity of obtained results. Thus, for a single experimental setup under the same conditions, the number of fragments formed for the same size drop during its freezing varied from zero to a few hundred. Similarly, under the same laboratory conditions, studies observed that only a fraction of the droplets shattered, whereas the other fraction did not produce any

15

fragments. This suggests that the laboratory experiments might contain hidden non-controlled parameters, which hindered obtaining reproducible results for each freezing droplet.

440   One of these parameters may be the orientation of the crystallographic axis of the INP with respect to the droplet surface at the moment of nucleation (Fig.10a,b,c). Since the growth rate of ice along the *a*- and *c*-axes is different (Macklin and Payne, 1968), the process of the droplet filling with the ice network during the recalescence stage may create a non-uniform distribution of temperature, and ultimately affect the symmetry of the ice shell. In the case of a polycrystal INP it is expected that during the recalescence stage a droplet will be filled by the ice network more uniformly (Fig.9c) as compared to a monocrystalline INP (Fig. 10ab).

445   Humidity of the surrounding environment may be another hidden aspect affecting SIP. Depending on the humidity level, the droplet may either grow or evaporate prior to nucleation. This may create additional temperature gradients at the droplet surface depending on its diameter. The near-surface temperature gradients may either hinder or facilitate the formation of the ice shell.

          The topology of liquid volumes inside the freezing drop may also be an important factor for SIP. Thus, the

450   cracking rate may be affected by the symmetry of the ice shell as well as the displacement of the liquid core with respect to the droplet center (Fig. 10d,e). The tensile stress formed in the ice shell is also expected to depend on how liquid water volumes are distributed across the freezing droplets: inside one big (Fig. 10d,e) or multiple small volumes (Fig. 10f). Unfortunately, no attention was given to this effect in previous laboratory studies.

455   There are a number of other parameters which received little attention in laboratory experiments that include: (a) size distribution of ice fragments, (b) minimum size of splinters, which may form during fragmentation, (c) minimal size for droplets to shatter, (d) effect of the angle between the *c*-axis and the droplet surface on ice shell formation, (e) humidity of the air.

          Growing evidence from in-situ observations (e.g. Korolev et al. 2004, 2020; Rangno, 2008; Lawson et al.

460   2017;) suggests that fragmentation during droplet freezing is an important SIP contributor to the concentration of cloud ice particles. Unfortunately, the diversity of laboratory results related to fragmentation during drop freezing hinders the development of a quantitative description and refined theory of this mechanism in order to use in cloud simulations. A variety of parameters and fragmentation types makes the experimental studies and quantification of this mechanism a challenging and intricate problem.

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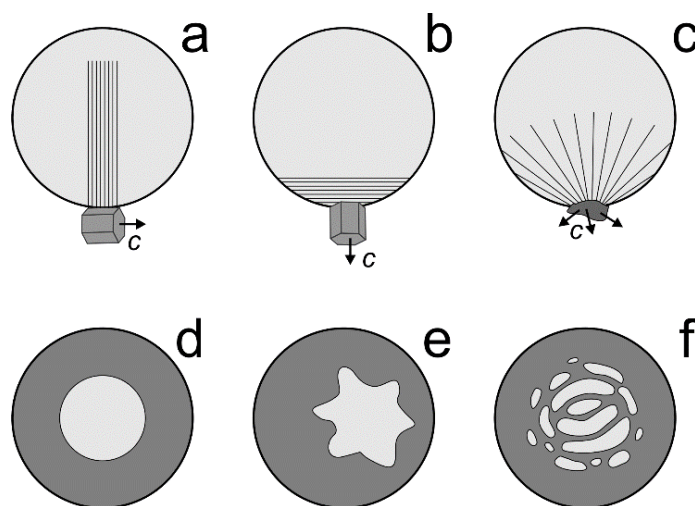
Figure 10. A conceptual diagram showing different possibilities of freezing of a supercooled droplet after nucleation by (a) monocrystalline INP with c-axis parallel to the droplet surface; (b) monocrystalline INP with c-axis perpendicular to the droplet surface; (c) polycrystalline INP. The visuals in (d,e,f) show various possible topologies of liquid zones formed during freezing: (d) idealized spherical liquid volume symmetrically centered with the ice shell (frequently used in numerical simulations of droplet freezing); (e) non-symmetrical liquid volume displaced towards the ice shell wall; (f) multiple disconnected liquid volumes.

3. Splintering during riming

3.1 Efficiency of rime splintering

Splintering during ice particle riming is another mechanism that can explain SIP. Macklin (1960) observed splinter production in a small wind tunnel during the collection of droplets on an icing rod with 0.6cm diameter at temperatures -5C< T_a <-20°C. The droplet diameters in their size distribution varied from several to 140μm (MVD~67μm) and their speed changed from 2m/s to 12m/s. A microscopic examination revealed long spicules and a few micrometer-sized ice features formed on the surface of the rod. The small fragile formations were hypothesized to be a source of the splinters. The ice crystal concentration during experiments was frequently observed to increase by a few orders of magnitude, reaching values of the order of 10^{-1}cm^{-3} at temperatures as high as -5°C.

Latham and Mason (1961) observed riming of freezing droplets on the hailstone simulator, accompanied by the ejection of ice splinters. They established that the splinter production varied with the air temperature, drop diameter and impact velocity. A maximum production rate of 14 splinters per droplet, was observed in droplets with diameter 70μm, impacting at 10 m/s at a temperature of -15°C.

