We thank Referee #1 for his comments to improve the quality of the paper. Below we will give a point-by-point answer to the individual comments.

COMMENT:

Page 8923, line 22 – remove "also"

ANSWER:

Will be removed.

COMMENT:

Page 8924, line 10 – Tg' is defined in the next sentence; however, I would like to see

the definition first.

ANSWER:

Will be re-arranged as follows:

"A useful quantity for the interpretation of the temperature-dependent ice nucleation behaviour observed in AIDA is the temperature T_g' that is defined as the intersection between the curves of the glass transition temperatures, T_g, and the ice melting temperatures, T_m, in the state diagram (Zobrist et al., 2008). This is because the AIDA expansion runs are started at almost ice-saturated conditions in the chamber volume (see Sect. 3), so that the composition of liquid citric acid aerosol particles prior to the expansion runs adjusts approximately to that defined by the T_m curve. For citric acid, T_g' is at about 212 K. For a chamber temperature above 212 K, the aerosol particles were therefore initially in a liquid state, resulting in homogeneous freezing of the citric acid aerosol particles in the corresponding expansion run at a relative humidity similar to liquid sulphuric acid droplets. Below 212 K, a much smaller number fraction of the now glassy citric acid particles heterogeneously nucleated ice over a range of relative humidities below the homogeneous freezing threshold."

COMMENT:

Page 8925, line 13 - This is a really long sentence. I would break this up into 2 sentences.

ANSWER:

Will be re-phrased as follows:

"After the ice cloud from this first expansion run had sublimed, we started a second expansion run at the same temperature and with these cloud processed aerosol particles. In this second run, ice nucleation was detected at very low supersaturation levels well below that required for homogeneous freezing."

COMMENT:

Page 8925, line 8 - Does Murray et al. (2010) give a hypothesis for why the glassy citric

acid particles do not heterogeneously nucleate ice on a first expansion run when the initial temperature is close to Tg'? If so it would be nice to see that explanation in the introduction in case the reader is not familiar with Murray et al. (2010).

ANSWER:

No, this issue was not discussed in depth by Murray et al. (2010). It is only evident from the ice nucleation results presented in Fig. 2 of Murray et al. (2010) that the expansion run 5 that was started only slightly below Tg' led to a much smaller heterogeneous ice mode (as indicated by the size of the circles) compared to run 3 that was started deeply in the glassy regime.

COMMENT:

Page 8926, line 26 – When the aerosol particles are cooled below Tg', the particles vitrify. Wouldn't this greatly increase the equilibration times? It says decrease on page 8927. And if the ambient RH does not equal the water activity of the particles, how do you know where points X1 and Y1 lie on Figure 1?

ANSWER:

Yes, this increases the equilibration times, we will correct this error on page 8927. The trajectories in Fig. 1 are plotted as a function of the relative humidity of the air, RH_w, and not as a function of the water activity of the particles. In the AIDA chamber prior to an expansion, RH_w is controlled by the ice coated chamber walls and the starting points of the trajectories thus approximately lie on the T_m curve. We will clarify this extending our statement on page 8926, line 19:

"As described in Sect. 3, the inner walls of the AIDA chamber are coated by a thin ice layer in preparation for the expansion runs. Prior to an expansion, RH_w is thus controlled by the ice coating on the chamber walls and the starting points of the two trajectories are given approximately by the points X1 and Y1 on the ice melting point curve."

COMMENT:

Section 3.1 – Is this entire section needed? I feel like most of the particulars are discussed in other publications. Perhaps this section can be moved to a supplemental section or removed all together. Of course the particulars to this specific manuscript should be left in. Maybe the particulars would go in what is now section 3.2?

ANSWER:

As also suggested by Referee #2 (specifically addressing the too detailed TDL description), we have greatly condensed the information from Sect. 3.1. We still think that it is easier for the reader to split the information from Sect. 3 into two sub-chapters. The revised Sect. 3.1, however, will only include a brief description of the AIDA chamber and the humidity measurements, the methodology of expansion cooling, as well as the final paragraph with the additional instrumentation (OPCs, FTIR, light scattering). This is the minimum information needed for a reader previously unfamiliar with

AIDA to understand the experiments and the measurement data presented in this manuscript. Unnecessary details of the chamber operation and the TDL system were removed. The new Sect. 3.1 is as follows:

