Reply to the reviewers, round 2

We greatly thank the reviewers and editor for their time evaluating our manuscript in this second round. In this round, we followed all of the suggestions. To help show exactly how we followed the suggestions, see the details below. We list the suggestions, and follow with our changes. These changes are marked with a "→" and the new text is highlighted yellow (deletions are not shown).

In addition, we carefully reread the entire manuscript, finding small changes such as an added word or two here and there, or a change in wording to improve consistentency, to help clarify the text.

Anonymous Referee #1

1) The length: Although the original manuscript was too long for the publication as an original article, now the revised manuscript becomes significantly shorter. Some people may think still lengthy, the present length would be within the acceptable range. I understand the authors' thought that they want to include other secondary habits in this paper. Then I leave the decision about the length to the editor.

→Thank you. We understand the difficulty of reading long papers. On the other hand, if organized well, the long paper can be easier than trying to keep track of two or three separate papers that overlap. Our aim is to reduce the overall length (from having several medium-length papers) and have all the connected material together. So, we tried to make the organization much clearer. It seems we have at least partly succeeded.

2) The authors' main point and the formation mechanism of the air pockets: Now I understand the authors' main point: the lateral-type growth has not been considered in ice-crystal growth models. Since the main point was not clearly explained in the original manuscript (it was fully impossible for me to find the point), I misunderstood that the authors' main point is the novelty of the lateral-type growth: the authors claim that the lateral-type growth cannot be explained by the traditional concept. Then I could not agree with this in my last review.

The addition of the section "2 Background" is also helpful in avoiding the wrong impression that the authors do not fully understand the fundamental growth mechanism of crystal growth.

My misreading of the original manuscript was also due to the usage of the words "lateral growth" and "lateral-type growth" without the careful definitions. I am a crystal growth physicist working outside the field of atmospheric science. In my field, "lateral growth" explicitly means the "lateral growth of elementary steps (and also that of bunched steps)" and does not mean "the areal growth on fully facetted faces". Without the definitions newly added at the end of the section 2, I could not understand the authors' meanings. The words "lateral growth" and "lateral-type growth" still remain throughout the manuscript. I believe that many crystal growth physicists will have the same

misreading. Then I strongly recommend that the authors should replace these words with a different word newly prepared, such as “lateral-areal growth” or “lateral-protrusion growth”.
→We changed the term in the title, abstract, introduction, and a few other spots to "lateral facet growth". This should be sufficient to prevent confusion with lateral growth of steps (which is similar, particulary with macrosteps). In some places, we have simply "lateral growth", but the reader has already been told it is about the spread of facets so the distinction should be clear throughout the text.

3) The “disorder of the surface” appeared at the end of the third paragraph of the section 2 Background: Here, the usage of the term “quasi-liquid layers” makes many readers easier to catch the point.
The following is not my review comment but my personal comment unconnected with this review: a recent review article (Y. Nagata, et al., "The surface of ice under equilibrium and non-equilibrium conditions", Accounts Chem. Res., 52, (2019) 1006-1015) will be also helpful to clarify the situation of ice surfaces; this article demonstrates that top-most bi-layers of ice basal faces are disordered above -90°C (and second-top-most bi-layers are also disordered above -16°C), however such disordered layers grow fully in the “layer-by-layer manner” like elementary steps.

→This is a good study to reference, and we agree that it is better to use the common term QLL. As a result, we rewrote the section as

"The BCF model of surface diffusion assumes that the mobile surface molecules are sparse and non-interacting. For ice, this assumption is suspect over much of the atmospheric temperature range. Specifically, the ice–vapor interface is widely thought to contain significant disorder, a phenomenon also called the quasi-liquid layer QLL (e.g., Rosenberg, 2005). A recent study finds that this "layer" is limited to two ice bilayers (~0.74 nm) below −2 °C, and less than half that below −16 °C (Nagata et al., 2019). Despite this layer's thinness, such a surface still deviates greatly from the BCF assumption. Nevertheless, the BCF model is often …"

Anonymous Referee #2

Summary and Main Comments:

