We thank the anonymous Reviewer for his/her encouraging comments. The Reviewer's suggestions were helpful and helped us to improve the quality of our manuscript. Below we give detailed answers to the general and technical comments. The Reviewer comments are *in italics* **and our reply** in blue standard text**.**

This manuscript reports the viscosity transition of α-pinene SOA generated during the CLOUD experiments at CERN at different temperatures using an optical method with a depolarization instrument. The method based on the different depolarizing properties of spherical and non-spherical shape of particles. The manuscript provides information of transition, which was defined as viscosity transition, from non-spherical to spherical shape when increasing RH (particles taking up water vapor). This study demonstrated an optical method that could be applied for the chamber studies on the phase transition of particles. The experimental methods are valid and the scientific approach and discussion are sound. The paper is well written and organized. I recommend it for publication with minor revision. Please see the following comments that the authors may want to consider in the revision.

General comment:

1, Previous studies as mentioned by the authors often have a definition based on physical properties, such as viscosity, for different phases, for example, liquid, viscosity < 100 Pa s. The definition of viscosity transition here in this manuscript is not very clear, the transition from non-spherical shape to spherical means transition from solid/semi- solid to liquid or solid to semi-solid? Spherical shape does not mean the particle is in liquid state. It is suggested to provide the clear definition of the viscosity-transition. The definition of viscosity-transition-RH is placed in the later part of the manuscript, P28594 line 16. The way it is defined is not really quantitatively; can "decrease significantly" and "reached a constant level" be quantitatively defined? I suggested make these definitions at the early part of the paper and indicate that these are instrumental defined, unless physical means/values are provided associated with this viscosity-transition for definition.

We agree with the Reviewer that in our case the change in the depolarisation properties does not necessary imply that the particles are liquid after the transition. Also, we do not directly measure the viscosity of the particles, but their shape. Thus, our viscosity-transitions as it was described cannot be related to any specific change in the viscosity. We can only estimate the viscosity of the particles based on previous studies (as suggested by the Reviewer) or based on the relaxation timescales (please see the answers below). In the revised manuscript we label the viscosity transition as viscosity/shape-transition, similar to Kidd et al. (2014). We hope that this illustrates the fact that the assumed change in the viscosity is based on the direct observation of the shape change. We define this term already in the Introduction and highlight that this transition is instrumental defined: "*We define an instrumental-based viscosity/shape-transition RH that describes the RH value, where the particles were observed to be spherical or isotropic according to our method.*" (p28580, line 13, old manuscript version).

Furthermore, to clearly state that our method can only detect the viscous state, but not measure the viscosity of the particles, several modification were made. Also any statements that the particle would be liquid after the measured viscosity/shape-transition were removed. These modifications include:

1. In the abstract, we modified lines 6-8: "*we deployed a new in-situ optical method to detect the viscous state of alpha-pinene SOA particles and measured their transition from the amorphous highly viscous state to states of lower viscosity*" and removed the word liquid in line 10.

2. Chapter 4.5: we added "or low-viscous" next to liquid at page 28594, line 25 (old manuscript version).
3. We added at the end of the first paragraph in conclusions that the method presented here does not provide a direct measure of viscosity: "*although the viscosity of the SOA particles cannot be directly determined from our measurements.*"

2, P28595, Line 8-23, as mentioned above the definition of viscosity transition is not clear, RHg indicates the transition from solid to semisolid, caution is needed when compare the viscosity-transition RH with RHg as authors also mentioned that the viscosity- transition RH is different from RHg. If I understand it correctly, most likely the viscosity- transition RH will be higher than RHg (how much higher depending on the definition of viscosity-transition RH). Line 18-19, it is lacking of logic here for reasoning the comparison with RHg, the transition timescale and atmospheric processes are the dynamic factors, they may affect the viscosity-transition RH measured in the chamber, but not the reasons for the comparison. Why the viscosity-transition RH is more relevant for atmospheric processes? P 28597, line 11-12, why it is directly relevant for GLASSY Transition of SOA, isn't this conflict with the statement in P28595, line 8-11.

We agree with the Reviewer and removed this paragraph. It is more suitable to estimate the viscosity in our case and compare our results to previous studies as suggested by the Reviewer. A new paragraph was added (please see the Author's reply below) that better discusses the physical meaning of our viscosity/shape-transition.

3, It is recommended to calculate or estimation the viscosity based on the transition time, particle size changes with additional assumptions. Based on these data it may be possible to estimate the viscosity of the particles or at least the changes of viscosity during the transition. There are a few more studies that maybe useful. Kidd et al (2014) provided viscosity information for a-pinene SOA generated at different RH, Wang et al (2015) estimated the viscosity of a-pinene SOA as a function of temperature and RH, Song et al (2015) summarized a nice set of the viscosity data for a-pinene SOA from previous studies for comparison.

