Atmos. Chem. Phys. Discuss., 14, C4758–C4763, 2014
www.atmos-chem-phys-discuss.net/14/C4758/2014/

Atmospheric Chemistry and Physics
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Interactive comment on "Mesoscopic surface roughness of ice crystals pervasive across a wide range of ice crystal conditions" *by* N. B. Magee et al.

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Received and published: 12 July 2014

Environmental Scanning Electron Microscopy (ESEM) is a rapidly developing microscopic technique allowing observations of many natural objects (microorganisms, atmospheric particles, etc.) in the wet environment preserving them from dehydration. The sensitivity of ESEM to the surface morphology and enhanced (with respect to optical microscopy) focal depth makes it highly attractive for the visual observations of nucleation and growth of ice crystals. I can only share authors' enthusiasm in watching and recording the ice crystals grow and develop all kinds of spectacular surface morphology. No doubt these observations will be useful for atmospheric ice research

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community, giving the notion of high complexity of "real life" ice crystals and stimulating further research. Sharing the zeal, I would still like to express my concerns about the insufficient attention given to the experimental conditions and somewhat farfetched conclusions drawn from the incomplete observational base. I hope that careful consideration of the issues I rise below would increase the quality and scientific impact of the manuscript.

General remarks

1. My main concern is about the role of the substrate and the absence of the background pressure in the mechanism of the ice crystal growth. Both issues are very briefly mentioned in the manuscript (as in page 8400 lines 8-11: "These images do show mesoscopic surface roughness, but suggest that the presence of air or different modes of internal heat transfer may significantly affect the character of surface texture development") but not discussed further with respect to the surface morphology of the growing regime of ice crystals. However, they can be very important for comparing the ice morphology observed in ESEM with ice growing under atmospheric conditions. For example, it is known that the diffusion coefficient Dw of water vapor in air-vapor mixture is inversely proportional to the background pressure. Using approximation given by Marrero and Mason in "Gaseous diffusion coefficients" in J. Phys. Chem. Ref. Data, v1. no1(1972) for near-vacuum conditions of ESEM (-20°C, 1 Torr) the value of Dw would be 120 cm2/s, which is 600 times larger than the diffusivity of water in air at atmospheric pressure (~0.2 cm2/s). At the same time, diffusivity of water is the important factor influencing, together with the supersaturation, the crystal growth mechanism (diffusion limited vs. attachment kinetics limited, see, for example K. G. Libbrecht, The physics of snow crystals, Rep. Prog. Phys. 68, 2005). Since no background pressure was provided in the specimen chamber of the ESEM, all crystals images reported in the manuscript have been obtained with crystals grown in the attachment kinetics limited regime, meaning that the growth of crystals is controlled by the rate of the incorporation and redistribution of water molecules within the surface of the ice crystal,

and not by the rate of diffusion of water molecules towards the crystal surface. The influence of enhanced diffusivity on the sublimation morphology of ice can be even more pronounced.

The role of the substrate is not so evident for the surface morphology of the growing ice crystals. In case of freely levitated crystals, the latent heat of solidification is removed only by thermal diffusion (0.025Wmˆ-1Kˆ-1 for air at standard pressure). Because this heat removal is slow, the heating of the ice crystal may slow down the growth, driving the growth into the heating-limited regime. This effect should be more pronounced for lower pressure, because of the much stronger flux of water vapor towards the surface (see, again, K.G.Libbrecht, 2005). In case of the crystal having thermal contact to a substrate, this heating limitation will be removed because of the much higher thermal conductivity of a substrate (from appr. 2 Wm-1K-1 (mica) to 400 Wm-1K-1 for copper) and ice (2.5 Wm-1K-1). Therefore, ice crystals having thermal contact with any substrate in pure water vapor environment would be growing in the kinetic limited regime already at low supersaturation, as opposed to the atmospheric ice crystals mostly growing in diffusion limited or heating limited regime under realistic atmospheric conditions.

