Reply to the reviewers

We thank the reviewers and editor for their time evaluating our manuscript. Here we reply to their helpful suggestions, pointing out changes made to the revised manuscript. First, the main issues:

The major criticism of reviewers 1 and 3 was the length or organization. We responded by reducing the length of the main body of the manuscript by about half. Our other large change is to the introduction, which reviewer 2 expressed a need for more general background. So, we broke the original Introduction into Introduction and Background, adding general information in Background. Except for Appendix A, which is needed for the test of facet spreading, all theory sections are moved to a separate manuscript. The remaining parts have been significantly reorganized, and much rewritten, to make reading easier. All detailed discussion of the secondary habits was moved to the appendix, and some figures have been revised to help clarify the content. Finally, the title has changed to better reflect the content.

Shown below are the complete reviewer comments, separated by our replies with a red dashed line and made distinct with larger font. Also, in yellow highlighting are changes in the revised manuscript.

Anonymous Referee #1 Received and published: 26 April 2019

The authors studied the formation mechanisms of air pockets and other secondary habits in snow crystals. The topics coincide with the scope of Atmos. Chem. Phys. Discuss., and the secondary habits of snow crystals are interesting from the fundamental viewpoint. However, first of all, this manuscript is too lengthy (26 figures are shown in the manuscript of 51 pages in total), and too many subjects are included in a mixedup way. Therefore, I need to say the presentation quality is poor. Second, the authors insist that the formation of corner air pockets (the main subject in this manuscript) cannot be explained by the traditional growth mechanisms based on lateral step motion, and by the morphological instability based on the inhomogeneous distribution of vapor density. Then the authors conclude that the lateral-type (protruding) growth, which is the key mechanism in this study, is a novel concept. However, I cannot agree with such authors' claims (for details, see the comment 2). Hence, I believe that the scientific significance of the present manuscript is also poor. Since the amount of revisions is significantly large and the conceptual revisions are necessary, I do not recommend the publication of this manuscript.

Our reply:

Thank you for your comments. We address these issues below. In brief, the manuscript is reorganized, with a main body half as long. The issue with comment 2) seems to be based on a misreading as explained below.

Following 1-3 are major comments.

1. Too lengthy: one paper should have one main claim. Then, I believe that the following topics should be presented in separated papers: # the formation mechanism of the corner air pockets, # quantitative discussion about the kinetics of the lateral-type (protruding) growth, and # secondary habits other than the corner air pockets (these topics can be also moved into supplementary information)

Our reply:

We followed your suggestions in part by moving most theory subsections to a separate manuscript. The secondary habits discussion is a crucial part of our main claim and remain in this paper, but due to their length we moved them to the appendices and simply summarize the findings in the main body. They remain in the paper because they give further support for the importance of lateral growth and provide numerous tests of the phenomenon, but do not interrupt the flow. More importantly for us, they may satisfy readers curiousity about how some very unobvious crystal forms can arise.

Our main claim is that lateral growth should be included in any complete model of ice growth. We stated this in the introduction and in the conclusions, but neglected to explicitely state it in the abstract. This omission has been corrected. Appearing at the end of the abstract:

Although these suggested mechanisms may presently lack quantitative detail, the overall body of evidence here demonstrates that any complete model of ice growth from the vapor should include lateral-type growth processes.

The title was also changed to better reflect the content.

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2. The formation mechanism of the corner air pockets: the authors mentioned that the normal growth via step motion and the standard hollowing theory based on the morphological instability caused by the inhomogeneous vapor density cannot explain the formation of the corner air pockets. Then, I shall explain the formation of the corner air pockets by the traditional concepts. The key is the morphology of a snow crystal at the beginning of the growth. 1) When a starting crystal is fully faceted, the local vapor density becomes maximum at the corner of the crystal, providing a hollow not at the corner but at the center of the crystal face, as the authors explained. 2) In contrast, when a starting crystal is partly rounded, the layer-by-layer growth of the faceted face (located at the center of the crystal) proceeds. Then a spreading edge appears as shown in Fig. 1b (marked by e) and Fig. 8c. Since the spreading edge shows an angular shape and the corner of the crystal is still rounded, the local vapor density at the tip of the spreading edge becomes higher than that at the rounded corner, providing the overhang as shown in Fig. 1d and Fig. 8d. After once the overhang was produced, the overhang is developed spontaneously (the authors call this process the protruding growth), and the corner air pocket is formed. These processes never violate the traditional concepts of the layer-by-layer growth and the morphological instability.

