

Anonymous Referee #1

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

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.

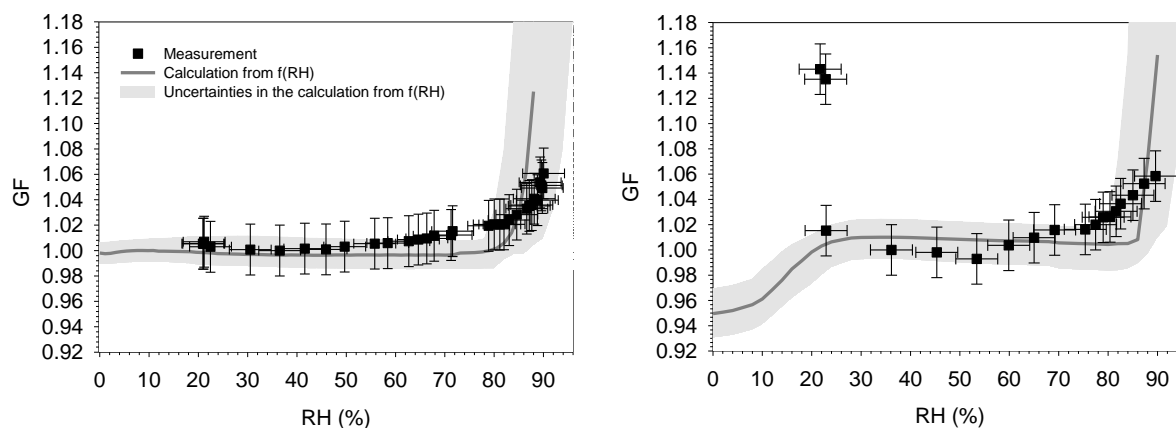


Figure 6: Humidograms showing measured (black symbols) and predicted GF (grey line) as a function of RH of SOA (a) after 1 hour of reaction and (b) after 14 hours of reaction. The grey area represent the uncertainties in the calculation of GF from $f(RH)$.

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).

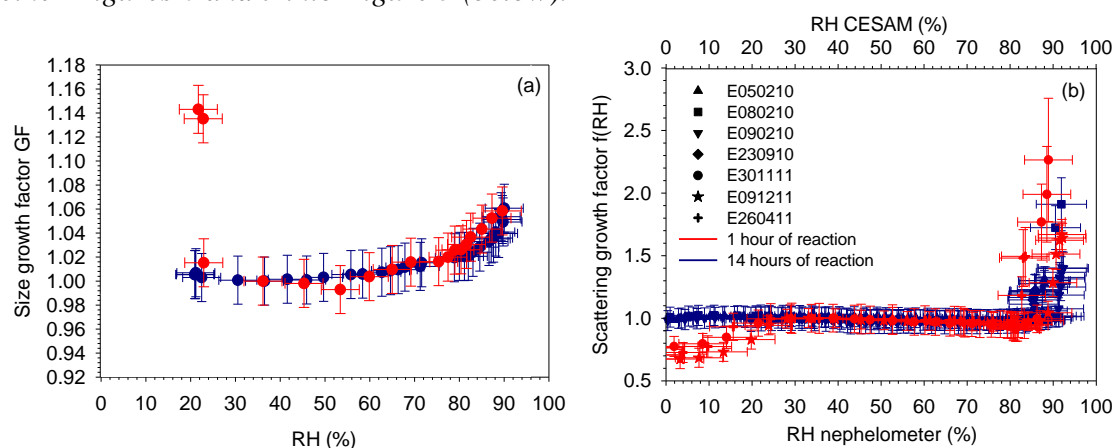


Figure 5. Humidograms of (a) size growth factor measured by the H-TDMA and (b) scattering growth factor measured by the nephelometer as a function of RH within the nephelometer (bottom axis) and the RH within the chamber (upper axis), for “fresh” SOA (after 1 hour of reaction) in red and for “aged” SOA (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 $[VOC]/[O_3] \sim 1$. This was performed in an effort to probe the influence of the evolution of the SOA chemical composition during formation (by gas-phase partitioning, 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.

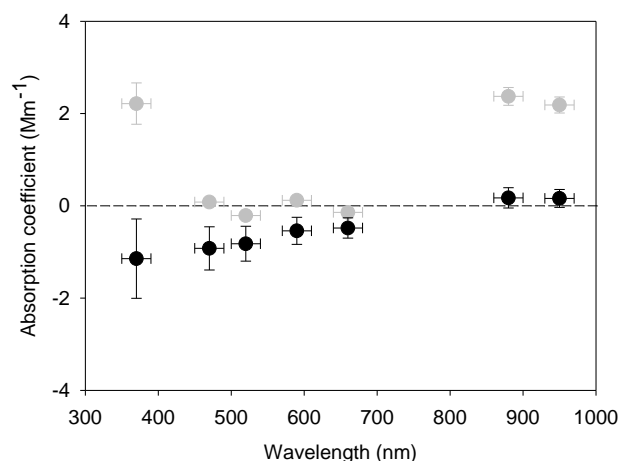


Figure a: Comparison of the absorption coefficient with (dark bots) and without (grey dots) the correction terms described by Collaud Coen et al. (2010)

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”.

