Comment on "Review of Experimental Studies of Secondary Ice Production" by Korolev and Leisner (2020)

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Abstract. This is a comment on the article 'Review of Experimental Studies of Secondary Ice Production' by Korolev and Leisner (2020, hereafter termed 'KL2020'), referring to the discussion about ice fragmentation. We argue that firstly, theoretical and modelling studies of SIP are possible even without any further laboratory experiments. Secondly, hypothetically our previous theoretical and modeling studies would still be valid even if all the previous laboratory/field experiments about fragmentation in ice-ice collisions were to be shown to be totally erroneous: these provided a both a theory for cloud glaciation delineating a phase-space for instability of ice concentrations (in 2011) and a universal formulation of fragmentation for any given collision with a sound theoretical basis, being a flexible framework into which future lab observations may be assimilated (in 2017). Thirdly, we argue that the only two lab/field observational studies characterising fragmentation in ice-ice collisions are not so erroneous as to prevent their use in representing this breakup in numerical models, contrary to the impression given in the review. Finally, a scaling analysis suggests that breakup of ice during sublimation can make a significant, albeit lesser, contribution to ice enhancement in clouds.

1 Introduction

The literature of secondary ice production was recently reviewed by KL2020. The focus of their review is on laboratory experiments. It is commendable that their review attempts to re-invigorate laboratory observations of the various types of fragmentation of ice, research that has in the last decade been oriented chiefly towards heterogeneous ice nucleation. Regarding observations of breakup in ice-ice collisions, only two prior lab studies have ever been published quantifying it (Vardiman 1978; Takahashi *et al.* 1995).

However, the unfortunate impression is given to the reader that numerical modeling and theoretical studies of breakup in ice-ice collisions are somehow currently impossible due to the fact that reliable data for laboratory experiments are critically missing at present. Even if the available data were unreliable or somehow unrepresentative of natural clouds, which we argue here is not the case (Sec. 4), some stand-alone theoretical and modelling studies are nevertheless possible by developing a formulation with a framework that solely relies on theoretical studies from classical mechanics of collisions and statistical physics:

- Yano and Phillips (2011) delineated the time-scale of explosive growth of ice concentrations and the conditions for such instability in terms of dimensionless parameters that provide a phase-space of the system.
- The role of stochastic variability among fragmentation events in promoting the onset of instability is elucidated theoretically by means of the Fokker–Planck equation (Yano and Phillips 2016).
- Phillips et al. (2017a) show a formulation based on principles of energy conservation and statistical variability of strengths of asperities, using a recent definition of the coefficient of restitution. This versatile framework was calibrated with the latest lab/field data but exists as a theory independently of such data.

The purpose of the present commentary is to point out some misunderstandings apparent in the review by KL2020, as a result. The focus is on Section 4 of KL2020, which covered this topic of fragmentation of ice by breakup in ice-ice collisions, because that is where we have made critical contributions both in theories and modelling recently. KL2020 have issued a corrigendum for how this section of KL2020 deals with three of our earlier studies about fragmentation of ice are cited in their corrected review (Yano and Phillips 2011; Yano *et al.* 2016; Phillips *et al.* 2017a). Before the correction, a fourth study (Phillips *et al.* 2018) was cited erroneously.

The structure of the comment is as follows. First, to enable the reader to follow the technical details of our arguments defending the studies criticized by KL2020, the next two sections summarize both lab/field experiments observing breakup in ice-ice collisions and how these inspired our subsequent theoretical and modeling contributions. That is followed by detailed comments on the review by KL2020 (their Section 4) and its corrigendum in Sec. 4. This describes how the review by KL2020 made several misunderstandings in their survey of prior lab studies underpinning our published modeling of fragmentation of ice in ice-ice collisions. The present comment describes these errors, which their Corrigendum has not rectified.

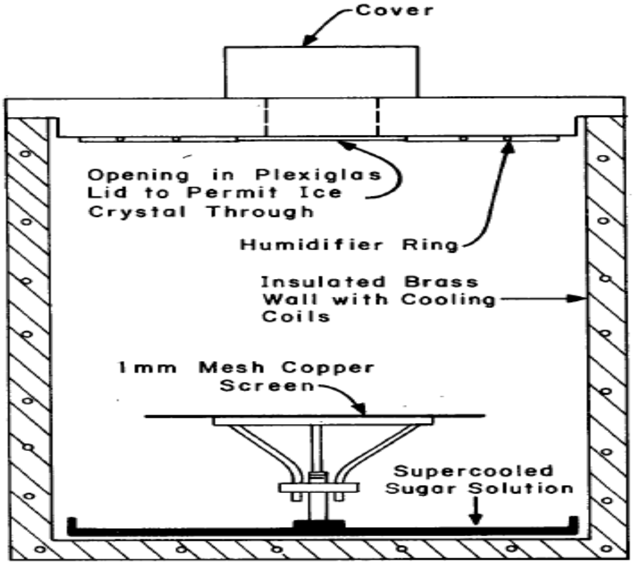

Figure 1: *The portable instrument used to determine the number of fragments from natural falling ice particles. It was placed outdoors on the ground with natural ice falling through the hole in the top and into the chamber, breaking on impact with the screen. From Vardiman (1978). © American Meteorological Society. Used with permission.*

