We would like to thank the Referees for their insightful and helpful comments on the manuscript. These comments have helped us to improve the manuscript. We have conducted additional work requested by the Referees and we hereafter answer all the comments.

Anonymous Referee #1

Since they refer to the same topic, comments 1, 4, 5 and 6 are discussed together.

1. Overall, this manuscript has the potential for eventual publication in ACP. However, I suggest a significant rewrite before reconsideration. In my opinion, a more convincing case needs to be made as to how combining simultaneous hygroscopic and optical measurements helps characterization of SOA properties relative to separate measurements, particularly given the authors' claim that "it is critical to simultaneously determine the hygroscopic behaviour of its size distribution and optical properties as well as their dependence on the chemical composition" (P10547, L16-17).

4. There is very little discussion directly relating the calculated scattering growth factor values, f(RH), to hygroscopic growth factor values at the same RH. In my opinion this should be a significant component of the manuscript to support the authors' claim that "it is critical to simultaneously determine the hygroscopic behaviour of its size distribution and optical properties" (P10547, L15-L16). In the current manuscript the size growth factors (Figures 5-6) and scattering growth factors (Figure 7) are presented and discussed separately for the most part.

5. One application/advantage of combined f(RH) and GF measurements seems to be the ability to assess possible residence time limitations (or lack thereof) in the HTDMA. On P10553 near the end of Section 2.3, the authors set up this discussion with the statement: "the two approaches of hygroscopicity measurements could lead to different results, which carry information on water transfer dynamics, possible particles reorganisation or phase transfer equilibrium establishment." However, this is not evident from the data presented in the current manuscript.

6. The authors could use measured f(RH), size distribution, real refractive indices of SOA and of water to calculate a growth factor from the optical measurements. This calculated growth factor could be plotted along with the HTDMA-derived GF as a function of RH for "fresh" and "aged" SOA. In my opinion this is a logical extension of the data presented in Figures 5-7 and I think could more evidently show if there are residence time limitations in the HTDMA-derived GF's. Does the "scattering growth factor" increase with aging? This was not clear in the current manuscript. If the authors are aware of other ways to combine the scattering and size GF data, those should be added to the discussion as well.

Response: - Relating the calculated scattering growth factor values, f(RH), to hygroscopic growth factor values, GF, at the same RH is a very interesting suggestion. It was also raised by Referee #2. We used Mie scattering calculations for homogeneous spheres to determine GF from f(RH). σ_{scat} *was calculated for different GF at specific RH. The optimal GF as a function of RH was determined so that the differences between measured* σ_{scat} *and those obtained using Mie calculations were minimized. Particles were assumed to be homogeneous spheres of uniform CRI. The CRI calculations were based on volume weighted refractive indices of SOA and water. Uncertainties on the theoretical GF were estimated from the standard deviation of the measured f(RH), the uncertainties on the f(RH) measurements and the uncertainties on the*

RH measurements. Figure 6 (below) shows the comparison between measured and predicted GF values for SOA at two different reaction times: for "fresh" SOA (after 1 hour of reaction), and for "aged" SOA (after 14 hours of reaction). For both reaction times, the model approach agrees well with the measurements above 30% RH, indicating no kinetic limitations for water uptake. For "aged" SOA (after 14 hours of reaction), the observed underestimation of the model below 30% RH might be due to the phase transition from a predominantly glassy state to a predominantly liquid state, as discussed in section 4.2.

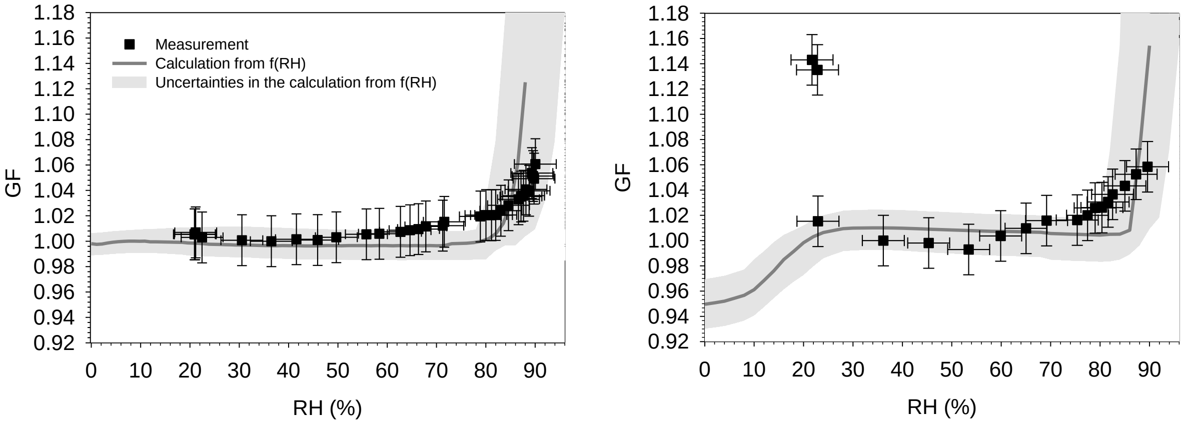


Figure 6: Humidograms showing measured (black symbols) and predicted GF (grey line) as a function of RH of SOA (a) after 1 hour of reaction and (b) after 14 hours of reaction. The grey area represent the uncertainties in the calculation of GF from f(RH).

This new data treatment has been added to section 3.3 as follows: "An important concern in measuring the hygroscopic properties of the particles is to allow sufficient time for particle-water vapor equilibrium. Various studies have discussed the possibility that insufficient time for humidification could result in an underestimation of the particles' water content (Chan and Chan, 2005; Saxena et al., 1995; Duplissy et al., 2009; Denjean et al., 2014). In this study, the residence time for SOA particles in the wet air stream was significantly longer when particles were humidified in-situ in the chamber. It took ~1 hour in the chamber, instead of 15 seconds in the H-TDMA. These two approaches can thus be complementary to carry information on water transfer dynamics of α-pinene-O_3 SOA. We used Mie scattering calculations for homogeneous spheres to determine GF from f(RH). σ_{scat} was calculated for different GF at specific RH. The optimal GF as a function of RH was determined so that the difference between measured σ_{scat} and those obtained using Mie calculations were minimized. Particles were assumed to be homogeneous spheres of uniform CRI. The CRI calculations were based on volume weighted refractive indices of SOA and water. Uncertainties on the theoretical GF were estimated from the standard deviation of the measured f(RH), the uncertainties on the f(RH) measurements and the uncertainties on the RH measurements. Figure 5 shows the comparison between measured and predicted GF values for SOA at two different reaction times: for "fresh" SOA (after 1 hour of reaction), and for "aged" SOA (after 14 hours of reaction). For both reaction times, the model approach agrees well with the measurements above 30% RH. It indicates no kinetic limitations of α-pinene-O_3 SOA for water uptake. For "aged" SOA (after 14 hours of reaction), the underestimation of the model below 30 % RH (Figure 6) might be due to a change in the physical state of SOA, as discussed in detail in section 4.2."

- Regarding the comment of the Referee on the sentence written P10547 L16-17, we agree that the measurements of GF and f(RH) must not be necessarily performed at the same time and we decided to delete the word "simultaneous" in the sentence.

- The Referee also underlines that the evolution of scattering growth factor with aging has not been discussed in the paper. However, this point is already discussed in the paper P14 L19-22: "All the experiments exhibited the same trend, but different values are observed at 90% RH. This can be explained by the different size distribution from one experiment to another. In particular, the proportion of particles larger than 100 nm is different resulting in a changing capacity in absorbing water (Biskos et al., 2006) and hence varying the observed f(RH)". We think that this comment may be due to Figure 6 which presents f(RH) for "fresh" and "aged" SOA separately. In order to facilitate the comparison of f(RH), Figure 6 has been modified to present the data at different reaction times. In addition, due to the Referee's comment that "in the current manuscript the size growth factors (Figures 5-6) and scattering growth factors (Figure 7) are presented and discussed separately for the most part", we decided to merge together Figures 6 and 7 into Figure 5 (below).