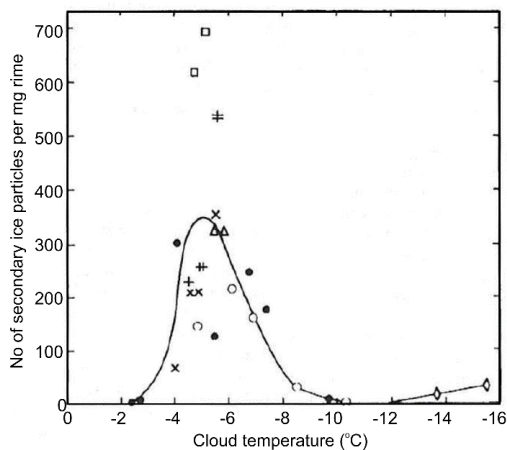Later, Hallett and Mossop (1974) and Mossop and Hallett (1974) observed splinter formation during riming in the cloud chamber with liquid water content ~1g/m^3 and droplet concentration 500cm^{-3}. They found that splinter production is active in the temperature range -8°C< T_a <-3°C. Furthermore, the rate of splinter production had a pronounced maximum at the air temperature of -5°C and the drop impact velocity 2.5m/s

(Fig.11). At these conditions, one splinter was produced per 250 droplets of diameter $D>24\mu m$. The phenomenon of splinter production during riming is usually referred to as the Hallett-Mossop (HM) mechanism.

Figure 11. The dependence of the number of splinters per mg of rime on ambient temperature at speed 2.7m/s obtained experimentally by Hallett and Mossop (1974).

495

Mossop (1978, 1985) found that the presence of droplets with $D<12\mu m$ in addition to those $D>24\mu m$ 500 increases the splinter production further. Saunders and Hosseini (2001) studied the splinter production in a wider range of impact velocities of up to 12m/s. They found that the maximum secondary ice ejection occurs at 6m/s with the number of splinters nearly five times lower than it was found in the Hallett and Mossop (1974) and Mossop and Hallett (1974) experiments.

Heymsfield and Mossop (1984) studied the effect of the rimer surface temperature on the production of 505 secondary ice particles. They found that raising the surface temperature of the riming particle by 1°C transposes the splinter production curve virtually unchanged to air temperatures 1°C lower. This led them to conclude that splinter production due to the HM-mechanism may occur at air temperatures lower than -8°C depending on liquid water content (LWC) and the rimer fall velocity, which are the main factors determining the surface temperature of the riming particle. This conclusion is consistent with earlier work by Foster and 510 Hallett (1982).

The quantification of the rime splintering production obtained from the experimental studies on Hallett and Mossop created a basis for various formulations of SIP parameterizations (e.g. Cotton et al. 1986; Meyers et al. 1997; Reisner et al, 1998 and others), which are widely used in numerical simulations of clouds.

3.2 Physical mechanism of rime splintering

515 Several studies are aimed at understanding the physical mechanisms responsible for splinter production. For instance, Macklin (1960) documented that fine ice structures formed during riming could be easily detached from the rimer and form splinters. One of these fine ice features are shown in Fig.12a. Choularton et

al. (1978, 1980) suggested that, if droplets D >25µm are accreted to the ice substrate by a thin neck, they will

520 minimize the heat transfer toward the rimer. This arrangement may induce symmetrical heat loss to the air,

which then leads to the formation of a complete ice shell around a droplet as it freezes. The freezing of liquid

will result in a pressure build up inside the droplet which may cause shell disruptions with subsequent

production of fragile protuberances of frozen water (Fig.12b). Mossop (1980) credited this hypothesis by

pointing out that the ice shell is weakened by the presence of ammonia and results in a reduced number of

protuberances and splinters. He also showed that the increased ammonia concentration in droplets results in a

525 reduction of the rate of splinter production. Griggs and Choularton (1983) suggested that the cut-off at about

-8°C is due to the more rapid growth of ice shell, which is too strong to be disrupted by the internal pressure.
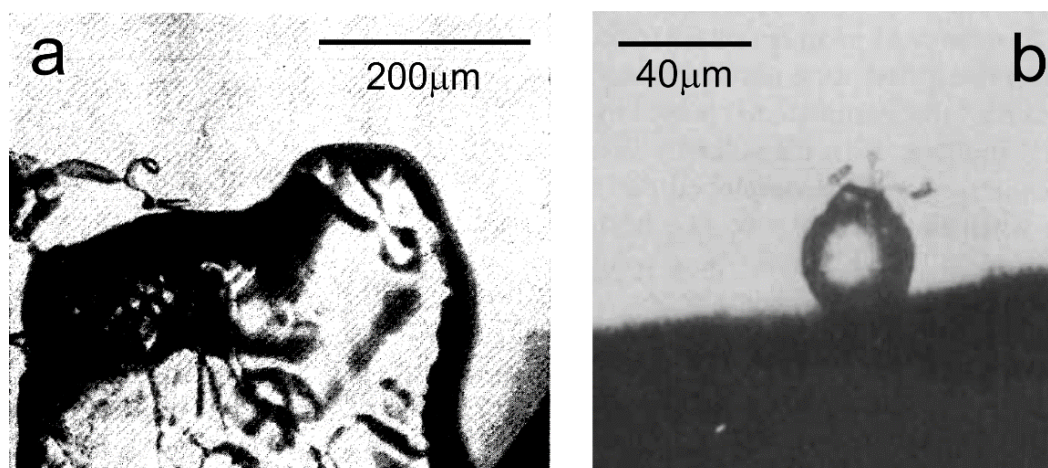
Figure 12. Pictures of the rime fragments: (a) frozen splash observed at T_a =-6°C and speed 6m/s (Macklin, 1960); (b) disruptions of protuberances formed out of a 35µm rimed droplet at T_a =-7°C and speed 1.5m/s (Choularton et al. 1980)

530

Dong and Hallett (1989) reported that after impact with ice, droplets tend to spread over the surface of ice

at all temperatures above -8°C. They concluded that splinter production by pressure build-up inside individual

frozen droplets is unlikely to be responsible for the shattering. They suggested that fragmentation is associated

with the stress build up within an accreted droplet. This occurs when the droplet experiences a temperature

535 gradient between the colder substrate and the surface of the droplet, freezing at 0°C. However, the absence of

protuberances, when the droplets rime onto an ice surface above -10°C, contrasts with the observation of

Choularton et al. (1980) who showed photographs of discrete frozen droplets together with protuberances

obtained in the temperature range -3°C to -7°C (Fig.12b).