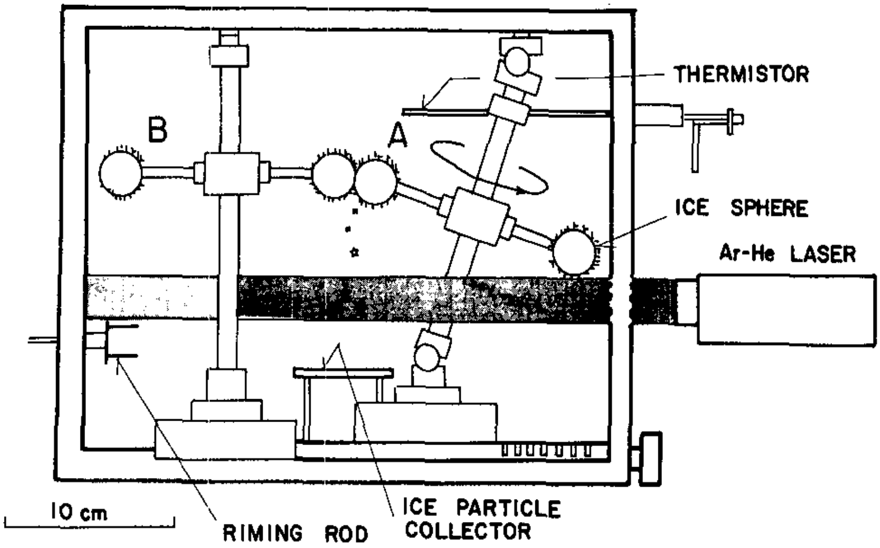

Figure 2: *Experimental apparatus of Takahashi et al. (1995) to observe fragmentation in ice-ice collisions: (A) ice spheres rotated at a tangential speed of 4 m s^{-l}, while (B) ice spheres were stationary. The ejected ice particles were collected on the plate below. Cloud droplets were supplied from the centre of the right wall. From Takahashi et al. (1995). © American Meteorological Society. Used with permission.*

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2 Published Laboratory/Field Observations of Breakup in Ice-Ice Collisions

Hitherto, as far as we are aware, only two laboratory/field studies observing fragmentation in ice-ice collisions have ever been done. First, Vardiman (1978) constructed a portable chamber deployed outdoors on mountainsides in USA during events of ice precipitation (Fig. 1). The chamber had an opening at the top through which natural ice particles (about 0.5 to 5 mm in diameter) fell, descending into the field of view of a camera and impacting a metal plate. Fragments were recorded and collected. The numbers of fragments per collision were measured for various types of ice particle:

- Unrimed dendrites
- Lightly/moderately rimed dendrites
- Heavily rimed dendrites
- Lightly/moderately rimed spatial crystals
- Graupel

For each morphological type, the number of fragments per collision was in the range of 1 to 100 for most sizes, increasing with size. This number was published as a function of momentum change on the metal plate (Vardiman 1978).

Second, Takahashi *et al.* (1995) observed collisions between two giant spheres of ice (2 cm diameter), one rimed (A) and the other unrimed (B), in a cold-box in a laboratory. Figure 2 shows how these were performed. The unrimed sphere was fixed while the rimed one was on a rotating arm. Both particles were intended to be representative of small and large graupel colliding in a real cloud after falling into weak LWC conditions, where the smaller one grows by vapour deposition predominantly and the other by riming mostly, as observed by an airborne video-probe (Takahashi 1993; Takahashi and Kuhara 1993). Those studies in 1993 reported high concentrations of ice observed for such collisions. Takahashi *et al*. (1995) in the lab experiment observed hundreds of ice fragments per collision (N), and provided a reduced estimate of N for natural graupel of more common millimeter-sizes (about 50). Takahashi *et al.* (1995) did this rescaling according to the peak collision force for the fallspeed difference at such natural sizes (0.5 and 4 mm). However, as was later evident from our formulation by Phillips et al. (2017a), (Sec. 3), Takahashi *et al.* appear not to have accounted for the reduced area of contact at these more natural sizes, possibly resulting in an over-estimate of N by about an order of magnitude.

Thus, both lab/field studies encompass a variety of types of ice morphology and energies of impact. Although perhaps incomplete in terms of representing the full extent of this variety in natural clouds (Sections 4, 5), the alternative option of ignoring such breakup when constructing cloud models would seem absurd in view of the prolific fragmentation they observed.