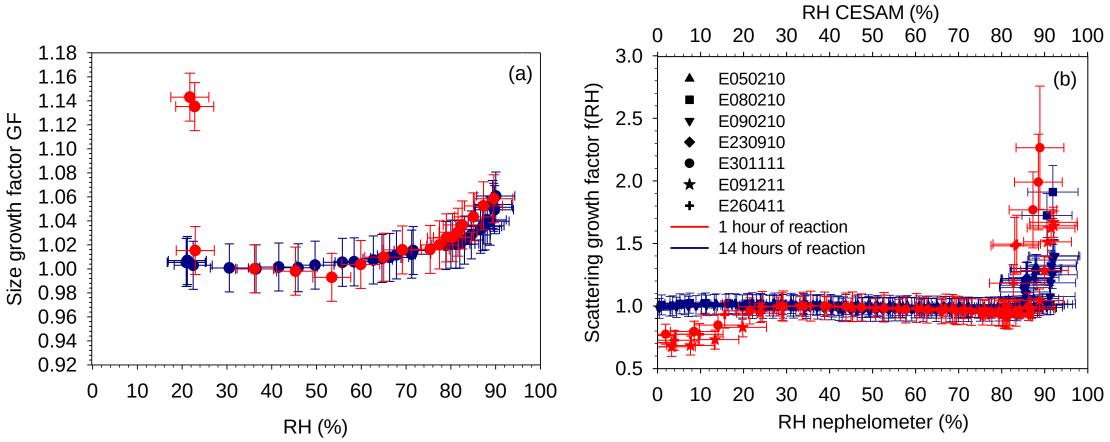


Figure 5. Humidograms of (a) size growth factor measured by the H-TDMA and (b) scattering growth factor measured by the nephelometer as a function of RH within the nephelometer (bottom axis) and the RH within the chamber (upper axis), for "fresh" SOA (after 1 hour of reaction) in red and for "aged" SOA (after 14 hours of reaction) in blue. For the calculations of f(RH) and GF, we used $\sigma_{scat}(RH_{dry})$ and $D_{p,m}(RH_{dry})$ at 40 %.

2. Figures and quantitative analyses focusing on how optical measurements enhance the hygroscopicity measurements (and vice versa) could significantly improve the manuscript. Because many of the results obtained by the authors have already been measured previously – in some cases by multiple researchers – at present it isn't clear to me what new information is gained by combining the measurement techniques that were used, aside from perhaps the XPS measurements of SOA surface chemical composition.

Response: As mentioned by the Referee, multiple studies have already investigated the hygroscopic and optical properties of α-pinene-O_3 *SOA. However, despite considerable progress in recent years, the studies on hygroscopic properties show disagreements: Saathoff et al. (2003) and Cocker et al. (2001) reported an increase of GF(90% RH) within 6 hours of reaction, while Warren et al. (2009) and Qi et al. (2010) reported a constant GF(90% RH). This has been stated in the introduction of the paper. It is also important to note that none of the previous studies have provided the O:C of α-pinene-*O_3 *SOA together with the GF.*

Understanding the evolution of hygroscopic properties of SOA during formation and its link with chemical composition was one important part motivation of our study.

Beyond the disagreements cited above, SOA was formed in previous studies under different initial conditions from ours. The authors used an excess of ozone to generate SOA within a few minutes and then investigated the effect of oxidation aging of SOA on hygroscopicity. The novelty of our study is to perform measurements of SOA on time scales up to 20 hours for the initial ratio of $[VOC]/[O_3]\sim1$*. This was performed in an effort to probe the influence of the evolution of the SOA chemical composition during formation (by gas-phase partionning, formation of oligomers,…) on the optical and hygroscopic properties of SOA.*

In addition, to our knowledge, only one measurement has been made on the evolution of the CRI of α-pinene-O_3 *SOA with time (Kim et al., 2010), but no correlation with the chemical composition was given. Here, we bring new data set on the optical and chemical properties of SOA on time scale up to 20 hours, which might be extremely useful for a better understanding of the evolution of the SOA during its formation.*

We agree with the referee that these information are not sufficiently highlighted in the manuscript. In an effort to clarify this point, the time scale of reaction and the initial concentration of α-pinene and ozone have been added in Tables 2 and 3 (also suggested by Referee #2).

3. I think that some of the discussion about SOA yields and functional group composition detracts from the main focus of the paper. Shortening or removing some of this discussion, along with a more comprehensive discussion relating optical and hygroscopicity measurements, should further improve the manuscript.

Response: Even if the SOA yields are not the main focus of the paper, we think that it is an important parameter since it gives information on the variability of the experimental conditions. It is particularly important because parameters from different experiments are compared together in the paper. We decided to keep the yield values in Table 1, but to move the literature comparison in the Supplementary Material in order to shorten this part.

Concerning the discussion of the functional groups, section 3.4.2. brings some issues on the decrease of the O:C in the SOA bulk. By looking at the functional groups, we concluded that the decrease of O:C could be due to an increasing partitioning of less oxidized semi-volatile compounds as the aerosol grew up. We think that this part is important to interpret the trend of the O:C and thus the evolution CRI with time, but we agree with the Referee that it could be shortened. We thus decided to move Figure 9 in the Supplementary Material and we combined a shortened version of section 3.4.2 with section 3.4.1.

7. There are many typos and grammatical errors that should be addressed. I also found two incorrect citations. Additional proofreading is required throughout the manuscript.

Response: In order to improve the English content of the paper, the manuscript has been read by two native English speakers. In addition, many corrections on typos and grammar suggested *by Referee #2 have been made. We hope that the quality of the paper has been significantly improved after these revisions.*

8. P10548, L21-L22: There are a few more recent papers that should be cited here:
Lambe, A. T.; Onasch, T. B.; Massoli, P.; Croasdale, D. R.; Wright, J. P.; Ahern, A. T.; Williams, L. R.; Worsnop, D. R.; Brune, W. H.; Davidovits, P. Laboratory Studies of the Chemical Composition and Cloud Condensation Nuclei (CCN) Activity of Secondary Organic Aerosol (SOA) and Oxidized Primary Organic Aerosol (OPOA). Atmos. Chem. Phys., 11, 8913−8928, 2011.
Wong, J. P. S.; Lee, A. K. Y.; Slowik, J. G.; Cziczo, D. J.; Leaitch, W. R.; Macdonald, A., and Abbatt, J. P. D., Oxidation of ambient biogenic secondary organic aerosol by hydroxyl radicals: Effects on cloud condensation nuclei activity. GEOPHYSICAL RESEARCH LETTERS, VOL. 38, L22805, doi:10.1029/2011GL049351, 2011.
Mei, F.; Setyan, A.; Zhang, Q.; and Wang, J.. CCN activity of organic aerosols observed downwind of urban emissions during CARES. Atmos. Chem. Phys., 13, 12155–12169, 2013.
Rickards, A. M. J.; Miles, R. E. H.; Davies, J. F.; Marshall, F. H.; and Reid, J. P. Measurements of the Sensitivity of Aerosol Hygroscopicity and the κ Parameter to the O/C Ratio. J. Phys. Chem. A, 117, 14120−14131, 2013.

Response: We thank the Referee for suggesting these papers. They are now added to the revised manuscript.

9. P10550-10551: It is not clear to me why it is necessary to go into this level of detail about aethalometer operation when the data are barely used except to confirm that the SOA doesn't absorb?

Reponse: We don't agree with the Referee on this point. Only a rigorous correction of the aethalometer data allowed us to conclude that SOA does not absorb light in the wavelengths 350-950 nm. This can clearly be seen in Figure a (below) showing the SOA absorption coefficients with and without the correction terms described by Collaud Coen et al. (2010). It can clearly be seen that, without the corrections, one could erroneously conclude that SOA absorb light at 350, 880 and 950 nm.