We estimated the viscosity during the transition based on the transition time and the size of the primary particle using the procedure described in Pajunoja et al., (2014). This leads to an estimation of viscosity of 10^7. We compared this viscosity value to observations listed by the Reviewer. However, the problem with comparing or relating to viscosity values from previous studies is that the studies are usually conducted at room temperatures. Therefore, the RH dependency of viscosity may not be trivially linked to our observations.

Consequently, a new paragraph was added to the chapter 4.5. "*As explained above, our method does not directly measure the change in the viscosity. However, we can estimate the viscosity of the α-pinene SOA particles based on the relaxation time scale and the particle size using the method described in Pajunoja et al. (2014). By assuming a primary particle size of 400 nm in an SOA aggregate, a relaxation time of 10 minutes and a surface tension of 35 mN/m (Pajunoja et al., 2014), we get an estimated viscosity of 107 Pas. This value describes the viscosity during the viscosity/shape-transition period. Previous studies (e.g. Renbaum-Wolff et al., 2013; Bateman et al., 2014; Kidd et al. (2014); Pajunoja et al., 2014) have studied the viscosity of α-pinene SOA particles at room temperatures. The studies have found that α-pinene SOA particles are generally in aqueous state already at RH> 70% (Song et al. (2014)). At colder temperatures we can expect the transition from viscous to aqueous state is shifted to higher RH (Wang et al. (2015)). As the measured viscosity/shape-transitions generally took place at RH< 70% the comparison with literature would suggest that albeit our SOA particles were spherical, they probably were not in* *aqueous state after the transition. Therefore, our measured viscosity/shape-transition would describe the change in the viscosity within a semi-solid material."*

Technical comments:

1, P28577, line 6, please provide full description of "CLOUD" and "CERN"

We added the full descriptions of these acronyms: "*In the CLOUD (Cosmics Leaving Outdoor Droplets) experiment at CERN (The European Organisation for Nuclear Research), we deployed...*".

2, P28578, line 25-27, please provide references for this statement

We added the reference Zhang et al., 2015.

3, P28582, line 10-12, please justify the assumption or provide references

There are several studies that have tried to retrieve the real refractive index of SOA particles (e.g. Schnaiter et al., 2005; Redmond and Thompson, 2011; Kim et al., 2010). However, most of these studies are performed at room temperatures and, where the SOA particles were likely in aqueous state. The reported refractive indexes for alpha-pinene SOA vary between 1.4-1.6. Furthermore, Kim et al. reported hat: "aerosol mass concentration, oxidation chemistry, temperature, and aerosol aging may all influence the refractive index". However, to our knowledge studies of the refractive index of viscous SOA particles have not yet been conducted. Therefore, we have to assume our refractive index and the best way to do this is to assume the refractive index of non-viscous alpha-pinene SOA.

The purpose for the modeling is to evaluate the magnitude of the depolarisation ratio and to show that small deviations from the spherical shape can induce measurable depolarisation. As the results are more qualitative than quantitative we don't see that the uncertainty in the refractive index will effect the main conclusions drawn from the modeling.

We removed the assumption that the Reviewer was referring to and modified the sentence in the lines 10-12 as follows: "*The refractive index of alpha-pinene SOA at near room temperatures has been shown to vary between 1.4 and 1.6 (Schnaiter et al., (2005); Kim et al., (2010); Redmond and Thompson, (2011)). As we do not have the information on the refractive index of viscous alpha-pinene SOA particles, we assumed in the modeling study a refractive index of 1.5+0i after Schnaiter et al., (2005).*"

4, P28585-28586, provide the full description of SMPS, CPC, ApicT etc.

We added the full description for Scattering Mobility Particle Sizer (SMPS), Condensation Particle Counter (CPC) and APicT (AIDA PCI in-cloud TDL).

5, P28586, What is the pressure in chamber during the experiments?

The chamber was always 5 mbar overpressure compared to atmospheric pressure. We added the following sentence to the second paragraph in section 3.2.2: "*The chamber was kept at 5 mbar overpressure to avoid under-pressure in the sampling instruments.*"

6, P28593, line 19-23, what do you mean "particle hardness"?