Emersic and Connoly (2017) studied microscopic riming events on an ice target using a high-speed video

540 recording. The droplet sizes ranged from 5 to 50 µm. It was found that the droplet behavior on impact depends

on the uniformity of the rimer surface. Thus, droplets tend to spread flat on flat ice surfaces at temperatures

associated with the HM-process, as was earlier observed by Dong and Hallett (1989). However, with

increasing rime depth, which is more commonly associated with graupel, growing rime spires protrude from

the surface into the airflow around the rimer. No protuberances or liquid ejection was observed during the

545   riming process, nor was any mechanical rime splintering event observed in approximately 1300 droplet

freezing events. Based on the results of their study, Emersic and Connoly (2017) hypothesized that the rime

spikes that develop with continuing droplet accretion could break off during particle tumbling or hinging by

small droplets.

3.3 Summary

550   The literature review showed that, apart from some early studies (Hobbs and Borrows, 1966; Aufrermaud

and Jonson, 1972), most laboratory experiments on the HM-process confirmed splinter production during

riming. However, there was no consistency in the rate of the rime splintering observed by different groups.

This discrepancy is most likely related to different laboratory setups and techniques used for splinter counting.

To conclude this section, it should be emphasized that after several decades of rime splintering studies, the

555   physical mechanisms behind this phenomenon are still under debate. Without clarifying the nature of this

process, a development of a physically based parameterizations for numerical simulations of clouds does not

seem to be feasible.

4.   Fragmentation due to ice-ice collision

560   Collision of ice particles may result in their mechanical fragmentation and production of secondary ice

(Langmuir, 1948, p.186). This hypothesis was stimulated by observations of ice particle fragments collected

during airborne (e.g. Hobbs and Farber, 1972; Takahashi, 1993) or ground-based (Jiusto and Weickmann,

1973) studies.

There were only two known laboratory works on collisional ice fragmentation. Vardiman (1978) explored

565   fragmentation of natural cloud ice particles on impact with a metal mesh. He found that "graupel is

surprisingly ineffective in generating fragments". However, light to moderate rimed spatial crystals are the

most efficient source of ice fragments. For planar crystals, the degree of fragmentation increases with the

degree of riming.

Takahashi et al. (1995) studied the dependence of mechanical fragmentation resulting from collision of 2cm

570   in diameter rimed ice spheres. The ice spheres were attached to the edges of 10cm long spinning metal rods.

The ice spheres were made to collide with each other at a speed of 4m/s. These speeds were used to simulate a

collision of 4mm diameter lump graupel with a density of 0.3-0.4g/m^3. The collisional force changed

incrementally from 20dyn to 500dyn. Takahashi et al. found that the number of fragments depends on the

degree of riming, temperature, and collision force. The maximum number of fragments per collision (up to

575   800) was observed at -16C.

It is hard to judge the consistency of the results obtained by Vardiman (1978) and Takahashi et al. (1995)

because of the differences in the experimental setups and environmental conditions. It is also difficult to

identify the degree of applicability of the rate of SIP obtained in these experiments to free falling ice particles grown in natural clouds.

580   Collisional ice fragmentation was also studied theoretically. Hobbs and Farber (1972), Vardiman (1978), Phyllips et al. (2017) studies were based on the consideration of collisional kinetic energy and linear momentum. Such considerations would be relevant only for cases of direct central impact. In a general case, angular momentum and rotational energy should be taken into consideration. Since oblique particle collisions are more frequent than central collision, the efficiency of SIP obtained in these works is expected to be

585   overestimated.

    The theoretical considerations of collisional fragmentation in Yano and Phillips, (2011), Yano et al. (2016) and Phillips et al. (2017) were based on the rate of ice production from Takahashi et al. (1995). A detailed analysis of the Takahashi et al (1995) laboratory setup indicated that the riming of ice spheres occurred in still air, which resulted in more lumpy and fragile rime compared to that formed in free-falling graupel. The

590   collisional kinetic energy and the surface area of collision of the 2cm diameter ice spheres also significantly exceed the kinetic energy and collision area of a few mm sized graupel. Altogether, it may result in overestimation of the rate of SIP, compared to graupel formed in natural clouds.

    It should also be mentioned that ice particle fragments observed in-situ (e.g. Hobbs and Farber, 1972) may be a result of particle breakups induced by the sampling instrument. Schwarzenboeck et al. (2009) identified

595   that 18% of observed incomplete dendrites are the result of natural fragmentation. The identification of natural fragments was based on the observation of the ice shapes near the expected break area, which were interpreted as "subsequent growth". However, it could be argued, that incomplete dendrites may naturally form because of growth suppression of one or more branches due to defects or dislocations on the crystal. Examples of 1-, 2-, 3- and 4-branched stellar and dendritic crystals with underdeveloped defected branches were documented by

600   Bentley and Humphreys (1962, pl.198-204), Auer (1970, Figs.9,28,30), Kikuchi and Uyeda (1978, Fig.2).

    In summary of this section, it can be concluded that the efficiency of SIP during ice-ice collisional fragmentation remains uncertain due to the lack of laboratory studies. No parameterizations of SIP due to ice-ice collisional fragmentation can be developed at that stage based on two laboratory observations, whose results are conflicting with each other. Additional laboratory studies are required to explore ice-ice collisional

605   fragmentation of free-falling ice particles with different habits. Ice fragments observed in-situ should be considered with caution due to potential particle breakups during sampling (see Section 8).