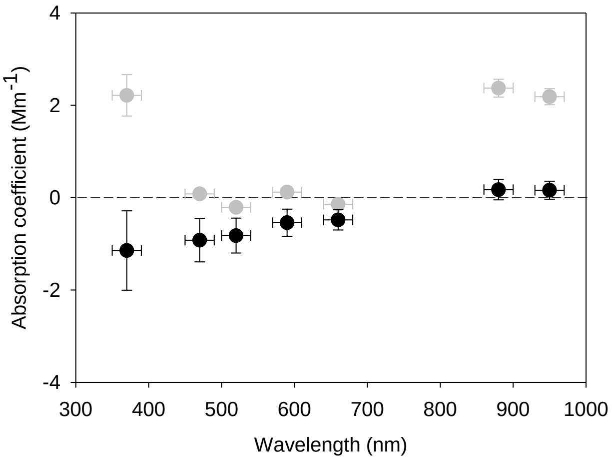


Figure a: Comparison of the absorption coefficient with (dark bots) and without (grey dots) the correction terms described by Collaud Coen et al. (2010)

10. P10557-10558, S3.1: This section could be shortened or moved to the Supplement because the results do not seem critical to the subsequent discussion of hygroscopicity and optical properties.

Response: As discussed in response to comment #3, this section has been shortened.

11. P10562-10563, S3.4.2: This section could be shortened or removed (or moved to the Supplement) because most of this is already well established in the literature. The novel result that the "surface" and "bulk" O/C ratios are different and vary differently with aging – is already presented in sufficient detail in S3.4.1.

Response: As discussed in response to comment #3, this section has been shortened.

12. P10567, S5: It might be useful to compare the extinction cross sections of α-pinene SOA with other aerosol species known to contribute to the direct effect, such as black carbon and biomass smoke.

Response: We have added a new Figure (Figure 9 below) to the paper to compare k_{ext} of α-pinene SOA with values used in global models for organic aerosol and those retrieved by Lang-Yona et al., 2010) for SOA generated from a mixture of biogenics (mostly terpenes) released directly from plants.
Section 5 has been completed as follows: "For comparison, k_{ext} of organic aerosols calculated using the CRI and GF as prescribed in global models are shown in Figure 9. Mie scattering calculations for homogeneous spheres were performed to calculate $k_{ext.}$ Number size distribution representing biogenic aerosol particles in the Amazon Basin (Martin et al., 2010) was used as an input in the Mie scattering calculations. For wet conditions, the CRI calculations were based on volume-weighted CRI of values of α-pinene-O_3 SOA and water. k_{ext} of SOA generated from a mixture of biogenics (mostly terpenes) released directly from plants (Lang-Yona et al, 2010) is also shown in Figure 9. Despite the wide range of variability of the CRI values assumed in global models for organic aerosols (1.45-0i to 1.6-0.003i), the values for k_{ext} were very similar for all models. The k_{ext} values retrieved in the present study for SOA after 14 hours of reaction agree with the values used for global models, while k_{ext} obtained for SOA at the beginning of the reaction are 2-7 times smaller. This work is a first attempt to assess the evolution of k_{ext} of SOA during its formation and aging. Our results suggest that the k_{ext} of atmospheric SOA is not static and a single size distribution and CRI does not appear sufficient to accurately model its direct radiative effect".

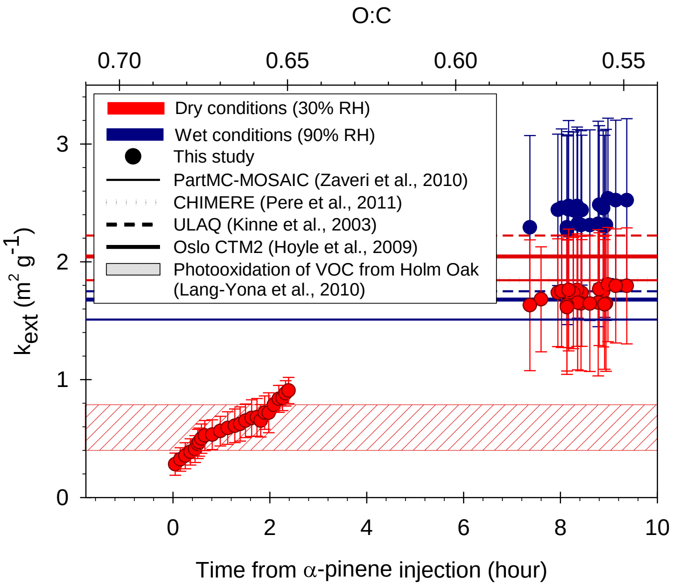


Figure 9. Mass extinction efficiency (k_{ext}) at λ=525 nm of SOA under dry (red dots) and wet conditions (blue dots) as a function of time and bulk O:C. These k_{ext} values are compared to those used for organic aerosols in several global models (lines) and those of SOA generated from a mixture of biogenics released directly from plants (dashed area) . Zaveri et al. (2010) assumed a CRI of 1.45-0i and GF of 1.23 , Pere et al. (2011) a CRI of 1.45-0.001i, Kinne et al. (2003) a CRI of 1.60-0.003i and GF of 1.09 (for ULAQ model) and Hoyle et al. (2009) a CRI of 1.53-0i and GF of 1.03. The hygroscopic growth of SOA was not taken into account by Pere et al. (2011).

13. P10573, L28: reference is incorrect.

Response: Corrected

14. P10577, L24: reference is incorrect.

Response: Deleted

15. P10589, F3: Is it necessary to show these results graphically if they are all zero within the uncertainty of the Aethalometer measurement?

Response: The Figure has been moved to the Supplementary Material.

16. P10592, F6: Can the authors clarify how the GF(90%) values in Figure 5 are calculated when this figure suggests that GF(90%) is approximately 175 / 190 = 0.92, rather than 1.02 – 1.07?

Response: We want to thank the Referee to point out this oversight in the paper. The GF(90%) has been calculated with $D_{p,m}(RH_{dry})$ *at 40 % RH. Below 40% RH, if the physical state of particles change (discussed in section 4.2.), the particles may appear at a higher mobility size. Using mobile diameter below 40% RH to calculate GF would underestimate the GF and overestimate the amount of solute in the droplets. As no information is available on how high the real GF should be at RH<40%, we used* $D_{p,m}(RH_{dry})$ *at 40 % RH to avoid GF less than 1.*

The following sentences have been added to Figure 5 as follows: "For the calculations of f(RH) and GF, we used $\sigma_{scat}(RH_{dry})$ *and* $D_{p,m}(RH_{dry})$ *at 40 % RH."*

17. P10593, F7: This figure is confusing. Panels (a) and (b) correspond to calculated f(RH) values after 1hr and 16 hr aging, but panel (a) shows different experiment dates than panel (b). Given that, it is not clear to me how to interpret trends in f(RH) over the course of an experiment because it doesn't seem possible to directly relate (a) to (b). Can the authors clarify this? Are there no experiments with nephthelometer measurements at the beginning and at the end on the same date from which there is a clear trend in σscat with aging?

Response: As stated in response to comment Referee #1, this Figure has been modified in order to facilitate the interpretation of the evolution of f(RH) with aging. The evolution of f(RH) is already discussed in the paper P14 L2.

There is no experiment with nephelometer measurements at the beginning and the end of the same date because these measurements implied to inject water vapor in-situ. At the end of a f(RH) humidogram, the experiment must be ended since the RH in the simulation chamber is 100%. This means that when humidograms are performed for "fresh" SOA, the experiment is stopped after 1 hour of reaction. Nethertheless, the experiments were performed under very similar initial conditions, as observed with the reproducible O:C, CRI and GF, and we consider that the f(RH) retrieved from different experiments can be comparable.
This last point has been added in the paper as follows: "No experiment with nephelometer measurements at the beginning and the end of the same date was available. However, the experiments were performed under very similar initial conditions as shown by the reproducible values of CRI and GF that are also comparable with each other. There is a first increase of f(RH) as RH increases from 0 to 30%. " (P14 L7).

