Response to Reviewer #2

General comments:

In this manuscript, the authors conducted flow reactor photooxidation experiments of toluene, and examined the SOA formation in the presence of initially wet or dry ammonium sulfate seeds. The approach followed that of Faust et al. (2017) to eliminate the effect of water on the gas-phase oxidation mechanisms, and allows experimenters to focus on the effect of aqueous vs dry seeds on gas-particle partitioning of organics. High resolution aerosol mass spectrometry was used to probe bulk composition, and various mechanisms were proposed to explain the changes in oxidation state, m/z fragment signal fractions and overall mass yields.

The results from this work are qualititatively consistent with those from Faust et al.

There are some questions regarding the oxidation mechanisms that lead to observed changes. My main criticism is that this work report mostly observational results, and lack any detailed mechanistic insights. The results are interesting, and can be explored more in order to provide useful information for both understanding the system and modeling the oxidation. I therefore suggest a few areas to look into, and the manuscript can be strengthened if the following questions are considered.

R: Many thanks for the suggestions. We emphasize that at moderate and atmospherically relevant RH, aerosol liquid water will exist and play an important role and it cannot be avoided, even for experiments started with dry AS seeds. The hygroscopic properties of AS and SOA naturally lead to the PRESENCE of water in particles in both dry and wet experiments. Our goal is not to provide mechanistic details of the reactions. Nevertheless, we appreciate the comments and suggestions of the reviewer to improve the manuscript.

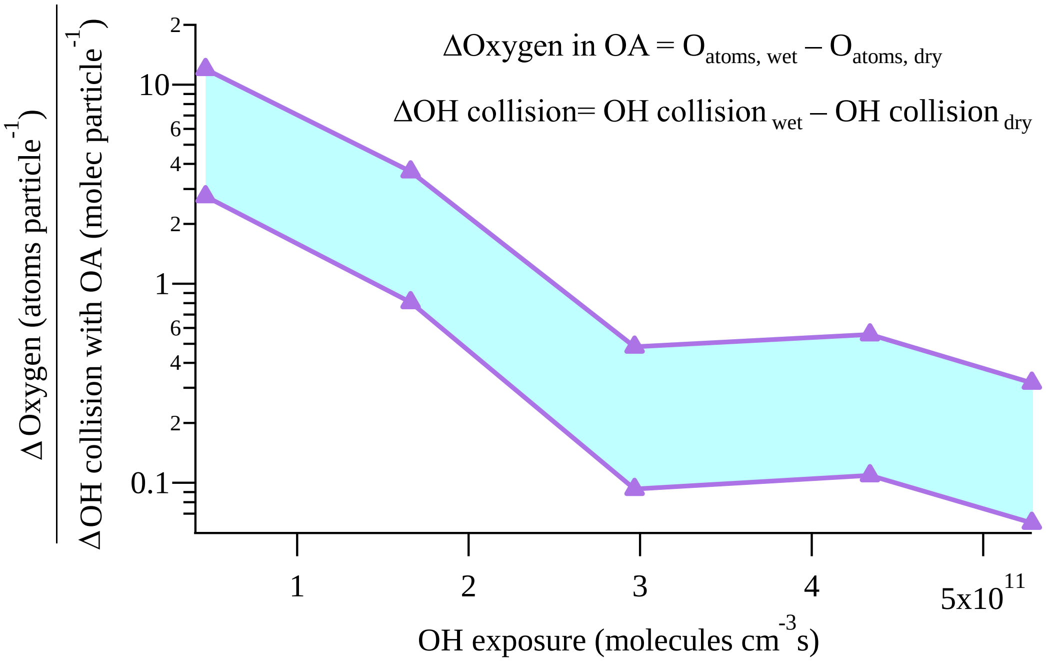
Major comments:

Q1: Oxidation mechanism: The major weakness in the paper is that it largely relies on bulk observation to probe mechanisms. While AMS is useful in obtaining bulk OA information and functional groups, the trends shown here are largely consistent with other aerosol aging observations in the lab or in the field. I am not sure if there are any novel insights in changes of m/z 43 and m/z 44, or the slope of the Van Krevelen diagram. It seems that the wet seed shows slightly different trends, but overall quite insignificant. The authors offer a few potential explanations (e.g. more OH aging, different gas-particle partitioning), but fail to go any deeper. If the explanation is more OH aging, then what is the equivalent change in OH exposure due to a wet seed (e.g. an OH exposure of 1 day at 65% RH is equivalent to an OH exposure of X days at 5%RH). Or, asking the question and framing the results in a different way: What is the increase in OH concentration in the aqueous phase that is required to explain the difference? Is this increase reasonable given the literature on aqueous OH production?

