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Atmospheric Chemistry and Physics Discussions

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

Anonymous Referee #2

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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 thermodynamic 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 morpholog-

ical 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.

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.

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?

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".

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?

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?

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).

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.

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.

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

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.

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.

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).

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?

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?

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.

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.

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?

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?

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

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?

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?

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?

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 characterization 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.

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!

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

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

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?

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.

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.

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.

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