5. Fragmentation due to thermal shock

    When a supercooled drop rimes on the surface of an ice crystal, it freezes, and its temperature rises to the

610   melting point (section 2). Some fraction of the latent heat released during freezing will be transferred into the ice crystal. This may cause a thermal shock at the location of the droplet attachments and fragmentation due differential expansion of the ice crystal (Koenig, 1963, p.35).

Dye and Hobbs (1968) observed during laboratory experiments that, when an ice crystal on some occasions became attached to a freezing drop, it would often break into 5 to 10 pieces as the drop froze. Sometimes, the
615   breakup of the crystal would occur when the drop cracked. On other occasions the crystal would break without any apparent changes to the freezing drop. They concluded that breaking up ice crystals, which collide with and nucleate supercooled drops, may play important role in increasing concentration of ice particles in natural clouds.

Hobbs and Farber (1972) observed in the laboratory shattering of a dendritic crystal into several pieces after
620   bringing it in contact with 2mm diameter supercooled drop. This observation is of considerable interest, for it suggests that the breaking up of ice crystals that collide and nucleate supercooled drops, may play an important role in increasing the concentration of ice particles.

From his lab experiments, Gold (1963) found that the surface temperature shock of 6ºC is necessary to produce the stress required for ice cracking.

625   Using thermoelastic theory, King and Fletcher (1976a) calculated thermal stresses in idealised ice shapes on impact with liquid droplets, when a small area was warmed to 0ºC. They concluded that a thermal shock mechanism is unlikely to be responsible for SIP at temperatures T_a>-5ºC.

King and Fletcher (1976b) conducted a series of experiments to study the effect of thermal shock on cracking of macroscopic polycrystalline spheres (D~2-3cm) and thick (Δh=1.7cm) and thin (Δh=1-2mm)
630   cylindrical plates with diameter b=5cm at temperatures down to -40°C. The cracking probability of ice plates versus temperature was studied for several ratios a/b=0.2, 0.4 and 0.6, where a is the diameter of the heated area. In such arrangement the thermal shock is expected to be more severe than that experienced by microscopic ice crystals during riming. Depending on the thicknessof the plates and the ratio a/b,the cracking temperature threshold varied from -5°C to -35°C. None of the plates fragmented or separated. King and
635   Fletcher concluded that thermal shock is unlikely to be an important ice multiplication mechanism at -5°C.

Experiments using thermal shock with macroscopic slabs and spheres by King and Fletcher (1976b) are not fully scalable down to microscopic monocrystalline ice particles. Moreover, the conclusions obtained in their studies are not consistent with the laboratory observations of Gold (1963), Dye and Hobbs (1968) and Hobbs and Farber (1972).

640   Despite the seeming feasibility of this SIP process to occur in natural clouds, this phenomenon got little attention from the cloud physics experimental community. Based on the previous experimental and theoretical studies, the efficiency of ice fragmentation due to thermal shock is expected to primarily depend on the air temperature, droplet size, ice crystal size and its habit. Unfortunately, none of these dependencies have been addressed experimentally. Therefore, the effect of thermal shock ice fragmentation on SIP remains
645   inconclusive.

6. Fragmentation of sublimating ice particles

Ice particle fragmentation and formation of secondary ice may occur during sublimation in undersaturated
650   cloud regions. Oraltay and Hallet (1989) studied evaporation of ice particles suspended on a fiber at a wind
speed emulating their fall velocity. They observed the fragmentation of dendritic ice shapes at subfreezing
temperatures only when relative humidity over ice was $RH_i<70\%$ and ventilation velocity 10 to 20cm/s.
However, no sublimation breakup was observed for columnar and plate-like crystals. Dong et al. (1994)
studied fragmentation of rimed ice and needles at $50\%<RH_i<90\%$, $-18°C<T_a<-5°C$ and ventilation speed
655   ~1m/s. In their experiments, they found that a few mm long rimed ice may generate up to 100 fragments
during evaporation at $RH_i<70\%$ within 1-2 minutes. Bacon et al. (1998) studied fragmentation of sublimating
ice particles suspended in electrodynamic balance inside a thermo-diffusional chamber at $85\%<RH_i<100\%$
and $-30C<T_a<0°C$. The observed fragmentation tended to affect prolate ice particles with an aspect ratio higher
than 3. An example of images of sublimating ice particle is shown in Fig.13. All three studies concluded that
660   breakup rates depend on temperature and humidity, but largely on the initial shape of the ice particle.
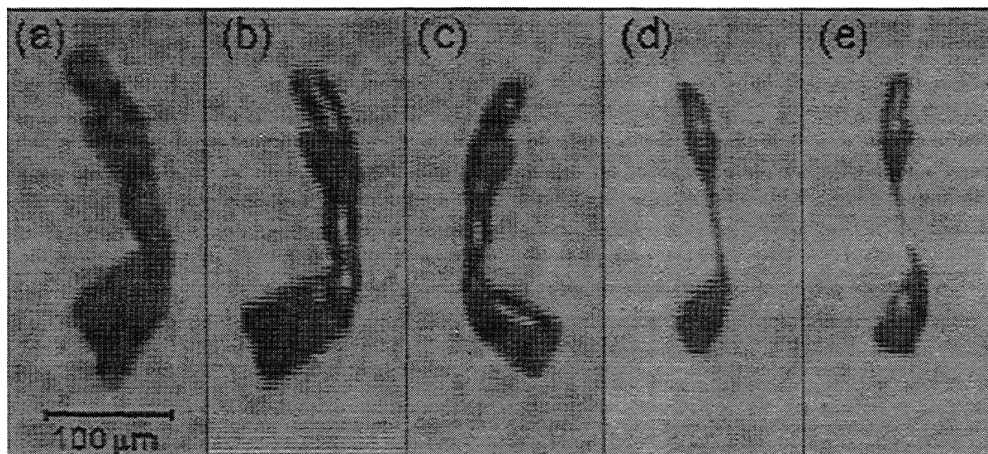
Figure 13. A sequence of images of a sublimating ice particle levitated in an electromagnetic balance. Time before
breakup (a) 6 min, (b) 4 min, (c) 2 min, (d) 20 s, and (e) at the moment of breakup (Bacon et al. 1998).