"The AIDA chamber is a cylindrical aluminium vessel of 4 m diameter and 84.3 m^3 volume which is located inside an isolating container whose interior can be cooled to 183 K by evaporating liquid nitrogen inside two heat exchangers (Fig. 2) (Wagner et al., 2006b). The temperature is controlled by ventilating air around the aerosol vessel and a mixing fan is continuously operating inside the vessel to ensure for homogeneous conditions in terms of temperature, relative humidity, as well as aerosol and ice particle number concentration throughout the chamber volume. The aerosol vessel is cleaned by pumping to a pressure below 1 hPa and performing a series of flushing cycles with particle-free synthetic air. Then, a specific amount of purified water from a heated reservoir is evaporated into the evacuated chamber. Water is added in excess to induce ice coverage of the inner chamber walls. After humidification, the chamber is refilled to ambient pressure with particle-free synthetic air. The particle background prior to aerosol injection is typically well below 1 cm^{-3}. Due to the ice covered chamber walls, the relative humidity of the air is close to an ice saturation ratio, S_{ice}, of unity, with S_{ice} denoting the ratio of the prevailing water vapour pressure, $p_w(T)$, to the saturation water vapour pressure over ice, $p_{w,ice}(T)$ (Murphy and Koop, 2005), at the same temperature. The water vapour pressure, both under cloud-free and in-cloud conditions, is measured in situ with high-resolution tuneable diode laser (TDL) absorption spectroscopy at near-infrared wavelengths between 1368 and 1372 nm with an uncertainty of ±5%. The absolute accuracy of $p_w(T)$ is mainly determined by the uncertainty of the line strength of the rovibrational water vapour transition that is scanned during the measurements (Wagner et al., 2008).

After the preparation of the chamber, dilute aqueous solutions of the investigated organic compounds were injected into the chamber as described in the Sect. 3.2. To investigate the ice nucleation ability of the generated organic aerosol particles, supersaturations with respect to the ice phase have to be established. This is achieved by expansion cooling, i.e., the AIDA pressure is reduced by controlled pumping, typically from ambient pressure to about 800 hPa. Depending on the pumping speed, humidification rates, dS_{ice}/dt, between about 0.02 and 0.3 min^{-1} can be controlled. The potential formation of ice clouds is detected by a comprehensive set of instruments whose details are described in a recent review and will therefore only briefly be summarised here (Wagner et al., 2009).

The number concentration of nucleated ice crystals is measured by two optical particle counters (OPC1 and OPC2, type WELAS2000, Palas GmbH Karlsruhe) that are located below the cloud chamber within its isolating containment. Both instruments sample from the chamber volume through vertically arranged stainless steel tubes and cover the size range from 0.7 – 40 μm (OPC1) and 2.3 – 104 μm (OPC2). The number concentration and size of ice particles can also be retrieved from in situ infrared extinction measurements that are performed between 6000 and 800 cm^{-1} at 4 cm^{-1} resolution by coupling a FTIR spectrometer (IFS66v, Bruker) to an open-path multiple reflection cell inside the AIDA chamber (Wagner et al., 2006a). Moreover, the nucleation and growth of aspherical ice crystals is detected by light scattering and depolarisation measurements. The instrument records the intensities of laser light of 488 nm wavelength that is scattered from the particles in the centre of the chamber in forward (I_{for}, scattering angle 2°) and backward (I_{back}, scattering angle 178°) direction. In the backward direction, the detected light intensity is polarisation-resolved with $I_{back,par}$ and $I_{back,per}$ denoting the backscattered intensities parallel and perpendicular to the incident polarisation state of the laser light. The depolarisation ratio, δ, is then defined as $\delta = I_{back,per}/I_{back,par}$. Apart from very small particles or particles with extreme aspect ratios, δ is different from zero for light scattering by aspherical particles (Mishchenko et al., 1996; Zakharova and Mishchenko, 2000). The formation of aspherical ice crystals by homogeneous freezing of liquid aqueous organic aerosol particles or by heterogeneous nucleation on glassy aqueous particles is therefore evidenced by an increase of the depolarisation ratio above the background level of about 0.02 – 0.03 that is observed in the presence of spherical seed aerosol particles. The small background level results from Rayleigh scattering by air molecules, minor misalignments in the optical set-up, as well as the only 99% degree of linear polarisation of the incident laser light."

COMMENT:

Page 8932, line 9 – I had to look the word envisaged up in a dictionary. Can a different

word be used here?

ANSWER:

Was changed as follows:

"The injection periods were adjusted to the designated experimental procedure. If repeated expansion runs with the same aerosol load or long cooling/heating periods of the AIDA chamber were intended, the initial number concentration was increased to account for the expected aerosol loss throughout the experiment."

COMMENT:

Figure 4 – There is a lot going on in Figure 4. I like the left 2 plots. Can some of

the right 5 plots be removed? In reality I am mostly paying attention to the Sice plot

(second one down) and the Nice/cm-3 (fourth one down). I know that the others plots

are there for completeness but can they be put into a supplementary section?