R1: AMS can provide insights to the overall evolution of OA and show the difference in bulk composition between initially dry and wet seeds without molecular level identification. The difference in SOA mass and composition between experiments with initially dry and wet seeds may be due to the enhanced gas-particle partitioning and/or enhanced OH aging in heterogeneous reactions. Since our experiments were conducted at 68%RH but not at 5%RH in both dry and wet cases, we cannot compare the results at different RH as proposed by the reviewer. Furthermore, though we cannot directly measure or calculate the OH concentration in the aqueous phase, we estimate the uptake of OH radicals to indirectly reflect the effects of enhanced OH aging on oxygen contents.

Specifically, we evaluate whether the enhanced uptake of OH radicals on initially wet AS seeds could explain the difference in oxygen contents, following the method of DeCarlo et al. (2008). We calculated R, the ratio of the difference in oxygen of OA between the initially wet and dry AS seed particles to the difference in the total number of OH collisions with OA at different OH exposures. To obtain R, the uptake coefficient (γ) of OH radicals was assumed to be 1 and 0.1/0.8 (lower/upper limit) for initially wet and dry AS seed particles, respectively (George and Abbatt, 2000). Note that as SOA formation takes place, the initially dry AS can become wet and the difference in γ between initially wet and dry seeds is reduced, especially at higher OH exposures. Without molecular level information on the organics, we assumed that each collision of OH with OA resulted in the addition of one oxygen atom to SOA. A value of R smaller than unity qualitatively indicates that the uptake of OH radicals can potentially explain the differences in oxygen contents in the dry and wet experiments.

The following figure (now Figure S6) shows that R is larger than unity at low OH exposures and smaller than unity at high OH exposures. This analysis suggests that the enhanced OH uptake may contribute to the difference in oxygen contents between dry and wet cases at higher OH exposures. At low OH exposures, the enhanced gas-particle partitioning may dominate the difference.



The following sentences have been added to the revised manuscript.

“We evaluate whether enhanced uptake of OH radicals on initially wet AS seeds could explain the difference in oxygen contents, following the method of DeCarlo et al. (2008). We calculated R, the ratio of the difference in oxygen of OA between the initially wet and dry AS seed particles to the difference in the total number of OH collisions with OA at different OH exposures. To obtain R, the uptake coefficient (γ) of OH radicals was assumed to be 1 and 0.1/0.8 (lower/upper limit) for initially wet and dry AS seed particles, respectively (George and Abbatt, 2000). Note that as SOA formation takes place, the initially dry AS can become wet and the difference in γ between initially wet and dry seeds is reduced, especially at higher OH exposures. We also assumed that each collision of OH with OA resulted in the addition of one oxygen atom to SOA. A value of R smaller than unity qualitatively indicates that the uptake of OH radicals can potentially explain the differences in oxygen contents in the dry and wet experiments. Fig. S6 shows that R is larger than unity at low OH exposures and smaller than unity at high OH exposures. This analysis suggests that the enhanced OH uptake may contribute to the difference in oxygen contents between dry and wet cases at higher OH exposures. At low OH exposures, the enhanced gas-particle partitioning may dominate the difference." (Line 398-414).

Q2: If the enhanced partitioning is due to availability of ALW, one can potentially explain the difference using Henry's law constants. What would the Henry's Law constants of the oxidation products need to be in order to show the difference in SOA yields between wet and dry seeds?