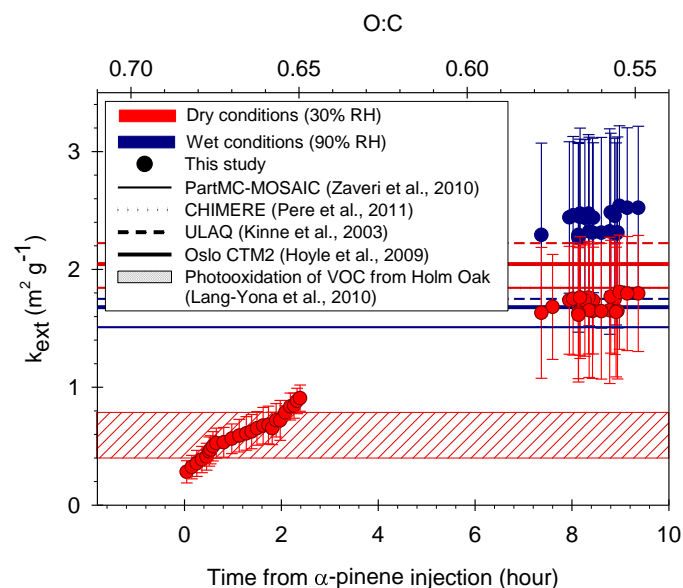


Figure 9. Mass extinction efficiency (k_{ext}) at $\lambda=525$ nm of SOA under dry (red dots) and wet conditions (blue dots) as a function of time and bulk O:C. These k_{ext} values are compared to those used for organic aerosols in several global models (lines) and those of SOA generated from a mixture 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 nephelometer measurements at the beginning and at the end on the same date from which there is a clear trend in σ_{scat} 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. Nevertheless, 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 σ_{scat} at a specific RH to σ_{scat} at 30 % RH to avoid values lower than 1”. Figure 7b shows clearly that $f(RH) < 1$ below ~25 % RH, down to a minimum $f(RH) \sim 0.7$. 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.

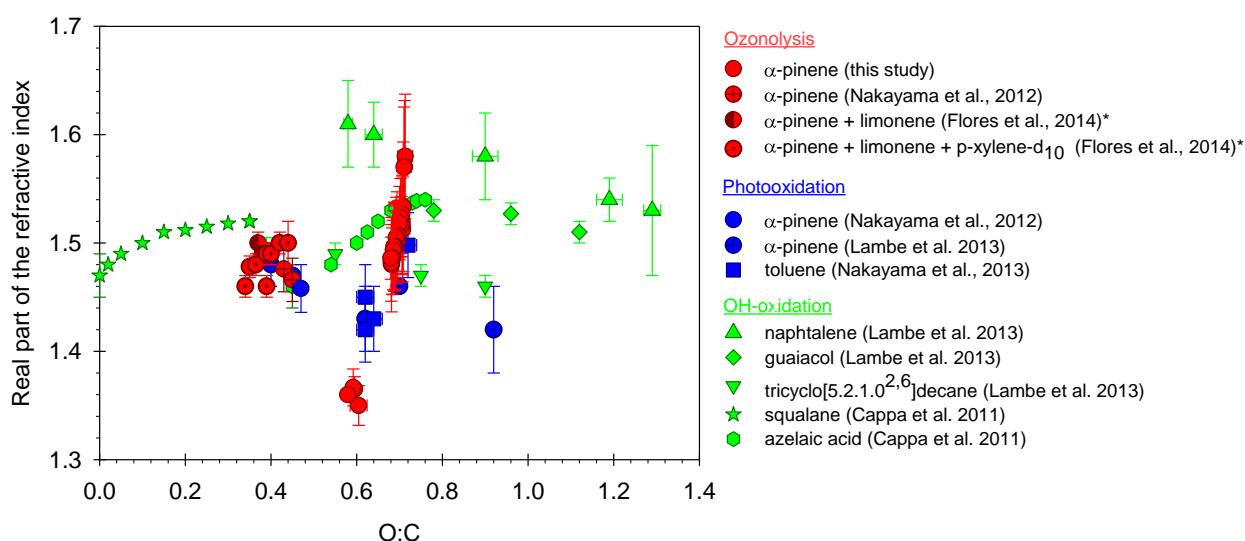
Response: As discussed in response to comment #11, this Figure has been moved to the Supplementary Material.

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 $\lambda=405$ nm

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

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