Atmospheric
Chemistry
and Physics
Discussions

Interactive comment on "Surface roughness during depositional growth and sublimation of ice crystals" *by* Cedric Chou et al.

Cedric Chou et al.

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Response to Referee #2

The manuscript "Surface roughness during depositional growth and sublimation of ice crystals" by Cedric Chou et al. describes a study of ice crystal growth and sublimation performed in a new experimental setup. The experimental apparatus uses a unique combination of devices (flow diffusion tube and 2D scattering instrument) thus assuring a novelty of the results. The study is also well planned and the setup is thoroughly characterized, both by CFD modeling and experimentally. The authors demonstrate a high level of understanding of the physics behind the experiment, even if the thermo-

dynamic parameters of the experimental system are not fully controlled. The paper is definitely worth being published, but must be thoroughly revised in many respects. I wish the paper were written more clearly. Some sections, as addressed below, require thorough editing. The relationship between the crystal evolution and its morphological complexity is, however, convincingly demonstrated. This work should trigger off studies of this phenomena with a better control of the supersaturation and optical control of the crystal morphology. I would, however, avoid naming the effect discussed in this manuscript the "surface roughness", because this implies a quite narrow range of texture features. The sublimation and regrowth of ice crystals often create a polycrystalline aggregate of tiny crystals that can have smooth surfaces. The ultimate example is the "Bucky ball" crystals as in Baran (2012). Should such aggregate be named "rough" or "irregular" or somehow else? Would scattering patterns on such crystals be identical? I would really like to see a thorough discussion of these issues in the introduction and a clear separation of "surface roughness" from the "morphological irregularity" throughout the manuscript. In some sense, this is already done by introducing the "combined roughness" based on the 2D scattering patterns analysis. The same should be done with respect to surface texture and geometry of the crystals, and the approach suggested in this manuscript (combination of microscope observation with scattering measurements) seems to be very promising for achieving this goal. Below please find my comments which I hope would be helpful in improving the readability of the manuscript. The parts in the manuscript I am addressing are identified by page and line number and the citation are given in italic.

We thank the Referee for a very thorough and encouraging review and many insightful comments, questions and helpful suggestions that allowed us to improve the manuscript. We list them below, together with our clarifications (in blue) and changes to the manuscript.

1. Introduction: How is the surface roughness defined and what is the quantitative

measure of surface roughness? In the introductory part, the irregularity of ice crystals seems to be treated in parallel with the concept of surface roughness. However, the manuscript is titled clearly "surface roughness... ". The introduction (and the manuscript) would very much benefit from a clear definition of surface roughness as compared to habit irregularity. It would be also very helpful if you could think of a way to introduce a quantitative parameter to characterize physical surface roughness.

We state clearly that fine surface roughness and large-scale irregularity are treated together. Further discussion of the meaning and significance of the measure of roughness goes beyond the scope of present work and is dealt with in detail in the cited articles (Ulanowski et al., 2012, 2014). Among others, it was pointed out that the lack of distinction between "roughness" and "complexity" from the point of view of 2-D scattering is likely to also apply to other light scattering properties. So the distinction may be to some degree artificial. Nevertheless, crystals in the experiments were relatively simple prisms, not complex ones, so the study is more directly relevant to surface roughness, hence our choice of emphasis.

2. Page 2 Line 31: "In the experiments, the ice crystals are fixed within the measuring volume and exposed to thermodynamic conditions... " were there many crystals in the sample volume?

We insert the text ", generally single,":

> "In the experiments, the ice crystals, generally single, are fixed within the measuring volume and exposed to thermodynamic conditions simulating single or multiple growth cycles at various temperature and saturation ratio."

3. Page 3, line 24. "...which ensures that the 22? halo scattering from ice prisms is

included." I am afraid a typical reader would not know what you are talking about. You can't expect anyone being familiar with refraction theory in hexagonal ice columns. Is this detail really needed here? The same line, replace "lower angles" with "smaller angles".

We alter the text to say "bright feature associated with the familiar halo occurring for ice prisms at the scattering angle of 22°". We do replace "lower angles" with "smaller angles", as suggested.