R2: The hydrophilic products should partition more readily into initially wet AS seeds than dry seeds and partially account for the difference in SOA yields. Both ALW and the Henry's law constant are relevant. Instead of focusing on the Henry's law constant alone, we focus on the uptake of glyoxal, a gas phase oxidation product of toluene oxidation, to illustrate the effects of enhanced partitioning of oxidation products on SOA yields. The following text has been added to the revised manuscript to estimate the effect of enhanced partitioning on SOA yields.

"The hydrophilic products should partition more readily into initially wet AS seeds than dry seeds and partially account for the difference in SOA yields. For example, as one of the important oxidation products, glyoxal was estimated to have an effective Henry's law constant of 4.52×10^{8} m atm^{-1} for our initially wet AS seeds due to the "salting-in" effect (Kampf et al., 2013), approximately 3 orders of magnitude higher than that in pure water (Ip et al., 2009). The uptake rate constant of glyoxal can be calculated as $(\gamma v\text{A})/4$, where γ is the uptake coefficient, v is the gas-phase velocity of glyoxal, and A is the total surface area of AS seeds. The uptake rate constant is 4.5×10^{-4} s^{-1} for initially wet seeds with $\gamma = 2.4\times10^{-3}$ estimated from glyoxal uptake in AS seeds at 68% RH (Liggio et al., 2005b). The average gas-phase glyoxal concentration was modeled to be 4.3 ppb at OH exposure of 0.47×10^{11} molecules cm^{-3} s using the Master

Chemical Mechanism v 3.3.1 (Jenkin et al., 2003; Bloss et al., 2005), which would result in approximately 1.6 μg m^{-3} of glyoxal in particle phase for initially wet AS seeds. If the particle-phase concentration of glyoxal was assumed to be 0 for initially dry AS seeds, the enhanced partitioning of glyoxal alone would account for 24.5% of the mass difference of SOA. Note that other hydrophilic products were not included in this calculation. This analysis suggests that the enhanced partitioning of hydrophilic products may play an important role in the difference of SOA yields at low OH exposures. As discussed above, the initially dry AS seeds approached wet seeds and reduce the differences between wet and dry SOA yields at high OH exposures. " (Line 317-336).

Q3: The approach used to calculate ALW separately for AS and toluene assume that they are linearly additive. In a metastable solution of ammonium sulfate, the ionic strength would be very high, and can affect the water solubility of the organics. Would it be valid to assume then the water uptake that cannot be explained by literature kappa of toluene SOA is associated with AS?

R3: The approach used in this study has been found to be adequate to well estimate the hygroscopic growth of inorganic and organic mixtures in laboratory and ambient studies, even at relatively low RH (Choi and Chan, 2002; Cheung et al., 2015; Svenningsson et al., 2006; Nguyen et al., 2016).

Q4: Experimental uncertainties: I am not sure if the experimental uncertainty in yields include only that from quantification of aerosol. The quantification of toluene reacted would play an important role as well, especially since the reported yields (with only uncertainty of SOA) have almost negligible uncertainty. In this work, toluene is not quantified, but the amount reacted is based on estimation of OH exposure. Other work in the literature quantifying yields measure the hydrocarbon precursor directly (using either PTRMS or GC methods). Offline quantification of OH exposure using SO2 and assuming light conditions are constant can be problematic. There needs to be a systematic investigation of the uncertainties, particularly that associated with quantifying hydrocarbon decay.

R4: The experimental setup follows Wong et al. (2015) and Faust et al. (2017). The flow conditions were exactly the same for initially wet and dry seeds. The calibration and toluene photooxidation experiments were conducted over three days. The light condition was not expected to change in such a short period. Furthermore, we focus on the relative SOA yields, which are not expected to be much affected by the uncertainties in toluene quantification since the initial concentrations of toluene and OH exposures were the same for both cases. In offline calibration of OH exposures, the addition of toluene would perturb the calculated OH exposures. The reduction in OH exposure due to the toluene + OH reaction was estimated to range from 15% at the highest OH exposure to 25% at the lowest OH exposure, using the method of Peng et al. (2016). Nevertheless, we assume that this reduction is the same for dry and wet seeds and will not influence the relative SOA yields.

For clarification, the sentence "*The addition of toluene may reduce the OH exposure.*" has been revised and now reads:

"The reduction in OH exposure due to the addition of toluene was estimated to range from 15% at the highest OH exposure to 25% at the lowest OH exposure, using the method of Peng et al. (2016)." (Line 157-160).