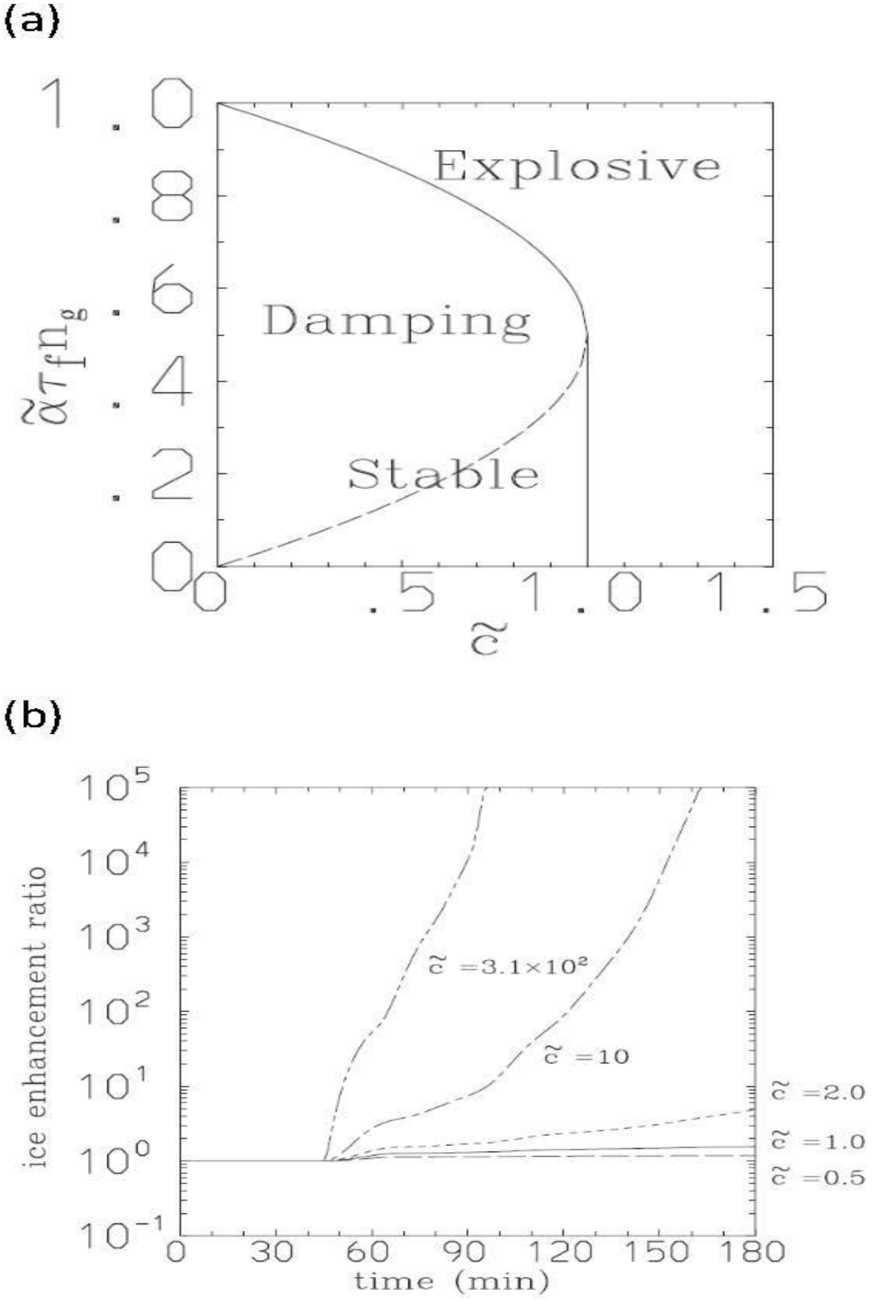

Figure 3: (a) The phase-space of stability for the 0D model of ice-ice breakup for a population of ice crystals, and small and large ice precipitation particles. The multiplication efficiency, $\tilde{c}$, (proportional to the number of fragments per ice-ice collision) and dimensionless initial precipitation concentration are the two axes. In (b) the time evolution of the ice enhancement is shown for various values of $\tilde{c}$. From Yano and Phillips (2011).

3. Our Theoretical and Modelling Contributions

Yano and Phillips (2011) and Yano *et al.* (2016) provided a 0D analytical model of a cloud. Both studies developed a theory of the nonlinear growth of ice concentrations with three species of ice: crystals, small graupel and large graupel. It was found that a sole non-dimensional parameter, $\tilde{c}$, which measures the efficiency of ice multiplication, characterizes the system in its 2D phase-space (Fig. 3a). $\tilde{c}$ is proportional to the number of fragments per ice-ice collision. A systematic investigation of the behavior of the system is performed by varying this nondimensional parameter. A tendency for explosive ice-multiplication is identified in a regime with $\tilde{c} > 1$.

The laboratory experiment by Takahashi *et al.* (1995), (Sec. 2), was referred to in both papers (Yano and Phillips 2011; Yano *et al.* 2016) solely for the purpose of obtaining an estimate for number of ice fragments per collision (N), which is a variable required for estimating the order of magnitude of the nondimensional parameter, $\tilde{c}$. The obtained estimate for a standard case of deep convection is $\tilde{c} \sim 300$, assuming the estimate rescaled by Takahashi et al. ($N \sim 50$). This standard value is far above the critical value of $\tilde{c}$ (unity) required for explosive multiplication. In this respect, those theoretical studies do not depend on any results of laboratory experiments in any critical manner. Figure 3b illustrates how a change of the number of fragments per collision by an order of magnitude only changes the time taken to attain an ice enhancement ratio of 100 by about 30 mins. This standard value is almost 3 orders of magnitude higher than the threshold of $\tilde{c}$ for onset of instability, namely unity (Fig. 3a).

Additionally, Yano and Phillips (2016) further considered a contribution of stochastic fluctuations of the ice-fragmentation number by collision. The paper shows that multiplicative noise effect induced by this stochasticity may lead to an explosive multiplication even under a subcritical state (i.e., $\tilde{c} < 1$). The system was characterized by the time-evolution of a probability distribution function in a phase-space according to the Fokker-Plank equation.

Yano and Phillips (2011) and Yano *et al.* (2016) used the original rescaled estimate from Takahashi et al. (1995) of N (50 splinters) for the standard case involving collisions among graupel particles of more typical natural sizes (0.5 and 4 mm). If this rescaled estimate ($N \approx 50$) is corrected with a value from our formulation in Eq (3) accounting for the reduced area of contact, then only a few splinters per collision is obtained. This implies that $\tilde{c} \sim 10$ which is still supercritical. However, the purpose of both of our papers (2011, 2016) was not to quantify the physics of breakup in a given collision at the particle scale; rather, they merely explored the general theory of overall effects on any cloud.

Next, Phillips *et al.* (2017a) created a formulation to predict the number of fragments of ice emitted in any collision of two ice particles, as a function of their sizes, velocities, temperature and morphology. It was based on the robust formulations from theoretical statistical physics considering energy conservation at the particle-scale. For a pair of colliding particles that initially are not rotating:

$$K_0 = K_1 + \Delta S + K_{th} \qquad (1)$$

Here K_0 is the initial CKE, while K_1 is the final kinetic energy of the system after impact in the frame of reference of the center of mass, consisting principally of the CKE of the colliding particles, but also their rotational kinetic energy. This coefficient of restitution (Wall *et al.* 1990; Supulver *et al.* 1995) is not restricted to head-on collisions and includes the possibility of oblique collisions with rotation afterwards. This, together with observed statistics of surface asperities, led to the formulation:

$$N = \alpha A(T, D \ldots)\left(1 - \exp\left[-\left(\frac{CK_0}{\alpha A(T,D,..-)}\right)^{\gamma}\right]\right) \qquad (2)$$

Here, N is the number of fragments per collision, K_0 is the initial collision kinetic energy (CKE) and α is surface area (equivalent spherical) of the smaller particle in the colliding pair, while $A(T, D \ldots)$, γ and C are empirical constants expressing how fragility depends on ice morphology and ambient conditions. Figure 4 shows how the general mathematical form of Eq (2) is consistent with independent data from Vidaurre and Hallett (2009) for a wide range of sizes at a given speed (130 m/s).

The empirical constants of the theory (Eq (2)) were constrained for observations of graupel-graupel collisions over a wide range of CKEs and impact speeds by the lab experiment from Takahashi *et al.* (1995), (Fig. 5). For other microphysical species, these constants were constrained by observations with a cloud chamber outdoors on a mountainside by Vardiman (1978), (Figure 1), (Sec. 2). Thus, splintering for each permutation of microphysical species in ice-ice collisions between snow, crystals, graupel and hail was predicted in our formulation (Phillips *et al.* 2017a).

