

Interactive comment on “Cloud chamber experiments on the origin of ice crystal complexity in cirrus clouds” by M. Schnaiter et al.

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Dear Authors,

Thank you very much for citing our papers in your work. That was indeed a very nice surprise because our works are usually being ignored by atmospheric scientists dealing with the formation and modeling of cirrus and PSCs. A reason for this could be that they are not able to comprehend a well-known-for-decades fact that ice is highly intolerant to impurities and, consequently, during ice nucleation and subsequent freezing of aqueous solutions a phase separation occurs into pure ice and a freeze-concentrated solution (FCS).

Below I give you a short list of our papers in which you will find pictures and videos

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(3, 5-7 in a list below) which demonstrate the freezing process in situ and show that indeed there is a freeze-induced phase separation (FIPS) into pure ice and FCS both in aqueous bulk solutions/formulations relevant for biotechnology and small aqueous drops of size and composition relevant for UT and polar stratosphere. After you have read our papers, I hope you will rewrite these sentences of yours and also add a couple of sentences into introduction about the FIPS in the atmosphere.

These are your sentences: “A possible explanation for this observation could be the formation of concentrated H₂SO₄/H₂O residuals on the ice crystal surface, which affects the regular crystal growth. Such a separation into solid ice and unfrozen residual solution was observed by calorimetric measurements on aqueous H₂SO₄ droplets in the cirrus temperature range 190K < T < 230K (Bogdan et al., 2006; Bogdan, 2006). Although emphasized by the authors, the formation of a complete H₂SO₄/H₂O coating of the crystal surface was not unambiguously proven by these studies. Further studies with homogeneously nucleated ice particles are necessary to investigate the impact of unfrozen H₂SO₄/H₂O residuals on the ice crystal complexity.”

Note, that freezing aqueous drops do produce spherical ice particles. This is easily observed in situ during freezing experiments on micrometer-scaled drops. The FCS-coating is very thin and, consequently, can exist around young cirrus ice particles (see papers 1,2 from the list below).

This is a short list of our papers:

1. Bogdan, A. and Molina, M. J.: Why does large relative humidity with respect to ice persist in cirrus ice clouds? *J. Phys. Chem. A*, 113, 14123-14130, 2009.
2. Bogdan, A. and Molina, M. J.: Aqueous aerosol may build up an elevated upper tropospheric ice supersaturation and form mixed-phase particles after freezing, *J. Phys. Chem. B*, 110, 12205-12206, 2010.
3. Bogdan, A., Molina, M. J., Tenhu, H., Mayer, E. and Loerting, T.: Formation of mixed-

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phase particles during the freezing of polar stratospheric ice clouds, *Nature Chem.*, 2, 197–201, 2010.

4. Bogdan, A. and Loerting, T.: Impact of substrate, aging, and size on the two freezing events of $(\text{NH}_4)_2\text{SO}_4/\text{H}_2\text{O}$ droplets, *J. Phys. Chem. C*, 115, 10682–10693, doi.org/10.1021/jp2007396, 2011.

5. Bogdan, A., Molina, M. J., Kulmala, M., Tenhu, H. and Loerting, T.: Solution coating around ice particles of incipient cirrus clouds, *PNAS*, July 2, vol.110, no.27, E2439, 2013.

6. Bogdan, A., Molina, M. J., Tenhu, H., Bertel, E., Bogdan, N. and Loerting, T.: Visualization of freezing process in situ upon cooling and warming of aqueous solutions, *Scientific reports* 4, 7414, DOI: 10.1038/srep07414, 2014.

7. Bogdan, A., Molina, M. J., Tenhu, H. and Loerting, T.: Multiple glass transitions and freezing events of aqueous citric acid, *J. Phys. Chem. A*, 119, 4515–4523, doi.org/10.1021/jp510331h, 2015. 8. Räisänen, P., Bogdan, A., Sassen, K., Kulmala, M. and Molina, M. J.: Impact of $\text{H}_2\text{SO}_4/\text{H}_2\text{O}$ coating and ice crystal size on radiative properties of sub-visible cirrus, *Atmos. Chem. Phys.*, 6, 4659–4667, 2006.

Interactive comment on *Atmos. Chem. Phys. Discuss.*, 15, 30511, 2015.

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