4. Page 3, line 25. "The camera images are digitized as 12-bit TIFF files..." you can't possibly mean that the camera produces analog images that have to be digitized afterward?

That is indeed what happens internally to the camera.

5. Page 3, line 30. Figure 2 does not show the fiber-optics illumination, could you show how it was located with respect to the sample volume?

The text mentions the illumination; we now add that it is "in-line" with the microscope tube and add extra detail in Fig. 2.

6. Page 4. Section 2.1.3 "Operating principle". The content of this section does not correspond to its title. Is that the operation principle of the whole setup or the flow diffusion channel? Before describing the simulation results, please explain exactly what has been simulated and what was the purpose of the simulation (I presume, the fast control of water-ice supersaturation in the vicinity of ice crystal located in LISA).

We create instead a new section entitled "Numerical simulations and thermodynamic characterisation". Our statement concerning the purpose of the simulations was prob-

ably too short. We change the text substantially as given below. The purpose of the steady state simulations was not to show the fast control of water-ice supersaturation. For that a transient model would be required. The response time of the system was a) calculated from mass flow rates and total volume of the system, and b) measured.

"The thermodynamic conditions at the tube outlet were extensively studied by means of computational fluid dynamics (CFD) simulations of the laminar flow tube, and by measurements of flow velocity, temperature and dew point at the tube outlet. Both the numerical simulations and the measurements have been done to characterise the experimental setup, as well as to demonstrate the fast control of temperature and supersaturation in the measuring volume.

The numerical simulations were done with the commercially available CFD code Fluent (Ansys Inc., USA). The Fluent model is a general purpose FVM (finite volume method) CFD model allowing the simulation of a wide range of small scale fluid flow problems. Here, the flow through the flow tube was simulated including a multicomponent treatment of the flow. The model accounts for the coupled processes of mass and heat transfer. With respect to the geometry and the laminar flow regime, the simulations were done on a 2-dimensional axisymmetric Cartesian grid by means of a pressure based steady state solver. Additional information about the numerical model, which has already been successfully applied to the characterisation of the laminar flow tube LACIS, can be found for example in Stratmann et al. (2004); Voigtländer et al. (2004); Voigtländer (2007) and Hartmann at al. (2011).

To illustrate the operating principle of the laminar flow diffusion channel, calculated thermodynamic profiles along the tube axis are shown in Fig. 3. ..."

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7. Some sentences don't make sense to me: "For a sufficiently high gas flow representing the residence time of the gas flow, the thermodynamic equilibrium between the wall and the gas flow will not be reached." Does the flow represent the residence time or vice versa? Please rewrite in a clear language.

We modify the paragraph by adding the text:

"If the residence time of the gas flow (controlled by the mass flow rate) is large enough, the gas flow cools down until thermodynamic equilibrium with the tube wall is reached. Conversely, for a sufficiently fast flow equilibrium will not be reached."

8. In Figure 3, please make the legends more clear. You should explain what the flow rates for various lines mean (total flow followed by the flow rates of dry and humidified flows at the inlet?). The green line in panel (b) has no dry/wet flow specification, why? Since the wall and inlet temperatures are the same in all panels, consider moving them into the figure caption.

9. Panel (b) of Figure 3 uses Kelvin as temperature units, but all other figures are in °C. For clarity, consider using the same units everywhere. The line showing the length of the tube (1 m) should be present in all panels, alternatively, you could consider truncating the simulation lines at 1 m axial position.

We change the figures according to the suggestions of the reviewer.

10. Line 23: "...this can be also done on a short time scale (about 5 s) by controlling the ratio of the dry and the wet sheath air flow while the total flow is kept constant." In the beginning, you mentioned that there was no separate aerosol flow along the center of the tube, so what is the "sheath flow" for? Was the humidity of the sheath

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flow controlled separately? Where is this 5-second estimation coming from? Was it measured or simulated?

We agree with the reviewer that the statement of "sheath air" could be misleading. The reviewer is right, there is no separate aerosol flow along the tube centre. There is only one particle-free gas flow along the tube. We delete the word "sheath".