Our reply:

Thank you for the suggested mechanism. We agree that the vapor-density gradients should promote protruding growth and that "traditional concepts" are involved. In the first paragraph of §3.6 (which the comment addresses), we are referring to hollows forming on a fully facetted face as being incompatible with the observed corner pockets. To reduce chances of the same misunderstanding, we changed the wording (now section 4.1.2):

Existing views on normal growth via step motion cannot readily explain corner pockets on fully facetted crystals. With normal growth, each pocket must have at one time been a hollow (lacuna or concave feature) before closing-off to enclose the air. And standard hollowing theory (e.g., Kuroda et al., 1977; Frank, 1982; Nelson and Baker, 1996) predicts that hollows form around a local vapor-density minimum, not at a corner where the driving force for normal growth is instead a local maximum. Moreover, the standard theory relies upon step clumping on a facetted surface.

We agree with the reviewer's ideas about protrusion initiation. But they were already in the manuscript—in the 2nd paragraph of §3.22. This section is now moved to section 5.1. The passage, from line 20

A possible answer to (1) is a large vapor-density gradient. Consider again the sketch in Fig. 2b. If the vapor-density contours closely parallel the surface, but "skim over" the inside corner **c**, then the vapor density would sharply decrease from **e** to **c** provided that this distance exceeded the vapor mean-free path. In such a case, the AST flux may build up nearer to **e** and not reach **c**, initiating the protrusion.

Thus, the reviewer simply misread what we had written. Hopefully, it is now clear. Finally, whether or not one refers to lateral-type growth as a "traditional concept" is immaterial (we never say it is not); the fact we emphasize is that it has not been considered in ice-crystal growth models, which is our main point.

In addition, the authors emphasize the importance of the diffusion of admolecules on the crystal surface (the surface diffusion of admolecules). Then the authors named this process "adjoining surface transport (AST)". I fully agree with the importance of the surface diffusion for the formation of the overhang and the subsequent protruding growth. However, the concept of the surface diffusion of admolecules is very traditional (firstly proposed by Frank and coworkers in the 1960s, and then experimentally proved by the growth of various crystals). Therefore, the authors should clearly show what is the authors' novel concept and what is not.

Our reply:

One paragraph in the original introduction was devoted to prior work on surface transport over crystal edges (page 8, paragraph starting on line 12). There are 8 references to prior work on the topic. So, we made no new additions to this discussion, as we feel the 8 citations are sufficient. Moreover, the overhanging aspect of protruding growth appears in none of these previous papers. The reviewer appears to be confusing surface diffusion with surface transport over the edge to the lateral-growth front. These are distinct processes that we have tried to further clarify in the new Background section.

Concerning the point that we should clarify what is new and what is not: We have followed the long-standing scientific practice of giving references to all relevant prior work except when long-established (e.g., kinetic theory of gases), with all else being presumably new. If we were to start saying "this is novel" every time we express a new result or idea, the reader would soon tire of the repetition and deem us arrogant. Even though some authors break with this practice and announce their result as "novel", most do not, and for good reason—that is the job of others to declare. Relevant changes to the manuscript are in our next reply.

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3. Throughout the manuscript, the authors should clearly explain what is the authors' new finding and what is not, with respect to phenomena and formulas as well. Followings are minor comments.

Our reply:

Whatever does not have a citation is thought to be new, as per standard scientific writing practices. However, several additions help clarify our contributions: In the introduction to the role of AST on secondary habits (Appendix B)

Most of these features and habits appear inexplicable with normal-type growth processes only, and only a few of them have even seen attempts at explanation.

And in the introduction to the main results (§4)

The following subsections survey, and partly explain, observations made in CC2, including previously unreported "corner pockets", "planar pockets", and "elongated edge pockets".

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4. The term "droxtal": since many readers (including me) are not familiar with this term, the authors need to explain it properly at the beginning.

Our reply:

In the revised manuscript, we define the term in the new background section, page 3 as

Atmospheric ice crystals generally begin with the simplest of shapes, a solid ice sphere, also called a droxtal.

