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Interactive comment on “Measurement of acidic ions and their qualitative effects on snow crystal morphology and the quasi-liquid layer” by T. N. Knepp et al.

T. N. Knepp et al.

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The authors would like to thank the referee for his/her helpful and insightful comments. We believe that the revised manuscript deals with all the reviewer's concerns. Here we delineate the changes to the manuscript as a result of the reviewer's comments, in the order that they were raised. The referee posed a question regarding the determination of the acetic acid content in the chamber. We have addressed this issue in the revised manuscript, p. 7, lines 200–203. The sample air was bubbled into impingers arranged in a series fashion, which contained sodium bicarbonate solution (at a relatively high concentration compared to the amount of acetic acid introduced from the chamber), effectively optimizing the collection efficiency for acetic acid. As the gas-phase acid passes through the basic solution it reacts to form its conjugate base anion, resulting

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in a product with no vapor pressure, thus quantitatively trapping the acetic acid. This method was tested using a certified acetic acid permeation source by flowing air from the permeation oven through the sampling apparatus, with the impinger solutions analyzed by ion chromatography. This analysis yielded a permeation rate that agreed with the source's certification within 10%, with a collection efficiency of >95%. The 0.2% dissociated acid cited in the text is in reference to its interaction with pure water droplets in the atmosphere, and not to be mistaken for the extent of dissociation in basic solution. Between acid-dosed experiments and clean air experiments the chamber was washed with 2M NaOH to remove adsorbed acetic acid, followed by a Millipore water rinse (as described on p. 6, lines 173-176 of the revision). The chamber was flushed with clean air, after being washed, for a period of one month before the clean air morphologies were again grown/observed. During this time the acetic acid content in the chamber air was observed to decrease to below the detection limit with time. Due to the slight positive pressure from the flow of clean air into the chamber, room air was not able to enter the chamber, thus preventing further contamination. Referee #2 also expressed concern regarding the temperature of onset of the quasi-liquid layer (QLL), and our estimation of the layer thickness of the QLL. We agree that the temperature of onset for the QLL is highly debatable, and dependent on the analytical method employed. However, the research presented in this manuscript is not intended to provide any elucidation on this matter. We chose -10oC as the "consensus" temperature of onset, based in part on the knowledge that there exists a distinct morphological transition temperature at -10C based on numerous reports. A focus of this research was to test the hypothesis presented by Kuroda and Lacmann (1982), which stated that the morphology change that occurs at -10C relates to the disappearance of the QLL at that temperature, and that the QLL thickness directly influences the snow crystal morphology. Kuroda and Lacmann suggested the temperature of onset of the QLL to be roughly -10C for the basal plane, which is roughly consistent with the literature observations, though quite variable. If one is only interested in transition temperature shifts based on varying QLL thicknesses, our data clearly show this to be the case

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(assuming that QLL thickness relates to the concentration of AA in equilibrium with the ice) regardless of the exact temperature of onset for pure water ice (as indicated on pp. 10-13 lines 292-382 of the revised manuscript). It was not our objective to determine with accuracy the temperature of onset for the QLL; we can only interpret our data in light of others'; previous research (e.g. Kuroda and Lacmann, and Sokolov and Abbatt). Further, the freezing point depression calculation was done with the best understanding of the information available to us. The additional references provided by the referee are indeed informative, present much good science, and were therefore cited in the text, on pages 3, 4, 8, and 15 of the manuscript. Further, to resolve the referee's concerns regarding air quality, we conducted further study of the chamber air for hydrocarbon, NO, NO_x, NO_y, NO₂, and aerosol content. The results of these measurements are provided on pp. 10-11, lines 301-316 of the revised manuscript. Most significantly, we were not able to reproducibly define the conditions leading to the lack of temperature dependent morphological diversity, though there clearly exists some set of conditions under which this exists, as stated on p. 10 lines 304-307 of the revision. During the last set of clean air experiments we did observe the typical morphological transitions as seen in "dirty" air, in addition to the new clean air morphologies. In other words, these new clean air morphologies were not reproducible. The chemical conditions in the chamber were observed to be the same in all cases, to within our limits of detection, and we thus can not currently explain this variation between the clean air and the more accepted temperature-dependent morphologies. However, we can state that there does exist a set of conditions in which the temperature-independent clean air morphology is apparent. Clearly, more work needs to be done here to elucidate the exact chemical/physical conditions in which this occurs and to further understand the relevant ice surface physics. We thank the referee for pointing out technical errors and inconsistencies in the manuscript, which have been corrected in the re-submitted manuscript (see title, p. 6 lines 156, 160). To our knowledge the difference in results, if any, between synthetic and natural hair/string is unknown, though these are certainly interesting questions but which were not a subject of this study. We chose acetic acid

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for several reasons, i.e. the ease with which it can be handled and measured, it is relatively safe to work with, it is an important tropospheric acid, and the precedent of working with this acid in snow crystal experiments (e.g. Libbrecht, 2005), as stated on p. 4 lines 115-117 of the revised manuscript. We thank the reviewer again for his/her insightful comments and help in making this a better manuscript. We acknowledge, in agreement with the referee, that there remains a significant amount of work in this field with many potentially fascinating discoveries to be made. Though we can not explain these new clean air morphologies on a molecular level we feel it important to share this new observation with the scientific community so that it might be further investigated/understood.

Interactive comment on Atmos. Chem. Phys. Discuss., 9, 735, 2009.

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