

Reviewer #1

Section 3.1: In the discussion of cloud cover, as indicated by satellites, the authors should provide the exact details of the cloud data (parameter value, time of data, and spatial area of data obtained relative to the sample site) and discuss the satellite retrieval(s) used.

The presence of clouds was determined by examining satellite pictures provided by Sat24. We used the view over Europe at 11:00 LT. The text has been updated to indicate this and Lines 219-222 now read as “Cloud cover, as indicated from satellite measurements, showed that the days preceding Period A were generally cloud free whereas clouds developed west of the ground sites preceding Periods B, C, and D (not shown). The presence of clouds was determined by examining satellite pictures set to the view of Europe at 11:00 LT provided by Sat24 (<http://en.sat24.com/en/eu>).”

Section 3.1: In addition to the previous comment, were there reports of precipitation in Period B to the west or anywhere near the measurement site? In other words, was wet scavenging important in explaining any aspect of the data?

Wet scavenging was not likely important as very little precipitation was observed during the study period. There was little evidence of precipitation west of the site and measurable precipitation was recorded at SPC only on the afternoons of 23 June and 6 July. The text has been updated to reflect this and Lines 223-225 now read as “Also, wet scavenging was not likely important as there was very little precipitation at SPC or west of the site during the entire study period. Only two cases of light rain lasting ~30 min, which occurred on the afternoons of 23 June and 6 July, were recorded at SPC.”

Section 3.1, Lines 11-25 on pg 35493: the discussion about correlation coefficients is useful, but I am not sure what rationale went into these three categories (>0.7 , $0.4-0.7$, <0.3). How do these relate to statistical significance on a standard students t-test table for the respective degrees of freedom used to generate the best fit lines? More discussion is required about the choice of these three categories.

We feel R^2 values are more informative for looking at the data since a R^2 value tells the fraction of variance in the y variable explained by the x variable. The three bins of R^2 values, therefore, are intended to quantify broad ranges of explained variance. p-values can also be useful for examining the statistical significance of a relationship, but even relationships where little variance is explained can be significant and thus not terribly interesting to discuss. For example, performing a t-test on both WSOC vs. OOA-4 for Period A (now Figure 10g) and Period C (now Figure 10h) would suggest their relationships were significant (Period A WSOC - OOA-4 = 3.07 $>$ t-test = 0.89 and Period C WSOC - OOA-4 = 0.75 $>$ t-test = 0.35). However, the R^2 value for Period A is 0.04 and Period C is 0.64, respectively. Therefore, while the t-test shows both relationships are statistically significant, the R^2 values are telling us more about the importance of the relationship between WSOC and OOA-4. Lines 238-248 now read as “We first will compare all four periods to examine for evidence of aqSOA. Then we will provide a further examination of aqueous aerosol tracers and WSOC for the two periods with similar air flow

(Periods A and C). Our analysis will largely be based on least square regression correlation analysis to examine the relationship between various species and provide a general approach to examine for evidence of aqSOA. We have chosen to examine R^2 values as opposed to p-values since R^2 values can provide a useful tool for explaining the amount of observed variance in a dependent variable that is explained by variation in an independent variable. p-values merely indicate whether a relationship is statistically significant without information about the amount of variance explained. To help categorize the fraction of variance explained, we consider a high correlation as R^2 values greater than 0.7, a moderate correlation as R^2 values between 0.3 to 0.7, and a low correlation as R^2 values less than 0.3.”

Figure 2: for Period B there seems to be a gap in the ALW data. Why is that? Also, why aren't data used (in the context of Figures 3-5) for the next couple of days when large changes in ALW are observed?

The gap in the ALW data is due to missing PILS-IC data. The caption of Figure 2 (now Figure 3) has been updated to indicate this and it now reads as “**Figure 3.** Times series of hourly averaged measured (a) WSOC, (b) calculated ALW, (c) RH, and (d) Temperature at SPC. Any gaps in ALW are due to missing PILS-IC data. The dashed vertical lines indicate midnight local time (UTC+2). Periods A, B, C, and D are also indicated.”

As suggested by the reviewer, we have also expanded the analysis to cover the periods before and after the original Period B. The original Period B is now called Period C. The new Period B covers 30 June, 1-2 July and Period D covers 6-7 July. The new Period B has moderate ALW and Period D has the highest ALW observed during the study. This allows comparison of different scenarios to help strengthen our results. Figures 3-5 (now Figures 4-6 and Figures S8-S9) have been updated to include all four Periods. The discussion about these figures in Section 3.2 has been updated and Lines 251-281 now read as “WSOC is shown as a function of RH for the times of RH increasing (Fig. 4a and 4b) and decreasing (Fig. 4c and 4d) during Periods A, B, C, and D. For Periods B, C, and D, WSOC had no relationship with RH. Only during the times of increasing RH did Period A have a relationship of increasing WSOC with RH, consistent with local aqSOA formation. This can further be illustrated by examining the correlation of WSOC vs. organic aerosol (OA), aerosol liquid water (ALW), and RH for Periods A, B, C, and D during the times of RH increasing (Fig. 5 and S8). In general, WSOC had a strong relationship with OA, but only Period A additionally had a moderate correlation of the WSOC with both ALW (Period A $R^2 = 0.65$ vs. Period B $R^2 = 0.15$, Period C $R^2 = 0.29$, and Period D $R^2 = 0.01$) and RH (Period A $R^2 = 0.39$ vs. Period B $R^2 = 0.01$, Period C $R^2 = 0.12$, and Period D $R^2 = 0.07$). The good correlation between WSOC and ALW is in agreement with a previous smog chamber study that found that ALW is a key determinant of SOA yield [Zhou *et al.*, 2011]. This also supports a recent study that observed ambient aqSOA formation during the nighttime as evident by the increased partitioning of gas-phase WSOC to the particle-phase with increasing RH [El-Sayed *et al.*, 2015]. The study by El-Sayed *et al.* [2015] found the increase in the fraction of total WSOC in the particle phase (F_p) at the two highest RH levels (70-80%, >80%) to be statistically significant compared to the F_p values at RH < 60%. The main focus of their work was to investigate if the uptake of gas-phase WSOC to aerosol water occurs through reversible or

irreversible pathways. The data suggested the aqSOA was formed irreversibly. We investigate this with our data in section 3.3.2.