The 5 s estimation comes from a calculation, but was also measured by observing the ice crystals. For the calculation, we simply considered the total volume of the system downstream of the humidifier (not only the flow tube) and the flow rates.

11. Figure 4 and discussion thereof on page 5 casts many questions in conjunction with the data of figure 6: What are the solid lines: interpolation of numerical model results or something else? Was there wet flow in the model calculations and what were the wall boundary conditions? It appears to me that the measurements have been conducted under dry conditions, without ice coating the walls of the flow tube. Is that the case? Any idea why the measurements and model calculations deviate from each other at low wall temperature? Please address these issues thoroughly.

The solid lines in Fig. 4 and Fig. 6 are an interpolation of the experimental data - we add a note in the caption of the Fig. It is correct that there was no wet flow for the temperature characterisation data shown in Fig. 4. This means, the investigations have been done under dry conditions (by using pressurized air with a dew point slightly below -40°C). Using dry conditions holds for both, numerical simulations and measurements. The wall boundary condition in the simulations was zero flux. The reason for determining the temperature at dry conditions was that temperature measurements are not trivial under cold and wet conditions. For example, icing at the temperature sensor might occur. We know from wet simulations (with $S = 1$ at the wall boundary) that the temperature at the tube outlet is not significantly influenced by the presence of water

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in the system.

Furthermore, we also think that the differences between experimental data and simulation results are caused at least partly by the measurement technique. Especially at low flow velocities the temperature sensor, which was positioned in the optical measuring volume of LISA, several millimetres below the tube outlet, might not give true values. We spent much time using different types of sensors (various Pt100 and thermocouple sensors) to find out which one gives the best results for our application. In conclusion, even if the sensor is precisely calibrated (e.g. in an ethanol bath against a reference PT100 sensor), the difference between measurements and simulation results is probably due to technical measurement issues.

Measurements and simulations shown in Fig. 6 have been done for wet flow conditions. To minimize the effect of icing at the walls, the measurements were done only for a short time after the wet flow was switched on.

12. If the only purpose of Figure 5 is to demonstrate that IRIS "...can be used over a broad temperature range", please consider moving it into a supplementary material. It does not contribute to instrument characterization above what has been shown in Figure 4. Besides, it is unclear what are the solid lines on the color mapping.

We follow the suggestion of the Referee and move Fig. 5 into the supplement. We also add a figure to the supplement showing flow speed (measurements and simulations) data at the tube outlet. We change the text accordingly, delete the corresponding sentences (p. 5, l. 2-4) and add instead :

"Additionally, an extended data set of temperature measurements is shown in the supplement material."

and in the previous section we change the last sentence to (p. 4, l. 31):

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"Measured and calculated flow velocities were found to be very similar (see supplement material)."

13. Figure 6 and the discussion thereof on page 5: for the sake of comparison, please keep the same colors as in Figure 4 (red for T_wall = -40°C and so on).

There isn't one to one correspondence between the conditions in the two figures, hence the different colours. However, we follow the suggestion of the Reviewer and improve the Figs. for the revised version.

14. The deviation of measured RHi values from FLUENT results is striking, although FLUENT apparently makes a good job reproducing the flow temperature at the outlet (I am referring to the figure 4, the case of T_wall = -30°C). There, FLUENT underestimates the temperature only by 1K, which translates into 10% difference of the water vapor pressure at this temperature but not into 20% as suggested by Figure 6! Also, why don't you show FLUENT results for other wall temperatures?

As stated in the reply to Point 6, the purpose of the simulations was to design the experiments. Therefore, the model was simplified in several ways. We reduced the flow simulations to a multicomponent (water + air), but single phase (gaseous phase) problems. Water phase transition was considered as a sink only. This means, the growing ice layer at the tube wall was not simulated. In the simulations, the wall boundary condition was defined by setting the walls to saturated conditions with respect to ice (100% at the wall temperature). Hence we are not able to simulate the temporal change of the saturation (and temperature) profile. However, one can imagine that a growing ice layer may act as an insulator increasing the temperature gradient. Additionally, the dry air flow was considered to be completely dry in the simulations (water mass fraction of zero), while the dew point of our pressurized air was in reality between about -50°C and -40°C. Simulation results are shown and compared to experimental

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data for the case of Twall = -30°C because most the the experiments have been done at this temperature. For the data shown here, it is therefore the most relevant temperature value. Furthermore, measurements are getting more and more challenging at even lower temperatures. As stated above, temperature measurements at -40°C might be biased due to technical limitations. The same holds for dew point measurements. Therefore, we would expect that the differences between experiments and simulations results are a even bigger at -40°C.

