We thank the anonymous Reviewer for his/her and encouraging comments. We found that the comments helpful and by answering them we were able to improve the quality of our manuscript. Here, we give detailed answers to the comments. The Reviewer comments are *in italics* and our reply in blue standard text**.**

Järvinen et al. introduce a new method of probing the viscosity of SOA as a function of RH and temperature, and they apply this method to determine information related to the viscosity of SOA generated from the oxidation of alpha-pinene. The experiments are novel and extremely important, as they provide laboratory data related to the viscosity of SOA at cold temperatures. Currently this type of information is very limited, yet this type of information is needed to understanding ice nucleation by SOA in the atmosphere. Although the experiments are excellent and the information provided is crucial, I do have comments on the data interpretation that should be addressed before publication. Once these issues are addressed, I highly support publication in ACP.

We thank the Reviewer for this very positive general comment. For the revised version, we have improved the data interpretation based on the Reviewer's suggestions. We are more careful with the term "viscosity-transition" and have renamed it as "viscosity/shape-transition". We now clearly state already in the Introduction that the method does not directly measure the viscosity of the particles, but their shape. The change in the particle shape can be related to changes in the viscosity. We estimated the viscosity of the particles during their transition and have derived a viscosity of 10^7 Pa s. This value was compared to literature to estimate the physical meaning of our observations. In the revised manuscript we, therefore, omit assuming that the particles were in an aqueous state after the transition. We also do not discuss the results in the context of glass-transition. We hope that the revisions have brought the clarity to the manuscript that the Reviewer was looking for.

Comments on interpretation of experimental results:

1) Page 28596, line 20. Consider replacing "for the phase transition" with "for the aspherical to spherical transition" since the authors have not proven that they have measured a phase transition. For example they haven't proven that they are measuring the relative humidity for the glass to semisolid phase transition (which occurs around 10ˆ12 Pa s) or the semisolid to liquid transition (which occurs at 100 Pa s).

We agree with the Reviewers suggestion and changed the phase transition to "the transition from aspherical to spherical".

2) Conclusions, line 4. Would "aspherical to spherical transition" be more appropriate than "viscosity transition"? Viscosity is expected to change (i.e. transition) with RH in a continuous manner for most RH values. For example at warm temperatures and high RH values, as the RH is increased the viscosity is expected to change (i.e. transition) yet the experiments described here are not sensitive to these changes.

The Reviewer is correct, when pointing out that we do not directly measure the viscosity transition, but shape change. This shape change can be related to a viscosity transition, as discussed above. To be correct, we modified the second sentence in the conclusions following: "*Here, we estimated the viscosity of α-pinene SOA at different atmospherically-relevant temperatures and relative humidities using a new optical and non-invasive in-situ method*". In the following sentence, we explain what the method measures directly: "*We used near-backscattering depolarisation to measure the asphericity of the SOA particles and their shape change to spherical that took place at viscosity of around 10^7 Pa s.*"

3) Page 28587 line 1. Consider removing "during a phase transition" since the authors are not necessarily probing a phase transition. Rather they could be just sensitive to when the viscosity falls below a certain value.

We removed "during a phase transition" and just state "during RH increase".

4) Abstract. The authors claim that they are measuring the transition from the amorphous viscous state to a liquid state. However, the experimental observations (non- spherical to spherical transition) may occur at viscosities much higher than a liquid state, which has been defined in the atmospheric literature as a material with a viscosities less than 100 Pa s.

We agree with the reviewer. The added viscosity investigations showed that the alpha-pinene SOA particles had a relatively high viscosity of 10^7 Pa s during their shape-transition. Therefore, we cannot assume that after this transition the particles would have been liquid, i.e. have viscosity less than 100 Pa s. We modified this sentence in the abstract following: "*we deployed a new in-situ optical method to detect the viscous state of α-pinene SOA particles and measured their transition from the amorphous highly viscous state to states of lower viscosity*".

5) Page 28595, line 9, the authors refer to the full deliquescence relative humidity. Please define what they mean by full deliquescence relative humidity. Also, how do they know that the so-called viscosity transition occurs near the full deliquescence relative humidity?

We removed this paragraph discussing the full deliquesce RH. Instead we estimated the viscosity of the alpha-pinene SOA particles and discussed our findings in the view of previous studies.

6) Page 28595, line 11-13. Here the authors are discussing the timescale for water diffusion within the particle bulk. The authors should estimate what this time scale is based on recent published diffusion coefficients for water in SOA and include these estimated time scales in the manuscript. Based on these values is water diffusion the rate limiting step in their experiments? See [Price et al., Chemical Science, 2015; Lienhard et al., ACP, 2015].

This is a very interesting question from the Reviewer. However, we see that this question is out of the scope of this paper, as we concentrate more on the optical properties of the alpha-pinene SOA particles than their water uptake properties. Following the study of Price et al. (2015) equilibrium is reached in seconds for 240 K, which would suggest that the water diffusion is not limiting our experiments.

