Response to the Comment of Reviewer 1

We would like to thank Reviewer 1 for their comments and helpful suggestions. We reply to the individual points below.

General Comments

This manuscript describes the modification of the commercial instrument SPIN (SPectrometer for Ice Nuclei), which is a device for measuring ice nucleation activity of laboratory-prepared as well as natural ice-nucleating particles. In particular, by modification of the compressor system for cooling the measurement chamber of the instrument, the authors extended the temperature range of the SPIN device. The functionality in the entire (now extended) range of humidity and temperature is studied using model laboratory aerosols of ammonium sulfate and silver iodide. Overall, this is a valuable approach, although at the low temperature there seems to be an offset from the theoretical homogeneous ice nucleation curve. The authors speculate that this may be due to the fact that the aerosol particles do not reach equilibrium before freezing, see comment (6) below. I find this somewhat unsatisfactory and suggest that the authors spend more thought on this (and maybe, if possible, supply some additional sensitivity measurements). Moreover, because the paper's goal is allowing other scientists to make the same modifications to their SPIN instrument, I suggest a more detailed description/listing of the individual steps in order to become unambiguous.
Formally, the paper text, length, and figures are appropriate. However, I have a request regarding wording: apparently, the authors mix up vapor pressure and partial pressure and I request a correct and consistent usage of these terms, see comments (1) and (3) below.
In summary, the manuscript provides a useful approach and technical modification of an existing instruments, which I consider to be publishable in Atmospheric Measurement Techniques after the comments below have been considered in a revised version.

Scientific Comments

(1) *Page 2, Line 17; and caption to Fig.1: "Under steady state conditions a linear temperature and vapour pressure gradient establishes between the plates." I think the term vapor pressure is not used correctly and consistently. Vapor pressure is a property of a liquid or solid, and partial pressure is a property of a gas (mixture). The term vapor pressure is used here with its meaning of partial pressure. The IUPAC definition is: "For a mixture of gases the contribution by each constituent is called the partial pressure." In the caption of Fig.1 the term saturation pressure is used, which actually is the (saturation) vapor pressure.*

Following the reviewers recommendation, we replaced "vapour pressure" with "water vapour partial pressure" throughout the text when referring to the gas phase.

(2) *P.4, Figure 2: I would prefer that the figure directly indicated the modifications in the setup, either by colors or by shading etc. As it is now, the modifications are not evident to me. Given that these modifications are the essential novel part of this study, I also strongly recommend a more-detailed point-by-point listing of all modifications, so that any other SPIN user can follow and repeat it with their setup immediately. The latter may be provided in an appendix or supplement.*

To make the needed changes more evident, we added references to Fig. 2 and list the steps in the text as follows:
To reach lower temperatures, the SPIN cooling system has been modified by reconnecting the cold wall, cascade compressor system to deliver R116 refrigerant to both wall plates. The configuration of the modified setup is shown in Fig. 2. In practice, three modifications to the cooling system are needed:

1. At the upper part of the chamber, a junction is added to the high pressure liquid R116 line (lower stage, yellow line in Fig. 2) to connect it to the warm wall plate in parallel to the cold wall.
2. The refrigerant outlet lines of cold and warm wall, where the refrigerant exits the wall plates in the form of low pressure gas (lower stage, blue line in Fig. 2) are joint together to return to the cold 2 compressor.

3. Accounting for the increased volume of R116 needed to cool both wall plates, an additional expansion volume (14ℓ steel tank) is added to the return line (lower stage, blue line in Fig. 2), to give room to the gaseous refrigerant and prevent overpressure when the system is not running.

(3) *P.4, L.6/7: "decreasing absolute vapour pressure". What is an ABSOLUTE vapor pressure? I guess you mean total partial pressure, do you? See comment (1) above.*

As can be seen in Fig. 1, second row, the water vapour partial pressure at a certain relative humidity decreases with decreasing temperature, causing slower ice crystal growth. We changed the sentence accordingly.