Phillips *et al*. (2017b) applied the formulation in the ‘aerosol-cloud model’ (AC) to quantify the role of ice-ice collisional breakup for a convective storm observed by radar and aircraft over the US High Plains (STEPS). Ice-ice breakup generated over 99% of all non-homogeneously nucleated ice particles, and was needed for agreement of ice concentrations predicted by AC with aircraft observations, which were corrected for artificial shattering biases. Only with our scheme for breakup included were various observations (e.g. supercooled liquid water content) of the clouds reproduced. This discovery was never mentioned in the review by KL2020.

Curiously, Phillips et al. (2017b) found that the graupel-snow collisions were orders of magnitude more prolific in generating splinters than collisions among only graupel/hail. Thus, they argued that the 0D analytical model of Yano and Phillips (2011) and related theory of instability were directly applicable to graupel-snow collisions, except with the small ice precipitation

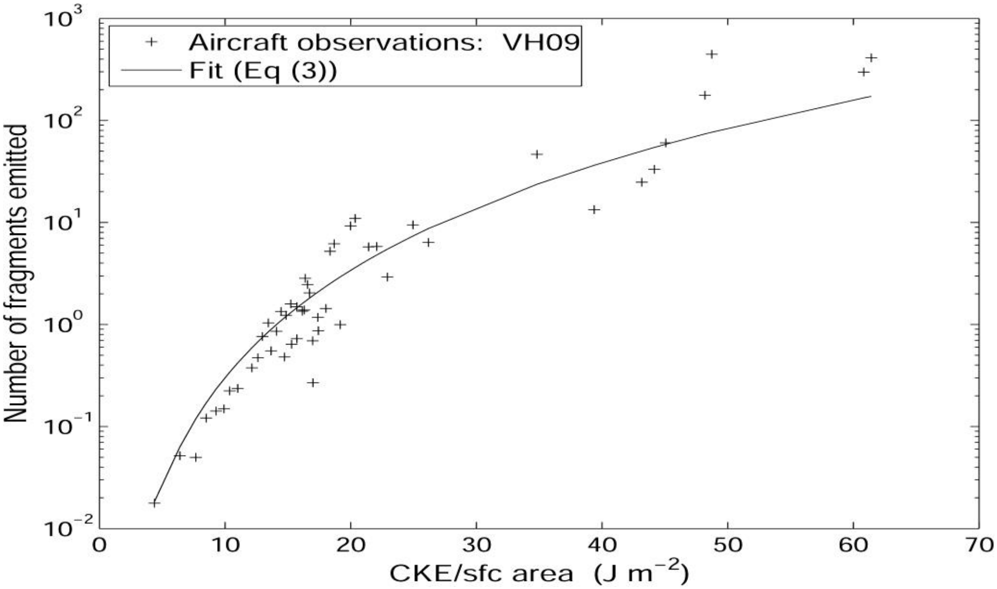

Figure 4: *Measurements by Vidaurre and Hallett (2009) of numbers of fragments from ice crystals of many sizes (10–300 µm) impacting the formvar replicator flown at 130 ms^{-1} through clouds above Oklahoma, plotted as a function of the ratio of CKE to surface area of the ice particle (see Eq (2)). From Phillips et al. (2017a). © American Meteorological Society. Used with permission.*

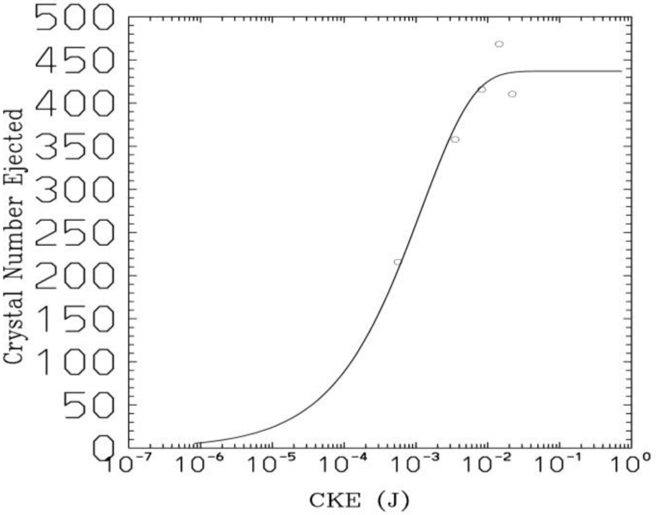

Figure 5: *Measurements of numbers of fragments per hail–hail (2-cm diameter) collision (circles), as a function of initial CKE, which we inferred using Hertz theory from the published data of the experiment by Takahashi et al. (1995). From Phillips et al. (2017a).* © American Meteorological Society. Used with permission.

