I thank the reviewer for his/her thoughtful comments and the corrections, which I address below in italic.

The manuscript by Marcolli reviews previous laboratory experiments on the pre-activation of aerosol particles by the pore condensation and freezing (PCF) mechanism. The PCF mechanism has been introduced by the same author two years ago as a potential ice nucleation pathway for heterogeneous ice formation at temperatures below 235 K and relative humidities below water saturation. Under such conditions, heterogeneous ice formation was before conceptually ascribed to deposition nucleation without involving liquid water. Depending on the temperature for melting and the relative humidity for sublimation, ice trapped in pores from a first nucleation event can facilitate ice crystal growth in a second nucleation event, i.e., lead to pre-activation. The author first describes the conditions for pre-activation in terms of pore size and RH for several pore geometries. Then, previous literature on pre-activation is summarized, and the data are analyzed by categorizing them with respect to the experimental set-up, the aerosol type, and the pre-activation conditions. Finally, potential scenarios where pre-activation could contribute to ice formation in the atmosphere are outlined.

The manuscript is generally well written and the previous literature thoroughly reviewed. The pre-activation topic, as first emerged around 1950 but then neglected for decades, has received new attention in the past years – and it is a valuable effort and therefore fits well within the scope of ACP to critically review the current state of knowledge in order to stimulate further experimental and theoretical work on this issue. I therefore support the publication in ACP, but have some concerns, as outlined below, which should be addressed before final publication.

Major comments:

1) My first major concern is the widespread use of the term “PCF mechanism” to account for all pre-activation experiments and trajectories discussed throughout the manuscript. If I understand correctly, the PCF mechanism was proposed as kind of challenging hypothesis against the classic view of deposition nucleation at temperatures below 235 K. And there are good reasons for this hypothesis, most importantly the well-documented, strong increase of the ice nucleation efficiency of numerous types of aerosol particles just below 235 K. But the manuscript’s title “Pre-activation of aerosol particles by pore condensation and freezing” implies that in all considered examples the PCF mechanism accounts for initial ice formation and pre-activates the particles. In some cases, the pore ice might indeed be formed by the PCF mechanism, but there are numerous examples where there is no need to explicitly invoke this theory. For example, for all wet trajectories discussed in Figs. 1-3, initial ice formation and potential pre-activation occur by droplet activation and immersion freezing somewhere on the particle surface. At least if I understand correctly, ice formation by immersion freezing is not supposed or required to happen directly inside the pore. The susceptibility to pre-activation would then just depend whether ice propagates into the pore or not (e.g. inhibited by a narrow pore opening of an ink-bottle-shaped pore), but is not explicitly caused by the PCF mechanism as defined above. Also for the dry trajectories where initial ice formation occurs at colder temperatures, there is no need to exclusively infer the PCF mechanism as the formation pathway for pore ice. Wouldn’t it be possible that certain pores or void spaces in an aggregate particle can also be filled with ice in a “conventional” deposition nucleation pathway? Instead of writing e.g. on page 9, line 15-16 “*. . .* reviewed under the presumption that pre-activation occurred by the PCF mechanism”, I would like to see a more general statement that the experiments are analyzed under the assumption that pre-activation is due to the formation and retention of ice in pores, but that there are various mechanisms by which pore ice can be formed, one of them being the newly proposed PCF mechanism.

I agree with the reviewer. I changed the text in the manuscript accordingly.

The revised manuscript carries now the title: “Pre-activation of aerosol particles by ice preserved in pores”

2) As a second major issue, I would like to see a bit more discussion on the sublimation of ice in pores and whether and for how long ice could survive even in an ice-subsaturated environment. Are there any experimental or modeling studies on that issue? The author argues on the one hand, for example for the dry trajectory shown in Fig. 1, that ice immediately sublimates in the 8 nm-sized cylindrical pore after RHi drops below ice saturation during adiabatic heating. On the other, the generally better pre-activation efficiency observed in the cold stage experiments is always explained by the hypothesis that pore ice is conserved in voids between the substrate and the particle, even if conditioning occurs at RHi values well below 100%. But why should ice located between the particle and the substrate be more stable against sublimation at ice-subsaturated conditions compared to the case where ice is retained in pores within the particle or between particle aggregates? If there is no valid argument for this, such

a definite conclusion as e.g. on page 25, line 17-18 cannot be drawn.

The timescale is indeed an important issue. However, there is no simple way to calculate or estimate the sublimation rate of ice in pores. There are some recent papers that treat this question. Based on these studies, I added a new section 3.2 to the revised manuscript with the title "Kinetics of pore ice sublimation".

Minor comments:

Page 4 & 5 in general: Please also indicate in the main text how the ice and water vapor pressures were calculated, I only discovered this information in the Figure captions.

I added the following sentence to the first paragraph of section 3.1: "Ice saturation and water saturation are calculated with the parameterizations from Murphy and Koop (2005)."

Page 5, Sect. 3 in general: There is frequent reference to the melting temperature of ice in pores throughout the discussion (computed with Eq. 4). Maybe it would be useful to include of graph of the pore-diameter-dependent melting temperatures, similar as in Marcolli (2014).