(4) *P.5, Figure 3b and abstract and P.8, L.11: According to Fig.3b, the total experimental range to measure ice nucleation (indicated by the hatched area) is extended by 14 K at maximum when compared to the original SPIN range, but certainly less than the 20 K given in the abstract as well as in the conclusion (P.8, L.11). Please correct accordingly.*

The stated "more than 20K increase in measurement range" is based on experience and depends on ambient factors as well as operator's decisions. We consider it a good estimate, to give a SPIN owners an idea of what to expect if they decide to do the modification. Shown as hatched area in Fig. 3(a) and Fig. 1 below, in theory (neglecting heat transfer from the ambient), the temperature range achievable at the lamina position increases between 30K at $RH_{ice} = 100\%$ and 14K at RH_{ice}=190%. There are two practical factors to consider: First, operating SPIN in a laboratory or field station at around 293K ambient temperature limits the lowest achievable wall plate temperatures to 5-10K above the boiling point of the refrigerant due to imperfect insulation of the wall plates. Exemplary measurements conditions are shown in Fig. 1 below. Decreasing the cooling rate and ambient temperature can reduce this offset, but are often impractical. Secondly, not all refrigerant changes phase when the chamber is operated at a temperatures too close to the boiling point of the refrigerant. This causes incompressible liquid refrigerant to flow back to the compressor, causing what is known as "liquid slugging" that destroys the compressor. The "safe" temperature range of operating the SPIN chamber is therefore offset, both in the original and the modified configuration, by 5-10K from the maximum. Taking the two factors into account, shifts the calculated ranges (hatched in Fig. 1, below), 5-10K towards higher temperatures and larger ice crystals.
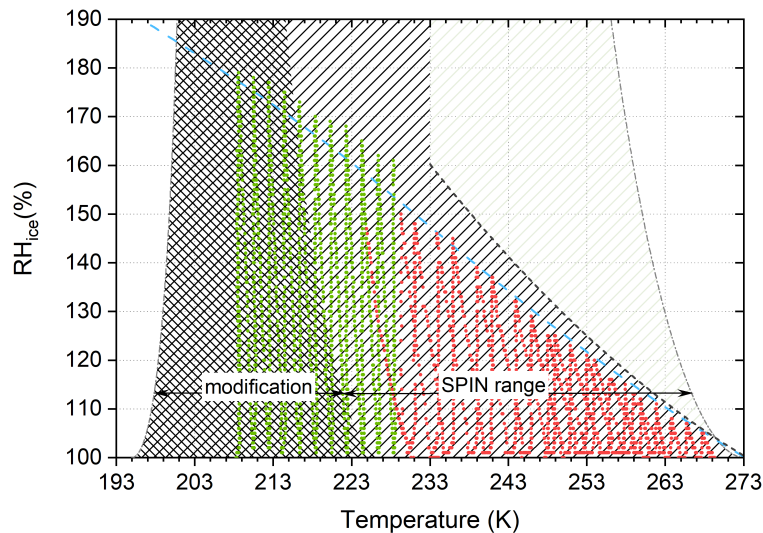

Figure 1. Comparison of the typical range of experimental conditions probable with the original SPIN (red points) to conditions after the modification (green points, lower 20 K of data shown in Fig. 4(a)).

2

The "limiting conditions" from ice crystal grow in Fig. 3(b) assume spherical ice growth (equations are now added in appendix A) and are therefore a lower limit for ice growth within the residence time in SPIN. Time dependent ice nucleation on the other hand can introduce a spread in growth time of the forming ice crystals and thereby their size distribution. It is an operator's decision which sizes are counted as ice crystals. From practical experience, the modification increased the measurement range by > 20K at low RH_{ice} and > 15K at high RH_{ice}. The temperature range to conduct RH-scans at constant T is increased by > 20K. The statement of a gain in temperature range of over 20K is therefore kept as is in the conclusion. In the abstract we now state the lowest temperature (208 K) at which we performed experiments, instead.

(5) *P.6, L.10: "and 1% AF curve derived using the parametrization of Koop et al. (2000)" How was the Koop-line calculated for 1% activation? The line will depend upon the time interval for which the aerosol particles are exposed to the T and RH conditions. What time interval was used for the calculations?*

The equations to calculate the 1% AF using the parametrisation of Koop et al. (2000) have been added in appendix B. We use the total residence time of exposure to the respective conditions (residence times slightly change with operating conditions and are calculated individually at each point in the RH-T space) to obtain the droplet size. The line of homogeneous freezing in Fig. 4(b) gives an estimation for the lowest RH_{ice} at each temperature where homogeneous freezing can cause 1% of droplets to freeze.

