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Atmospheric Chemistry and Physics
Discussions

Interactive comment on "Pre-activation of ice nucleating particles by the pore condensation and freezing mechanism" *by* R. Wagner et al.

Anonymous Referee #2

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General comments

Wagner et al. describe experiments in the AIDA cloud chamber on a microphysical process termed "pore condensation and freezing mechanism" (PCF). This process leads to preactivation of certain types of aerosol once they have temporarily experienced temperatures some degrees lower than 237 K, the spontaneous freezing limit of pure water. The process works even although neither water nor ice saturation was achieved during the cooling process. The reason for this is the negative Kelvin effect in appropriate pores or between structural elements of the aerosol particles which substantially lowers the saturation water vapour pressure so that condensation can proceed at ambient vapour pressures much lower than saturation (with respect to a plane water

surface).

The paper is quite interesting but hard to read. The reason for this trouble is the great amount of detail that must be provided for every experiment. Fortunately, there are several appendices where additional details are transferred to. Because of this it is particularly necessary that the theoretical principles are explained clearly. In the following I would like to make some suggestions for modifications that would to my view make the theoretical part more concise, hence clearer.

The paper is an appropriate contribution to ACP.

Specific comments and suggestions

Description of the theory

In principle, the PCF process is easy to understand. First we need condensation of liquid water in a pore under water-subsaturated conditions. This is possible because of the negative Kelvin-effect (i.e. Kelvin effect with negative radius of curvature), which effectively lowers the saturation vapour pressure. The radius of curvature is determined by the temperature and the actual relative humidity. It is proportional to the pore dimension, and thus it sets a maximum pore dimension for which condensation is just possible. Then we need freezing of the liquid, which is only possible if the pore size is larger than the critical ice germ size, again dependent on temperature, which sets the lower pore radius. Thus the range of pore sizes is set by the two described conditions. Although all this is mentioned in the paper, it is to my taste too dispersed and thus the reason why only a certain pore size range can cause the observed effects can easily be overlooked. For instance, the abstract only mentions the appropriate range, 3-8 nm, without any explanation. For readers that only read the abstract it is not possible to understand why exactly this is the appropriate range. Nobody will remember the values later, but a concise explanation can be memorised. I suggest that such a short summary of how the processes set the size boundaries of the pores is provided, perhaps in 4.1 before the details with formulas etc. are explained in the following subsections. In the abstract, one sentence like "this range is set by a combination of requirements from the negative Kelvin effect for condensation and a critical size of ice germs for ice growth" will suffice.

Other comments and questions on Section 4

1) Marcolli (2014) introduced the term "inverse" Kelvin effect, and it is used here as well. But the notion "inverse" is not explained, neither by Marcolli nor here. Probably it refers to the negative curvature of the water meniscus in a pore, and then it might be better to use "negative" Kelvin effect. As it also induces condensation at negative (!) supersaturation, not inverse supersaturation, I suggest to replace "inverse" by "negative".

2) Why is it the surface tension between water and air that appears in Eq. 1 and not the surface tension between water and the pore wall? And how is the radius of curvature of the water meniscus related to the pore size. I have the impression that these two (generally different) quantities are taken to be equal in section 4.1.1.

3) Point B* in the diagrams: How can water in the pores freeze heterogeneously when there is a quasi-liquid layer between the ice and the pore wall? Doesn't this exclude the possibility of heterogeneous nucleation?

4) I was wondering why condensation in the pores that commences already at your starting T (253 K) in pores with D<11 nm, does not lead to a size range 3-11 nm. Instead, the maximum pore dimension is 8 nm, wich refers to 237 K. How can one explain this. Does the water in the larger pores (8-11 nm) evaporate before it can freeze?

Minor comments

1) P. 29001, ll. 4-5: "an even smaller fraction" should be quantified.

2) P. 29001, ll. 14-15: correct hyphenation of "represent".

3) P. 29002, ll. 10-11: "INPs that have crystallized...". It is unclear whether these are ice crystals or other crystals that contain crystal water.

4) Section 3.1: It is reported that the *unprocessed* CBV400 starts nucleating ice at RHi=102% and that this is probably not due to deposition nucleation but due to pre-activation following the PCF process pathway. I do not understand this argument: when the aerosol is unprocessed, as stated, how can it simultaneously be pre-activated?

Figures: As water saturation is important for your explanations, RH with respect to liquid water should be indicated as well in your figures (in addition to RH wrt ice).

Figure 5c: red hatching not visible in my printed version.

Interactive comment on Atmos. Chem. Phys. Discuss., 15, 28999, 2015.