Figures 6 and S9 show the correlation of WSOC vs. nitrate, oxalate, and sulfate for the times of RH increasing. Nitrate and WSOC are strongly correlated only during the times of RH increasing for Period A. Early morning nitrate peaks were observed at SPC during the first part of the study, but were absent at the upwind Bologna site (Fig. 7). The occurrence of these peaks overlaps with Period A. (Note, the nitrate event observed on 6 and 7 July during Period D will be discussed in Sect. 3.4.) This additionally suggested that the nitrate formation or the ammonium-nitrate-ammonia-nitric acid equilibrium at SPC was locally controlled since the back trajectory analysis indicated both the SPC and Bologna sites were sampling similar upwind air masses to each other in each period (Fig. 2). Therefore, the correlation with locally formed particulate nitrate suggests local formation of WSOC. (Note, increased nitrate also results in higher ALW at the same RH.) This argues that aqSOA formation was predominately local during Period A.”

It should also be noted that we updated the overview section to explain the expanded analysis and Lines 206-218 now read as “Therefore, our analysis will focus on comparing these two different halves of the study. Given our interest in examining for evidence of aqSOA we picked four periods with varying levels of WSOC and ALW. We also picked cases with both sites sampling similar air masses on a given day. Period A represents the first half of the study and covers 19-21 June. Period A has elevated WSOC and moderate ALW. As indicated by the difference in the length of the back trajectories [*Draxler and Rolph, 2013; Rolph, 2013*] shown in Fig. 2, Period A occurred during the end of a stagnation. Period B (30 June, 1-2 July), Period C (3-5 July), and Period D (6-7 July) represent three different cases in the second half of the study. Period B has moderate ALW, Period C has low ALW, and Period D has the highest ALW observed during the study. As indicated by Fig. 2, all three of these periods represent typical background conditions influenced by regional transport, but with slightly different flow patterns. The flows of Periods A and C are most similar. Due to changes in the WSOC concentrations and a non-consistent flow pattern on a daily basis, no periods between 23-29 June were examined.”

Figure 2: Avoid having numbers overlap on the y-axis for the two panels.

The y-axis has been fixed. The updated figure is shown below.

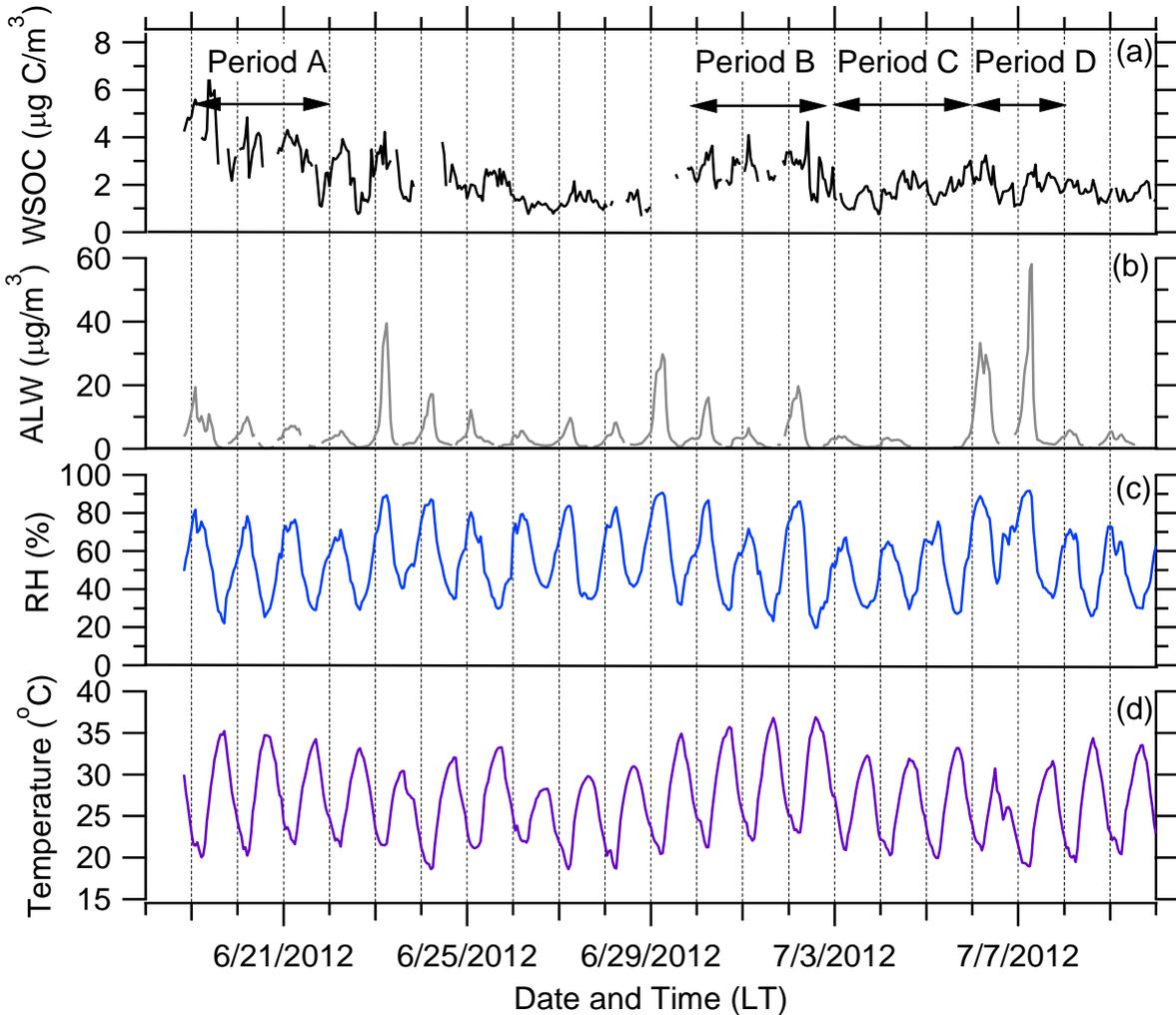
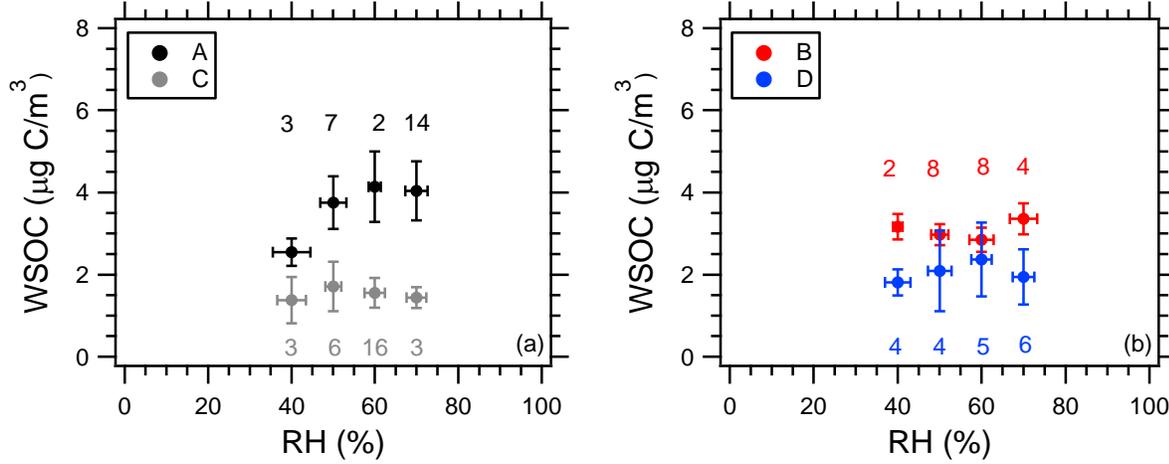