18. P10593, F7: I am confused by this statement in the figure caption: "The scattering growth factors are calculated as the ratio of σscat at a specific RH to σscat at 30 % RH to avoid values lower than 1". Figure 7b shows clearly that f(RH) < 1 below ~25 % RH, down to a minimum f(RH) ~ 0.7. Is this a real trend? Please discuss or clarify.

Response: We thank the Referee to point out this mistake. The legend of the Figure has been modified as discussed in response to comment #16.

19. P10595, F9: This figure could be removed (or moved to the Supplement) for reasons mentioned in previous comment #9.

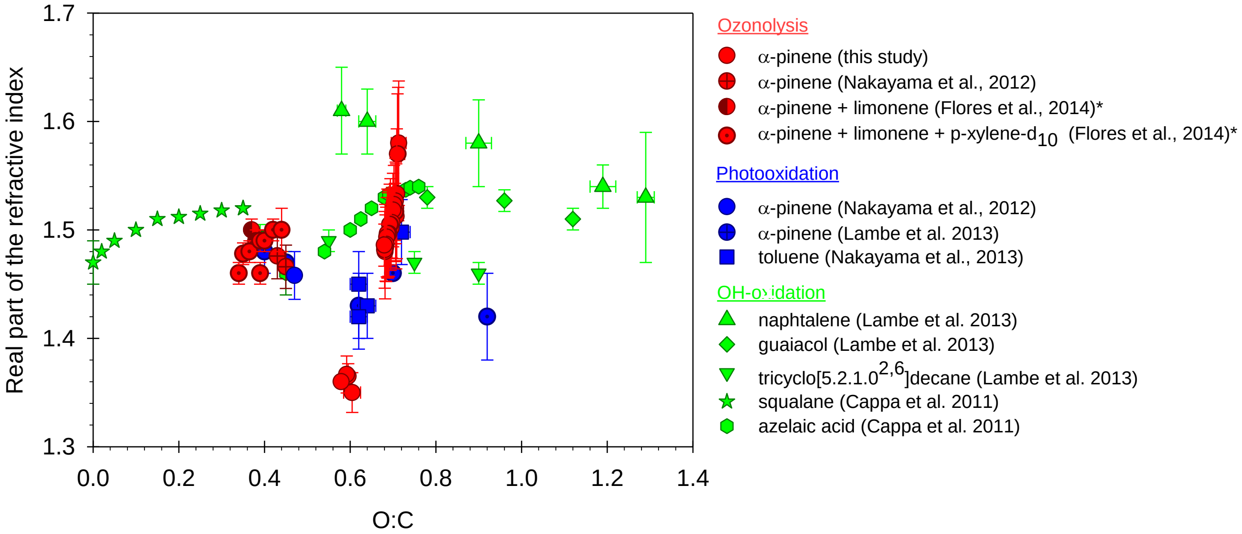
Response: As discussed in response to comment #11, this Figure has been moved to the Supplementary Material.

20. P10596, F10: Some combination of the size growth factor, scattering growth factor, and k_{ext} could also be plotted as a function of O/C to illustrate the relationships between the different parameters as a function of chemical composition.

Response: We don't think that it would be relevant to plot the GF and f(RH) as a function of O:C since we observed that these parameters remained constant with time. As discussed in response to comment #12, Figure 9 has been added in the paper to plot k_{ext} as a function of O:C.

21. P10596, F10: It might be useful to add literature data to this figure from the Nakayama et al. (2012), Lambe et al. (2013), and Liu et al. (2013) studies cited in the manuscript.

Response: We added in this Figure (see Figure 8 below) all the references cited in section 4.1. We did not include the study of Liu et al. (2013) since the authors did not provide O:C.



* *measured at λ=405 nm*

Figure 8: Real part of the refractive indices at λ=525 nm of SOA as a function of the bulk O:C. In our study, representative error bars represent ±1σ in replicate measurement.

<u>*References:*</u>

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Anonymous Referee #2

General comments:

In general, the topic of the manuscript is crucial for the restriction of uncertainties of aerosol direct effect in climate models. There are still lots of open questions in this field, related to the hydroscopicity and scattering effect of SOA particles. The manuscript has potential results for the publication in ACP but in my point of view, drastic revisions would be demanded to the manuscript before acceptance. The title is a bit misleading since the main focus of the manuscript is to investigate the effect of residence time on hygroscopicity and optical properties of SOA particles. Hygroscopicity and optical properties of pure α-pinene derived SOA have already reported widely in previous studies (cited in the manuscript). If the present title is not rephrased, strengthening of the relation between hygroscopicity and optical properties is needed (interpretation, figures etc.).

Response: We have decided to strengthen the link between optical and hygroscopic properties as this was also pointed out by the Referee#1. In the revised version, we have used Mie scattering calculations for homogeneous spheres to determine the size growth factor GF from the scattering growth factor f(RH). σ_{scat} *was calculated for different GF at specific RH. The optimal GF as a function of RH was determined so that the differences between measured* σ_{scat} *and those obtained using Mie calculations were minimized. Particles were assumed to be homogeneous spheres of uniform CRI. The CRI calculations were based on volume weighted refractive indices of SOA and water. Uncertainties on the theoretical GF were estimated from the standard deviation of the measured f(RH), the uncertainties on the f(RH) measurements and the uncertainties on the RH measurements. Figure 6 (below) shows the comparison between measured and predicted GF values for SOA at two different reaction times: for "fresh" SOA (after 1 hour of reaction), and for "aged" SOA (after 14 hours of reaction). For both reaction times, the model approach agrees well with the measurements above 30% RH, indicating no kinetic limitations for water uptake. For "aged" SOA (after 14 hours of reaction), the observed underestimation of the model below 30% RH might be due to the phase transition from a predominantly glassy state to a predominantly liquid state, as discussed in section 4.2.*
These results have been added to section 3.3 in the manuscript.

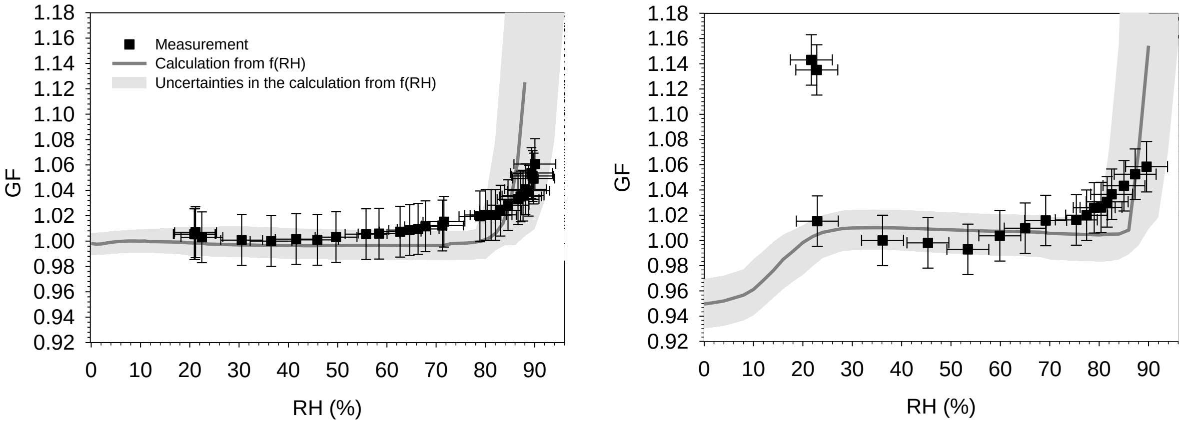

Figure 6: Humidograms showing measured (black symbols) and predicted GF (grey line) as a function of RH of SOA (a) after 1 hour of reaction and (b) after 14 hours of reaction. The grey area represent the uncertainties in the calculation of GF from f(RH).

The other main result in the manuscript is the different O:C at the surface of SOA particles. This should be highlighted more since it has not been reported before. The explanation of the XPS analysis should be more comprehensive. It is conceivable that the particles evaporate within the XPS preparation and analysis which could change the O:C remarkably. Moreover a controller measurements of O:C of totally homogeneous SOA particles with the instruments (AMS & XPS) would make the O:C surface/bulk analysis much more reliable. The question about the offset between the analysis methods remains open.