The term is no longer used in the abstract, as we now refer to the droxtal center as

small circular centers in dendrites

The term is also defined in the figure with droxtal images. Before, this was Fig. 2, now it is Fig. 1:

Figure 1: Crystals at different stages between large droxtals (just-frozen droplets) and prisms

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5. The section 1.2 gives the impression that the authors do not fully understand the fundamental growth mechanism of crystal growth. The concept of the surface diffusion of admolecules (AST) is widely accepted in the crystal growth of wide variety of materials: not only for the metal whiskers, but also for semiconductor crystals, molecular crystals and ice crystals as well (as studied by Hallett, Mason et al, Kobayashi, and Asakawa et al.) Hence, for me, the application of the surface diffusion to the lateral and protruding growth by Yamashita (2015) does not look a significant revision, since the lateral and protruding growth can be explained easily, as shown in my above-mentioned comment 2.

Our reply:

We do not see the relation here between the first sentence and the rest of the paragraph. We agree that surface diffusion is widely accepted, and cite these and other authors for work on AST in the Background section. But surface diffusion is not AST: AST is defined as adjoining surface transport to the lateral "growth front" as stated on lines 11-14 of the abstract. AST was defined elsewhere as well. To help reduce this confusion, we briefly list the relevant definitions now at the end of the background section. The previous section 1.2 has been expanded to the new Background section, so this "impression" should no longer be present.

Concerning comment 2, we address that confusion above. However, whether or not the reviewer considers lateral and protruding growth a "significant revision", the fact remains that it had not been considered as an important aspect of ice growth from the vapor. Lateral-type growth does not appear in the ice-growth literature, and AST is generally ignored. For example, at the end of the new section 5.2.1, after briefly summarizing how AST-driven lateral growth can explain seven secondary habits and features, we add

A recent review of ice growth from the vapor suggested that AST may be unnecessary for understanding ice growth forms (Libbrecht, 2005). The above examples suggest otherwise, instead arguing that many oft-observed secondary features may be inexplicable without the AST mechanism.

Getting back to the reviewer's first sentence, do we "fully understand the fundamental growth mechanism of crystal growth"? Does anyone?

By the way, it is impossible to obtain the reference Yamashita 2015. The page numbers of the references Yamashita 2013 and 2016 should be 165-176 and 393-400, respectively: the page numbers of 23-33 and 15-22 are those in the issues 60 (3) and 63 (5).

Our reply:

Yamashita 2015 is a conference proceeding paper. Indeed, such papers are often hard to obtain, but not impossible (we found it). The reference serves the purpose of giving credit to the originator of the idea. Given that this is a common practice in scientific papers, we keep the reference. We have a few other cases like this, but the vast majority of our references are easy to access.

Thank you for the corrected page numbers for Yamashita 2013, 2016. The Tenki journal has two page numbering systems on each page, and, being unfamiliar with this journal's system, it seems we picked the wrong one. This has been corrected.

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6. The authors should show the schematic illustration of the new crystal-growth apparatus (CC2) in this study, since the reference Swanson and Nelson 2019 is still in preparation.

Our reply:

The manuscript is under review and available for viewing, so we did not include a drawing here. We updated the reference so readers can view the apparatus. Please see the link in the new references:

Swanson, B., and Nelson, J.: Low-Temperature Triple-Capillary Cryostat for Ice Crystal Growth Studies. Atmos. Measurement Techniques, https://doi.org/10.5194/amt-2019-137, 2019.

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7. In the section 3.4, from Fig. 4, I cannot understand the difference between the expanding boundary of the basal face and growing macro-steps. The authors also should clearly explain the kinetic models I, II and III in the main

text (of a separated paper), since the quantitative discussion has no meaning without obtaining the complete understanding of the models.

Our reply:

A macrostep exists as a large step within a facet and it arises from the clustering of smaller steps. Neither apply here. The case here, as we try to clarify in the main text and the appendix, is a facet expanding over a rough, round surface. The rough, round surface is not yet a facet, and thus our case is not a macrostep; moreover, the facet edge did not arise from step clustering. To help clarify, we added the following text where we discuss the plot:

…, but it is a reasonable fit to the initial cross-section profile. This profile is that of a flat facet out to a radius $r < a$, and a curved profile between r and a where the crystal had rounded during sublimation. (Refer to Fig. A1 for further details.)