Figure 3: add error bars in the RH axis too for each marker. Also, it would be useful to report the number of points used for each marker in the two panels. For example, is the 80% RH point in panel A based on very few points compared to the other markers? And why isn't there a point at 80% RH for the bottom panel for Period A. The decrease in RH should start at 80% if the increase in RH ended near 80%. It seems as though if the RH increased to 80% that one can be picky as to which panel that marker is placed in and obviously it looks much better in the top panel to make the case for the reported conclusions. This is an issue that needs to be discussed in a revision.

Errors bars on the RH axis have been added to the figure as suggested. The number of points used in each RH bin has also been included in the figure. The updated Figure 3 (now Figure 4) is shown below. The 80% point in Period A was due to only one point, which is why it was included in only the RH increasing figure. However, this point does not drive the observations. Therefore, to show how robust the results are we have redone the analysis going from 40% to 70% RH and then from 70% to 40% RH. All relevant figures and text in the paper have been updated accordingly for this reanalysis.

RH Increasing



RH Decreasing

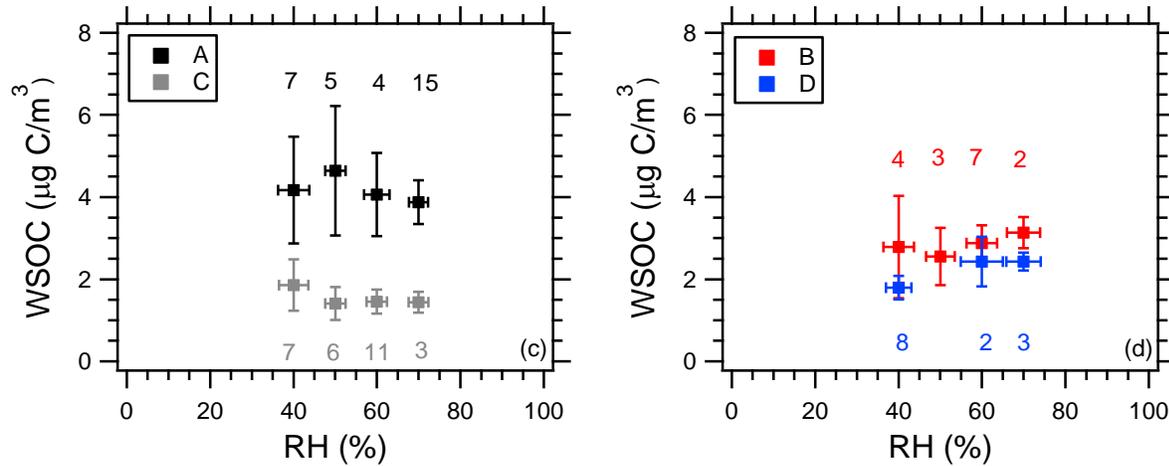


Figure 8C: what would the correlation be if the outlier point farthest to the left is omitted and are there any special characteristics associated with that datapoint? In the absence of that point it could be argued that a somewhat similar relationship exists as compared to panel D.

We believe this is in reference to Figure 5c (now Figure 6c) as Figure 8 is a time series. The outlier point does not dramatically change the correlation. If the outlier point is not included in the fit the R^2 value remains low, changing from 0.16 to 0.32. The point itself does not appear to have any special characteristics. It looks like the oxalate concentration could possibly be a bit lower than would be expected at its WSOC concentration, but we double checked even the chromatogram and it appears to be correct. Therefore, the point is still included.

Figure 8A-B: it seems that the same general positive trend exists in both panels. The issue in panel B could be that there is a bit more noise and it has (what appears to be) fewer data points.

We believe this is also in reference to Figures 5a and 5b (now Figures 6a and 6b). We agree that due to the differences in the axes the same general trend does appear in both plots. There is a bit less data in Figure 5b due to the missing anion data during the original Period B. Therefore, we have supplemented AMS nitrate and sulfate data into the new Figures 6b and 6f. However, this does not change the lack of relationship between WSOC and nitrate, the R^2 value changes from 0.18 to 0.01 when including the AMS data.

Figure 9: Panel B shows the presence of 2 outlier points to the middle-right that reduce its correlation. Having more datapoints would help in this case as it is unclear as to what explained those outlier points. The same applies to Panel B; note also that the right value on the x-axis of panel d is cut off.

The two outliers appear to be an artifact from the PMF analysis. However, they do not significantly change the correlation. If those two points are removed then the R^2 value for WSOC vs. OOA-1 (Figure 9b, now 10b) changes from 0.02 to 0.09 and the R^2 value for WSOC vs. OOA-2 (Figure 9d, now 10d) changes from 0.03 to 0.14. Also, the x-axis on Figure 9d (now 10d) has been fixed.

Table 2/3: These tables seem somewhat distracting in my view and I am not sure how important they are to the discussion in the paper. The authors should strongly consider incorporating discussion of those tables to a larger extent if they think they should be kept.

We have removed Table 2. However, since Table 3 includes the results from the two different fits used to perform the multilinear regression to determine the contribution of each AMS ME-2 factor to the WSOC, we have moved Table 3 to the supporting information (now Table S2).

-It would be useful to know exactly the times corresponding to when RH was increasing and decreasing for the plots shown in Figures 3/4/5. Some discussion about what other factors varying during those two periods of time would be helpful to show that the authors have considered all possibilities affecting their organic aerosol data and why their stated conclusions are the most obvious reason as to why different results exist.