ANSWER:

In addition to the S_{ice} and N_{ice} sub-panels , also the first plot with p and T is necessary to comprehend the temporal order of an experiment (start of pumping, stop of pumping, start of refilling etc.). Concerning the two remaining sub-panels, the OPC scatter data and the depolarisation data, we consider the scatter plots from the OPCs as the most illustrative way to display the overall ice nucleation behaviour in the expansion runs, e.g. to demonstrate the absence of ice formation in run 1E (Fig. 6), to display the dual nucleation event in run 3H (Figs. 10&11), to evidence the appearance of the pre-activation behaviour in run 4B (Fig. 12) and its absence in run 5B (Fig. 13). There are only a few occasions where the OPC scatter data are maybe obsolete. But for the sake of a uniform graphical representation of the AIDA data which is introduced in Fig. 4 (right panel) and then maintained throughout the entire manuscript, we would strongly propose to keep the OPC subpanels in all figures.

The same reasoning, to a lesser extent, also holds for the depolarisation data. Admittedly, they are not explicitly discussed in all of the expansion experiments, but they are also referred to in the general discussion in Sect. 4.5 (page 8950, line 15) and we would therefore also like to show these data and not only explain them in words. In our opinion, it is not particularly practicable to transfer only the depolarisation sub-panels to a supplementary section because without the other subpanels showing p, T, and S_{ice} etc. the time evolution of the depolarisation data cannot be fully understood.

COMMENT:

Figure 5 (left panels) – I was hoping that the colored traces (second panel) would be labeled as in Figure 4. I had to keep looking back to remind myself what the colors meant.

ANSWER:

Yes, we will add the labelling once again in Fig. 5 so that the reader gets accustomed to the colour coding.

COMMENT:

Figure 5 (left panels) – Now I see why the authors wanted to put the depolarization ratio panel in the figures. If I am not mistaken, it is to point out the reason for a few ice crystals forming prior to homogeneous nucleation. I still think that this can be explained in words and without a full on panel in the figure.

ANSWER:

See our discussion from above.

COMMENT:

Figure 7 (bottom panel) – The black and the brown line are hard to distinguish from one another. Can a different color be used?

ANSWER:

Yes, we will use a different colour (olive-green). Fig. 14 will be changed accordingly.

COMMENT:

Page 8942, line 5 – I think the authors mean ". . .a typical pair of homogeneous freezing and pre-activation runs, respectively."

ANSWER:

Exactly, we will include the "…, respectively."

COMMENT:

Page 8942, line 10 – Do the authors have a hypothesis concerning why ice forms at an Sice value of 1.05 during the 3C pre-activation run and 1.12 in run 2C? It seems the only difference is the starting temperature (224 K in 2C and 230 K in 3C).

ANSWER:

We have addressed this issue in the overall discussion on page 8954, line 3ff, speculating that the longer time period between the homogeneous freezing and the pre-activation run in Exp. 2 could explain the different threshold compared to Exp. 3. This could be due to the sublimation of potential ice embryos in pores or a modification of the surface properties. But we forgot to mention that also the starting temperature in run 2C was different. We will therefore extend our discussion on page 8954, line 3ff as follows:

"The nucleation threshold in the pre-activation run 2C, however, was slightly higher than in typical pre-activation runs conducted only 30 min after the homogeneous freezing run (e.g. run 3C). On the one hand, this might be simply due to the different starting temperatures between run 2C (224 K) and run 3C (230 K). On the other, potential ice embryos in pores might sublime when the particles are exposed to $S_{\mathrm{ice}} < 1$ for long time periods or the surface properties of the porous glassy aerosol particles might change, thereby explaining the different nucleation thresholds between run 2C and run 3C. Further experiments are needed to confirm whether this was just a singular observation or a general trend which might even get more dominant for longer time periods of one or several days before the second freezing cycle and/or exposure to ambient relative humidities lower than 70% with respect to ice."

COMMENT:

Wilson et al. 2012 is referenced a multitude of times (9 times by my count) and some of the analysis of the results are dependent on this paper. Has Wilson et al. been peer reviewed? If not I would like to see this paper published in ACP before Wagner et al. is published.

ANSWER:

It was arranged that the Wagner et al. and Wilson et al. manuscripts appeared at the same time on the ACPD discussion forum and that they were properly cross-referenced. The same will certainly be arranged for the final manuscript versions.

Moreover, following the suggestion from Referee #2, we will include an overview table with the results from the Wilson et al. study that are referred to in our manuscript. This enhances the readability of the manuscript because most of the Wilson et al. 2012 citations become obsolete. The details of these changes are outlined in our response to Referee #2.