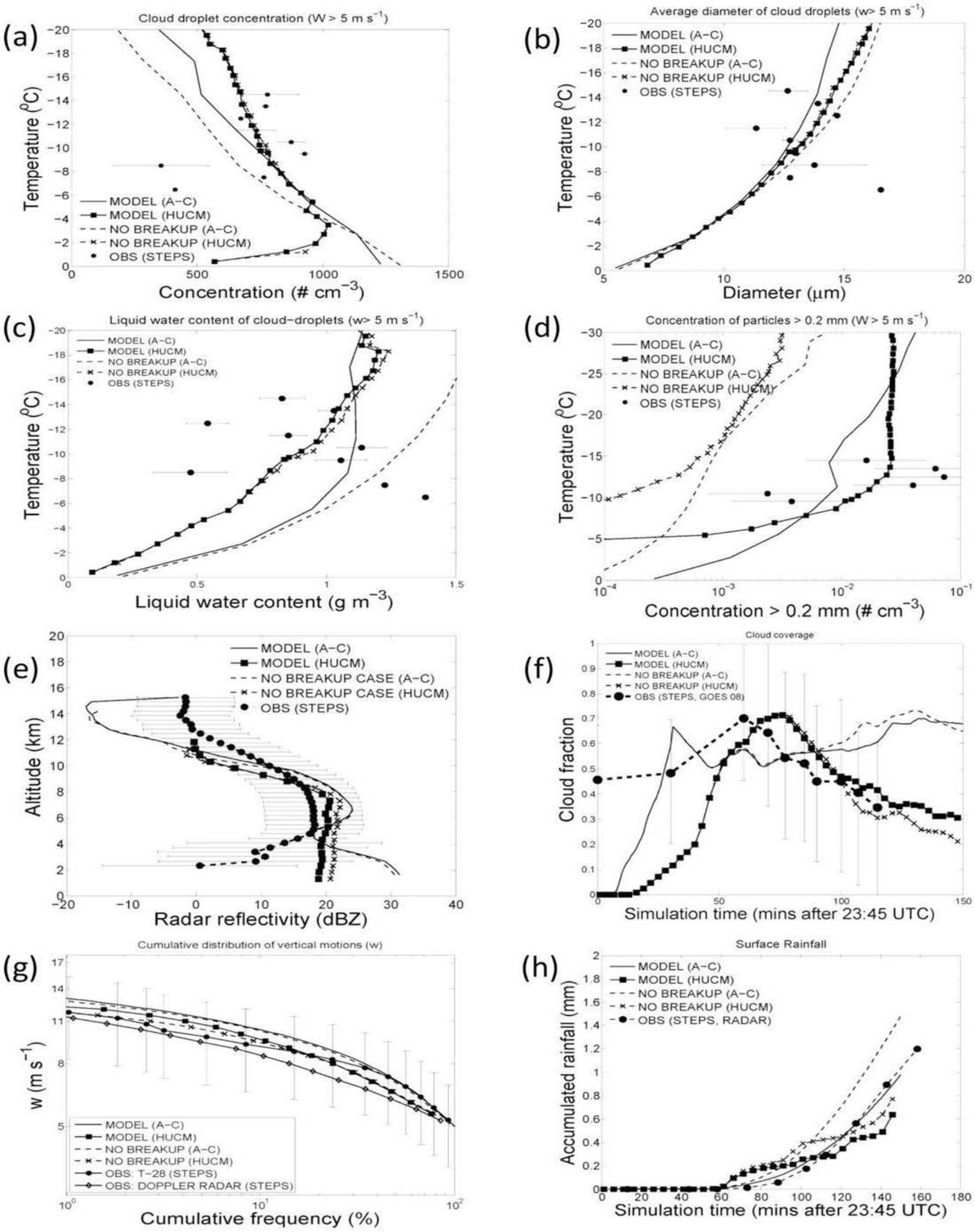

Figure 6: Validation of the aerosol-cloud (AC) simulation with and without breakup in ice-ice collisions, using aircraft data, radar data and satellite data. Quantities shown are cloud-droplet concentrations and mean size, liquid water content, ice concentration, radar reflectivity, cloud fraction, ascent statistics and surface accumulated precipitation from (a) to (h). Ice concentration is shown logarithmically in (d), both with and without breakup. Another simulation of the same case by HUCM model is also shown. Reproduced from Phillips et al. (2017b). © American Meteorological Society. Used with permission.

species defined as "snow" (> 0.3 mm; crystals and their aggregates) instead of "small graupel". Phillips et al. (2017b) estimated the corresponding value of multiplication efficiency for graupel-snow collisions as $\tilde{c} \sim 10$ for the cold-based convective storm (STEPS).

There were no tune-able parameters to adjust in that simulation by AC (Phillips et al. 2017b). Two cloud models with contrasting architecture simulated the same case (a mesoscale convective system 100 km wide). Ours was hybrid bin/bulk microphysics, the other was pure spectral microphysics (Hebrew University Cloud Model [HUCM]). Both models allowed the same conclusion to be reached: only by including ice-ice collisional breakup could the order of magnitude of the ice concentration observed be reproduced. Thus, there was no possibility of tuning the CCN or IN activity. Equally, the vertical velocity statistics observed by aircraft were realistically reproduced and so there was no chance of tuning the instability of the initial sounding (CAPE) to produce the semblance of agreement. Over a dozen quantities were validated against the coincident aircraft, ground-based and satellite observations (Fig. 6), including particle size distributions (Phillips et al. 2017b, their Figure 5).

The aerosol-cloud model took as input the coincident observations (IMPROVE) of the mass concentrations of the 7 chemical species of aerosol. The CCN activity was predicted and then validated by Phillips et al. (2017). The active IN was predicted from the observed dust, soot and organic concentrations by the empirical parameterization (Phillips et al. 2008, 2013), which has been independently validated for various other observations in our earlier papers. The CCN and IN activity spectra predicted by AC were then given to initialize HUCM.

Finally, regarding an entirely different SIP mechanism, by pooling published lab observations of freezing drops in free-fall, a formulation was created for secondary ice production by fragmentation of raindrops freezing quasi-spherically ('Mode 1'), (Phillips *et al.* 2018). By theoretical considerations, an alternative type of secondary ice production from collisions of a raindrop with a more massive ice particle was represented ('Mode 2'). The formulation for both modes was applied in a detailed bin microphysics parcel simulation and shown to reproduce aircraft observations of cloud glaciation for a tropical convective case. Note that the SIP mechanism discussed by Phillips et al. (2018) has no connection whatsoever with the topic that it was erroneously cited for by KL2020, namely breakup in ice-ice collisions (see Corrigendum).

4. Comments on the Review Paper and its Corrigendum

4.1 Breakup in Ice-Ice Collisions

4.1.1 Criticisms by KL2020 of Both Lab/Field Experiments and of our Theories/Modeling

This is what was written by KL2020 (their Section 4) about fragmentation in ice-ice collisions, after corrections from the corrigendum, about both lab/field experiments by Vardiman (1978) and Takahashi et al. (1995), (Sec. 2) and our theoretical works:

> *"Collisional ice fragmentation was also studied theoretically by Hobbs and Farber (1972) and Vardiman (1978). These 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 overestimated.*
>
> *The theoretical framework of collisional fragmentation developed in Yano and Phillips (2011), Yano et al. (2016), and Phillips et al. (2017*[a]*) was calibrated against experimental results of Vardiman (1978) and Takahashi et al. (1995),* [sentence A]. *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 collisional kinetic energy and the surface area of collision of the 2 cm diameter ice spheres also significantly exceed the kinetic energy and collision area of graupel whose typical size is a few millimeters. Altogether, it may result in overestimation of the rate of SIP, compared to graupel formed in natural clouds.*
>
> *…**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***. [sentence B]*"*

Sentence A would have been more accurate if it had mentioned that Yano and Phillips (2011, 2016) and Yano *et al*. (2016) provided a theory of only cloud glaciation and did not focus on fragmentation at the particle-scale. They did not use data from Vardiman (1978).