We have added a table to the supporting information (Table S1) that now lists the exact dates and times used to create the times of RH increasing and decreasing in each Period. A copy is shown below. We also have expanded Table 1, which provides the concentrations of the various aerosol and gas-phase species measured in each period. The updated table is shown below. We also added some text in the overview section to point out that the only differences across all of Period A and the other periods are elevated OA, WSOC, and NO_x concentrations. Lines 225-228 now read as “Table 1 provides a comparison of the various concentrations and parameters observed during all four periods. With the exception of WSOC mentioned above, only the OA and NO_x (nitric oxides) concentrations across all of Period A are noticeably elevated compared to Periods B, C, and D.”

Table S1. Dates and times for the times of RH increasing and decreasing during Periods A, B, C, and D.

Period	RH Increasing	RH Decreasing
A	18 June at 20:00 – 19 June at 01:00, 19 June at 20:00 – 20 June at 06:00, 20 June at 21:00 – 21 June at 07:00	19 June at 02:00 – 19 June at 12:00, 20 June at 03:00 – 20 June at 11:00, 21 June at 00:00 – 21 June at 12:00
B	29 June at 19:00 – 30 June at 06:00, 30 June at 19:00 – 1 July at 06:00, 1 July at 21:00 – 2 July at 07:00	30 June at 04:00 – 1 July at 12:00, 1 July at 01:00 – 1 July at 08:00, 2 July at 02:00 – 2 July at 10:00
C	2 July at 21:00 – 3 July at 07:00, 3 July at 23:00 – 4 July at 06:00, 4 July at 20:00 – 5 July at 07:00	3 July at 03:00 – 3 July at 11:00, 4 July at 01:00 – 4 July at 12:00, 5 July at 05:00 – 5 July at 11:00
D	5 July at 19:00 – 6 July at 07:00, 6 July at 16:00 – 7 July at 03:00	6 July at 01:00 – 6 July at 15:00, 7 July at 07:00 – 7 July at 14:00

Table 1. Average concentrations of aerosol and gas-phase species along with various meteorological parameters observed during the times of RH increasing and decreasing during Periods A, B, C, and D. NA = not available

	OA ($\mu\text{g}/\text{m}^3$)	WSOC ($\mu\text{g}/\text{C}/\text{m}^3$)	Glycolate ($\mu\text{g}/\text{m}^3$)	Acetate ($\mu\text{g}/\text{m}^3$)	Formate ($\mu\text{g}/\text{m}^3$)	Chloride ($\mu\text{g}/\text{m}^3$)	Sulfate ($\mu\text{g}/\text{m}^3$)	Oxalate ($\mu\text{g}/\text{m}^3$)	Nitrate ($\mu\text{g}/\text{m}^3$)	Sodium ($\mu\text{g}/\text{m}^3$)	Ammonium ($\mu\text{g}/\text{m}^3$)	Potassium ($\mu\text{g}/\text{m}^3$)	Magnesium ($\mu\text{g}/\text{m}^3$)	Calcium ($\mu\text{g}/\text{m}^3$)	ALW ($\mu\text{g}/\text{m}^3$)
Period A RH Increasing	8.93	4.73	0.28	0.40	0.43	0.13	3.49	0.24	2.91	NA	NA	NA	NA	NA	6.81
Period A RH Decreasing	9.63	5.09	0.30	0.33	0.47	0.17	3.23	0.23	5.61	NA	NA	NA	NA	NA	7.29
Period B RH Increasing	4.06	2.87	0.22	0.24	0.24	0.09	3.22	0.12	1.67	0.01	1.04	0.43	0.10	0.37	4.21
Period B RH Decreasing	3.78	2.89	0.22	0.24	0.23	0.09	2.69	0.11	1.56	0.01	1.04	0.48	0.09	0.13	4.34
Period C RH Increasing	2.05	1.55	0.24	0.28	0.23	0.11	2.80	0.13	1.18	0.04	0.92	0.51	0.11	0.26	2.89
Period C RH Decreasing	2.01	1.54	0.22	0.32	0.23	0.10	2.75	0.12	1.28	0.04	0.94	0.54	0.09	0.06	2.64
Period D RH Increasing	2.89	1.92	0.17	0.18	0.21	0.11	3.38	0.12	1.31	0.02	1.07	0.48	0.10	0.32	4.10
Period D RH Decreasing	3.02	1.99	0.19	0.19	0.24	0.14	4.89	0.13	3.56	0.03	2.00	0.55	0.10	0.20	7.90