The following text was also added for clarification.

"The flow and light conditions were the same for initially wet and dry seeds. Therefore, the quantification of toluene would not introduce uncertainties to the relative SOA yields described in Section 3.1 as the initial concentrations of toluene and OH exposures were the same for both cases." (Line 174-177).

Minor comments:

Q5: Abstract: Should be less focused on specific details of the experiments. I suggest taking some of the numbers out (unless it is a really important number that, for example, a modeler can use or another experimentalist can directly compare). Rather, there may be broader implications other than these results warrant further study. What are the detailed mechanistic insights? What further developments are needed to better understand water uptake? Just a few sentences would suffice.

R5: Some parts of the abstract have been rewritten as suggested.

"*At an OH exposure of 4.66×10^{10} molecules cm^{-3} s, the ratio of the SOA yield on wet AS seeds to that on dry AS seeds was 1.31±0.02. However, this ratio decreased to 1.01±0.01 at an OH exposure of 5.28×10^{11} molecules cm^{-3} s. The decrease in the ratio of SOA yields as the increase of OH exposure may be due to the early deliquescence of initially dry AS seeds after coated by highly oxidized toluene-derived SOA.*" now reads "The ratio of the SOA yield on wet AS seeds to that on dry AS seeds, the relative SOA yield, decreased from 1.31±0.02 at an OH exposure of 4.66×10^{10} molecules cm^{-3} s to 1.01±0.01 at an OH exposure of 5.28×10^{11} molecules cm^{-3} s. This decrease may be due to the early deliquescence of initially dry AS seeds after coated by highly oxidized toluene-derived SOA." (Line 27-33).

"*Our results suggest that AS dry seeds soon turn to at least partially deliquesced particles during SOA formation and more studies on the interplay of SOA formation and ALW are warranted.*" now reads:

"Our results suggest that inorganic dry seeds become at least partially deliquesced particles during SOA formation and hence ALW is inevitably involved in the SOA formation at moderate RH. More laboratory experiments conducted with a wide variety of SOA precursors and inorganic seeds under different NO_x and RH conditions are warranted." (Line 44-48).

Q6: Line 36: m/z 29, 43, 44 are specific to the aerosol mass spectrometer (AMS).

R6: "obtained using an aerosol mass spectrometer (AMS)" was added. (Line 40-41).

Q7: Line 47-48: these references might not quite reflect the current state of knowledge. For example, Tsigaridis et al. (ACP, 2014) did a model intercomparison and found that the secondary nature of OA can be reproduced, but not the overall amount. Shrivastava et al (ACP, 2011) shows that the total SOA amount can be reproduced, but require some model tuning. Overall, there may be too many studies to cite for a simple argument here, but the de Gouw et al. and Volkamer et al. studies are quite out of date at this point.

R7: SOA models usually include update of the volatility basis set (VBS) formalism to treat gas-particle partitioning and multi-generation oxidation (Shrivastava et al., 2011; Tsigaridis et al., 2014), increased SOA yields that account for vapor wall loss in smog chambers (Zhang et al., 2014; Hayes et al., 2015) and additional SOA precursors such as S/IVOCs (Robinson et al., 2007). These updated models can better reduce the gap between the modeled and observed SOA, but have resulted in over-prediction of SOA at long aging times. It remains unclear whether these updated models improve the simulation of SOA for the right reasons. Here, we pointed out that the presence of ALW may influence the SOA yields, which is not well treated even in the updated models. To reflect the current state of knowledge, we added the following sentence to the revised manuscript.

"The updated models incorporating the volatility basis set (VBS) formalism (Donahue et al., 2006) can better predict the observed SOA, but SOA formation still remains under-constrained (Shrivastava et al., 2011; Tsigaridis et al., 2014; Hayes et al., 2015; Ma et al., 2017)." (Line 57-60).

Q8: Line 60: I would also add that understanding water uptake of SOA is important for estimating its loss by wet deposition, which is highly unknown at this point.

R8: The following sentence has been added as suggested.