Regarding our theoretical and modeling work, contrary to these quoted paragraphs from KL2020, our formulation of fragmentation of any given ice-ice collision (Phillips *et al*. 2017a) is in fact not based on simple curve-fitting of laboratory/field results but rather has a robust theoretical basis (Sec. 3). In fact, our overall theoretical formulation itself is developed in a general sense, as a versatile framework independent of any particular laboratory experiment. The fundamental quantities determining fragment numbers per collision are not merely particle size and temperature *per se*, but rather are the initial CKE and contact area (Eq (1)). Equally, regarding our 0D analytical model of cloud glaciation, the theoretical results reported in

Yano and Phillips (2011) and Yano *et al.* (2016) do not depend on laboratory results in any sensitive manner. A lab observation of N is merely used for an estimate a value of the non-dimensional multiplication-efficiency parameter, $\tilde{c}$, along with several other values so as to illustrate how natural deep convection is in an unstable regime with respect to ice multiplication. That investigation (Yano and Phillips 2011; Yano *et al.* 2016) was not fixed to this estimated value, but instead was performed over the full range of possible values of this non-dimensional parameter ($\tilde{c}$). In short, their review continues to give a misleading picture of our work, even after the corrigendum.

Equally, regarding the lab/field studies of breakup, KL2020 still negatively criticise the experimental results of Vardiman and Takahashi *et al.* (1995), which provided estimates of some parameters in the theoretical formulation by Phillips *et al.* (2017a), even after their corrigendum. In particular, their contentious claim has not been retracted about the impossibility of applying both lab/field studies for any formulation (sentence B). Their statement (sentence B) effectively casts doubt on the integrity of our formulation for no reason. Their negative criticisms of both lab/field experiments (Takahashi *et al*. 1995; Vardiman 1978) are exaggerated, because our theory (Phillips *et al*. 2017a) allows us to relate numbers of fragments to the fundamental determinative quantity, namely initial kinetic energy, for any collision. This allows us to quantify the errors inherent in published measurements from various simplifications in the design of both experiments. The errors, when quantified, transpire to be quite minimal in terms of numbers of ice particles emitted from any collision, for the following reasons.

First, as noted in the above quote, KL2020 criticize Vardiman (1978) for failing to include rotational energy from oblique collisions in their theory. Korolev and Leisner (2020, personal communication) also extend this criticism to include a similar omission in actual observations by Takahashi *et al.* (1995) and Vardiman (1978). Yet it can be shown that the final rotational energy is only a small fraction of the initial CKE. For any sphere of size, R, and mass, M, its moment of inertia is $I = (2/5)MR^2$. Consider a strongly oblique collision between two spheres of different sizes and assuming very frictional surfaces of both. During the rebounding part of the period of contact, the ratio of rotational to translational kinetic energy of the smaller sphere is approximated by that of a sphere rolling on a flat surface: 2/5. At the other extreme, for a head-on collision between such spheres there is zero rotation afterwards. Thus, by this assumption of very frictional surfaces, the average fraction of initial CKE converted to rotational energy, accounting for all possible angles of collision, would be of the order of 10%. In fact, this must be an upper bound (10%), since Supulver *et al.* (1995) observed slow ice-ice collisions of 5-cm ice spheres in the lab and found friction is insignificant, implying little transfer to rotational energy in nature.

Consequently, the error in either the measured or predicted numbers of fragments per collision, introduced by artificially preventing any rotational evasive response on rebound, is minimal. That omission only weakly affects the energy available for fragmentation during impact (Eqs (1), (2)). This available energy is related to the difference between initial and final total kinetic energies. For example, a 10% change (the upper bound noted above) in the initial CKE would correspond to only a 1% change in fragments emitted, in view of our analysis of observations by Takahashi *et al*. at various impact speeds (Phillips *et al.* 2017a), (Fig. 5). Thus, the omission of rotational evasive rebound from oblique impacts in Vardiman's theory, and in either of the two lab/field experiments, cannot seriously have biased the fragmentation per collision.

Equally, the effect from fixing the target (ice sphere or metal target) in both experiments by Vardiman (1978) and Takahashi *et al*. (1995) can be shown to be unimportant for any energy-based formulation of fragmentation such as ours. Essentially, since the target was rigidly fixed by metal fixtures in both experiments, the effect on the collision is *via* a drastic increase of the inertial mass of one of the colliding particles (m_1 and m_2). The "particle" becomes the planet Earth or effectively infinite mass. The general equation for the initial CKE for a relative speed of V (along the line of collision) is

$$K_0 = \frac{1}{2}\frac{m_1 m_2}{m_1 + m_2} V^2 \qquad (3)$$

Fixing one of the particles is expressed by $m_1 \to \infty$ and then $K_0 \to (1/2) m_2 V^2$. As an example, consider two identical particles $m_1 = m_2 = M$ that collide when free. The initial CKE is then $K_0 = (1/4) M V^2$. Now we fix one of the particles, and $K_0 = (1/2) M V^2$. Consequently, the effect from fixing one of the particles on the nature of collision is represented by the CKE being doubled. Whether or not both particles are free does not alter the fact that CKE always provides the initial energy in the centre of mass of the colliding pair, which is the source of energy for deformation (e.g. fragmentation). CKE is always the fundamental quantity. Thus, since the formulation by Phillips *et al.* (2017b) related the number of fragments to the CKE in the lab experiment, this relation is universal to any type of collision irrespective of whether both colliding particles are free.