15. Does the non-linearity of the measured RHi data reflect the time evolution of the flow temperature field, as discussed on page 5, starting from the line 14? Have the measurements been taken by stepwise increasing the wet flow? Would you expect a different behavior if the wet flow was decreasing instead?

Yes, one reason for the non-linearity of the measured RHi data is the time evolution of the temperature field. A second reason is the wall loss of water vapour at high RH. Water vapour is transported to the tube wall forming an ice layer. This sink also depresses the supersaturation. And yes, it is correct that the measurements were taken by increasing the wet flow.

The temporal change of the temperature is not linear. We observed faster changes at the beginning of the wall ice formation. At high RHi ice is quickly formed at the wall. Therefore, at high RHi there is not much difference if the measurements starts with a high wet flow. However, at low RHi it is. We did measurements with decreasing wet flow showing a difference.

16. Page 6 line 6: I believe the correct name is Gray Level Co-occurrence Matrix (GLCM). I don't know what a "co-matrix" is. What is the definition of the image "texture"? How is it different from "brightness distribution"? A typical atmospheric scientist would know little to nothing about it.

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The Reviewer is right, while the term co-matrix is often used for brevity in the literature, it is more correct to say "Co-occurrence Matrix"; we make a change in the text. See also response to Point 1.

17. Page 6 line 10 and on: since the concept of GLCM and its features is the central one in the manuscript, it would be nice to include a definition of GLCM "energy" which is used in the equation 1 to calculate the "combined roughness" but is not defined anywhere in the manuscript. This is even more so because the cited paper (Ulanowski et al., ACP 2014) does not provide any explanation of "energy" either, referring to the original paper by Haralik et al, 1973. However, Haralik et al. have not used the term "energy" among the statistical descriptors of image texture. It is therefore impossible for the reader to track down the definition of the term "energy" based on the provided information. Clarification of this issue is strongly advisable.

Incorrect: Haralick et al. do define "energy", just that this initial paper does not call it such. In any case, readers wishing to implement the measures can easily locate literally thousands of relevant references. However, for clarification we insert the text:

"(also known as uniformity, or angular second moment)"

18. A follow-up question to equation 1: The term "combined roughness" and the way it is discussed later suggest that this quantity describes both the irregularity (that is, the degree of deviation from a pristine habit) and the true physical surface roughness. Is that correct and if yes, what is their relative contributions?

This question cannot be answered at this stage, as the properties are not separable and anyway cannot be defined unambiguously. Further discussion is in the reference cited.

C11

19. Page 6, line 21 and on: The method of size determination should be described in much more detail as it is given in the present form. For one, it is not clear at all if the size of the ice crystal has been always determined based on the analysis of the speckle area alone. The optical setup includes a microscope and the example microscope pictures definitely show that they were good enough to determine the size of the crystals within a few micron accuracy. This, however, is not mentioned explicitly. Even if the speckle area provides the necessary accuracy of size determination, one would certainly want to validate this method against the old-fashioned visual examination? This brings me to a question how exactly the size of the crystal has been retrieved from the speckle area? The only explanation in the manuscript is at the end of section 2.3, stating: "In addition, the size of the ice particles, which is inversely proportional to the average area of speckle spots, is retrieved". However, the citing paper (Ulanowski et al., 2012) shows clearly that the relationship between the speckle area and size does not follow the simple inverse law (their equation 12 and figure 5). If the functional form of the relationship is not known, the only possibility that is left is to construct a calibration curve from the measurements where the crystal size is retrieved independently (using, for example, the optical microscope). Could you show such a curve? What are the uncertainties of size determination based on the speckle area analysis?