Response: It is possible that some volatile compounds evaporate during the XPS analysis due to the low pressure in the instrument. However, we observed in our companion paper (Denjean et al., 2014b), an increase of the O:C after heating by the evaporation of semi-volatile components of SOA. This indicates that the O:C at the surface at the SOA reported in the present study would be an overestimation of the real values and strengthen the clear different O:C at the surface and the bulk SOA particles.
This point has been added in the new manuscript as follows: "Some volatile compounds could evaporate during the XPS analysis due to the low pressure in the instrument. In our companion paper (Denjean et al., 2014b), an increase of the O:C has been observed after heating by the evaporation of semi-volatile components of SOA. Therefore, the O:C at the surface of the SOA was certainly overestimated in the present study." (P16 L18)

Performing new measurements with homogeneous particles is clearly a control that should be pursued in future work. However, for the comparison, we considered an uncertainty of ±30% on the O:C of the bulk SOA, which may be largely overestimated compared to the experimental variability. In addition, as discussed above, the O:C of the surface of SOA was certainly an overestimation of the real value, which suggests a clear difference of O:C between the surface and the bulk SOA.

Indeed, there is high interest related to the core-shell chemical structure and more detailed analysis of mass spectrums from the two methods could improve the paper significantly. Nevertheless, this is not related to the optical properties in the manuscript at all, so, what is the main purpose of the surface/bulk – composition analysis in this manuscript?

Response: We plotted the spectra provided by both methods for "fresh" SOA (at the beginning of the reaction) and for "aged" SOA (after 14h of reaction) (Figure S4 and Figure a, shown below). Figure S4 shows that the AMS mass spectra were dominated by m/z 44 for 'fresh' SOA, while the strongest signal was observed at m/z 29 for 'aged' SOA. As discussed for Figure S5 (which shows the evolution of f_{43} and f_{44}), it indicates an increase of less oxidized semi-volatile compounds in the particle phase with aging.
We will complete section 3.4. as follows: "The evolution of the AMS mass spectra during the reaction is shown in Figure S4 (Supplementary Material). The AMS mass spectra were dominated by m/z 44 for 'fresh' SOA, while the strongest signal was observed at m/z 29 for 'aged' SOA. Figure S5 also shows that f_{44} decreased while f_{43} increased with time. These observations indicate an increase of less oxidized semi-volatile compounds in the bulk particle phase with aging.

The XPS spectra (Figure a below) shows an increase of the intensity of the C-O peak with time. However, this trend is very hard to interpret since the C-O pic refers to many oxygenated *compounds (alcohols, aldehydes, carboxylic acids, peroxides, ethers). Therefore, it is not possible to provide any clear conclusion on the evolution of the functionality at the surface of SOA. We decided to not include this figure in the paper since it does not provide any further information on the chemical composition of the surface of SOA.*

The measurements with nephelometer and aethalometer provide information on the optical properties of the bulk SOA. Therefore, the correlation between optical properties and O:C of the surface of SOA has no interest here. In contrast, the hygroscopic properties of particles can be strongly influenced by the chemical composition at their surface. This is the reason why the O:C of the surface is only related to the hygroscopic properties in section 4.1.

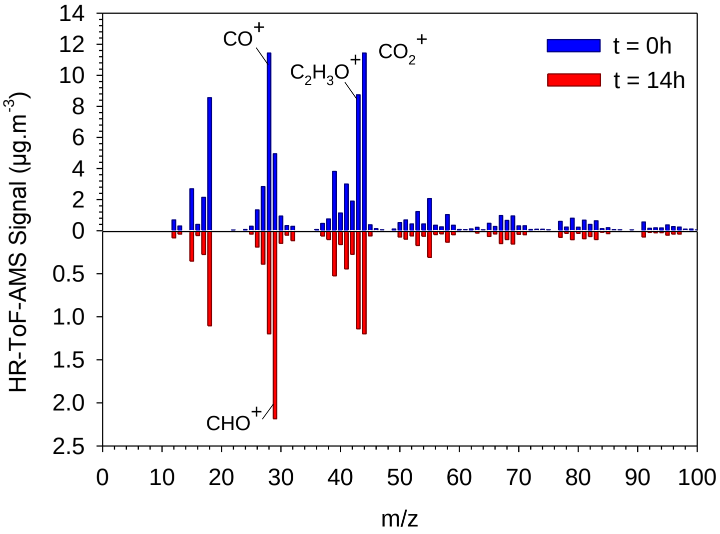

Figure S4. AMS mass spectra of α-pinene-O_3 SOA after 1 hour of reaction (red) and after 14 hours of reaction (blue).

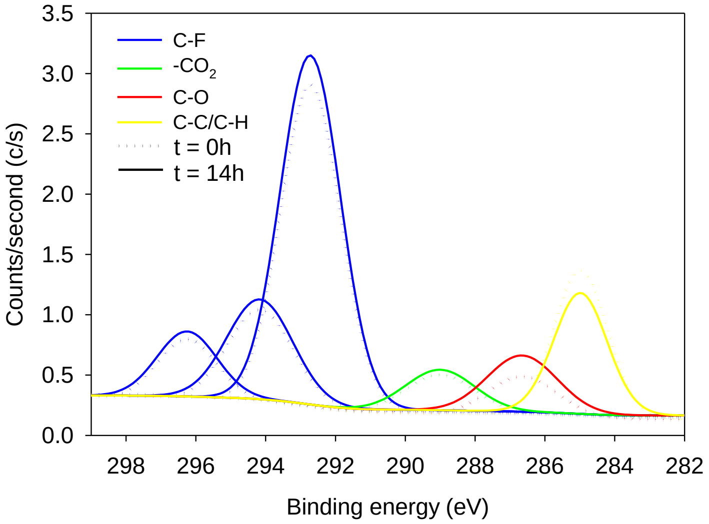

Figure a. Temporal evolution of the of the XPS spectra in the C1s region (normalized to the integrated peaks area of the –CO_2, C-O and C-C/C-H) of SOA after 1 hour (solid lines) and after 14 hours of reaction (dashed lines).

Additional proof reading is required throughout the manuscript.

Response: In order to improve the English content of the paper, the manuscript has been read by two native English speakers.

Comments in more details:

P1L9: Affiliation information incomplete

Response: The affiliation has been replaced by "Leibniz Institute for Tropospheric Research, Permoserstr. 15, 04318, Leipzig, Germany"

P1L22: "Physical, hygroscopic, optical and chemical properties of …" Hygroscopic and optical properties ARE physical properties

Response: The sentence has been modified as follows: "The SOA formation and aging were studied by following their optical, hygroscopic and chemical properties".

P1L25: "under various relevant atmospheric conditions, including relative humidity (RH)" In terms of variability, RH was the only variable in the system and mainly it was kept <30%RH which doesn't make it so atmospheric relevant.

Response: This sentence is not included in the new version of the paper.

P1L27-28: "…the dependence to RH of the particle size (*GF*)…" This is misleading, it would rather be: "…the dependence of RH on the hygroscopic growth factor (HGF) of particles…"

Response: This sentence has been replaced by "the effect of RH on the particle size (size growth Factor, GF) and on the scattering coefficient (scattering growth factor, f(RH))".

P2L10: "adsorption" should be absorption since the O:C wouldn't decrease adsorption / hydrophilicity.

Response: replaced

P2L12-13: "We postulate that this change could be due to a change in viscosity of the SOA from a more glassy state to a more liquid state" For this postulation I do not find any data to make an interpretation like this.

Response: The change in the viscosity of the SOA has been discussed in details in the discussion section (P19L8 - P20L11). This result is based on the change of both mobility diameter and scattering coefficient from 0 to 30% RH observed for aged SOA.

P2L29: "…the mixtures found in the atmosphere..", I would replace atmosphere with "atmospheric SOA" since the atmospheric gas phase compounds are not making the large uncertainties directly.