Indeed, the coefficient of restitution for head-on collisions is observed to be an intrinsic property of the materials of the colliding particles. It is practically independent of their masses. Whether the inertial mass of one of the colliding pair is infinite, by it being fixed, is irrelevant to the value of the coefficient. This is why a fixed ice target sufficed in lab studies to observe the coefficient of restitution for free ice particles in planetary disks by Bridges *et al.* (1984), Hatzes *et al*. (1988) and Sepulver *et al.* (1995). When Takahashi *et al.* (1995) applied their lab data (sphere of 2 cm in diameter colliding with fixed target) to a collision between two spheres that are free of 0.5 and 4 mm, the ratio of the assumed masses is about 1000 and the CKE is practically the same as if one of them had been artificially fixed. Consequently, the fixing of the target in both lab experiments introduces no significant error for our formulation (due to its fundamental basis on CKE) or for Takahashi's estimate for natural collisions between large and small graupel (applied by Yano and Phillips 2011).

Second, it is not true that any "*overestimation of the rate of SIP*" must arise from the fact that "*2 cm diameter ice spheres also significantly exceed the kinetic energy and collision area of* [most natural] *graupel*", as KL2020 still claim. The formulation of Phillips *et al.* (2017a) represents the fundamental dependence of fragmentation with both CKE and surface area of contact realistically (Sec. 3): the numbers of fragments per collision observed by Takahashi *et al.* (1995) for a wide range of impact speeds (or CKEs) were realistically reproduced by our theory (Phillips *et al.* 2017a), (Fig. 5). The fact that we fitted the theory to the giant spheres of the lab experiment of Takahashi *et al.* (1995) does not make it erroneous for smaller sizes, because our formulation (Eqs (1), (2)) has a sound theoretical basis on the conservation of energy and is universally applicable to particles of all sizes and morphologies. CKE is always the fundamental quantity irrespective of the nature of the collision and the coefficient of restitution (defined in terms of energy) is an intrinsic property of the materials comprising the particles, as noted above.

Third, although the unrimed ice sphere in each collision observed by Takahashi *et al.* (1995) would be expected to have a "*fragile*" surface, they selected this combination of surface morphologies (rimed and unrimed, depositional and riming growth respectively) because they judged it to be representative for real in-cloud situations of ice multiplication that they had observed directly. Previously, fall-out of large graupel into regions of weak LWC with small graupel had been observed *in situ* by Takahashi (1993) to generate copious ice crystals—with a novel video probe. High concentrations of ice crystals co-located with graupel had been observed in the mixed-phase region of deep convective clouds with the video probe flown on a balloon by Takahashi and Kuhara (1993), providing indisputable evidence of ice morphology and size. Such measurements by balloon are without the artificial shattering biases afflicting some airborne probes.

Consequently, Takahashi *et al*. (1995) argued that, in natural clouds, small graupel after forming through dominant riming growth can encounter predominant vapour growth when falling into such weak-LWC regions with LWC less than 0.1 g m^{-3}. Even if such parts of any convective cloud are limited, the in-cloud motions must tend to mix the crystals throughout most subzero levels of the cloud over time. The inherent nonlinearity of ice multiplication occurring by such breakup, involving growth of splinters to become ice precipitation by a positive feedback (Yano and Phillips 2011), must make such active parts of the cloud disproportionately influential compared to their volume.

In summary, both lab/field observational studies of breakup in ice-ice collisions are not so erroneous as to prevent their application in the simulation of ice multiplication in natural clouds.

4.1.2 Other Possible Criticisms of Both Lab/Field Experiments

Since sentence B (above quote) is such a dismissive and contentious claim, it is valid to ask what other criticisms could be made to support it beyond those expressed by KL2020. We identify here a few possible criticisms and then argue that they too lack credibility.

First, it could be argued that somehow the weight of the fittings supporting the moving ice sphere in the Takahashi *et al*. (1995) experiment must have biased the measured fragment numbers. Indeed, inspection of published diagrams of their apparatus reveals a third ice sphere on the opposite end of the metal nearly horizontal rotating bar (about 13 cm long, 3 mm wide) for the main moving rimed sphere (Takahashi *et al*. 1995), (Fig. 2). It has the same size as the other two spheres but does not collide when they do. We estimate that altogether the moment of inertia for the main rotating sphere ("A" in Fig. 2) about the central axis supporting the bar, was approximately doubled by including this extra sphere and the weight of the moving fixtures. Since the total initial CKE equals the rotational kinetic energy of the ice sphere plus attachments (proportional to this moment of inertia) turning on the axis (almost vertical in Fig. 2), the initial CKE is roughly doubled by the extra mass. Hence, the extra weight for the rotating sphere acts to bias the measured numbers of fragments by only about 10% in light of observations by Takahashi *et al*. (1995) at various impact speeds (Fig. 5).

Equally, it would be wrong to suggest that our formulation somehow did not consider rotational effects or that it is somehow ill-posed by not treating rotation explicitly. Rotational KE is included in Eq (1) underpinning our formulation and is only a small fraction of the total KE in any impact, as proven above.

Second, there is the criticism originally made by Phillips *et al.* (2017a) of the Vardiman (1978) experiment: ice particles were weakened by sublimation prior to collection outdoors, as they fell through the ice-subsaturated environment below cloud-base into the chamber on the mountainside where they broke up (Fig. 1), (Sec. 2). Phillips *et al.* reported that the prediction of N from Eq (2), which for graupel-graupel collisions is fitted to only the Takahashi *et al.* (1995) observations, would be much lower (by about half an order of magnitude) than if Eq (2) were fitted only to observations by Vardiman (1978) for collisions of graupel, for the same size (the largest he observed). Yet in reality this was never a grave problem for our formulation because Phillips *et al.* (2017a) then applied a correction factor to the fragility coefficient, C, in Eq (2) to yield a match at this size between both predictions. Phillips *et al.* then applied this same correction factor to the formulation for other microphysical species (graupel-snow and graupel-crystal collisions) when constraining them with the Vardiman (1978) data alone. Prior sublimation artificially boosting the observed fragmentation of natural ice falling onto the mountainside was invoked as the reason for this empirical correction by Phillips *et al.* (2017a).