7) In previous publications the time scale for aspherical particles to transition to spherical particles has been used to estimate viscosities [Zhang, Sanchez et al. ACPD, 2015; Pajunoja et al. Aerosol Sci. Tech. 2014]. Have the authors considered estimating viscosities from their observables? I think the current paper would be clearer if the authors estimated viscosities from their results.

We used the method in Pajunoja et al. (2014) to estimate the viscosity of the SOA particles base on the transition time to spherical. This estimation is showed and discussed in the chapter 4.5. We agree that adding this estimation, we can bring clarity to the revised version and better discuss the physical meaning of the shape transition.

Experimental issues:
1) Page 28586. Line 16-17. The authors state that two thermocouples near the wall were excluded

due to wall temperature effects. I assume this means that the temperature close to the wall was colder? The experiments involved adding a flow of humidified gas and mixing the flow in the chamber with a fan. Is it possible that the temperature and relative humidity was different close to the walls, and the aspherical to spherical transition occurred close to the walls, not in the middle of the chamber? If this was the case, the temperature and RH reported may be different than the temperature and RH at which the aspherical to spherical transition occurred?

Excluding the two thermocouples closest to the wall is necessary in cloud expansion experiments, when we cool the chamber volume but the chamber walls stay at a constant temperature. This creates a temperature gradient that could bias the measurements. However, in the experiments presented in this paper such expansion experiments were not conducted, so excluding the two thermocouples next to the wall is not necessary. We checked the differences between the thermocouples and came into a conclusion that they agree quite well with each other. The maximum temperature difference they showed was 0.2 K. This difference is the same order as the measurement uncertainty of the thermocouples. Therefore, we don't think that significant temperature gradients existed during the experiments that could have led to biases. We removed the phrase " *excluded due to wall temperature effects"* so that the reader is not mislead.

Comparison with Koop et al. 2011:

1) Page 28595, Line 20-25: I do not understand the argument that the measured "viscosity transition" can be compared to the relative humidity of the glass transition (RHg) in atmospheric context. The glass transition occurs at a viscosity of roughly 10^12 Pa s. To argue that the "viscosity transition is comparable to the glass transition, I think one would need to argue that both transitions occur at roughly the same viscosity? Can the authors estimate that viscosity at which their aspherical to spherical phase transition occurred?

We estimated the viscosity during the transition, and indeed the shape transition occurs at lower viscosities. Therefore, we removed this statement.

2) Page 28595, line 27-29. "Our results suggest the relationship between the transition C11151

RH and the temperature is more linear than predicted by Koop et al. 2011. This may be especially relevant at high RH, where we can expect that the transition might take place at a much higher temperature than predicted by Koop". I suggest the authors should modify this text since all the experimental results shown here are within the estimates presented by Koop et al. 2011 (green shaded region in Figure 11).

We modified the sentence the following: "*Moreover, our results suggest the relationship between the transition RH and the temperature is more linear than predicted by Koop et al. (2011), although measurements at colder temperatures are missing. To understand the viscosity transition of alpha-pinene SOA particles at high RH in the cirrus regime, measurements at colder temperatures are recommended.*". We also removed the sentence: "*where other studies might suggest that it relax*" from atmospheric implications.

Minor points:

2) Page 28581. Line 4. Should "isotropic" be deleted?

We agree that the word "isotropic" is not necessary here and, thus, we removed it.

3) Page 28587, line 15. "the SOA particles were generated and grown in the chamber by continuous oxidation of alpha-pinene with ozone". Should this be ". . . by continuous oxidation of

alpha-pinene with ozone and OH radicals"?

We did not limit the formation of OH radicals, so the Reviewer's suggestion is correct. We modified the sentence accordingly.

4) Figure 5. In the figure caption it is stated that the linear depolarisation ratio stayed below the detection threshold during the entire experiment. However, at times between 6:00 and 7:00 it looks like the linear depolarisation reaches as high as 0.1. This seems to be well above the LOD. Please discuss.

The signal between 6:00 and 7:00 is purely noise as the particles were too small to scatter enough light to the detectors. Only after 7:00 we had enough signal to reliably determine the depolarisation ratio. However, the Reviewer is correct, when pointing out that we do not mention the noise in the text. In the revised version we added a sentence to the figure caption: "*At the beginning of the growth (6:00-7:00) the SOA particles were too small to efficiently scatter light and the signal shown in panel (c) at that time is mostly noise. After the particles were larger than 100 nm, the linear depolarisation ratio stayed below the detection threshold.*"

5) Was figure 8 discussed in the main text? If not, should it be removed from the manuscript?

The figure 8 was discussed in the third paragraph in the chapter 4.3. The purpose for this figure was to show that circular depolarisation ratio is always higher than linear, i.e. it could be potentially more sensitive to particle shape.