665   During in-situ observation of metamorphosis of shapes of sublimating ice particles in natural clouds,
Korolev and Isaac (2004) came to a conclusion that ice particle fragmentation during sublimation does not play
an important role in SIP.

In order for the ice fragments formed during sublimation to result in ice multiplication, they have to re-
enter back into a supersaturated cloud region. Since small ice fragments have lower terminal fall velocity, their
670   residence time in the undersaturated environment may be long enough to result in their complete evaporation
before they can re-enter a supersaturated environment. This appears to be a significant limitation of the SIP
mechanism due to sublimation breakup. This mechanism is also unlikely to explain explosive concentrations

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of small ice crystals frequently observed in convective and stratiform frontal clouds (e.g. Lawson et al., 2017; Korolev et al, 2020).

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7. Activation of INPs in transient supersaturation around freezing drops

Muchnik and Rudko (1961) and Dye and Hobbs (1968) reported observation of a halo of small droplets formed around freezing drop immediately after the moment of its nucleation. Dye and Hobbs (1968) explained the origin of small droplets by the activation of CCNs in the region of high transient[1] supersaturation formed

680   around freezing droplets. After ice nucleation, the droplet surface temperature T_s rises to 0°C. Under the condition that the surrounding air has T_a<0°C, the surface of the freezing drop acts as a source of water vapor to a colder environment. The resulting water vapor diffuses radially outward. Depending on the air humidity, it may create at some distance from the droplet a region with supersaturated air. Nix and Fukuta (1974) developed a theoretical framework for the calculation of the supersaturation field around a stationary freezing

685   drop. They showed that maximum supersaturation increases with the decrease of T_a and the increase of drop size.

Cheng (1970) attempted to explain the origin of small droplets due to their ejection from the freezing drop. However, this explanation was challenged by Hobbs (1971). Rosinski et al. (1972) also described laboratory results refuting Cheng's interpretation of the halo around freezing drops.

690   Later, Gagin (1972) proposed a mechanism explaining SIP due to activation of INP in high transient supersaturation area around freezing drops. He argued that high supersaturation may result in activation of insoluble INPs, which normally do not activate at typical cloud supersaturation levels (SS <1%).

Rosinski et al. (1975) studied activation of silver iodide and soil particles placed on a flat plate at different distances from 2mm freezing drops. They found that silver iodide nucleated as water at temperatures T_a>-

695   9.8°C, and as ice at T_a <-9.8°C. Soil particles with sizes 20μm and 40μm nucleated as water at temperatures -20°C and -16°C, and as ice at colder temperatures, respectively. Rosinski et al. concluded that "production of ice particles by-condensation-followed by-freezing in a parcel of a cloud containing large freezing drops is orders of magnitude higher than by contact nucleation".

Gagin and Nozyce (1984) suspended 1mm-2mm diameter drops inside a gradually cooling chamber. The

700   drops froze at a mean temperature -6.5°C as they contained silver iodide. Complete drop freezing occurred in 5-6 minutes, when the ambient temperature decreased down to -10°C -12°C. From the aerosol in the ambient air they found that during drop freezing - on average 1.6-2.1 ice crystals were activated around freezing drops. The nucleation of ice crystals was attributed to supersaturation sensitive INPs.

Nix and Fukuta (1974) performed calculations for a quasi steady state case, where the transient

705   supersaturation was determined by molecular diffusion. In laboratory experiments of Rosinski et al. (1975) and

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[1] Some studies use the term "transitional".

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Gagin and Nozyce (1984) the transient supersaturation in addition to the molecular diffusion was also contributed by a mixing with a convective flow, induced by the temperature difference between the drop surface and environment. None of the above studies accounted for ventilation effect for free falling drops. In this regard, it was not clear whether the obtained results are applicable to natural clouds, since the mixing will

710 occur in the wake of falling drops, and it will be mainly determined by turbulent mixing.
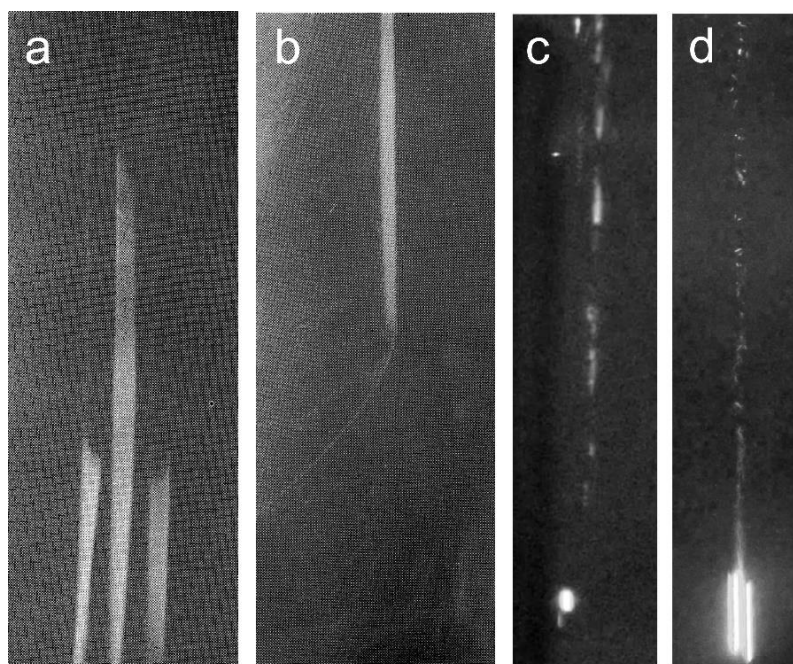
Figure 14. The trajectories of falling drops with D =100μm visualised by nucleation of CCN and INP in the high supersaturation formed around freezing drops at the moment of (a) initiation of freezing and (b) by the end of freezing at ambient temperature $T_a \approx$ -65°C (Iwabuchi and Magono, 1975). Nucleation of water droplets and ice particles in the