We now discuss macrosteps in a few places in the new Background section, clarifying their difference with a spreading facet.

Finally, we keep the appendix here to explain the calculations for the plot, but as suggested will have further details of the model in a separate paper.

By changing the value of h/xs arbitrarily, one can easily fit the experimental data. Hence, here the authors need to explain the causes of the change in the value of h/xs (I believe that the cause of the change is the evolution of macro-steps) and also whether the change is appropriate or not. The authors also need to discuss the values of h and xs.

Our reply:

Yes, changing h/xs allows us to fit the data. We made no claim to the contrary and write that accurate measurements with a different apparatus are needed. Nevertheless, the qualitative trend in the fitted curve is consistent with the cross-sectional profile of the crystal when the facet began expanding. It has nothing to do with macrosteps because the expanding facet is not a macrostep. The cause of the change in h is shown in Fig. A1, and explained in the caption:

At a later time t', the value of h is larger (light shading) due to the advancement to $r(t')$, making a larger distance between the rough surface and basal surface at c.

Of course, it is likely that some normal growth on the basal facet occurred that contributed to h, but this rate of growth in this case is negligible compared to the rate of lateral growth, also making the contribution to *h* negligible. (Other cases may differ, so we do not claim this contribution can always be ignored.)

Without an accurate measure of the crystal profile and an established value for *x*s, providing separate numbers for *h* and *xs* here is largely pointless; the trend is clearly consistent with the profile, but accurate measurements are needed. The whole point of the comparison is to show that the AST is the only process capable of explaining the data. Nevertheless, we added the following discussion of *h* and x_s:

A reasonable estimate of height *h* upon reaching the edge is 1–5 μm. With this range, the fit in Fig. 6 (inset) predicts h/x_s = 0.3, giving x_s = 3–17 μm at this temperature, which is comparable to the value of about 2 μm found by Mason et al. (1963).

But to emphasize the main point here, the last paragraph was modified slightly as

Nevertheless, the observed behavior clearly shows that mechanisms I and III cannot explain the observed lateral growth. Only growth driven by a flux of surface mobile molecules, the AST mechanism, from the facet to the lateral-growth front is capable of fitting the observations.

Also, to help reduce confusion on this issue, we removed the two intermediate curves with high supersaturations from the plot. Now there are just three curves for the three models, all at the same supersaturation.

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8. In the section 3.9, the authors should explain the impossibility and instability much more in detail.

Our reply:

Detailed examination of impossibility and instability theories are covered well in the literature (though the term 'impossibility' is not used elsewhere). However, we agree that a little more explanation and rewording will help. In this section, after the 2nd sentence, we have the following rewrite that expresses the key differences between these two types of behavior:

In the standard treatment, however, the hollow occurs when the gradient in supersaturation needed for uniform growth can no longer be compensated for by the step density (e.g., Kuroda et al., 1977; Frank, 1982). In other words, normal growth of the entire facet becomes impossible, which is different from being unstable. In this "impossibility" case, one expects the hollow initiation and shape to be nearly identical on identical faces in a nearly uniform environment as well as being highly reproducible when other crystals grow under the same conditions. If merely an unstable phenomenon, then a sufficiently uniform, constant condition may be expected to circumvent the hollowing. Conversely, if hollows do form, then their initiation and shape should differ between identical faces due to minute differences in conditions. We suggest here that inclusion of lateral-type growth processes predicts qualities of unstable growth at low supersaturations, leading to hollow close-off and terracing features.

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9. The sections 3.10-3.22: If these sections have scientific significance, the authors should explain them in separated papers. If their scientific significance is not so large, the authors should move them into the supporting information.

Our reply:

These sections provide explanations for commonly observed ice-crystal forms and secondary features, making them significant to anyone interested in the causes of crystal shapes. Please compare to any paper (*of thousands*) that have been written about dendrite branching or some of the cited ones dealing with trigonal growth.

As these crystal forms and features had not been satisfactorily explained by other mechanisms, the explanations here involving lateral-type growth provide support for the importance of lateral-type growth, the main point of our paper.

We understand that the volume of information makes reading in one push difficult, so we moved these subsections to the appendix, instead summarizing them in the new subsection 5.2.1. We also added text and two figures showing images of two such crystal forms to more clearly motivate the need for explanation.