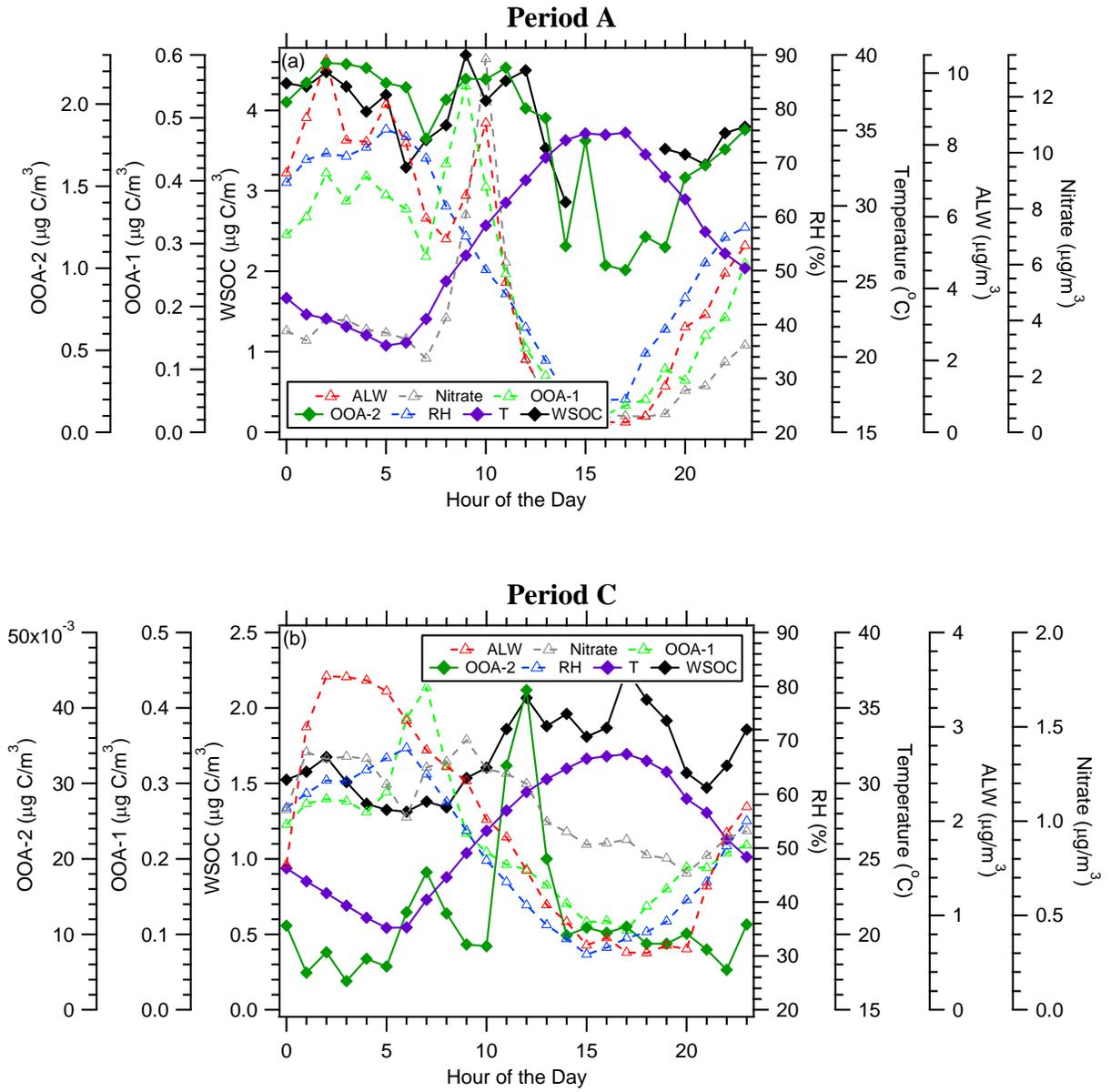
	Ozone ($\mu\text{g}/\text{m}^3$)	NO _x ($\mu\text{g}/\text{m}^3$)	SO ₂ (ppb)	Benzene ($\mu\text{g}/\text{m}^3$)	Toluene ($\mu\text{g}/\text{m}^3$)	Xylene ($\mu\text{g}/\text{m}^3$)	Glyoxal (ppb)	T (°C)	RH (%)
Period A RH Increasing	47.42	28.90	0.65	0.21	1.21	0.26	0.05	24.47	64.49
Period A RH Decreasing	63.70	17.75	1.14	0.27	1.78	0.34	0.09	26.09	57.66
Period B RH Increasing	76.6	10.94	0.68	0.19	0.83	0.53	0.06	26.74	60.87
Period B RH Decreasing	51.6	9.30	0.69	0.29	1.43	0.66	0.07	26.2	61.20
Period C RH Increasing	61.29	9.72	0.40	0.17	1.18	0.40	0.05	23.31	60.60
Period C RH Decreasing	75.40	8.08	0.51	0.17	1.11	0.44	0.07	25.02	53.88
Period D RH Increasing	87.21	8.93	0.30	0.12	0.52	0.23	0.05	25.63	63.45
Period D RH Decreasing	93.73	5.12	0.38	0.15	0.85	0.28	0.07	27.32	54.92

Reviewer #2

p. 35497 lines 3-19: This argument distinguishing between OOA-1 and OOA-2 needs to be clarified, and requires more data to be shown in support of the distinction. The authors state in line 12 that OOA-1 correlates with WSOC during increasing RH in period A, but not period B, as shown in Figure 9. This also appears to be the case for OOA-2. Furthermore, the authors state in line 5 that both OOA-1 and OOA-2 increase with RH and WSOC throughout period A, but do not show this data. They state in line 10 that OOA-1 drops in late morning when the RH declines (perhaps visible in Figure 8a?), and use this to argue for reversible aqueous formation of OOA-1. Then, the authors “illustrate” this, reversibility by appealing to increasing RH data. They state that OOA-1, but not OOA-2, correlates with RH during times of increasing RH during period A, but fail to show this data, either. These observations are then used to make inferences about the different natures of OOA-1 and OOA-2, but too little data distinguishing the two (beyond the O/C ratio) has been shown to make the argument convincing.

In order to better explain and illustrate the differences between OOA-1 and OOA-2 we have added diurnal profiles of WSOC, OOA-1, OOA-2, RH, temperature, ALW, and nitrate for Periods A and B (now Period C). This figure, shown below, helps to better show the relationship and timing of when OOA-1, OOA-2, RH, and ALW decrease. The discussion has also been updated and Lines 344-356 now read as “The multilinear regression analysis performed on the Period A measurements suggests that the largest water-soluble fractions are exhibited by OOA-1 and OOA-2, whose concentrations were observed to increase along with RH and WSOC for all the days in this period of the campaign. Due to the very different absolute average concentrations, the second factor (OOA-2) provided the largest contribution to WSOC, accounting for more than one third of the total water-soluble organic carbon concentration. Interestingly, the diurnal trend of OOA-1 indicated that its partitioning to the aerosol phase was largely reversible, and its concentrations declined steeply in the late morning hours when RH and ALW decreased (Fig. 12a). In the same hours of the day, the OOA-2 concentrations were largely unaffected by RH indicating (a) that OOA-2 mainly accounted for oxidized compounds stable in the aerosol phase and (b) that boundary layer growth is not the reason for the decrease in OOA-1 as this should have affected all factors. OOA-1 and OOA-2 can therefore be hypothesized as two aging stages of aqSOA formation during Period A.”

Figure 12



It would be appropriate to reference the paper by Jian Yu et al. (2005), who identified the correlation between aerosol oxalate and sulfate, either in the last paragraph of the introduction or with the Sorooshian reference at the bottom of p. 35494.

The *Yu et al.* [2005] paper has been added. Lines 127-130 now read as “We also look at the relationship of oxalate with sulfate and gas-phase glyoxal; oxalate and sulfate are both produced by cloud processing and glyoxal is a known precursor to aqSOA formation [*Yu et al.*, 2005; *Tan et al.*, 2009; *Ervens and Volkamer*, 2010; *Lim et al.*, 2010; *Sorooshian et al.*, 2010].” and Lines 286-288 as “Oxalate and sulfate are known tracers for aerosol formation through cloud processing [*Yu et al.*, 2005; *Sorooshian et al.*, 2010], although sulfate does also have a substantial, albeit slower, gas-phase formation pathway [*Seinfeld and Pandis*, 2006].”

This reviewer would like to see more connection or comparison made between the results of this study and the closely related work of El-Sayed (2015), briefly references in line 12 of p 35494.

