Review of "New investigations on homogeneous ice nucleation: the effects of water activity and water saturation formulations" by Baumgartner et al.

Verdict

I recommend that the paper be published subject to major revisions.

General comments

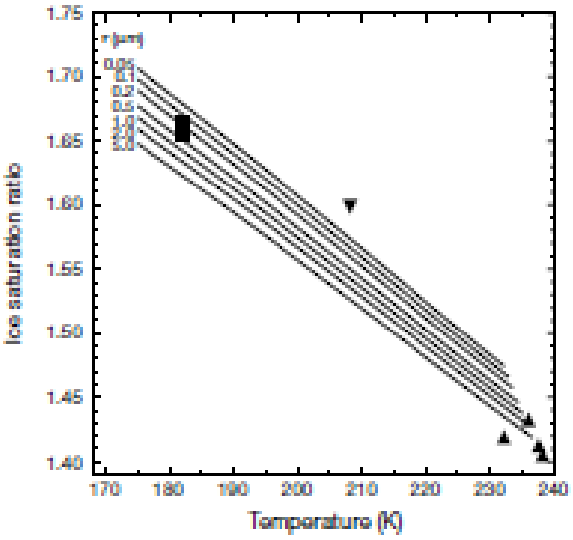
At the start of the paper, it needs to be clarified that there are two broad types of homogeneous freezing: that of solution droplet aerosols (CCN) and that of cloud-liquid/rain. It also needs to be clarified that the paper is only about the former.

An intellectual weakness of the paper is that there seems to be a rather ad hoc trial of various combinations of water activity and vapour pressure schemes, with a recommendation of an optimal combination without clear arguments independently for why these schemes are best. So, the reader may infer that there is a likelihood of a balance of compensating biases which could be unsettled if a different chemical species of aerosol (not sulphate) or different thermodynamic conditions were simulated.

Need to argue more about whether the Nachbar scheme is more accurate than the Murphy and Koop scheme for vapour pressure. Yes, the Nachbar scheme is from recognition of a new phase of water. But what do the authors think about the likelihood of this assumption being correct ? What new information has come to light that supports this assumption ?

It would be good to comment on AIDA measurements by Mangold et al. (2005) presented at EGU about *lower* humidities for freezing of ammonium sulphate than predicted by Koop. Was this an anomalous observation perhaps ?

For modellers who use the Koop scheme, we freeze CCN aerosols at temperature- and size-dependent critical saturation ratios. How should we modify our schemes ? Modellers have typically simply created a lookup table of these critical ratios from the published plot of Koop et al. (2000):



Can a simple numerical fix be recommended ? If a line of best fit can be given for the "best fit" of Fig. 15a for r = 0.05 um, do we just shift the line downwards for larger sizes ?

In summary, this paper represents a significant advance in knowledge. More discussion of the merits of the various schemes (Luo, Nachbar, Carslaw etc) is needed. Also an awareness of the place of homogeneous aerosol freezing among other types of homogeneous freezing needs to be conveyed. Above all, modellers need to be told how to upgrade their lookup tables (e.g. saturation ratio as function of size and temperature) in light of these important results from AIDA.

Specific comments

Line 24: the statement is too narrowly centered on aerosols: "*In contrast, homogeneous nucleation refers to the spontaneous freezing of pre-existing solution particles*". In fact, homogeneous nucleation of ice refers to spontaneous freezing of any drop, whether a solution droplet ('homogeneous aerosol freezing') or a liquid cloud- or rain-drop ('homogeneous freezing of cloud-liquid or rain').

Line 363: It is interesting that, qualitatively, a similar sort of dependency on updraft speed is found with homogeneous freezing of cloud-droplets near -36 degC. With these too, the largest freeze first during ascent and an ascent-dependent fraction of all the cloud-droplets will freeze with the rest evaporating due to the ice particles lowering the humidity. There is preferential freezing of the smaller cloud-droplets. So the number of ice particles initiated increases with updraft speed. Phillips et al. (2007, JAS) parameterized this and showed that it has an order-of-magnitude impact on mesoscale averages of the ice concentration aloft.

Line 573: The paper suggests an optimum combination: "Carslaw's method for the water activity in the nucleation routine and Luo's method in the Köhler equilibrium routine, in combination with Nachbar's formulation of the saturation vapor pressure". But what are the independent reasons for thinking that Carslaw's method is better than alternatives for water activity ? Why is Luo's method better than

alternatives for the Kohler equilibrium routine ? Why is Nachbar's formulation best ? Need to provide some independent reasoning or evidence.

Could there be a serendipitous compensation of opposing biases among these three schemes, giving the impression of realism for the wrong reasons ?

Technical details

Figure 13: this has wrong entries in the legend (lines instead of symbols) and multiple panels have identical titles and legends without clarity from the figure caption.