This paper reports on measurements of the growth of single faceted ice crystals grown on a capillary in a new cryogenic chamber. The chamber allows for crystals to undergo growth and sublimation cycles with imaging that has enough detail so that approximate rates of growth can be derived from the data. The data show clear indications of the formation of entrapped air pockets in crystal corners and edges among other interesting features. These pockets appear after periods of sublimation. While the appearance of these pockets has been noted in prior measurements and observations, the authors of this article provide an explanation for the existence with of these pockets with a theory of lateral facet growth by protrusions driven by the flux of molecules across an adjoining facet. The authors show that a physically plausible model of protrusion growth driven by this adjoining flux of mobile surface molecules can explain the rate of lateral facet spreading derived from the data. The authors then make use of theories of lateral growth, and protrusion growth in particular, to provide qualitative explanations (or perhaps hypotheses) for the development of various secondary habits of ice crystals. This article is the first attempt (that I am aware of) to quantify and explain the lateral growth of crystal facets. At present, there are no quantitative or qualitative models of lateral growth used in the atmospheric sciences. Ice crystal growth models currently used in the atmospheric sciences fit into only two categories: (1) Models that are designed for growth that is normal to the facet. In other words, steps formed by nucleation or by outcrops of dislocations propagate parallel to the facet, causing the facet to grow outward, normally (what the authors refer to as normal growth). These models are used to study the development of single crystalline habits, and they are sometimes used to understand laboratory growth data. (2) Models that treat the growth of ice as if the surface doesn't matter. These models use the capacitance framework, which is not strictly appropriate for faceted growth. These models have been used to interpret laboratory growth data and are used ubiquitously in atmospheric cloud models. Hence, and as the authors themselves point out, there has been no focus in the atmospheric science community on facet growth that is lateral, however it is clear that this sort of growth must be important. For instance, facets must develop over time anytime an ice crystal nucleates either from a frozen droplet or from a solid aerosol nucleus. This process must include the lateral spreading of facets, a process for which we have very little data and no quantitative models. One can easily imagine where lateral spreading may be important: There are numerous laboratory measurements of the growth rates of ice crystals that begin from a nucleated ice particle. These crystals all probably undergo a period of facet development where lateral spreading, and perhaps protrusions, are likely important. However, lateral growth is never considered when laboratory growth data are interpreted (because, of course, no model of this sort of growth exists at the present time). One can also imagine that lateral growth is important in the modeling of atmospheric clouds: The overall mass growth rates of crystals that are growing laterally are likely quite different than normal growth of crystal facets, and probably very different than the capacitance model growth rates. Substantial differences in the crystal growth rates would naturally lead to impacts on model simulated cloud properties including numbers of nucleated ice crystals, and the mass and thermal energy budget of a cloud layer (through latent heating and crystal sedimentation out of the cloud layer). At the present time, the community lacks measurements, ideas, and theories (even simple ones) to advance the way we think about growing ice crystals and the impacts they may have on clouds. This paper is a nice first step in examining lateral growth and its potential impacts on a variety of complex crystal forms, and I think the paper will stimulate the thinking of those interested in advancing our methods of modeling ice in the laboratory and the atmosphere. I am therefore eager to see this paper published in some form, and I would suggest minor revisions: The paper is quite clearly written and is well argued within the constraints of the available data. While the paper is shorter and clearer than the original discussion paper (I perused this paper as well during my review), and the science appears quite sound, I do have a number of suggestions and questions (see below) related to the presentation of the material, and this is the reason for my recommendation of minor revisions. The above summary of the paper is, of course, my current understanding of the material and I hope that I have not misunderstood the authors' ideas and intent.

→Thank you for this nice summary. It provided useful ideas for revisions made to the introduction sections as mentioned in (1) below.

Enumerated Comments:

(1) General comment on the introduction/background: While the introduction is quite clear and well written, I do think it may be hard for those who are not quite familiar with the theories of ice crystal growth to place the results here into an atmospheric context. Ice crystal growth theorists and laboratory scientists who measure the growth of crystals will probably be able to grasp the concepts presented in the present paper, but those outside of these areas may have more difficulty even though the material is of general interest (in my view). Perhaps adding a few sentences that place these results into a broader atmospheric context would be worthwhile. I do not think that adding this is critical to the paper, it is a suggestion that may help interested readers see the possible implications of these results.

→We added a more general introduction to snow and ice as a new first paragraph, with emphasis on factors that may (eventually) involve applications of the present research. Thus, the entire first paragraph is new, with the old first paragraph shifted to being the 2nd paragraph etc.

(2) Line 18, pg2: I may have missed it, but I do not think that I saw the definition of the initialism "BCF" given earlier in the text.

→We reworded it to show the source of the initials: "The most widely used model for the growth of crystal faces from the vapor is the "BCF" model (Burton, Cabrera, and Frank, 1951; see Woodruff, 2015, for updates and history)."

(3) Line 33, pg2: "all thick surface regions leading growth" is a little awkward. I would suggest rewording.