(6) *P.6, L.15: "aqueous aerosol do not reach equilibrium before freezing in SPIN" This is indeed a possibility, and maybe at the lowest temperatures diffusional limitations within the liquid droplets may also become relevant. However, this non-equilibrium state before freezing hampers the accuracy and applicability of SPIN, in particular in the extended low temperature regime. I would hope for more investigations as this is the main additional range of the SPIN instrument introduced in this work.*

At this point we would like to consider the discrepancy an interesting observation and only speculate on the reason. From the scatter of literature data included in Fig. 4(b) it can be seen that deciding whether the experimental data or parametrization (partly based on this data) are more robust is difficult. If droplets were not in equilibrium, they would be smaller and more concentrated, resulting in the 1% AF line to shift towards higher RH. This would explain the observed offset at the lower temperatures. Alternatively, Cziczo and Abbatt (1999) argued against substantial excursion from equilibrium even at short timescales and low temperatures, leaving the interpretation that the Koop et al. (2000) parametrization has a small, eventually solute dependent (Swanson, 2009) offset. We disagree that the applicability of SPIN to investigate homogeneous freezing is affected. The SPIN setup can provide measurements to investigate and refine the current understanding, be it diffusion of water into particles or ice nucleation.

Minor and Technical Comments

(7) *P.1, L.15: "At intermediate temperatures (236K < T < 273K) heterogeneous ice nucleation above water saturation. . ." I believe this should be AT OR BELOW water saturation, rather than ABOVE it. Or do you mean to ". . . above ICE saturation"?*

We changed the wording to "... close to water saturation..."

(8) *P.1, L.24: Here and at many other places in the text "Often, the dependency of ice nucleation on T, RH by a specific mechanism" I do not like the notation T, RH within a sentence. I guess you mean ". . . on T and RH by a . . ."? Please reword and use consistently throughout text.*

"T, RH" was replaced by "T and RH" when used within a sentence.

(9) *P.2, L.29-30: I assume these are the boiling points at standard or ambient pressure, correct? Please refine wording.*

We added that boiling points are given at atmospheric pressure (1 atm).

(10) *P.3, Figure caption 1: Reference Murphy and Kopp (2005) is misspelled.*

Corrected.

(11) *P.4, L.7: "4 m in 10 s residence time" Replace "in" by "within".*

Corrected.

(12) *P.6, L.3: "are reports of $(NH_4)_2SO_4$ forming ice heterogeneously at cirrus temperatures" This is only correct for non-deliquesced (i.e. effloresced) particles. I suggest to add the deliquescence line of $(NH_4)_2SO_4$ to Fig 4b.*

We added the temperature dependent DRH line to Fig. 4(b).

(13) *P.8, L.11: "We describe a mechanically easy modification" Maybe "simple" is better than "easy"?*

We replaced "easy" with "simple".

(14) *P.8, L.18: Replace "AgI" by "AgI particles"*

Corrected.

(15) *P.8, L.23: There is no link provided to the repository.*

Unfortunately our data repository is not operational yet. The data are available upon request from the author.

References

Cziczo, D. J. and Abbatt, J. P. D.: Deliquescence, efflorescence, and supercooling of ammonium sulfate aerosols at low temperature: Implications for cirrus cloud formation and aerosol phase in the atmosphere, J. Geophys. Res., pp. 13 781 – 13 790, doi:10.1029/1999JD900112, 1999.

Koop, T., Luo, B. P., Tsias, A., and Peter, T.: Water activity as the determinant for homogeneous ice nucleation in aqueous solutions, Nature, 406, 611–614, 2000.

Swanson, B. D.: How Well Does Water Activity Determine Homogeneous Ice Nucleation Temperature in Aqueous Sulfuric Acid and Ammonium Sulfate Droplets?, J. Atmos. Sci., pp. 741 – 754, doi:https://doi.org/10.1175/2008JAS2542.1, 2009.