Response: The sentence has been replaced by "... due to the complexity of the atmospheric SOA …"

P3L1: "residence time in the atmosphere" …lifetime?

Response: changed

P3L4: "atmosphere" …atmospheric SOA?

Response: changed

P3L12-13: "Some models…" incomplete sentence

Response: The sentence has been rephrased by: "Some models assume that SOA absorbs weakly solar radiation, and set the imaginary part of the CRI near 0.006, while others ignore the absorption by SOA"

P3L19-21: "α-pinene ozonolysis is one…and its ozonolysis is…" Ozonolysis and its ozonolysis

Response: The sentence has been re-written as follows: "The ozonolysis of α-pinene ozonolysis is one of the most well studied SOA systems (α-pinene-O_3 SOA), as α-pinene is a significant biogenic VOC in many regions, and its ozonolysis plays an important role in SOA formation".

P3L22: α-pinene-O_3 has to be defined here.

Response: Done

P4L1-3: What are the reasons for the simultaneous measurements?

On further consideration, we decided to delete the word "simultaneous" in the sentence cited by the Referee since the measurements of GF and f(RH) were not necessarily performed at the same time.

P4L2&L9 Word “their” refers to SOA particles or physical properties or aging?

Response: The sentence has been as follows: “it is critical to determine the hygroscopic behavior of both the size distribution and optical properties as well as the dependence of these properties on the chemical composition”

P4L15: I assume you can control more than RH, also T, p, gas phase concentrations etc.

Response: Yes, this is right, we added these details in the sentence: “under various relevant atmospheric conditions (temperature, relative humidity, pressure, gas phase concentration, etc.)”.

P4L20 + all the other acronyms in the text: Acronyms with multiple capital letters should not be written *italic*.

Response: Corrected

P4L27: σ should be defined here.

Response: We added the definition: “the ratio of the scattering coefficient (σ_{scat})”

P5L12: “over more..”

Response: Already replaced in the latest version of the paper by “for more”

P5L18: Accuracy of Vaisala RH sensor is typically +/-3% over the whole RH range which should be mentioned.

Response: The accuracy is written in the following sentences: “The sensor was calibrated prior to the experiments. The RH accuracy was ±1.9% up to 90% RH and the temperature accuracy was ±0.1°C at 20°C.”

P5L28: “…at flow rates 3/0.3 Lpm” The aerosol flow in TSI-CPC3010 is 1 Lpm (+/-10%), not 0.3 Lpm.

Response: We will add in the paper details of the experimental setup as follows: “The aerosol flow was diluted with filtered air before entering the CPC, in order to maintain the nominal flowrate at 1 Lpm in the CPC. The dilution air flow was sucked from the simulation chamber *to avoid any pressure gradient in the SMPS." (P6L30) and "Size distributions were corrected by the SMPS software for [...] the dilution of the aerosol flow before entering the CPC".*

Moreover, the response time of CPC3010 is ~1-5 sec., has it taken into account in SMPS scans (i.e. what was the scanning time)?

Response: We added in the new version of the manuscript (P5L31): "The SMPS scanning time was 2 minute 15 sec in total".

P5L30: PSL is not defined in the text

Response: The definition was already added in the new version of the paper: "using monodisperse PolyStyrene Latex spheres (PSL, Duke Scientific)"

P7L20: replace *-signs with dots in the equation

Response: Replaced.

P8L1: Equation (3): Why there are sum and differential in a same equation? The sum could be an integral or then differentials could be increment deltas (Δ). The same equation is used in previous paper, Denjean *et* al. (2014).

Response: We thank the referee to point out this error. We replaced the sum by an integral.

P8L18: H-TDMA is defined already at P8L13.

Response: Deleted

P8L21: I would not use a manner of representation: *Dp,m(dry)*, whereas I would prefer to use *D*p,m(RHdry) or something similar. "dry" is not a variable, RH is. (Note: Compare also the style between these representations)

Response: $D_{p,m}(dry)$ *was replaced by* $D_{p,m}(RH_{dry})$.

P9L1: There is accuracy for RH of +/-1%. Accuracy of the sensor is 3% and then in P15L1 you show accuracy of 4.2% for humidity. This is not totally logical.

Response: We realize that the sentence was not clear. In this sentence, "+/-1%" is not a reference to the accuracy for RH, it refers to the ability of the H-TDMA to hold a constant RH (+/-1%) at a nominal value (90% RH). We change the sentence as follows: "In this study, the H-TDMA was typically operated at a constant high RH (± 1%) of 90%".

The uncertainty of 4.2% RH associated with the RH measurements was calculated from the weighted average of RH uncertainties of the aerosol flow and sheath flow (+/-3%). We completed the sentence (P14L19) as follows: "the RH uncertainty was based on the weighted average of the RH sensors uncertainties at the entrance of the H-TDMA"

P9EQ(4): incorrect font and design (*italic* etc.)

Response: The design of all the equations has been changed

P9L10: "A the end…" …At?

Response: deleted in the new version of the paper.

P9L19: "H-TMDA" should be H-TDMA

Response: corrected.

P9L19-22: I do not find the result from the previous Denjean *et* al. (2014): 101 "…which carry information on water transfer dynamics…etc."

Response: In Denjean et al. (2014a), we applied the two approaches (H-TDMA and in-situ humidification of particles) to investigate the hygroscopic properties of ammonium sulfate particles. We observed that ammonium sulfate particles humidified for a few minutes in the chamber displayed a different behavior than with the H-TDMA: A continuous increase of D_p *and* σ_{scat} *was observed from RH values as low as 30% RH, while these parameters stayed constant until 80% RH with the H-TDMA. We showed that layers of water were adsorbed on ammonium sulfate particles far below deliquescence for RH values as low as 30 % in the chamber. The two approaches of hygroscopicity measurements can thus be complementary to carry information on water transfer dynamics.*

P10L27-29: Reformulation of this sentence is needed.

Response: The sentence was re-written as follows: "Further chemical analyses were performed by collecting SOA on PTFE filters (Zefluor, 47mm diameter, 2 µm pore size, Pall Life Sciences), which were cut to the size of the collector using ceramic scissors."

P10L33 L min-1 is used instead of Lpm. The same unit has to be used everywhere in the text.

Response: Corrected

P11L1: Why didn't you collect the samples at different times, for example an initial sample and after 14 hours?

Response: This is exactly what we have done in this study: we sampled at different times of the reaction from 30 minutes to 17 hours. To clarify this point, we have added the following sentence: "Filter samplings were performed in the chamber at different reaction times from 30 minutes to 17 hours at a nominal flow rate of 2 Lpm for a sampling time varying between 30 min to 2 hours, depending on the total SOA volume concentration".

P12L1: The first sentence is not needed here. In this part of the text it is too general.

Response: Deleted

P13L7: …but up to 2 times lower compared to other studies" This sentence needs citations.

Response: The citations will be added: "but up to 2 times lower compared to other studies (Wang et al., 2011; Griffin et al., 1999; Cocker et al., 2001; Saathoff et al, 2009; .Shilling et al 2008)".

P14L27: "A" should be "The"

Response: Changed

P14L27: The message of the first sentence is not clear to me. Is this a general overview or did you do also humidograms in this study?

Response: It is a general sentence to introduce the results obtained with the first approach (HTDMA measurements). The sentence was changed to: "The first insight in the hygroscopic behaviour of SOA was brought by measuring humidograms of SOA with the HTDMA".

P14:L27 H-TDMA

Response: Corrected

P15L2: "SOA remained hydrophobic…" This is NOT true. GF values are higher than 1 which indicates that indeed, they are hydrophilic! Nevertheless you cannot say if the water is on the surface of the particles (adsorption) or inside the particles (absorption). Restriction in water uptake of SOA particles doesn't mean that they are automatically hydrophobic.

Response: We wanted to emphasize in this sentence that the GF stayed rather low compared to very hydrophilic compounds. We agree that the sentence was confusing and we will delete it.