Perhaps what we failed to express here is that the extreme complexity of ice-crystal growth in air warrants a variety of approaches. At this stage, we need to at least know which processes to include in a crystal growth model. No model has yet included lateral-type growth. By including evidence that such a growth process can explain numerous micron-scale features in vapor grown ice (the secondary features), this paper shows just how prevalent the lateral-type growth is.

If getting the relevant growth processes right is not significant, then we do not know what is.

Anonymous Referee #2 Received and published: 30 April 2019

The paper “Air pockets and secondary habits in ice from lateral-type growth” presents extensive collection of micrographs of growing ice. I am very pleased by its esthetical beauty. I think it should be published for its experimental value, regardless the theoretical explanations. The explanation of air pockets formation is elegant. I cannot judge on its correctness. Nevertheless, I have some suggestions stemming mostly from the fact that I am not an expert in the field of ice grow from the vapor phase and thus I would welcome some introduction and generalizations. That may be a case for most of the readers, though. I suggest the paper be published after considerations the comments.

Our reply:

Thank you very much for recognizing the extent and significance of the work in this paper and making these suggestions. We have added some more general background in the beginning, breaking the introduction into a shorter introduction section that gives our motivation, similar to before, and a new background section that covers the more general topic (or "generalizations").

I would think that such extensive work deserves broader introductions and connections to what “general ice knowledge” may cover. I started form Hobbs: Physics of ice Ch. 8: I found the description of the growth in the direction of c and a-axis easier to understand then to consider the basal face and prismatic face in presented manuscript. Can both description be shown in the pictures?

Our reply:

Yes. We revised the sketches in Fig. 1 (now Fig. 2) to include different views and the principle axes as expressed in Hobbs. As mentioned above, we have broadened the introductory section, adding a section 3 "Background". This section explains the normal growth directions.

In Hobbs (ch. 8.3) the linear growth (here named normal) is defined as normal to crystallographic face. Is the here discussed lateral growth perpendicular to both c-axis and a-axis? Would not that be more exact definition than that given on the line 5 of page 2?

Our reply:

Lateral growth of a basal face is growth perpendicular to the c-axis, but the same cannot be said for the prism face. However, the maximum dimensions normal to the c-axis tends to be in the a-axes directions as shown in the revised Fig. 1, now Fig. 2.

We also improved the sketches in this new Fig. 2 to help explain what we mean by lateral growth. Also, in the Introduction, we help to clarify lateral growth:

The rate is often called the linear growth rate (e.g., Lamb and Scott, 1972), but to help distinguish this face-normal growth from face-lateral (or areal) growth, we refer to it as the normal growth rate.

Also, we try to clarify the types of lateral growth with a bulleted list of definitions at the end of the new Background section.

I think, that schema in Figure 1a suggests that the droxtal has 8 prismatic phases –should not there be only 6 of them?

Our reply:

There are just two prism faces in back. The figure has been revised to help address your previous concern, and now makes the six prisms clear by showing the top view at right in the new Fig. 2.

I had some previous knowledge of "snow morphology diagram", where temperature and humidity is decisive for the shape of snowflakes. Thus I was surprised that the current paper does not describe the humidity in details or does not attempt systematic study of the influence of temperature and humidity. Is reasonable to suppose the dependence? Is AST necessarily needed for the observations or would the vapor deposition normal to a-axis be sufficient?

Our reply:

Lateral-type growth should depend on temperature and humidity. We plan to investigate this experimentally, and also hope that this paper spurs others to investigate as well. We now mention some expected dependences. For example, in the Background section, pg. 3, lines 3-5, we mention the temperature dependence of x_s:

Experiments reported in the 1960s indicated that x_s on the basal face varied dramatically with temperature, changing by a factor of 5–7 between about −7 and −12 °C (Mason et al., 1963; Kobayashi, 1967). Although the exact values of x_s may be disputed, both studies independently found the values to be largest in the tabular regime, smallest in the columnar.