Third, it could be argued that somehow the variation of rime density was not explored adequately in both lab/field experiments. Cloud-liquid properties and the impact speed of cloud-droplets when accreted were variables not varied by Takahashi *et al.* (1995). However, the standard conditions for riming in the experiment by Takakashi *et al.* (see Corrigendum of KL2020) are representative of those in which large graupel are created in clouds. When the duration of vapour growth and riming of both ice spheres was varied from 5 to 15 mins, the number of fragments measured by Takahashi varied by a factor of about 2.

Again to conclude, contrary to sentence B of the review, in view of such errors, the measurements of fragment numbers in ice-ice collisions by Vardiman (1978) and Takahashi *et al*. (1995) are not so defective or unrepresentative as to render them useless for estimating coefficients in a theoretical formulation. Naturally, these coefficient values can easily be corrected as more accurate laboratory measurements become available in future, but without changing the formulation itself in any manner.

4.2 Sublimational Breakup

On the topic of sublimational breakup, KL2020 conclude that "*this mechanism is also unlikely to explain explosive concentrations of small ice crystals frequently observed in convective and stratiform frontal clouds*". The reason given is that ice fragments may disappear by evaporation before they can be recirculated. Although that is indeed a major limitation, in reality during descent there must be continual emission of fragments and their continual depletion by total sublimation. This causes a dynamical quasi-equilibrium with an enhanced fragment concentration. So any mixing of downdraft air into the updraft will transfer air with the enhanced ice concentration into the updraft. Also, the entrainment of dry air and turbulence can cause a subsaturated environment, leading to the breakup, followed by possible mixing of fragments into convective ascent.

One can perform a scaling analysis: Oraltay and Hallett (1989) observed rates of emission of the order of $F \sim 0.1$ fragments per second per parent dendritic crystal (a few mm) initially, during sublimation at relative humidities with respect to ice of about 70% or less. Such humidities would be attained in an adiabatic parcel descending at about 2 m/s from about -15 degC initially with about $n = 3$ L^{-1} of crystals initially (2 mm). If each fragment takes a minute (τ) to disappear by total sublimation, then the equilibrium number of fragments per parent particle is $F\tau \sim 0.1 \times 60 = 6$. Dong *et al.* (1994) observed rates of emission of $F \sim 0.3$ fragments per second per parent graupel particle (a few mm) initially. In a similarly subsaturated downdraft, there would be an equilibrium number of fragments per parent of $F\tau \sim 20$.

The pivotal point here is that such a quasi-equilibrium concentration is maintained throughout the entirety of the subsequent descent after being reached. Thus, any recirculation of downdraft air into the surrounding convective ascent would transfer air enriched in fragments for their subsequent vapour growth and survival—*irrespective of the timing of this recirculation during the descent, regardless of whether the prior descent is shallower or deeper*.

5. Conclusions

The review of secondary ice production by KL2020 (their Section 4) continues to depict our work (Yano and Phillips 2011; Yano *et al*. 2016; Phillips *et al.* 2017a) in a distorted manner, even after their Corrigendum. In particular, KL2020 made a contentious claim (sentence B above; Sec. 4) about the impossibility of developing any model formulation based only on the two existing experimental datasets of breakup in collisions of ice (Vardiman 1978; Takahashi *et al.* 1995). That claim is false and arose because KL2020 did not appreciate that our formulation of such breakup is based chiefly on an energistic theory coming from theoretical physics that is universally applicable to all collisions (Phillips *et al.* 2017a). The truth is that this formulation can be delivered even without quoting the results from any laboratory experiments. KL2020 then supposed that any unrepresentative aspect of the collisions in the experiment will somehow cause problems for calibrating the formulation, when in reality the formulation is based on such fundamental quantities that this is not a problem.

In reality, the two laboratory studies of fragmentation are not so erroneous as to prevent their use in calibrating theories of fragmentation in any ice-ice collision, such as ours. As noted above, our formulation does not necessarily require any such calibration. As far as we are aware, the only major issue with both lab/field experiments is a possible bias from sublimation of natural ice particles outdoors prior to their fragmentation observed by Vardiman (1978). But Phillips *et al.* (2017a) knew about this possible bias and corrected for it when calibrating their theory (Sec. 4.1.2). Anyway, the bias was not enough to alter the order of magnitude of N. It is a moot point whether the supposed sublimational weakening of these observations might actually have been representative of natural graupel aloft within clouds, since episodes of subsaturation with respect to ice are likely along the trajectory of any graupel particle while in-cloud.

More crucially, the errors in the breakup rate per collision from the formulation, quantified by Phillips *et al.* (2017a) as a factor of about 2 or 3, soon become immaterial in the context of explosive multiplication of ice concentrations in a natural cloud (Yano and Phillips 2011, 2016). Whether the number of fragments is under- or over-estimated by an order of magnitude does not alter the fact that explosive growth of ice concentrations by orders of magnitude will occur anyway (Sec. 3; Fig. 3).

For any ice multiplication somehow involving mixed-phase conditions (e.g. growth of fragments by riming to become graupel), the explosion occurs until an upper limit is reached (related to onset of water subsaturation), with little sensitivity to the breakup rate. Both published lab/field experiments we used are sufficient to allow a formulation that produces a simulation of observed cloud properties in agreement with aircraft observations of ice concentrations and many other related properties (Phillips *et al.* 2017b).

Finally, the possibility of sublimational breakup contributing to observed ice enhancement cannot be dismissed as easily as the review paper suggests. In reality, a dynamical quasi-equilibrium is established between emission and total sublimation of fragments, so that any region of sustained subsaturation with respect to ice will develop an enhanced ice concentration persisting throughout the descent. The enhanced ice can then be transferred into regions of ascent subsequently for growth. An order of magnitude of ice enhancement is possible from sublimational breakup in convective downdrafts and in their vicinity within the cloud. This quasi-equilibrium ice concentration was overlooked by KL2020.

In summary, the only two lab studies about breakup in ice-ice collisions hitherto (Vardiman 1978; Takahashi *et al.* 1995) are not so erroneous as to prevent their use in treating this process both in theoretical studies and numerical modeling of clouds, contrary to the claim by KL2020.

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