P15L31: Error estimations of O:Cs are straight from the Aiken *et* al. (2007), no any statistical part?

Response: The uncertainties given by Aiken et al. (2007) may be overestimated compared to the experimental variability and even experimental reproducibility observed in this study. In fact, we estimated the experimental uncertainties to be ±0.01 from the standard deviation of the experimental values.
This point has been added in the text as follows: "The uncertainties in bulk O:C given by Aiken et al. (2007) may be overestimated compared to the experimental variability and even experimental reproducibility observed in this study. In fact, we estimated the experimental uncertainties to be ±0.01 from the standard deviation of the experimental values."

P16L1-8: This is not enough to explain the decrease of O:C over aging time.

Response: The decrease of O:C ratio over aging was discussed in details in the section 3.4.2 of the old manuscript (this section is now combined with section 3.4.1., as suggested by Referee #1). This part only aimed at comparing the bulk and surface O:C ratios.

P16L10-16: So bulk O:C is decreasing and surface O:C is increasing but after hours they are ~equal (within errorbars). So it also means that bulk O:C inside the particles is decreasing effectively even more than 0.68 => 0.55. Have you calculated how much should it be in the core of particle if the surface O:C is 0.33 versus 0.46?

Response: The XPS allows the measurement of the O:C of the particles to a depth less than 10nm. This means that, for a particle of D_p*=200 nm, the O:C at the surface contributes at* $\frac{\frac{4}{3}\pi.10^3}{\frac{4}{3}\pi.100^3}.100 = 0.1\ \%$ *of the O:C of the bulk, which is negligible.*

P16L19: The section 3.4.2 does not give any new aspects or answers to the reader. It could be shortened and combined with 3.4.1.

Response: The objective of section 3.4.2. was to bring some issues on the decrease of the O:C in the bulk SOA. By looking at the functional groups, we concluded that the decrease of O:C *could be due to an increasing partitioning of less oxidised semi-volatile compounds as the aerosol grew. We think that this part is important to interpret the trend of the O:C and thus the evolution CRI with time, but we agree with the Referee that it could be shortened. We thus decided to move Figure 9 in the Supplementary Material and we combined a shortened version of section 3.4.2 with section 3.4.1.*

P18L10: The following could have an important role in this case: Alfarra, M.R., *et* al. Water uptake is independent of the inferred composition of secondary organic aerosols derived from multiple biogenic VOSs, ACP, 143 13, 11769-11789, 2013.

Response: We thank the Referee for this paper. We have completed the 2^{nd} *point in the interpretation of the stability of GF in section 4.1. as follows: "2) Other factors than the bulk O:C can control the water uptake of SOA: recently, Alfarra et al. (2014) reported a positive correlation between hygroscopicity of particles and their degree of oxidation for SOA produced from the photooxidation of α-pinene, β-caryophyllene, linalool and myrcene, but not for limonene SOA. They suggested that other factors such as solubility, surface tension, molecular weight, density and particle phase are likely to be playing important roles in controlling GF values."*

P18L14: "out" should be our?, "Gf" should be GF.

Response: corrected.

P18L20: "The variation of GF due to the chemical composition change may not be detected due to sensitivity of the H-TDMA." Indeed, but WHY you are then analyzing hygroscopicity at all? In my point of view, lot more of data with different O:C, VOC concentrations, different precursors and wider GF range would be required.

Response: The aim of the paper is not only to find relations between hygroscopic properties and chemical composition of SOA, but also to see if these properties evolve during the aging of α-pinene-O_3 *SOA. As stated in the introduction, previous studies show disagreements on the evolution of the hygroscopicity of α-pinene-*O_3 *SOA and here we provide new data under different experimental conditions. It is also important to note that none of the previous studies have provided the O:C of α-pinene-*O_3 *SOA together with the GF.*
We modified the sentence as follows: "The variation of GF due to the chemical composition change is smaller than the sensitivity of the H-TDMA"

P18L22: You cannot say "less hydrophilic", also solubility can play a role, not only hydrophobicity.

Response: Deleted

P18L23: I would remove the word “significantly”.

Response: Deleted

P18L25: adsorption should be absorption?

Response: Changed

P19L6-18: You could calculate the effective change of the shape factor of SOA particles which would explain “the shrinking effect”.

Response: The referee suggests estimating the change of the shape of SOA. This calculation would assume non-sphericity for the SOA particles under dry conditions, which is not the case, as shown by the TEM imaging described below (Figure S6 of the manuscript).

P19L15: I would replace “merge into single spheres” with “coalescence as it is in Pajunoja *et* al. (2014).

Response: Changed

P19L16-19: The residence time for α-pinene SOA in Pajunoja et al. (2014) seems to be up to two months so the particles may be agglomerates with shorter residence times (the time they are not coalescenced yet).

Response: We thank the Referee for this information. In order to investigate the shape of SOA, we performed TEM measurements of SOA after 1 hour and 14 hours of reaction. SOA particles were collected on copper TEM grids. 50 particles have been analyzed by TEM. Figure S6 shows an example of SOA particles after 1 and 14 hours of reaction. Only spherical particles has been observed in the samples. This indicates that the coagulation taking place at the beginning and after 14 hours of reaction resulted in spherical coalesced particles

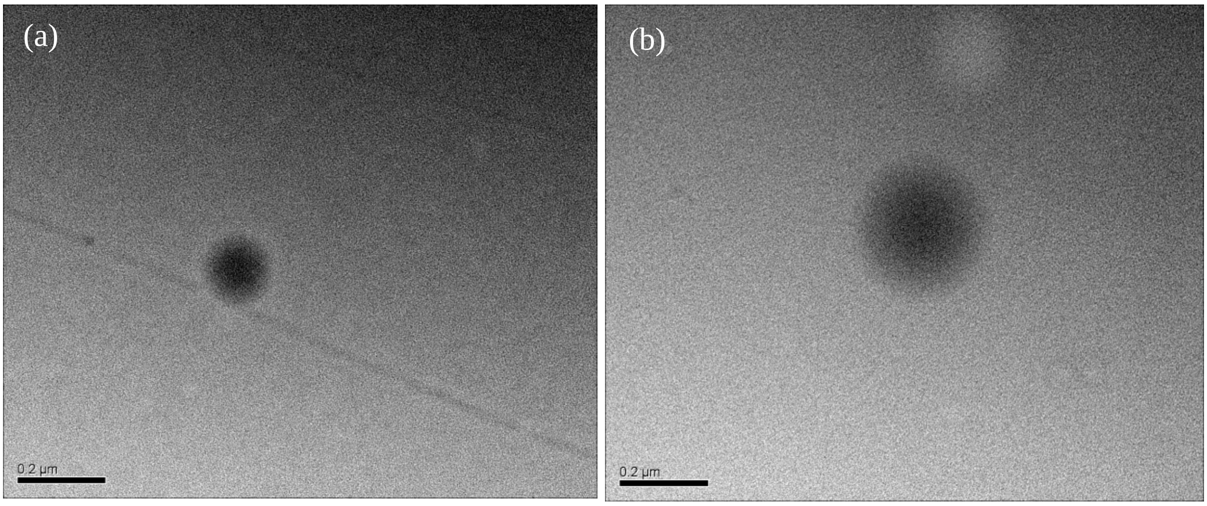

Figure S6. Examples of TEM images of α-pinene-O_3 SOA (a) after 1 hour and (b) 14 hours of reaction.

These measurements have been added in section 4.2. which has been changed as follows: "Electron microscopy analysis was used to investigate the shape of SOA particles. SOA particles after 1 hour and 14 hours of reaction were analyzed by transmission electron microscopy (TEM) (Figure S6 in the Supplementary Material). Experimental details are given in the Supplementary Material. In total 50 particles were analyzed by TEM. Figure S6 shows an example of SOA particles after 1 and 14 hours of reaction. Only spherical particles have been observed in the samples. This indicates that the coagulation taking place at the beginning and after 14 hours of reaction resulted in spherical coalesced particles".