"In addition, understanding water uptake of SOA is important for estimating its loss by wet deposition, which is not well constrained." (Line 72-74).

Q9: Line 97: is the silica gel diffusion dryer manufactured by TSI? If so, consider including the model number. Is the 30% outlet RH experimentally verified? I would imagine the outlet RH would be a function of the inlet RH.

R9: It is a homemade one. The outlet RH was verified to be lower than 30%. The inlet RH should be stable during the experiment as the flow rate was stable.

Q10: Line 103: Is it correct that the ALW is estimated using the method described later? If so, please mention.

R10: Yes. "see Section 2.4" was added for clarification. (Line 119).

Q11: Lines 140-146: Based on the OH exposure calculated, what is the amount of toluene consumed?

R11: The reacted amount of toluene is now provided in Table 1. The following sentence has been added:

"The reacted and final concentrations of toluene were calculated from the OH exposure and the rate constant of the reaction between toluene and OH (Atkinson and

Arey, 2003) (Table 1).” (Line 172-174).

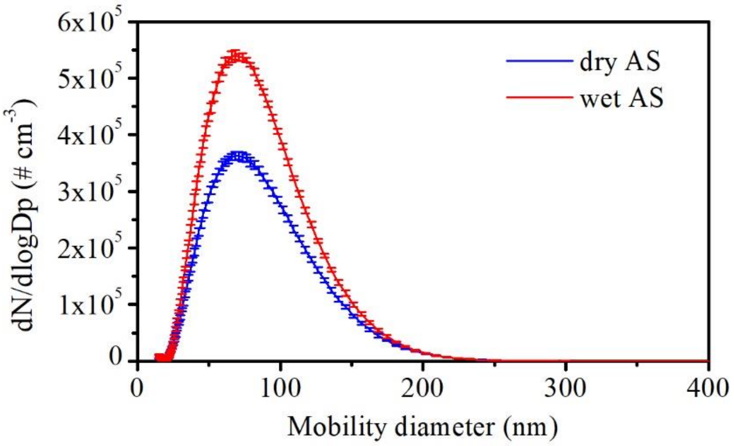
Q12: Line 166-167: is this filtered air flow with or without toluene and/or oxidation products?

R12: The filtered air flow was without oxidation products. These oxidation products were expected to have a negligible influence on the concentrations of major gases, e.g. N_2, O_2, and CO_2.

Q13: Line 177: Just to make sure that sulfate is indeed coated with SOA, it would be great to show size distributions: Either a unimodal SMPS number size distribution showing no nucleation, or the PTOF on the AMS showing that organics and sulfate are in the same mode.

R13: As shown in the following figure (now Fig. S2), at the OH exposure of 0.47×10^{11} molecules cm^{-3} s, the particle number distributions for both cases are unimodal, indicating no nucleation. The following sentence was added to the revised manuscript.

“The unimodal size distributions of particle numbers show the SOA formation on AS seed particles without much nucleation mode particles (Fig. S2)” (Line 198-200)



Q14: Line 187: Is it really evaporation of the organics? Would 8% mass loss due to evaporation be enough to show up in shifts in particle mode or median diameter (it would be around a ~2% change in diameter)?

R14: We cannot rule out other possibilities based on the dataset. Hence we said that it is possibly due to reversible partitioning of the SVOCs in the original text. The ~2% shift in particle mode diameter was not enough to be captured by AMS or SMPS.

Q15: Line 252: I suggest keeping all OH exposure numbers to 10ˆ11 molec cm-3 s. For example, I recommend changing 4.66 x 10ˆ10 to 0.47 x 10ˆ11.