A discussion about the *El-Sayed et al.* [2015] paper has been added. Lines 262-269 now read as “This also supports a recent study that observed ambient aqSOA formation during the nighttime as evident by the increased partitioning of gas-phase WSOC to the particle-phase with increasing RH [*El-Sayed et al.*, 2015]. The study by *El-Sayed et al.* [2015] found the increase in the fraction of total WSOC in the particle phase (F_p) at the two highest RH levels (70-80%, >80%) to be statistically significant compared to the F_p values at RH < 60%. The main focus of their work was to investigate if the uptake of gas-phase WSOC to aerosol water occurs through reversible or irreversible pathways. The data suggested the aqSOA was formed irreversibly. We investigate this with our data in section 3.3.2.”

Reviewer #3

1. Page 35488, line 16-21: Lee et al. (2012) experimentally demonstrated formation of aqueous SOA through photo-oxidation of real cloud water samples. It is recommended to include this reference here.

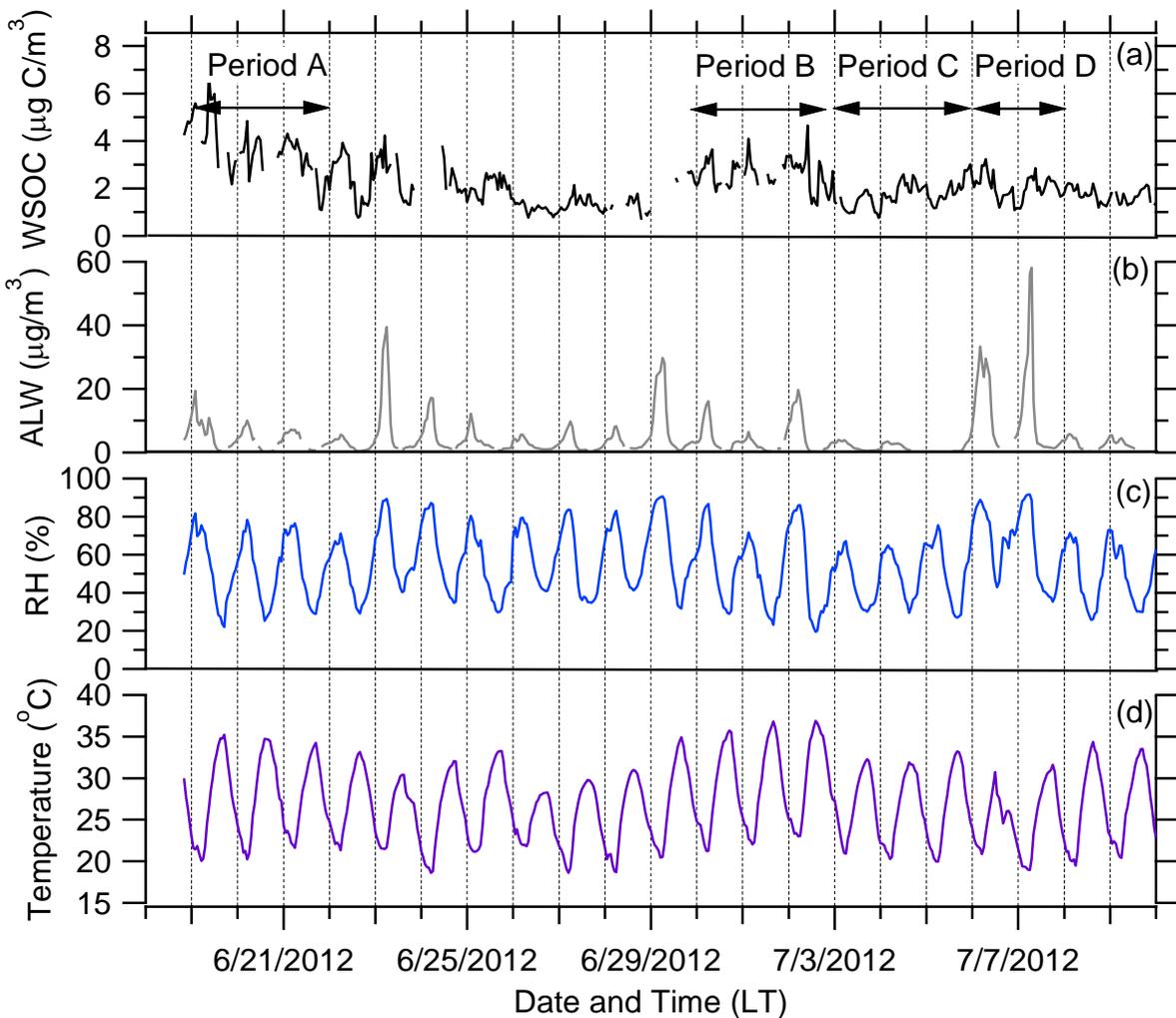
The *Lee et al.* [2012] reference has been added. Lines 99-103 now read as “These products can remain in the particle phase after water evaporation, forming what is termed aqueous secondary organic aerosol (aqSOA) (e.g. [*Blando et al.*, 2000; *Altieri et al.*, 2006; *Carlton et al.*, 2007; *de Haan et al.*, 2009; *Galloway et al.*, 2009; *Ervens and Volkamer*, 2010; *Sun et al.*, 2010; *Lee et al.*, 2012; *Monges et al.*, 2012; *Nguyen et al.*, 2012; *Tan et al.*, 2012; *Gaston et al.*, 2014]).”

2. Page 35492, line 17-25: The justification of using Period A and B to represent the first and second halves of the study, respectively, is unclear. Are the two selected periods defined based on meteorological conditions and/or aerosol chemical compositions? In particular, the mass loading of nitrate and ALW content in Period B are very different to those observed in the rest of second half. Furthermore, the authors mention that Periods A and B had similar air mass origins in general (line 20-22), which is somewhat contradict to the following sentence highlighting that Period A occurred during the end of a stagnation event and Period B represents typical background conditions influenced by regional transport (line 23-25).