715 wake of falling with D≈2mm (c) $T_a \approx$ -20°C, $RH \approx$60%, 0C< T_a<2°C; (d) T_a =18°C, RH=60%, T_d=10°C In these experiments AgI and of the Snomax were used as ice nuclei (Prabhakaran et al., 2020).

Iwabuchi and Magono (1975) in their experiments on freezing electrification documented formation of fog along the trajectories of 90μm-160μm free falling freezing drops at T_a=-65°C (Fig.14ab). They used the fog

720 formation to identify the start and end moments of the drop freezing.

Prabhakaran et al. (2020) conducted experiment with a 2mm diameter free falling drops in the environment with T_a=-18°C and RH=60%-80%. The experiments were conducted using Snomax and AgI aerosols induced in the ambient air. The drop temperatures varied in the range 0°C<T_{dr}<+20°C. The high drop temperatures were used to enhance supersaturation and exaggerate ice nucleation in the undersaturated air. The free-falling

725 drops formed fog trails consisting of activated cloud droplets and ice (Fig.14cd).

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Nix and Fukuta (1974) also pointed out that hailstones during wet growth have a surface temperature close to 0°C, and therefore they may act as a source of high supersaturation. Under such conditions, hailstones may activate many more supersaturation sensitive INPs than a freezing droplet, since the affected volume in such a case will be much larger. Fukuta and Lee (1986) performed calculations of supersaturation around falling

730   graupel with different sizes (2mm, 4mm, 6mm) at different ambient temperatures (-10°C, -20°C, and -30°C). They found that bigger graupel with larger sweeping volume has lower maximum supersaturation. Thus, over 2mm and 6mm falling graupel maximum supersaturation with respect to water at -10°C, -20°C and -30°C reaches approximately 10%, 40%, 100% and 5.5%, 23%, 35%, respectively. The finding that for falling

735   freezing drops maximum supersaturation is decreasing with the increase of the drop size is opposite to that for stationary drops in Nix and Fukuta (1974).

Chouippe et al. (2019) performed direct numerical simulations of a free-falling ice sphere in a humid air accounting for heat and mass transfer. This study was focused on the study of accuracy of numerical simulation. It confirmed the conclusion obtained in previous studies that supersaturation increases with the

740   increase of the temperature difference $\Delta T = T_s - T_a$. However, the effect of graupel size on supersaturation was not discussed there.

It is worth noting, that high transient supersaturation may form not only over a particle with a surface temperature $T_s > T_a$. Similar effect may also occur over a graupel or hailstone, for which the surface temperature did not reach equilibrium, and remains colder than the ambient air. Thus, Schaefer and Cheng

745   (1971, Fig.1a) observed initiation of ice around a simulated graupel with temperature colder 5°C than the ambient air temperature. Unfortunately, no other details of the experimental setup were available from their work.

The above studies suggest that the activation of INP is expected to grow with the increase of difference between $T_{dr} - T_a$. However, Baker (1991) argued that even if T_a is low enough, the volume around freezing

750   droplet is too small compared to regions with high concentration of ice. Therefore, INP activation in transient supersaturation around freezing drops have a low significance for SIP. This, result is conflicting with the conclusion obtained in Rosinski et al. (1975) and Prabhakaran et al. (2020).

The studies described above provide experimental and theoretical support that activation of supersaturation sensitive INPs in the wake of free-falling freezing drops, wet hailstones or riming graupel is one of possible

755   mechanisms of SIP. Unfortunately, due to limited experimental studies, the effect of INP activation around falling hydrometeors cannot be quantified and employed in cloud simulations. The future laboratory studies should be focused on the quantification of the effect of $T_{dr} - T_a$ of a free-falling hydrometeor on the INP activation.

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8.   Spurious enhancement of ice concentration during sampling

Airborne in-situ measurements is the main source of information about the concentration of ice particles in natural clouds and the environmental conditions associated with SIP. The accuracy of measurements of small ice particles is of great importance for the closure of SIP parameterizations and feedbacking laboratory studies. In this section we discuss the issues of in-situ cloud particle sampling, which may result in artificial enhancements of measured ice concentration.

At the initial stage of regular cloud observations with optical particle probes (Knollenberg 1981) it was found that small ice particles were observed in all ice clouds including precipitating and undersaturated cloud regions where existence of small particles conflicted with their small fall velocity and rapid sublimation, respectively. Such observations required developing additional mechanisms to explain the omnipresence of small ice crystals.

The hypothesis of enhanced ice concentration induced by airborne instruments has been discussed over a long period of time. Larger ice particles may bounce off the forward probe's tips or inlet, and shatter into smaller fragments. After rebounding, the shattered fragments may travel into the sample area and cause multiple artificial counts of small ice. Cooper (1977) was the first to recognize a potential significance of particle shattering and suggested filtering the shattered artifacts based on the characteristically short interarrival times between successive particles passing through the probe's sample volume. Several following works based on comparisons between several airborne instruments (Gardiner and Hallett, 1985; Gayet et al., 1996) or analysis of the particles' interarrival time (Field et al., 2003) posed the question of whether the observed high concentration of ice particles is real or an artifact.