Instead of reproducing the Figure of Marcolli (2014), I prefer to explicitly refer to it at the end of Section 2. Moreover, the pore melting temperatures are indicated in Figures 1 – 3 of this review by the high temperature end of the dashed portion. Water in pores of 2 nm and 1 nm remains liquid. This is now explicitly stated in the figure captions of Figs. 1 – 3.

Page 6, lines 15-16: This is one occasion where the immediate sublimation of pore ice at RHi below 100% is assumed (see comment above). But later on (e.g. page 13, line 1 or page 14, line 14,15), it is argued that ice in spaces between a particle and a substrate could trigger ice crystal growth even for RHi « 100% during conditioning.

For the discussion of the trajectories, thermodynamic equilibrium was assumed. This is stated at the beginning of Sect. 3.1. To make this clearer, the statement on page 6, lines 15 – 16 is changed to: "Therefore, in a cylindrical pore of 8 nm, no persistent pre-activation occurs for T < 233 K because of the sublimation of the pore ice."

When irregularly formed particles are deposited on substrates in cold stages, voids with narrow opening may form between the substrate and the deposited particles. These voids can swell when they fill with liquid water and should be able to keep ice below ice saturation analogously to the case of swelling pores discussed in Fig. 3. Thus, a thermodynamically stable state is assumed and the pore ice should be preserved permanently.

Page 7, lines 10 – 12: I do not understand the line of argument here.

This sentence is improved in the revised manuscript. It now reads: "However, a cylindrical pore with 4 nm diameter should have a similar ability of pre-activation".

Page 15, lines 3 – 5: Here, it might be good to refer to the Adler et al. (2013) study, where the formation of porous particles upon freeze-drying was clearly shown with the microscope images.

I added the following sentence to the revised manuscript: "This hypothesis was confirmed by Adler et al. (2013) who showed that freeze drying leads to highly porous particles."

Page 17, line 15: See above: Why should particularly this ice between particle and substrate survive long exposure to dry conditions and then contribute to pre-activation?

I explain this hypothesis better by reformulating: "When irregularly formed particles are deposited on substrates in cold stages, voids with narrow openings may form between the substrate and the deposited particles. These voids are likely to swell when they fill with liquid water and should be able to keep ice below ice saturation analogously to the case of swelling pores discussed in Fig. 3."

Page 23, line 1 ff: In such a case (when the ash particles are not pre-activated at RHi < 100%), pre-activation would then require a preceding ice nucleation event with the ash particles and not just cooling to low temperatures where pore water could freeze at ice-subsaturated conditions. This would certainly lower the relevance of pre-activation.

I agree. The ash of the Eyjafjallajökull *eruption needs to have gone through a cirrus cloud to become pre-activated. Indeed, cirrus clouds were present at that time (Schumann et al., 2010). However, the porosity of volcanic ashes is variable. In other cases, pre-activation might also be possible at ice-subsaturated conditions.*

Page 24, line 8: Could you include references that meteoritic particles have proven to be poor INPs? I recall e.g. the study by Saunders et al. (2010) where nanoparticles of iron oxide, silicon oxide and magnesium oxide were considered as relatively efficient INPs at T < 220 K.

Saunders, R. W., Möhler, O., Schnaiter, M., Benz, S., Wagner, R., Saathoff, H., Connolly, P. J., Burgess, R., Murray, B. J., Gallagher, M., Wills, R., and Plane, J. M. C.: An aerosol chamber investigation of the heterogeneous ice nucleating potential of refractory nanoparticles, Atmos. Chem. Phys., 10, 1227-1247, doi:10.5194/acp-10-1227-2010, 2010.

Biermann et al. (1996), and Mason and Maybank (1958) investigated the ice-nucleating ability of meteoritic material. I added these citations to the revised manuscript.

Page 26, line 13: I actually like the speculations about the scenarios where pre-activation could contribute to atmospheric ice formation in Sect. 6, but a statement in the summary section like "are likely to influence ice cloud formation" is not enough substantiated and should be more clearly denoted as a hypothesis. The same holds for the statement on page 25, line 17-18 as outlined above.

I agree that these are speculations and I weaken the statement in the summary and conclusions section as requested by the reviewer.

Technical corrections:

Thanks for the corrections

Page 1, line 1: aerosol *corrected*

Page 1, line 19: . . . is perfectly sheltered . . . *corrected*

Page 3, line 10: humidities *corrected*

Page 4, line 29: wrong Greek symbol for the density of liquid water *corrected*

Page 5, line 1: use Greek symbol for the surface tension *corrected*

Page 6, line 2: sublimating pore ice *corrected*

Page 6, line 11: liquid water within the pore evaporates *corrected*

Page 7, line 23/24: ice crystal sublimates *corrected*

Page 13, line 19: maybe it is meant: among the few samples *yes, corrected*

Page 26, line 4: relevance . . . depends *corrected*

Page 43, line 1-2: maybe it is meant: Tcond – conditioning temperature *yes, corrected*

Page 46, line 4: shouldn't it be 238 K ? *yes, corrected*

Interactive comment on Atmos. Chem. Phys. Discuss., doi:10.5194/acp-2016-837, 2016.