The method is described in detail and validated in the reference cited, which also includes a calibration curve. However, for clarification we insert the text:

"and is used throughout the present work to determine crystal size. The size measured in this way represents the diameter of equal area circle projected along the line parallel to the laser beam."

See also response to Points 20 and 26.

20. On the other hand, the visual inspection is claimed to be used to "[...] compensate

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for temporal changes of the thermodynamic conditions caused by the ice formation at the tube wall" by adjusting the flow rate if the crystal growth slows down. These should be explained more clearly: were the microscope images used to control the growth rate of the ice crystal AND the speckle area analysis used to measure the crystal size in parallel? How do these two methods compare?

Since the characterization of the thermodynamic conditions was not precise enough to establish the point of equilibrium between the crystal and the vapour, we had to observe the crystal to find out whether it was growing or not, i.e. if it was in equilibrium. This established the point of reference. But otherwise the images were not used for adjusting the conditions. To clarify, we replace the last, potentially misleading sentence with the text:

> "In this way the settings corresponding to the point of equilibrium between the crystal and the vapour can be found, to act as a reference point."

The two methods cannot be directly compared because the crystal dimensions "seen" by the two methods are orthogonal (one is parallel, the other perpendicular to the laser beam).

21. Page 6 line 25: "[...] and the amount of speckle represents crystal roughness." This is one example where the roughness should be clearly defined. Are you talking about the roughness of the surface or "combined roughness", which if I understand correctly, is the crystal irregularity plus surface roughness?

This was addressed previously, the roughness measure combines both. And the statement cited is a qualitative explanation, simply aiding the reader in the interpretation of unfamiliar 2-D scattering patterns.

C13

22. Section 3 Results and discussion.

23. Figure 7 is a beautiful example of the 2D interference pattern produced by smooth and rough crystals. Could you show the corresponding microscope images of the crystals responsible for them?

While we agree with the Reviewer that the inclusion of images with Fig. 7 would be interesting, optical microscopy images (or cloud probe ones) do not reveal sufficient detail of surface roughness. Anyway, in this case parallel microscopy was not obtained, as the patterns were generated in a conventional cloud chamber. We now add an explanatory sentence in the caption:

> "The patterns were produced using the SID3 instrument in the AIDA cloud chamber during growth at low (left) and high (right) supersaturation (Schnaiter et al., 2016)."

24. Page 7 line 26: "Fast growth can moreover lead to the creation of defects and ionization, ..." what exactly do you mean by "ionization"? Charging, creation of the local or surface charge?

Please see the multiple references cited, which deal with this broad topic, as well as the Conclusions and the Reviewer's own Point 32.

25. Section 3.1. It is stated several times in the manuscript that the supersaturation could not be determined precisely due to the instability of the thermodynamic conditions in the flow tube. You are, nevertheless, able to estimate the supersaturation with an accuracy of around $\pm 5\%$ (as in lines 3 - 4 on page 8), which is not that bad for a highly dynamic system. Given the amount of effort that has been put into character-

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ization of the flow tube and the fact that the supersaturation is indeed the key factor controlling the morphology of the ice crystals, I would suggest that you rewrite the characterization section clearly stating the range of supersaturation and the accuracy you could achieve but avoiding saying that the supersaturation could not be controlled. This creates unnecessary distrust in your results and shifts the focus of the discussion away from the physical mechanisms of surface roughening.

We did not say that supersaturation could not be controlled. In section 3.1 we state that "supersaturation ... could not be determined precisely". However, this may be misleading, so we now say "directly" instead, changing the sentence to:

"Since the supersaturation controls the growth rate but could not be determined directly with high accuracy in our experiments, ..."

26. Page 8 lines 8-9: "The crystals can be compared directly as they grow from 20 μm to 29 μm, after fitting trend curves using LOESS". What trend curves? What is LOESS? Was the size of the crystals determined from the speckle area analysis? What was the accuracy of such determination? Could you provide the confidence intervals for the "LOESS" fit? Would there be any growth in the confidence intervals for the "slow" growth case? What does "raw" in the legend of figure 8 means: measurement points, raw data? Please be more specific and more careful in presenting the results!