In other parts of the manuscript, we refer to this temperature dependence for the basal as being potentially important for various features, such as the two-level structure of planar crystals, capped columns, and trigonals. But the values for the basal have not been verified by other experiments and we do not have measurements for the prism. For protruding growth, another length scale may be important as well, the migration distance on rough faces, which may depend on temperature and supersaturation, but we have no theory or experiments to guide us. We mention this in the new Background section. Thus, we do not attempt anything like the snow crystal habit diagram for lateral-type growth features. However, we now mention the snow-crystal habit diagram in the introduction:

The primary and secondary habits depend on temperature and humidity as often portrayed on the habit diagram. This diagram has generally remained the same since Ukichiro Nakaya first proposed it (Nakaya, 1954), though some extensions and modifications have come from subsequent studies (e.g., Hallett and Mason, 1958; Takahashi et al., 1991; Bailey and Hallett, 2004; Takahashi, 2014).

It would be nice to shortly connect current observations to those of "classical" snowflakes formations. Is there AST mechanism needed there? I would appreciate if some discussed term are more explained and/or shown in the pictures (droxtal, adjoining facets, basal and prismatic facets in Figure 2).

Our reply:

We make several connections to the classical stellar crystal, but do not suggest that the main features such as the branches and sidebranches are due to lateral growth. In the main body, we make the connection to corner pockets in classical snow crystals in Fig. 8 and section 4.5, as well as discussing the two-level structure in section 5.2. Several other aspects of the classic snow crystal are addressed in Appendix B. We now list the common lateral-growth terms at the end of the Background section.

I think the abstract should be modified according the final content. Currently, I find some disagreement between it and the content of the manuscript. Also the name of prof. Yamashita in the abstract does not seem appropriate to me.

Our reply:

We are not sure what "some disagreement" refers to here, but we have assumed the reviewer means that some findings are not explicitly mentioned in the abstract. In response, we added our model fit to the abstract and mention other results from the experiments:

Further experiments revealed other types of pockets that are difficult to explain without invoking AST and protruding growth. We develop a simple model for lateral growth on a tabular crystal in air, finding that AST is also required to explain observations of facet spreading.

and added some words to help clarify our applications to observed secondary features:

Applying the AST concept to observed ice and snow crystals, we argue that AST promotes facet spreading, causes protruding growth, and increases layer nucleation rates. In particular, depending on the crystal shape and conditions, combinations of these lateral-type processes with normal growth can help explain presently inexplicable features and secondary habits such as air pockets, small circular centers in dendrites, hollow terracing and banding, multiple-capped columns, scrolls, trigonals, and sheath clusters. For dendrites and sheaths, AST may increase their maximum dimensions and round their tips. Although these applications presently lack quantitative detail, the overall body of evidence here demonstrates that any complete model of ice growth from the vapor must include these lateral-type growth processes.

About mentioning Prof. Yamashita in the abstract, we realize that the practice is not common. But it is done (see e.g., some abstracts from the physicist J. A. Wheeler, and the one by Frenkel that we cite), and in this case we prefer to have his name. He has promoted the idea of AST and protruding growth for years, often communicating with one of us, but has had difficulty writing his results up for an English journal. We reference all of his relevant work in the main text, but some readers only read the abstract and we feel that he deserves recognition for the concepts on the front page lest readers mistakenly think we originated the concepts of AST and protruding growth. But to help address your concern, we shortened the mention of his name in the abstract.

Anonymous Referee #3 Received and published: 7 May 2019

This paper reports an experimental study of ice growth from vapor in air, with a focus on the formation of air pockets and secondary habits. It ultimately looks to explain a wide variety of experimental observations on lateral-type growth and looks to relate the observed behaviour to a surface flux of water molecules, which the authors call "adjoining surface transport" (AST). As a non-expert this specific field (experimental studies of ice growth from vapor), I found this paper rather difficult to read, and I felt I learned very little in reading it. I found it to be not well written, and a bit of a jumble of data and ideas with no clear narrative. Hence, I think it could be considerably improved by shortening (i.e. less would be more here) and organizing the material better.

Our reply:

We are sorry to read that you learned very little from it. We agree that the original was hard to read completely through due to the length of the main body. To improve the narrative, we have reduced by half the main body, largely reorganized the paper, rewriting many parts, adding more motivation for the work, removed most theory sections, and added to the introduction a background section to put the work in context of standard crystal growth theory.

Applications to secondary features, which occupied much of the main body of the original, no longer appear before the summary, instead appearing in Appendix B, beginning with a list of the content. The main point is now emphasized more, which should also improve the narrative.

Also, the paper often seems to read more like a review, where it was often unclear where the authors' work started and ended. Perhaps after considerable revision with work might be appropriate for publication.