P19L20-P20L22: For me this concentrates too much on glassy state of particles since there are no results indicating highly viscous material. Evaporation of semi-volatiles and effect of wall losses in the chamber are not discussed enough and should be analyzed much more carefully! Partial vapor pressures of VOCs and other gas-phase compounds are changing which are affecting gas-to-particle equilibrium. Furthermore if the particles are assumed to be highly viscous they would need much more than 1 hour to equilibrate. Thus, the increase of RH at the end of the experiment could be way too fast for the viscous particles!
Kokkola H, et al. (2014) The role of low volatile organics on secondary organic aerosol formation. ACP, 14(3), 1689–1700 176
Zhang X., et al. (2014) Influence of vapor wall loss in laboratory chambers on yields of secondary organic aerosol

Response: The Referee suggests two interesting interpretations of the humidograms of GF and f(RH) below 40% RH: 1) the gas/phase partitioning and 2) the particles wall losses in the chamber.
1) Concerning the evaporation of semi-volatile compounds, even if it can lead to a decrease of GF, it cannot explain the increase of f(RH). f(RH) is positively correlated to the total mass concentration of particles and a decrease of the particles' size would decrease the total mass concentration and f(RH).
2) The particle wall losses in the chamber would lead to a decrease of the total number concentration and a decrease of f(RH). This is not what we observed during our experiments and it cannot explain the humidograms below 40% RH.

We changed the section 4.2. by adding the following sentences: "Although evaporation of semi-volatiles in the chamber could lead to a decrease of the observed mobility diameter, it cannot explain the increase of f(RH) below 40% RH. Furthermore, the observed decrease of the mobility diameter cannot be attributed to losses of particles to the walls of the chamber, since it would lead to a decrease of f(RH)".

The Referee also suggests that the time for humidification (1 hour) could be too short for humidifying the SOA regarding its viscosity. From the data obtained in this study, we cannot provide a fully answer on this point. As discussed previously, we have conducted new calculations to combine GF measurements (obtained with a humidification of 10 sec in the H-TDMA) and f(RH) measurements (1 hour for humidification) and observed a good agreement with the two approaches. This indicates no change for water uptake process during the first hour of humidification.

P33L1: Table 1: Initial α-pinene concentrations could be added to this because it has an effect on O:C and hygroscopicity of SOA particles.

Response: We are not able to give the exact value of initial α-pinene concentrations since the ozone was first introduced in the chamber and as soon as α-pinene was introduced, it immediately reacted with ozone. Because the FTIR measurement of α-pinene concentrations provide a response time of 5 minutes, the first measurement after α-pinene introduction shows a concentration already significantly lower than the real initial one (see Figure 2a). We added in the legend of Table 1: "All experiments started with ~ 200 ppb of α-pinene.".

P35L1: Table 3: This table would need also O:C, (α-pinene concentration), and estimated time after ozonolysis. I think also that this table is not obligatory since the values are quite similar. More than this table, the table where are GFs as a function of residence time would make better sense.

Response: We thank the Referee for this suggestion. Initial concentrations of α-pinene and ozone, and the residence time of the SOA in the chamber have been added in table. We did not include the O:C, since none of the previous study determined this parameter.

Reference	*GF(90%RH)*	*Residence time*	$[\alpha\text{-pinene}]_{initial}$ *(ppm)*	$[O_3]_{initial}$ *(ppm)*
This study	*1.02 - 1.07 (±0.02)*	*2 h*	*0.20*	*0.25*
	1.02 - 1.07 (±0.02)	*20 h*		
Prenni et al. (2007)	*1.01 - 1.07 (±0.02)*	*2 h*	*-*	*excess*
Qi et al. (2010)	*1.09*	*30 mn*	*0.05-0.10*	*0.30-0.34*
	1.09	*7 h*		
Saathoff et al . (2003)	*1.08 (±0.01)*	*1 h*	*0.06*	*0.5*
	1.11 (±0.01)	*6 h*		
Warren et al. (2009)	*1.02 - 1.16 (±0.02)*	*30 mn*	*0.05*	*0.3-0.5*
	1.02 - 1.16 (±0.02)	*6 h*		

To be consistent, these parameters have also been included in Table 2, as follows:

Reference	*Real CRI*	*Residence time*	*λ (nm)*	*[α-pinene]$_{initial}$ (ppm)*	*[O$_3$]$_{initial}$ (ppm)*
This study	*1.60 (± 0.02)*	*10 mn*	*525*	*0.20*	*0.25*
	1.33 (± 0.02)	*19 h*			
Kim et al. (2010)	*1.45 (± 0.05)*	*2.5 h*	*670*	*0.50-5.00*	*0.10-1.00*
Kim and Paulson (2013)	*1.39 (± 0.02)*	*< 30 mn*	*532*	*0.13-0.17*	*0.50*
	1.52 (± 0.02)	*4 h*			
Liu et al. (2013)	*1.498 (± 0.002)*	*38 s*	*550*	*4.00*	*52.2*
Nakayama et al. (2010)	*1.41 (± 0.02)*	*2-3 h*	*532*	*0.10*	*2.00*
Nakayama et al. (2012)	*1.47−1.48 (± 0.02)*	*2-3 h*	*532*	*0.10*	*1.09-2.57*
Redmond and Thompson (2011)	*1.49 (± 0.04)*	*-*	*532*	*-*	*0.5-1.00*
Schnaiter et al. (2003)	*1.44*	*1.23 h*	*>350*	*0.06*	*0.47*
Wex et al. (2009)	*1.45*	*2 mn*	*visible*	*-*	*excess*

P35L1: Table 3 & P42L1: Figure 6: Where is the point 175nm/190nm? If the residence time in the Table 3 is not listed the table is misleading.

Response: The GF(90%) has been calculated with $D_{p,m}(RH_{dry})$ at 40 % RH. Below 40% RH, if the physical state of particles change (discussed in section 4.2.), the particles may appear at a higher mobility size. Using mobile diameter below 40% RH to calculate GF would underestimate the GF and overestimate the amount of solute in the droplets. As no information is available on how high the real GF should be at RH<40%, we used $D_{p,m}(RH_{dry})$ at 40 % RH to avoid GF less than 1.

The following sentences have been added to Figure 6 as follows: "For the calculations of f(RH) and GF, we used $\sigma_{scat}(RH_{dry})$ and $D_{p,m}(RH_{dry})$ at 40 % RH."

P43L1: Figure 7: Are the legends correct? The day numbers are different so how have you interpreted the data? Also in the Fig 7b, the relative f(RH) <1 at <30% 194 RH but in Figure 6b the HGF is absolutely the highest. Why is so?

Response: There is no experiment with nephelometer measurements at the beginning and the end of the same date because these measurement implied to inject water vapor in-situ. At the end of a f(RH) humidogram, the experiment must be ended since the RH in the simulation chamber is 100%. This means that when humidograms are performed for "fresh" SOA, the experiment is stopped after 1 hour of reaction. Nethertheless, the experiments were performed under very similar initial conditions, as observed with the reproducible O:C, CRI and GF, and we consider that the f(RH) retrieved from different experiments can be comparable.
This last point has been added in the paper as follows: "No experiment with nephelometer measurements at the beginning and the end of the same date was available. However, the

experiments were performed under very similar initial conditions as observed with the reproducible CRI and GF and can be compared with each other" (P14 L17).

The trend of f(RH) and GF is already discussed in section 4.2. We postulated that it could be due to a change of the viscosity of SOA.

P44L1: Figure 8: This figure would need a plot b) where is a comparison between AMS & XPS methods for homogeneous SOA particles. Figure 8 should also be explained in the text since the decrease of O:C as a function of time is "odd".

Response: Performing new measurements with homogeneous particles is clearly a control that should be pursued in future work.
The decrease of O:C with time was already discussed in section 3.4.2. (now grouped with section 3.4.1) from the interpretation of the functional groups.

P44L2: "These bulk O:C ratio are…" need to be reformulated (should be plural 200 etc.), O:C is a ratio itself so now it means "bulk ratio ratio…".

Response: Corrected

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