To my understanding, the specific thermodynamic conditions in ESEM (absence of the background pressure and thermal contact to a substrate) is also the reason why no systematic dependence of the surface morphology was observed on the degree of supersaturation and growth rate. In fact, the authors correctly recognize the growth mechanism dominant in the experiments as a 2D nucleation, which is specific for the attachment kinetics limited growth regime (see, for example, the textbook “Crystals: Growth, Morphology and Perfection” by Ichiro Sunagawa, Cambridge University Press, 2007).

This conclusion does not reduce the need to understand und quantify this mechanism of ice crystal growth nor it diminishes the value of the presented observations. My main point is that one must be very careful extrapolating the observation results obtained in the ESEM towards the atmospheric conditions. Please consider including some more careful discussion of this issues in the revised version of the manuscript. However, I fully support the suggestion made by authors, that "…the next steps should focus on efforts to examine and quantify roughness in crystals transported from cirrus-analog environments and development of a mixed air/vapor capability for ice crystal growth in ESEM", but the thermodynamics underlying this conclusion should be elaborated more carefully.

2. In contradiction to your statement (page 8398, lines 15-16), "We are confident that vapor pressure values were precisely and accurately controlled and reported;…", there seems to be an inconsistency between the ESEM conditions reported in the text and the pressure and temperatures values shown on the ESEM images. For example, the crystal shown in Figure 2 is claimed to be held at equilibrium at -23°C, and the pressure of water vapor that can be read on the ESEM image is 7.02e-1 Torr. However, the saturation value of water vapor with respect to ice at this temperature is 0.58 Torr (I'm using parameterization from Buck, A. L., New equations for computing vapor pressure and enhancement factor, J. Appl. Meteorol., 20, 1527-1532, 1981). That would mean supersaturation of 1.21, instead of 1 as would be required by the equilibrium condition. Same story with Figure 4, panel C: pressure reads 0.19 Torr and reported temperature 39°C, but even if I assume the maximum possible temperature offset of 2.5K, the saturation vapor pressure for 36.5°C would be 0.143 Torr, not 0.19!. At the same time the crystal in the panel is clearly evaporating. Was some other independent pressure measurement employed or the temperature offset is stronger than you admit? A temperature and humidity calibration with e.g. deliquescence of NaCl would be highly recommendable, as well as placing additional temperature sensor on the surface of substrate.

3. I suggest that more details should be provided for the diffusion chamber experiment. How did you control the water vapor pressure at this temperature (it is below 4 Pa at -50°C)? How did you manage to nucleate ice at supersaturation below RHi of 110%?

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Have you been measuring the pressure and temperature inside the containment cell? How many ice crystals were collected into the cell for transport? Would that be possible to observe the crystal growth in the diffusion chamber visually (by means of a microscope) – that would provide necessary information on the growth regime!

Minor comments:

1. You might want to mention the method of formvar (polyvinil acetal resin) replication of ice crystals to study its surface morphology (introduced by Vincent J. Schaefer, Science, New Series, Vol. 93, 1941). This method, combined with scanning electron microscopy, offers quite a detailed insight into the surface morphology and habit variability of atmospheric ice crystals. Some works involving SEM date back already to 1950th, first visualization of different roughness of basal and prismatic facets by (Kuroiwa, J. of Glaciology v.8 no. 54, 1969). At that time the link between etched pits and defects in ice has been suggested. A short review of this method would be a valuable addition to the manuscript (introduction and literature survey).

2. “Wide range of ice crystal conditions” (in the title and elsewhere) is not very precise statement. In view of discussion of the different growth regimes (see general remarks) would it not be better to reduce it to “wide temperature range”?

3. You have mentioned many different substrates in section 2.1, but never discussed any of them afterwards. At the same time nucleation of ice on natural mineral substrates is highly interesting, especially as a potential source of induced crystallographic defects in the nucleus at the early stage of growth. Could you report any substrate dependence?

4. I wonder if there is any sense in reporting the roughness measure obtained for such small areas of the crystal (section 3.1.2). To my understanding, the main idea of defining roughness is to provide a measure for the whole population of crystals grown under similar conditions. This measure could be used then for correctly predicting their optical properties. How representative is the shown value for the single crystal and for

the ensemble?

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