LOESS is a well-established numerical technique, and is described and referenced earlier in the text. As for size, see Points 19 and 20. Concerning the "confidence intervals", these would not carry any information relevant to the behaviour of the observed crystal, as they are the outcome of secondary noise sources such as digitized image noise or mechanical vibrations, as pointed out in section 3.

27. Page 8 lines 17 - 28. I support the idea that nucleation of stacking disordered ice

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can be responsible for the formation of irregular crystals, but how does this relate to the surface roughness? I might remind the authors again that the title of the manuscript is "Surface roughness during depositional growth..."

Please see response to Point 1, it is stressed several times that we do not distinguish between fine and coarse. roughness.

28. Figure 9: Please use conventional way for naming the axis. The variables "droughness" and "dsize" are not defined anywhere in in the text. Besides, what size is that: radius, diameter, characteristic size...?

The axes are defined as rates in the caption, and the variables "roughness" and "size" in the text, see also response to Point 19 above. However, for clarity we alter the axis labels to "d(roughness)/d(time)" and "d(size)/d(time)".

29. Page 10 lines 8 - 9. "Careful examination of the retrieved crystal size shown in Fig 10 indicates markedly slower growth in later cycles, despite similar supersaturation levels". To my opinion, this is stretching the imagination too far. There are only two growth cycles delivering comparable data, and the difference in the growth slope can be caused by anything else. How similar are the supersaturation values? Was the size change confirmed by optical microscope? Why does the same behavior not show up in Figure 11?

The supersaturations were the same. We bring this observation to the readers attention as it is potentially important, and we do not claim it as a "fact", hedging our bets with words like "appears to" etc. However, we cite similar behaviour observed in other systems. Moreover, concerning Fig. 11, we beg to disagree, as similar behaviour can be seen in crystal growth rate. So we are prepared to stand by our statements, and further work will confirm or contradict them. As for the microscopy, see response to

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30. One more comment on this point. To my understanding, the growth rate based on the "optical size", as derived from the speckle area analysis, is directly related to the rate of growth of a volume equivalent diameter (or any other characteristic size describing the envelope dimension of the crystal). The growth rate based on such equivalent diameter is directly proportional to the mass growth rate. As "combined roughness" increases (as you have shown nicely), the ratio of surface to mass increases too, meaning that creating more surface in case of a growing complex crystal does not contribute to mass growth in the same way as in case of a growing pristine hexagonal column or plate. What implication this effect would have for the atmospheric phenomena is a question which, I am afraid, cannot be answered without thorough modeling of crystal growth with the cloud microphysical feedbacks.

No, it is not "volume equivalent diameter" - see Point 19. However, we are happy to support the rest of the Reviewer's comment.

31. Page 11 line 2-3: "It is very likely, as shown in our experiments, that at higher supersaturation rougher crystals will develop at the expense of smoother ones." I strongly doubt it. What would be the mechanism of such competition? Would you expect the pressure difference above smooth and rough surfaces? If not, why would rough crystals grow preferentially if both rough and smooth crystals are exposed to a supersaturated water vapor? Please clarify this statement or remove from the discussion.

We merely reiterate that our experiments show higher roughness at high supersaturation, and that this is also likely to occur in the atmosphere. However, to avoid misunderstanding, we change the words "develop at the expense of" to "tend to develop instead of".

C17

32. Page 11 line 22 and on: "Finally, we note that rough ice surfaces are associated with stronger electrical charging (Caranti and Illingworth, 1983; Dash et al., 2001; Dash and Wettlaufer, 2003), hence the presence of roughness may influence storm electrification". This is indeed very interesting link that is worth discussing in more detail. Could you say a few words explaining what mechanism underlay this phenomenon? I think this is the most far leading mechanism among other atmospheric applications.

This is indeed an intriguing possibility, that is why we speculatively mention it. However, we consider wider discussion to be beyond the scope of the present work, and we instead refer the readers to several references cited in sections 3.1 and 3.3.