The two periods were originally picked because they have similar transport patterns. In response to reviewer comments, we have now updated our analysis to include two more periods. The original Period B is now called Period C. The new Period B covers 30 June, 1-2 July and Period D covers 6-7 July. The new Period B has moderate ALW and Period D has the highest ALW observed during the study. We have also tried to clarify our meaning of similar air mass origins by instead talking about the flow direction. Lines 206-218 now read as “Therefore, our analysis will focus on comparing these two different halves of the study. Given our interest in examining for evidence of aqSOA we picked four periods with varying levels of WSOC and ALW. We also picked cases with both sites sampling similar air masses on a given day. Period A represents the first half of the study and covers 19-21 June. Period A has elevated WSOC and moderate ALW. As indicated by the difference in the length of the back trajectories [*Draxler and Rolph*, 2013; *Rolph*, 2013] shown in Fig. 2, Period A occurred during the end of a stagnation. Period B (30 June, 1-2 July), Period C (3-5 July), and Period D (6-7 July) represent three different cases in the second half of the study. Period B has moderate ALW, Period C has low ALW, and Period D has the highest ALW observed during the study. As indicated by Fig. 2, all three of these periods represent typical background conditions influenced by regional transport, but with slightly different flow patterns. The flows of Periods A and C are most similar. Due to changes in the WSOC concentrations and a non-consistent flow pattern on a daily basis, no periods between 23-29 June were examined.”

3. Page 35493, line 8-10 and Figure 2: Please add time series of ambient temperature and RH in Figure 2 for better illustration. Please also briefly explain how the RH variations can be used to diminish the influence of dilution and mixing. I wonder if the RH increasing period represents the period with a stable nocturnal layer.

Figure 2 (now Figure 3) has been updated to include temperature and RH. The new version of the figure is shown below. Also, our idea behind looking at the RH increasing vs. decreasing period is exactly as suggested, that the RH increasing period would represent a time with a stable nocturnal boundary layer. Lines 229-236 have been updated and read as “Each period will be examined in terms of the times when RH increased from 40 to 70% (times of RH increasing) and then when the RH decreased from 70 back to 40% (times of RH decreasing). This was done to try to diminish the influence of dilution and mixing on SOA concentrations and measurements of other key variables, since measurements of a conserved tracer were not available. The idea being that the times of RH increasing would represent a time with a stable nocturnal boundary layer. The switch in regimes on average occurs at 05:00 LT, but varied from 03:00 to 08:00 LT. Therefore, the times of RH increasing primarily occurred in the dark. “



4. Page 35494, second paragraph: It is recommended to discuss the potential formation mechanism of particle nitrate. In particular, NO_x concentration in Period A was higher than that observed in Period B. It is well known that NO_x can be lost at night to form N_2O_5 , which can

further react with water on aerosol surfaces to yield nitric acid. Increasing ambient RH may actually increase nitrate concentrations in particle-phase through N₂O₅ hydrolysis, that subsequently enhances ALW content for aqSOA production. In addition, it is possible that some nitrate can be formed in the residual layer at night and then convectively mixed after boundary layer break up, resulting in strong nitrate peaks (with relative low ALW probably because of the low RH after mixing) observed at around 9-11am.

The question of how the nitrate forms overnight is very interesting. We had a similar hypothesis that the elevated NO_x in Period A is contributing to the nitrate peak observed around midnight. Likely the second peak in nitrate observed in morning around 07:00 LT is due to mixing from aloft as the boundary layer breaks up. However, we prefer not to add any discussion to the text about this as we don't have any additional data to convincingly support these hypotheses.

5. Page 35495, line 6-17: I agree with the authors that oxalate is not a unique marker for aqSOA. As highlighted in the manuscript, previous laboratory studies have shown that photo-oxidation of glyoxal generates oligomers as major products through radical-radical reactions in aerosol water when OH radical concentrations is one the order of 10e-12M. However, oxalate can be largely produced in aerosol water at a lower OH radical concentration (10e-13M) likely due to insufficient organic radicals concentration for oligomers formation (Lee et al., 2011). Considering the uncertainty of OH radical concentrations in aerosol water, it is inappropriate to rule out the possibility of oxalate production in aqueous aerosol particles.

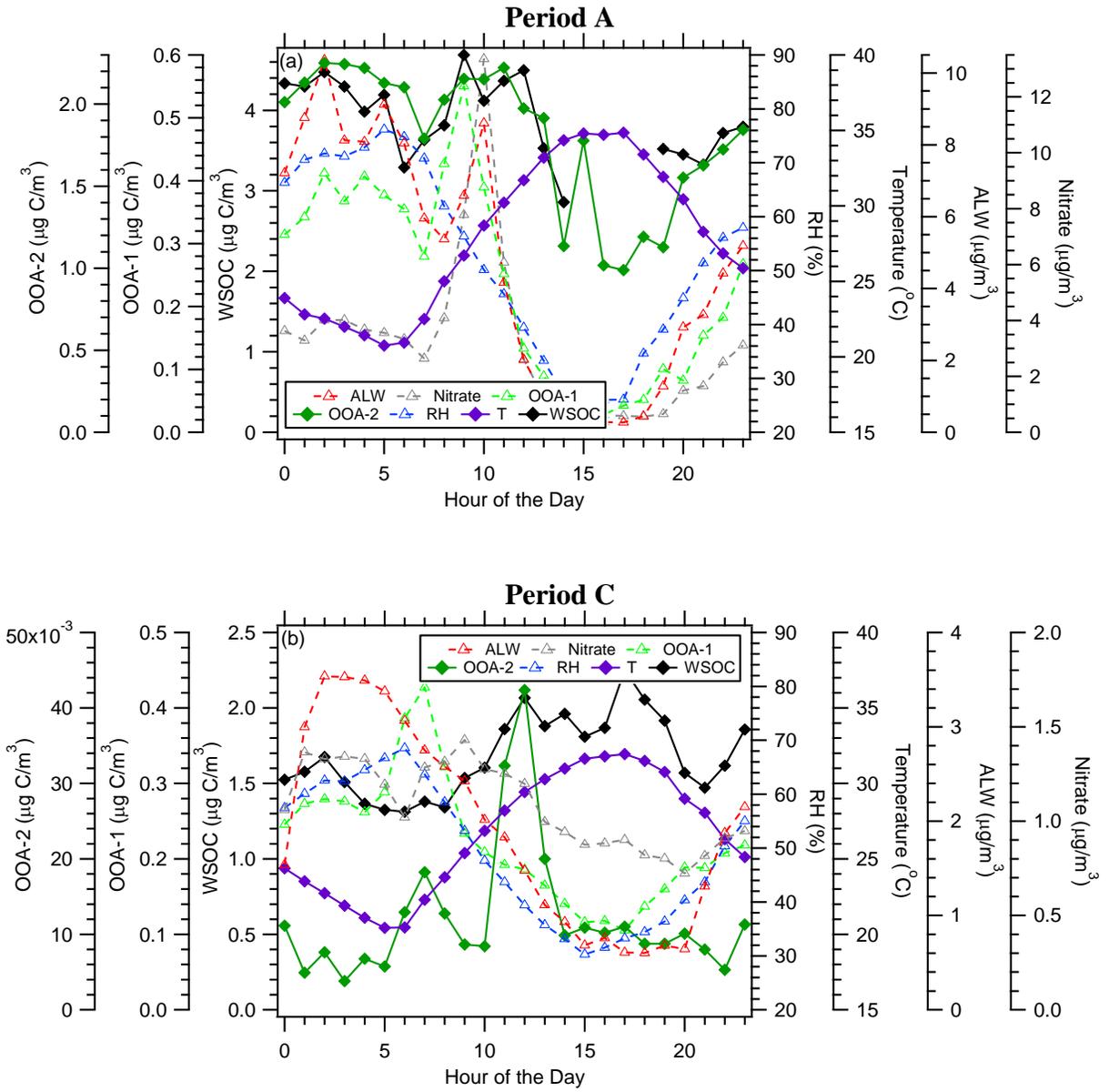
We have updated the discussion about oxalate to include the *Lee et al.* [2011] reference. We also now just mention that oxalate is not a universal marker for aqSOA, but do not go further to discuss it being a tracer for chemistry in clouds vs. wet aerosols. Lines 288-305 now read as “As shown in Fig. 8a and 8b for Periods A and C, during both the times of RH increasing and decreasing, there is a positive linear relationship between oxalate and sulfate (R^2 ranged from 0.39 to 0.68). The association between oxalate and sulfate but not oxalate and WSOC in Period A suggests that the local aqSOA formed in wet aerosols during Period A has little effect on oxalate. This result supports the supposition that oxalate is not a universal marker for aqSOA. This is further illustrated in our data by examining the correlation of oxalate vs. gas-phase glyoxal, a known precursor for aqSOA [*Tan et al.*, 2009; *Ervens and Volkamer*, 2010; *Lim et al.*, 2010], and ALW (Fig. 8c-f). Laboratory experiments suggest a relationship between oxalate and gas-phase glyoxal when there is in-cloud processing as oligomers have been proposed to be the dominant products from processing in aerosol water when hydroxyl radical concentrations are on the order of 10^{-12} M [*Lim et al.*, 2010; *Tan et al.*, 2010]. Oxalate could be produced in aerosol water at lower hydroxyl radical concentrations, such as 10^{-13} M, due to insufficient organic radical concentrations for oligomer formation [*Lee et al.*, 2011]. Although the hydroxyl radical concentrations are unknown, there is only a relationship between oxalate and gas-phase glyoxal for Period C during times of RH decreasing ($R^2 = 0.44$), which is when clouds were observed west of the site. In addition, there is no important relationship observed between oxalate and ALW for either period (all $R^2 < 0.17$).”