Our reply:

We understand how one might read the paper in this way because we have examined many observed crystal types. And in a sense, it is a review of our own work going back six years. But whether one considers it a review or not, it is all tied together by the clear evidence that lateral-type growth is needed to understand many ice-crystal forms. That this point should be emphasized is given by the fact that a previously published review considered it unnecessary to include AST in growth models. We now explicitely state this at the end of section 5.2.1:

A recent review of ice growth from the vapor suggested that AST may be unnecessary for understanding ice growth forms (Libbrecht, 2005). The above examples suggest otherwise, instead arguing that many oft-observed secondary features may be inexplicable without the AST mechanism. Additional cases, including aspects of primary habit, are briefly examined in the appendix as well. The arguments are mostly qualitative; nevertheless, they may help stimulate new measurements of x_s, further observations, and more detailed modeling of these interesting crystal forms.

However that review examines cases of normal growth for the primary habit and "morphological instability", but does not consider some of the secondary features like we do.

Concerning where our work started and others' ended, we reply to this point in a previous reply: we have followed standard procedure and given references for any work or results that are not ours.

Finally, we have made considerable revisions, as suggested.

I was also unable to make sense of their physical model.

Our reply:

For the examination of facet spreading in Fig. 5 (now Fig. 6), the model we used is the BCF model as justified in the Background section. Starting in the first paragraph:

The most widely used model for the growth of crystal faces from the vapor is the BCF model (Burton et al., 1951; Woodruff, 2015). This model supposes that a given molecule in the vapor above a faceted surface strikes the crystal surface and become temporarily trapped in a mobile state until either desorbing back to the vapor or migrating along the surface and reaching a more strongly bound state at a step edge….

Then at the end of the third paragraph

…Hence, at least as a first approximation, it is still useful to compare observed behavior of ice to the BCF model and make use of measured x_s values.

The basic physical model involves AST, which is not new. From the old Introduction, now near the end of the new Background:

The types of lateral growth here are driven by AST. Evidence for AST on ice is indirect, partly coming from early studies of spreading ice layers on covellite (Hallett, 1961; Mason et al., 1963, Kobayashi, 1967). In these studies, the rates of approaching micron-scale layers, also known as macrosteps (arising from clustering of smaller steps or contact between crystals of differing height) changed in a way consistent with a flux of molecules over the top edge of the layer. The concept has long been applied to the growth rates of metal whiskers (e.g., Sears, 1955; Avramov, 2007), but rarely applied to ice.

Most theory sub-sections have been removed (put in a separate manuscript). The details of the model calculation for Fig. 6 are in the first appendix. In the rest of the manuscript, we are only arguing qualitatively for AST effects. The reason is two-fold: 1) calculations of an AST for most secondary features would require much more space in a separate paper (e.g., for the needle crystals) and 2) the key parameter x_s is poorly constrained, particularly for the prism face. Our hope is that this study stimulates work on the topic, allowing such quantitative treatment in the future.

It has now be very well established that the surface of ice features a layer water molecules that exhibit mobility (this is sometimes referred to as the quasi-liquid-layer – QLL; see Rosenberg, Phys. Today 2005, 58, 50; Li and Somorjai, J. Phys. Chem. C 2007, 111, 9631; Björneholm et al., Chem. Rev. 2016, 116, 7698). While different experiments report different thickness for the QLL, they generally agree on its presence, and that its thickness (and other properties) are strongly temperature dependent. One would expect any surface flux to then depend on the thickness of the QLL and the mobility within the layer (which will also be strongly temperature dependent). Thus, AST should exhibit strong temperature dependence, and one would expect to observed "protruding growth" only when the effect of AST is large compared with the rate of vapor deposition (which will depend on the level of supersaturation).