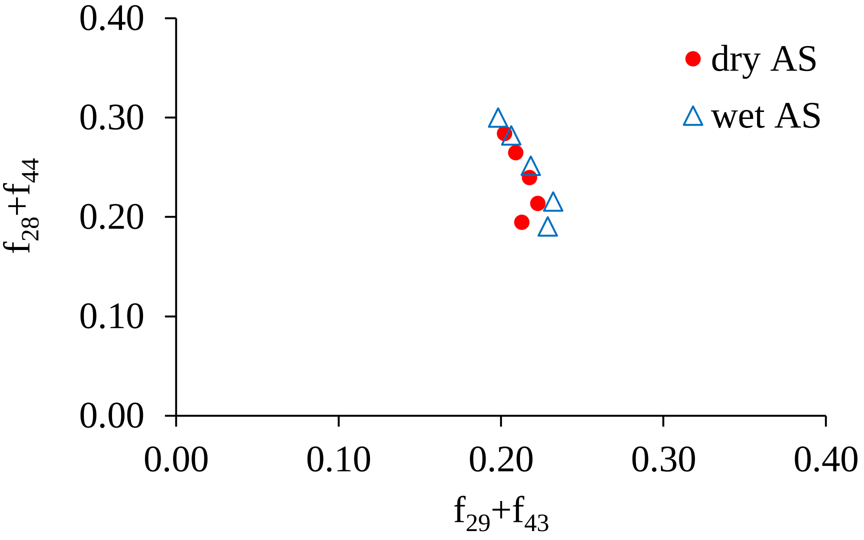
R15: Revised as suggested.

Q16: Line 290 – 292: The authors should be cautious about observations of oligomers on the AMS. The relatively low signal above m/z 80 does not mean there is little to no oligomerization. The AMS vaporizer at 600 C causes extensive thermal decomposition.

R16: Although the AMS can cause extensive thermal decomposition, previous studies suggest that m/z > 80 can be easily observed when the oligomers are abundant (Price et al., 2014; Gilardoni et al., 2016; Faust et al., 2017). We deleted this statement due to the lack of solid evidences.

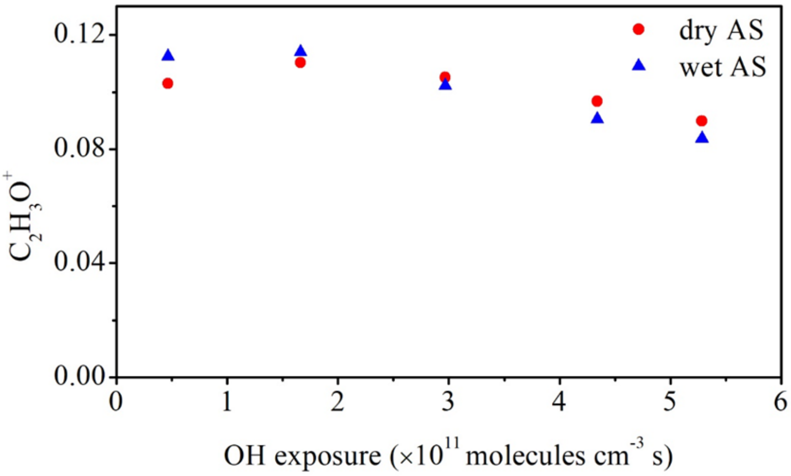
Q17: Line 298-303: Would a plot f(28+44) vs f(29+43) look different from f44 vs f43? The conversion of alcohols and aldehydes to acids described in the previous paragraph is fundamentally driving the trend of f44 and f43. So these two paragraphs are both conveying a similar observation, which is expected given the extensive literature on AMS description of aerosol aging. So I suggest condensing the discussion.

R17: As shown in the following figure, the f(28+44) vs f(29+43) plot looks similar as f44 vs f43 plot. The previous paragraph is focused on the comparison of the mass spectra between the lowest and highest OH exposure while this paragraph shows the overall evolution of f44 vs f43 from the lowest to the highest OH exposure, so we would like to keep the original discussions.



Q18: Line 314-315, and Fig. S4: The differences seem very minute to me and may not be statistically significant. The y-scale is misleading, and should start at zero.

R18: Fig. S4 has been revised accordingly as follows. The difference in the abundance of $C_2H_3O^+$ between dry and wet AS seeds is small but there is quite an obvious trend as OH exposure increases.



Q19: Line 314-315: If the trend is due to enhanced partitioning of water soluble organics to ALW, wouldn't the

R19: This issue of solubility of WSOC is likely addressed in response to Q1 and Q2.

Q20: Line 327: It looks like the subscript "C" in OSC is not capitalized, where it has been capitalized in other instances in the manuscript.

R20: Revised.

References:

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