6. Page 35497, line 9-14: Please explain the connection between Fig. 9a (i.e., correlation of WSOC with OOA-1 during the times of RH increasing for only Period A) and the argument in

line 9-11 (i.e., OOA-1 reversibility and its concentrations declined steeply in the late morning hours when RH and ALW decreased). Similar to comment 3, It is difficult to follow the description here without a timer series of ambient RH and temperature in Figure 8.

In order to better explain and illustrate this we have added diurnal profiles of WSOC, OOA-1, OOA-2, RH, temperature, ALW, and nitrate for Periods A and C (Figures 12), which are shown below. These help to better show the relationship than our previous explanation, which has been removed. Lines 344-356 now read as “The multilinear regression analysis performed on the Period A measurements suggests that the largest water-soluble fractions are exhibited by OOA-1 and OOA-2, whose concentrations were observed to increase along with RH and WSOC for all the days in this period of the campaign. Due to the very different absolute average concentrations, the second factor (OOA-2) provided the largest contribution to WSOC, accounting for more than one third of the total water-soluble organic carbon concentration. Interestingly, the diurnal trend of OOA-1 indicated that its partitioning to the aerosol phase was largely reversible, and its concentrations declined steeply in the late morning hours when RH and ALW decreased (Fig. 12a). In the same hours of the day, the OOA-2 concentrations were largely unaffected by RH indicating (a) that OOA-2 mainly accounted for oxidized compounds stable in the aerosol phase and (b) that boundary layer growth is not the reason for the decrease in OOA-1 as this should have affected all factors. OOA-1 and OOA-2 can therefore be hypothesized as two aging stages of aqSOA formation during Period A.”

Figure 12



7. Page 35498, Section 3.4: It is recommended to add some discussion regarding OOA-1 and OOA-2 formation outside of Period A and B. In particular, significant amounts of OOA-1 and OOA-2 were produced during July 6-7, which are comparable to those observed in Period A. Therefore, the weak correlations between WSOC and nitrate (also OOA-1 and OOA-2) are probably due to the large contribution of background organic aerosol. In addition, the temporal variations of OOA-1 and OOA-2 looks very different. OOA-1 was formed and correlated well with nitrate throughout the whole sampling period. If OOA-1 and OOA-2 represent volatile/semi-volatile aqSOA and stable aqSOA respectively, can the authors comments on the atmospheric conditions that can produce OOA-1 but not OOA-2?

We have added discussion about the formation of OOA-1 and OOA-2 outside of Period B. This is done in conjunction with the addition of the analysis of two more Periods. The original Period B is now called Period C. The new Period B covers 30 June, 1-2 July and Period D covers 6-7 July. Diurnal profiles of WSOC, OOA-1, OOA-2, RH, temperature, ALW, and nitrate for these two new periods are included in the supporting information (Figure S10, shown below). Lines 357-363 now read as “Interestingly, some OOA-2 is also produced in Periods B and D. Although the concentrations levels of OOA-2 observed are similar between Periods A and D, OOA-2 concentrations are much more sustained across the day in Period A. In addition, as illustrated in the diurnal profiles for these periods (Fig. S10) the OOA-2 follows along more closely with OOA-1, RH, and ALW in Periods B and D, likely due to the differences in meteorology and/or chemistry of these periods compared to Period A. Regardless of these differences the observations all still point to the strong relationship between OOA-1, OOA-2, and ALW.”

We also added text to point out that the original Period B is an example of a case where OOA-1 is produced, but not OOA-2. Lines 370-373 now read as “Similar to Period A, here again the times when RH and ALW were high showed relatively high concentrations of OOA-1 (Fig. 12b), which represented an additional (though small compared to OOA-4) contribution to WSOC. Period C provides a case where significant OOA-1 is formed, but not OOA-2.”

Figure S10