Korolev and Isaac (2005) documented OAP-2DC, OAP-2DP, and HVPS images of fragmented precipitation size ice particles as a direct evidence of the existence shattering. However, it did not clarify the origin of the enhanced concentration of small ice.

Field et al. (2006) applied an interarrival time algorithm to identify and filter out shattering artifacts in OAP-2DC and CIP measurements. It was found that after filtering artifacts, the OAP-2DC and CIP concentrations were reduced by up to a factor of four, when the mass-weighted mean size exceeded 3mm.

Heymsfield (2007), McFarquhar et al. (2007), Jensen et al. (2009), based on the comparisons between different airborne instruments, built up more evidence about the spurious enhancement of concentration of small ice particles.

Despite the growing evidence of the significance of the effect of shattering on ice particle measurements, the shattering hypothesis was not commonly accepted in the cloud physics community for many years. Many researchers argued that shattered particle fragments, after bouncing from the probe's upstream surface, shed along the surface of the arms or inlets, and that they could not travel several centimeters across the airflow at an aircraft speed of 100m/s to reach the probe's sample volume.

Direct experimental support for the shattering hypothesis has been provided by a series of high-speed videos of the ice particles behavior on impact the probe tips at an aircraft speed (Korolev et al. 2011, 2013b). These videos documented that after rebounding from the probe's tips, shattered small fragments can travel several centimeters across the airflow and reach the probe's sample volume (Fig.15).

800 Korolev et al. (2011, 2013a), Lawson (2011), Korolev and Field (2015) showed that the effect of shattering can be mitigated by using both antishattering K-tips (Korolev et al. 2013b) and the interarrival time algorithm (Field et al. 2006). It was also demonstrated that the interarrival time algorithm, when used alone, is not capable of identifying all shattering artifacts. Korolev et al. (2013a) showed that measured concentration of ice particles smaller 200μm can be enhanced due to the shattering effect by up to two orders of magnitude,

805 whereas the concertation of ice particles larger 400μm remains mainly unaffected.

Figure 15. A snapshot from a high-speed video showing a flow of shattered ice fragments rebounding from the hemispherical tip of the OAP-2DC particle probe. The shattered ice fragments deflected toward the sample area (bright
810 vertical band) are counted by the probe and artificially enhance the measured concentration. The video recording was performed in an ice spray at 80m/s in the Cox and Co. Icing Wind Tunnel Facility (for more details see Korolev et al. 2011, 2013ab).

Another source of artifacts in measurements of high concentration of ice particles by optical array probes
815 (OAP), is related to fragmentation of particle images when particles pass through the sample volume close to the edge of the depth-of-field (DoF) (Korolev 2007, Guélis eta al., 2019). A few one-two pixel images resulted from fragmentation of large out-of-focus images have an enhanced artificial contribution into particle concentration due to their very small sample volumes. This is a purely optical phenomenon, and it is relevant only to imaging particle probes. Currently, the problem of fragmented images is recognized by many research
820 groups. One of possible solutions of this problem is the exclusion of the first two or three size bins compromised by the ambiguity of the DoF definition and contamination by image fragments. Due to the extent

that particle counts from the first two or three size bins (smaller than 30 - 80μm depending on the OAP type) may significantly contribute to the ice concentration, a limitation is imposed on the measurements of total concertation of ice particles in SIP cloud regions.

825   These findings brought up a question whether early airborne studies of SIP were contaminated by shattering artifacts, which resulted in an artificial enhancement of the measured concentration of small ice. However, numerous recent in-situ measurements, which employed the antishattering techniques and updated processing algorithms, are in general consistent with the early SIP observations, and they also showed that in many clouds, ice particle concentrations are still much higher than the INP concentration (e.g. Crosier et al. 2011,

830   2014; Crawford et al. 2012; Stith et al. 2014; Lawson et al, 2015; 2017; Lloyd et al. 2015; Lasher-Trapp et al. 2016; Keppas et al. 2017; Ladino et al. 2017; Korolev et al. 2020; and others).
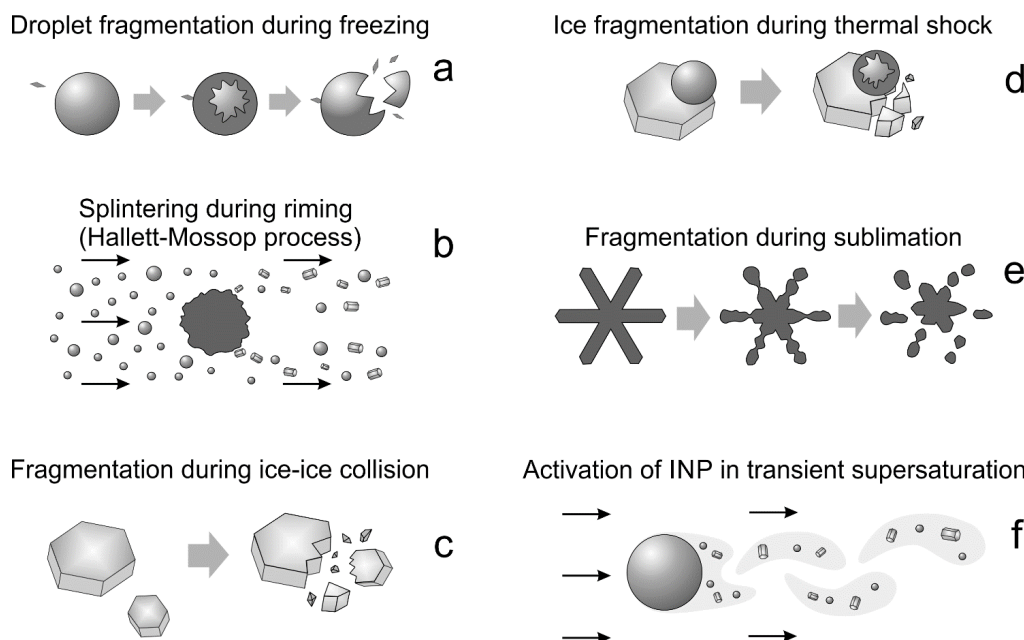
9.   Concluding remarks