By hardness we mean the ability to resist the deliquescence (higher deliquescence point). To avoid confusion, we removed the word "hardness" and modified the lines 19-23 as follows: "*The O/C ratio affects the particle deliquescence point; in the case of alpha-pinene SOA particles, we can expect the deliquescence point to be at higher temperatures with increasing O/C ratio (Berkemeier et al., 2014).*"

7, P28595, line 1, as mentioned above, the viscosity-transition RH is not quantitatively defined, is it possible that the observed wider transition RH is due to the arbitrally defined gray area as shown in Figure 4? If consider the uncertainty of RH as described in P28586 (7-13%), there is no significantly wider RH ranges as shown in Table 1. There may be one only considerable larger range is for -38 degree C (› 10%) as compared to 5-7% for higher temp.

The Reviewer is correct, when pointing out that we did not take into account the uncertainty range of the RH measurements. The measured partial water vapor pressure is always below 100 Pa, so we have an uncertainty of 13% at each temperature. When taking this into account, we cannot claim that the larger RH range in the measurements would not be due to measurement uncertainty. Therefore, we removed the second conclusion and the discussion related to it.

However, we disagree that the viscosity-transition is not quantitatively defined. The grey areas were not selected arbitrarily, but are based on clear changes in the depolarisation signal.

8, P28595, line 3, in Table 1, the viscosity-transition RH values are shown in ranges. it is not convincing that viscosity-transition RH has linear dependency of temp. Please provide fitting data or supporting information for this statement.

The near linear temperature dependency of the viscosity-transition is based on the measured transition RH, i.e. the RH value, where the particles appeared spherical according to our measurements (the highest RH in the range). A linear fit could be performed to the data points with a slope of -6.3 and an offset of 284. However, as we measure the viscosity-transition that is somewhat different than the glass-transition in Koop et al. (2011), we do not wish to provide a linear fit to the data, as the Reviewer pointed out. We modified the sentence on P28595, line 3, so that it is not generalized, and highlights that the linear dependency concerns only the measurements: "*The measured viscosity/shape-transition RH has near linear dependency of the temperature*".

9, P28596, line 5-7, SOA from difference precursors may behave differently. It is suggested to rephrase this statement and limited to a-pinene SOA as the manuscript only showed a-pinene SOA data.

We agree with the Reviewer that only alpha-pinene SOA was investigated and the method is not guaranteed to work with other SOA precursors. Therefore we modified the last paragraph in chapter 4.5. We added the word "alpha-pinene" before SOA in the first and in the second sentence and we removed the last sentences in the paragraph and replaced them. The revised paragraph states the following: "*Our method to detect the viscosity/shape-transition of alpha-pinene SOA is likely limited to laboratory experiments with high number concentrations promoting the formation of non-spherical SOA particle aggregates. Though our limited data do not support the hypothesis that SOA particles in the lower troposphere might be non-spherical, we have demonstrated the first non-invasive method to study the viscosity of SOA particles formed by ozonolysis of gaseous alpha-pinene. Moreover, the transition timescales that we observed, 10s of minutes, are relevant to those that we can expect in the atmosphere. Therefore, the viscosity/shape-transition RH that we report is relevant for the atmospheric processes. The present study focused on understanding the humidity effect on the viscosity transition temperature of a~single SOA component. The method could also be used in future studies to study SOA shape transitions of other SOA precursors.*"

10, P28597, line 22, "viscosity transition temperature RH at temperatures.." typo?

We removed the additional word "temperature". The corrected sentence is now: *"Generally, our results improve the understanding of the viscosity-transition RH at temperatures above the homogeneous freezing point."*

11, Fig.4, unit of particle concentration is missing.

We thank the Reviewer for noticing the missing units. We added in Figs. 4 and 5 "dN dlogD_p [cm-3]" above the color bar.

12, Fig.8 Caption, two "is" in last sentence.

We removed the additional "is".

Kidd, C., Perraud, V., Wingen, L. M., and Finlayson-Pitts, B. J.: Integrating phase and composition of secondary organic aerosol from the ozonolysis of a-pinene, P. Natl. Acad. Sci. USA, 111, 7552–7557, doi:10.1073/pnas.1322558111, 2014.

Wang, B. B.; O'Brien, R. E.; Kelly, S. T.; Shilling, J. E.; Moffet, R. C.; Gilles, M. K.; Laskin, A. Reactivity of liquid and semisolid secondary organic carbon with chloride and nitrate in atmospheric aerosols. J. Phys. Chem. A 2015, 119, 4498−4508.

Song, M., Liu, P. F., Hanna, S. J., Li, Y. J., Martin, S. T., and Bertram, A. K.: Rel- ative humidity-dependent viscosities of isoprene-derived secondary organic material and atmospheric implications for isoprene-dominant forests, Atmos. Chem. Phys., 15, 5145-5159, doi:10.5194/acp-15-5145-2015, 2015.