→We reworded and clarified as ", but for vapor growth, the leading fronts (i.e., outermost faces that define the maximum diameter and have the fastest normal growth) are usually facetted. Individual steps, and steps clumped into macrosteps, instead tend to have a rough edge as indicated by their curved perimeter (generally circular or spiral). Also, when the leading front is very thin, it may appear rounded."

(4) Line 34, pg2: "tend to have a rough edge" perhaps add "indicated by rounding", as you later point out. I think here you are trying to point out that vapor grown crystals are faceted, meaning that the surfaces are not "rough" but that individual steps can be rough as indicated by their rounded in appearance.

→We revised as "Individual steps, and steps clumped into macrosteps, instead tend to have a rough edge as indicated by their curved perimeter (generally circular or spiral)." We specified the perimeter to help clarify that the rounding is as viewed normal to the face.

(5) Line 17, pg 3: "Instead, atmospheric ice models usually…" This statement is definitely true for models like Wood et al. (2001), but ice models used for cloud simulations usually do not include any information about the crystal surface. The usual assumption is that the surface is at equilibrium and that no steps exist at all, since they use the capacitance model.

→Good point. We revised the passage, adding more explanation:

"Instead, atmospheric crystal-growth models usually assume a locally uniform vapor density near the step source and allow the vapor density to monotonically decrease or increase across the surface. (Most cloud models use the more extreme simplifications of the "capacitance model", which includes no detail of surface structure and assumes local equilibrium over the entire surface. But the recent work by Harrington et al., 2019 is a welcome exception.) As the crystal shape…"

(6) Line 18, pg 3: Should "density" be inserted in "vapor near the step source"?

→Yes, now added.

(7) Caption of Figure 1: The word "sizes" always seems ambiguous to me. Perhaps "diameters"?

→Yes, and the change has been made.

(8) Figures in general: A number of the figures show crystals grown in various chambers. I think it might be good to provide some more information on the environmental conditions: Temperature is sometimes given, but what about pressure and supersaturation?

→We now provide conditions as far as they are known for all captions.

(9) Line 9, pg4: I would insert "diameter" in the parenthetical "(~20 micrometers)" since Gonda's measurements were of the width of the frozen droplet.

→Yes, we added "diameter".

(10) Line 11, pg4: "…show these edges as rough.." Is this indicated by the fact that they are rounded? It might be worth it to point that out.

→Changed as " Figure 1a–d shows these edge fronts as rounded, indicating rough edge and hence an efficient collector of molecules.

(11) Line 13, pg4: I have real difficulties seeing the pyramidal facets on Fig 1a,b since the image is a little fuzzy. Perhaps an arrow could be used to indicate the location?

→We added arrows and modified the caption accordingly.

(12) Line 19, pg4: "nucleation of new growth layers". Is it possible that the facets are growing by dislocations instead of layer nucleation? Gonda and Yamazaki's (1984) paper shows the growth velocities of the a and c axes of their crystals, and the growth rates are quite close to each other (their Fig. 3). Given that the supersaturation in that case was between 1 and 2% it would seem that the growth would have to have been dominated by dislocations. Otherwise the axis growth rates

would have been different I would think (since the critical supersaturations for the basal and prism faces are around 0.5% and 2% respectively at Gonda's growth temperature of -15C).

→Yes, though still unknown, it seems likely that dislocations would be present on some faces. Our argument remains essentially the same, but to clarify we changed to " Thus, the propagation rate (and nucleation of new layers in the absence of a permanent step source) of surface steps will be reduced until the facet radius **m**–**e** exceeds the surface migration distance x_s."

(13) Figure 2: I very much like this figure. However, later on in the paper you discuss the vapor gradient near the protrusion. If it wouldn't make the figure too messy, it might be good to add in isolines of vapor density. I was able to follow your description of the structure of the contours, but an image would certainly help. Especially for those who are not familiar with the way vapor gradients may change near the surface of a crystal.

→Nice idea. We added an additional sketch to the figure to show qualitative features of the contours. To make better use of the revised figure, we also revised the relevant discussion in section 5.1 where we already referred to Fig. 2.

(14) Line 3-4, pg 6: I personally find it hard to see much in some of the images that Gonda and Yamazaki present in their papers. Would it even be possible to discern small air pockets given the image quality?