Our reply:

Thank you for pointing this out. We agree that the surface of ice has significant mobility and that many researchers have found evidence for a QLL. In the background section, we now briefly discuss the apparently disordered nature of the ice surface, its role in growth, and our reasoning for using BCF with the parameter x_s:

The BCF model of surface diffusion assumes that the mobile surface molecules are sparse and non-interacting. For ice, this assumption is widely believed to be violated over much of the atmospheric temperature range where the surface is thought to contain significant disorder (e.g., Rosenberg, 2005). Yet the BCF model is nevertheless often used to interpret experimental results (e.g., Sei and Gonda, 1989; Asakawa et al., 2014). A key parameter in the model is the mean migration distance x_s of a mobile molecule on the surface before desorbing, a distance that should differ between the basal (***b***) and prism (***p***) faces as well as depend on temperature. With interactions between these surface-mobile molecules (e.g., Myers-Beaghton and Vvedensky, 1990), x_s may also depend on supersaturation. In addition, the migration of surface vacancies may also affect x_s (Frank, 1993). Experiments reported in the 1960s indicated that x_s on the basal face varied dramatically with temperature, changing by a factor of 5–7 between about −7 and −12 °C (Mason et al., 1963; Kobayashi, 1967). Although the exact values of x_s may be disputed, both studies independently showed that the values are largest in the tabular regime, smallest in the columnar. Corresponding values for the prism have not been determined. Later, Nelson and Knight (1998) found a similarly sharp behavior in basal-face critical supersaturation between these temperatures. A possible link between these two parameters is clustering of the mobile species responsible for growth: when the temperature is such that clustering is strong, the critical supersaturation is low and surface-mobile molecules would become temporarily trapped in sub-critical nuclei, giving them very low mobility. Thus, the critical supersaturation would be low when x_s is low and vice-versa as found by experiments. The values of the measured critical supersaturations led Nelson and Knight to conclude that the surface was indeed disordered but " ...the [common] view of the ice surface as a liquid layer is not a useful idealization for crystal growth processes." Hence, at least as a first approximation, it is still useful to compare observed behavior of ice to the BCF model and make use of measured x_s values.

That passage also includes the review article by Rosenberg that you cited. Also, in the discussion of hollows and center-pocket formation, we mention a newer study of step dynamics in a disordered region (now in Appendix B.1):

(Neshyba et al. (2016) proposed a more detailed model of step dynamics for ice with a thick surface-disordered region, but it is not yet clear how a hollow would develop in that model.)

Finally, we consider that with or without a thick disordered layer, the AST process may continue. Even as isolated mobile molecules, the simple calculation using BCF gives a significant contribution to growth, the amount depending on the ratio of the surface-migration distance x_s to the edge thickness t. In the new section 5.1, at the start, we added

The microscale mechanism of facet spreading involves AST, which is the migration of molecules, first over the edge of the facet, and second with their finding a high-density of growth sites on the other side. The first may occur via isolated molecules or as a more cooperative phenomena in a thicker disordered region, but either case may be consistent with the observations here.

Concerning the temperature dependence of AST, this will largely depend on x_s, which is presently poorly constrained, and the thickness of the lateral-edge front. Other factors will be addressed in a follow-up paper.

In the section on the two-level formation on droxtals, now appendix B.3, we describe the temperatures that these are found and make the connection to the high x_s values measured by Mason et al. (1963) at these temperatures. In this section and elsewhere, we refer to their finding a temperature-dependence of x_s on the basal face.

Yet, the authors do a rather poor job of characterizing the conditions, temperatures and supersaturations, in the reported experiments. Moreover, in a carefully designed set of experiments (when one varies one of these, for example), it should be possible to see the effect become manifest. I would find such a set of data much more convincing. As a minimum, the authors should do a much better job of describing the conditions for each experiment, and then comparing and contrast the behaviour on the basis of these conditions.

Our reply:

We have included the conditions whenever possible. When they are only partly known, the examples are nevertheless useful to help support the roles of lateral-type growth. For example, in Fig. 1 (old Fig. 2), the main points of showing them are the spreading facets and the pockets. In this case, we give references to other cases with more precisely known conditions. For the case in Fig. 3, we give the temperature, supersaturations, and crystal sizes. For the case in the old Fig. 4 (now Fig. 5), we had the conditions in Appendix A, but now added them in the main text. Conditions were given for the crystal in Fig. 6 (now Fig. 7). We added the conditions in the caption of Fig. 12, but we empasize at the beginning that the behavior being reported occur over a wide range of conditions. This range will be explored quantitatively in a future study. In general, the effects of AST described here will be larger when the mean migration distance is larger than the edge-front or protrusion thickness. These quantities are not known very well, so testing the predictions will require their measurements. Our task here has been to establish that AST occurs and is worthy of further study.