9.1   General comments

835   Figure 16 shows a summary diagram with conceptual models of six SIP mechanisms discussed above.

The analysis provided in this work shows that the experimental studies are distributed quite unevenly between different SIP mechanisms. Most of the SIP experimental studies are associated with examining the mechanism of droplet fragmentation during freezing (32 publications[2]). A large number of laboratory works are dedicated to studying the rime splintering (HM-process) (22 publications). The other four mechanisms

840   received far too little attention from the lab research community: ice-ice collisional fragmentation (2 publications), thermal shock fragmentation (3 publications), sublimating ice fragmentation (9 publications); INP nucleation in transient supersaturation (4 publications). Even though none of the above mechanisms have a competed quantitative theoretical description, there is a reasonably good understanding of what physical processes are involved in these mechanisms with the exception of the HM-process. The situation regarding the

845   HM process is contradictory: on one hand, the parameterization of the HM-mechanism is widely used in cloud simulations and weather prediction models, on the other hand, there is no clear understanding of the physical processes underlying this mechanism. At the same time, none of the other five mechanisms is employed on a systematic basis in the weather prediction models.

The most striking outcome of this review is the diverse range of results obtained by different research

850   groups for each of the SIP mechanisms. This is one of the major issues hindering the development of parameterizations for numerical simulations.

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[2] A publication is considered related to a specific SIP mechanism if it includes *experimental* results related to this specific mechanism. Theoretical and in-situ observational works were not counted. Note, that some publications were not cited in this work.

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Droplet fragmentation during freezing

a

Ice fragmentation during thermal shock

d

Splintering during riming
(Hallett-Mossop process)

b

Fragmentation during sublimation

e

Fragmentation during ice-ice collision

c

Activation of INP in transient supersaturation

f

Figure 16. A conceptual diagram summarizing six SIP mechanisms (a) fragmentation droplets during freezing; (b) rime splintering (Hallett-Mossop process); (c) fragmentation of ice particles during ice-to-ice collision; (d) fragmentation of ice particles during thermal shock caused by a freezing drop attached to their surfaces; (e) fragmentation of ice particles during their sublimation; (f) activation of supersaturation sensitive INP in the transient supersaturation formed around freezing drops or wet graupel/hailstones.

9.2 Feasibility of SIP mechanisms

One of the important questions related to ice multiplication is whether all six mechanisms can occur in natural clouds.

The review of the lab experiments suggests that the mechanism of droplet fragmentation during freezing may be active across a wide range of temperatures. There is an increasing amount of evidence indicating universality of this mechanism, which may occur in both convective and stratiform clouds.

The rime splintering (HM) mechanisms require the presence of heavily rimed graupel with high fall velocity. Formation of such graupel is most likely to occur in convective mixed phase cloud regions in a quite narrow temperature range -8C<T_a<-3C.

Ice-ice collisional fragmentation requires a large separation of vertical velocities of ice particles to enhance kinetic energy of their collision. The most likely candidates for this process are lightly and heavily rimed ice particles. The formation of graupel usually occurs in mixed phase convective regions. Whether diffusionally grown ice particles may get fragmented colliding with each other remains unclear.

The theoretical analysis of the thermal shock fragmentation (King and Fletcher, 1976a) suggests that it requires precipitation size drops and temperatures colder than -10°C. Such conditions would be relevant for

875   mixed phase convective cloud regions where large drops could be transported by a vertical updraft to levels with cold temperatures.

Activation of SIP due to the fragmentation of sublimating ice requires spatial proximity of undersaturated and supersaturated cloud regions. In this case, secondary ice particles formed in the undersaturated cloud regions can be rapidly transported into the supersaturated regions prior their sublimation. Such conditions may 880   occur in cloud regions affected by entrainment and mixing with out-of-cloud dry air.

INPs activation in transient supersaturation requires precipitation size drops and high supercooling. As indicated above, such conditions are typical for convective cloud regions.

Out of six SIP mechanisms, the droplet fragmentation during freezing and INP activation in transient supersaturation mechanisms appear to be primary candidates for initial production of secondary ice at the early 885   stage of ice formation in convective clouds. The rest of the mechanisms require preexisting aged ice, and they may contribute in the ice concentration at later stages of cloud development.

9.3   The way forward

The large discrepancies within the experimental results obtained by different research groups necessitates 890   the development of laboratory setups that account for a variety of possible parameters that may be implicated in different SIP processes.

It is worth mentioning that the possibility of additional SIP mechanisms beyond the purview of this paper (Fig.16) remains unexplored. In this regard, studying the existence of other SIP mechanisms not described in this study is still on the agenda of SIP investigation (e.g. Knight 2012).

895   Because of the complexity involved in researching SIP, obtaining consistent results from independent research groups is an important task for SIP studies. This would require consolidating efforts across the cloud physics community. Laboratory investigations should go hand in hand with the development of theoretical descriptions of the SIP processes on a microscale level. This will create a foundation for physically based parameterizations for weather and climate models, which is the ultimate goal of all these efforts.

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Competing interests. The authors declare that they have no conflict of interest.

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