→We added "Given their small droxtal sizes and darkness of their images, one cannot rule out the existence of very small pockets, but their results show no indication of pockets of the scale seen in Fig. 1." We also try to clarify a closely related point in section 4.1.1 where we modified the text at the end as "Thus, although the phenomenon can appear on a range of crystal shapes, the corner radius may need to exceed a certain value for the corner pockets to either exist or become resolvable with standard microscopy. "

(15) Line 34, pg 6: Perhaps you could add "in a later study, both of us (JN and BS) began…" This would provide an implicit reference to BS as an author, since the initials BS are not otherwise defined. And this would be consistent with line 31 of the same page.

→Nice idea. We made the change.

(16) Line 2, pg 7: The initialism CC2 should be defined. Are the crystals grown at ambient atmospheric pressure in the new chamber? Can you provide a very brief explanation of how the supersaturation is estimated, since that is required for the growth calculation shown later.

→We revised as "For this work, we used a new crystal-growth apparatus, hereafter CC2, that improves upon the first "capillary–chamber" method in Nelson and Knight (1996)." The added part may help the reader remember the reason for the name CC2.

A few lines down, we added "…valve stopper. Briefly, the vapor source (ice, pure melt, or solution) has a surface area vastly greater than that of the observed crystal on a capillary. Thus, except very near the observed crystal (when air is present), the vapor density throughout the system is the equilibrium value of the vapor source from which we calculate far-field supersaturation. With this system,…"

In the next paragraph, we added "…thus complementing our CC2 results. In both the cloud chamber and CC2 experiments, the crystals grew in an atmosphere of air. Other…"

(17) Line 11, pg 8: "after the sublimation". It seems like the word "period" should be inserted here after sublimation.

→Yes. We added it.

(18) Line 12, pg 10: "unusually thin" Are they usually thicker? If so, how much?

→Unfortunately, we do not have clear sets of images of the more common "thick" cases here (we are preparing a more detailed study of hollows now), so we cannot give a quantitative or specific example. But this is a good point, so we added the sentence "That is, hollows often start by widening with a nearly circular rim shape (e.g., in hollow columns), whereas the hollows that preceded these planar pockets must have instead had a rim shape similar to a thick line segment before closing into pockets."

(19) Line 23, pg 10: "unusual for a crystal grown at such low supersaturation" Can a reference be provided here?

→We added "More typical cases for low supersaturation are shown in Figs. 7,9 and the literature (e.g., Nelson and Knight, 1996; Gonda, Sei, and Gomi, 1984)."

(20) Lines 5-8, pg 11: Using rings to determine the facet spreading is a nice idea. How is the location of each ring determined? It also might be good to provide an error estimate, which can then be used to provide an error estimate for the rate of spreading shown in Fig. 6.

→The estimated ring position was made by eye. We made a few small changes to this paragraph, also explaining as "The positions of these rings, simply estimated by eye, are marked in (f), with the time interval (units of 5 min) between marked positions in the upper right." The error estimates are addressed in the next reply.

(21) Concerning Fig 6: On a first glance, I thought that the partial grid behind the data points were actually error bars! However, it would be good to provide some estimate of the error. Since the supersaturation is used in the theoretical calculations, an error estimate on the supersaturation and the calculated growth would be good as well. Finally, the title of the figure led me to believe, initially, that the basal face radius was plotted, but it appears that this is a plot of the ring radius divided by the actual crystal radius. If this is correct, then you may want to clarify the title and, perhaps, use this as the y-axis label instead.

→We made most of these changes. We added error bars on the measurements. For the calculations, the rates for I and III are so small in comparison to observations that the uncertainty due to supersaturation uncertainty is within the thickness of the curves. For II, as this is a fit, we did not add the supersaturation uncertainty. We also clarified the title and added a sketch to clarify the variables. Also, we put "r/a" onto the y-axis label.

(22) Line 1, pg 15: This sentence was a little confusing to me. Should it read, "….was essential or if it was the greater amount of normal growth…"

→Agreed. We changed this to " As this is the only case we observed, it is hard to strongly argue a particular cause. One potentially important distinction from previous crystals with corner pockets is that the crystal in this case had a significantly greater rate of normal growth. We account for such normal growth by including S-type lateral growth along with the P-type in a possible mechanism argued in the next section."

(23) Line 20, pg 16: "…form near an edge or corner instead of one." I was a little confused by this sentence. Do you mean that there can be a single air pocket at a corner, but there can also be a pair of pockets near a corner but along the edge (as in Fig. 10h)?

→Changed to " Near the edge, the advancing fronts may generate pockets before converging, generating a pair of pockets instead of one. Figure 10e–h shows such a process." A few other small changes were made in the paragraph to help clarify the point.

(24) Line 19, pg 17: ""…the rim is narrower than that just inside the rim") This is a bit confusing and should probably be reworded.

→Now "(i.e., the rim radius is narrower than that just inside the hollow)".

(25) Figures 11 and 12: One way the discussion of these figures could be made a little clearer is if indications of various feature were made on the images themselves. For instance, on Fig. 12 one could indicate the "fan" like hollow in (a) and the flat terrace in (b).

→Done. We marked examples of center pockets, terraces, fanning, and corner pockets, referring to the markings in the text and captions.

(26) Line 19, pg 20: "analogous facet" is a bit ambiguous.

→Changed to " As with F-growth above, why would the thin front of the protrusion have a high density of growth sites that can efficiently collect all the AST flux and continue protruding?"

(27) Line 24, pg 20: "grown and sublimated in a pure vapor." It is my understanding that here you mean the situation where gas-phase diffusion becomes unimportant, which happens at very low pressures. However, at high temperatures aren't the vapor pressures high enough so that diffusion is still an issue? If so then perhaps one should add "near vacuum conditions."

→Even near melting, diffusion-driven vapor-gradient features such as hollows are not known to occur in a pure vapor. At least, we know of no such evidence. To help clarify this point, we added " in a pure vapor where such gradients are likely insignificant."

(28) Line 36, pg 20: "It would be less likely at the much lower-temperature protruding-growth effects found here." I found this sentence to be a bit awkward and suggest rewording.

→Changed to ", it would be less likely at much lower temperatures, such as for the corner pockets observed here near −30.0 °C."

(29) Line 30, pg 21: You may want to add a sentence or two here about why a small basal face is all that is needed.

→Yes. Changed as " Here, the appearance of a small basal face is all that is needed: AST from that face drives a small protrusion, and as the face grows, the rate increases due to the larger collection area and the protrusion extending into a region of higher vapor density. In this way, a large plate can develop, even when starting from the side of a small rime droxtal."

(30) Lines 36-37, pg 21: It may be worth pointing out that scroll forms are also found at lower temperatures (below -20C).

→Thanks. Added " Scrolls also form below −20 °C, also being part of some polycrystalline types."

(31) Appendix A: I very much like the model of protrusion growth. It is relatively simple but appears to capture the main physical features of the lateral spreading of a facet. On Line 1, pg 25, I assume this is an infinitesimally thin disk?

→Yes, clarified as " an infinitesimally thin disc"

(32) Line 2, pg 25: "Shifting and normalizing" What do you mean by "shifting"? Is this just the subtraction of N_infinity? And you may want to be a little more specific about the normalization.

→Rewritten as " It is convenient to shift *N* and making the variables dimensionless as"

(33) Line 5, pg 25: "can be shown". Was this shown in Nelson (1994), otherwise it might be good to give a reference to the solution.

→Yes. We rewrote the part as

"… vapor density. To determine the normalized flux F_v', we first assume it is known and solve for $\Delta N'$. In Nelson (1994), it is shown that

$$\Delta N'(\rho', z') = -F_v' \cdot h_{td}(\rho', z') \ ,$$
(A3)

where the thin-disc basis function h_{td} is an integral of Bessel functions."

(34) Equation A5, pg 25: My understanding of this function is that it provides the appropriate basis

function for the area (r-x_s to a) over which the flux of vapor is non-zero. If this is correct, then it might be good to introduce this basis function in this way. Perhaps pointing out that the idea here is to define a basis function for the ring region over which the vapor flux is non zero.

→ Correct. We revised as " However, in the facet spreading case, the flux is non-zero only in the thin ring $r-x_s \le \rho \le a$, not the entire thin disc. So we consider now the "thin-ring" basis function h_{tr} defined as…"

(35) Line 16, pg 25: "derivative of this function" Since there are two functions above this line, the function being referred to should probably be specified. I assume it is Eq. A5.

→ Correct. Replaced "this function" with the function.

(36) Line 15, pg 26: "From Eq A3 and A7…" I presume that h_td is replaced with h_tr though? If so then this should be specified.

→ Correct. To clarify, we added 1) " As the facet spreading situation is most similar to this thin-ring case, we use only h_{tr} from here." to line 18 on page 25 and 2) clarified this definition " where $h_{tr}(r')$ is shorthand for the full expression plotted in Fig. A2 (i.e., $h_{tr}(r', 0, r', x_s')$)." Immediately after the equation.

(37) Line 5, pg 27: The refinements described here sound like they would produce a more precise model, but given that we lack detailed measurements it's probably not warranted. It seems to me that the present "simplified" model is well-suited for the measurements that were taken from the growth chamber.

→In response, we added " and should be suitable for the present measurements."