Referee's Comment in Italic Font; Author's response in Red and Manuscript revision in Blue without Italic Font.

The manuscript presents information on yields for the photooxidation of single ringed aromatic structures. The senior author has been measuring yields from aromatic hydrocarbons (AHCs) for more than 15 years and is well versed. In this particular manuscript, the structure of the aromatic hydrocarbon (AHC) has been varied to examine differences in the organic aerosol (OA) yield. Thus, a series of 12 alkyl-substituted C8 and C9 AHCs have been examined. For these experiment, the aerosol yield has been determined using the Odum two-product model. Other OA parameters examined include the ratio of aerosol mass spectrometry (AMS) peaks attributed to OA, the OAoxygen-to-carbon (O: C) ratio, the oxidation state (OS), density, and volatility. The authors conclude that changes in the OA chemical composition and volatility influences the yield typically by increasing the mass for increased oxidation. The authors also consider the atmospheric implications of this study.

(1)The study addresses an issue of perhaps abstruse importance. The oxidation of alkyl substituted AHCs and the formation of secondary organic aerosol (SOA) has been examined extensively over the last 20 years and this work appears to cover some old territory. Many of these topics were addressed in Odum et al. 1997a, b (authors' references) and the present manuscript provides a bit more insight.

Odum's work was very important and provided a practical way to simplify aerosol yields. However, work over last decade has suggested the importance of NO_x to SOA formation from aromatic hydrocarbons (e.g., Song, et al., 2005) with increasing aerosol formation observed for aromatics as initial NO levels are decreased. Lowering NO_x conditions from the earlier Odum work improves representation of ambient conditions. Therefore, a comprehensive reinvestigation including isomer effects on SOA formation at more realistic hydrocarbon and NOx conditions is needed. Further, as noted by the reviewer, additional instrumentation available provides more insights into the SOA formation.

Higher yields are observed in this work for low NO_x conditions than the earlier high NO_x conditions (e.g Odum, et al, 1997, Figure 1, M_0 =40, yield= ~0.03 or ~0.06; current work M_0 =40, yield 0.07-0.12). This paper demonstrates the molecular structure impact of aromatic hydrocarbons on SOA formation including impacts on SOA yield, chemical composition and physical composition. This is the first comprehensive analysis of SOA formation from aromatic isomers since the original Odum work on SOA from aromatics. The previous Odum work provides only very limited experimental work on isomers which is insufficient to determine molecular impact on SOA formation. (e..g, p-xylene and o-xylenehave only two experiments)

(2) Admittedly, the AMS was not around and the aerosol density from the volume distribution was considered to be unity in the 1990s. However, this work also represents a step backwards. Whereas Odum et al. 1996, 1997a sought to simplify aerosol yields, this work goes in the other

direction and makes an argument (at least implicitly) that the yields should be addressed more precisely, a contention that I don't feel has been justified (see Table 2).

This work provides yield information with greater precision to dig into the role of molecular structure in SOA formation from aromatic hydrocarbon. We can't agree more with the reviewer that a simplified curve is a more attractive method for the curve fitting. Prior to further simplification, it is necessary to identify the relative importance of aromatic structure (o, m, p; alkyl length) especially when looking to project these findings for additional aromatic isomers. We currently have a paper under review to further demonstrate a novel method to simplify SOA yield from aromatic hydrocarbons, which requires insight on the relative importance of aromatic molecular structure. This work here focuses on molecular structure impact before stepping largely forward to the general trends found in SOA formation from aromatic hydrocarbons. The limited data sets available in earlier years are insufficient to reveal the difference among isomers and therefore might provide some bias on the similarity among aromatic hydrocarbon SOA formation, especially when conducted under high NO_x conditions. As demonstrated in the manuscript, the difference among SOA from aromatic isomers, including SOA yield (for example, para position has significantly lower SOA yield compared with ortha and metal position), chemical composition and physical composition, does exist and should not be ignored by oversimplification. Therefore, this work is valuable to understand SOA formation from aromatic hydrocarbons before generalizing SOA aromatic yield trends.

(3)That said, the experiments appear to have been carefully performed and there certainly are enough of them. Unfortunately, the initial conditions are all over the map and makes it very difficult to get a sense of the reproducibility of a given experiment. Virtually, nothing is said about uncertainty.

There is only $\sim 10\%$ of experiments with HC/NO larger than 60 ppbC:ppb. The majority of experiments have similar NO conditions (see reply to referee #1 in Comment 4). Also, the similar impact of NO on radical and organic nitrate formation is demonstrated to be insignificant (see reply to referee #1 in Comment 4. The uncertainty of experiment is 6.6% based on ten repeat m-xylene experiments (please see referee #1 in Comment 3). Uncertainty in all analysis is included in the updated manuscript.

1. My major substantive comments regarding the manuscript are as follows: (1) these experiments hardly qualify as being under low NOx conditions. The removal of RO2 radicals competitively by NO or RO2 determines the regime that the reaction is in. I would consider the low NOx regime as conditions where the RO2 radical-radical reactions become more important than the NO reaction. As a rule of thumb, I would say that this is certainly at no more than 5 ppb of NO for the conditions of these experiments depending, of course, on the specific RO2 radicals from the precursor AHC.

(1) From a kinetics perspective, low NO_x is even lower than the 5 ppb (suggested by the reviewer) tending to occur at NO levels in the 15 to 50 ppt range. However, starting with NO_x levels < 50 ppt is substantially lower than practical experimental constraints for Teflon

environmental chambers due to offgasing of HONO from Teflon surfaces (Carter et al., 2005). These experiments are referred to as low NO_x experiments to be consistent with environmental chamber literature over the last decade, which is referring to the relative amount of NO_x at the beginning of the experiment when compared to initial VOC. The NO_x ranges in this work are more consistent with urban NO_x loadings than the earlier high NO_x experiments performed by Odum (Odum, et al., 1997 and 1996) and others. Also, it is not possible to use an initial NO concentration ~5ppb since the low NO_x concentration leads to a low reactivity of overall reaction and therefore forms less aerosol which is not atmospherically relevant.

Clearly the NO will compete for the HO₂ or RO₂ when there is sufficient NO. In other earlier work we demonstrate that SOA will not be formed until NO₂/NO>70 (Li, et al., 2015), which indicates that RO₂ majorly react with NO instead of HO₂ or RO₂ even at NO~5ppb under the range of NO we investigated. The important point is that the NO concentration is extremely low (<10ppt) during the majority of the photooxidation experiment (after onset of O₃ formation). NO_x mainly exists as NO₂ when PM is formed and HO₂+RO₂ instead of NO+ HO₂/RO₂ dominates.

In order to clarify the NO_x condition we actually used, we add the following information on Page 5 at Line 29 after "under low NO_x" add "(10-138 ppb)".

(2)The version of SAPRC referenced to Carter and Heo (2013) is specifically geared to ozone prediction, that is, conditions where NOx dominates early product generations (two, at most three). As stated in Carter and Heo, (2013; Atmospheric Environment) SAPRC-11 is not geared for PM predictions. For this to be the case, the importance of RO2 + RO2 reactions should be adequately predicted as should the SOA mass. That is the point of the model and to predict radical concentrations as they were an end in themselves. The experimental SOA values could then constrain the model. That said, I question how well SAPRC-11 predicts radical concentrations under low NOx conditions. (3) Several sections need a complete writing overhaul. Section 3.2.1 is barely comprehensible. Many sections in the Results and Discussion presents data (e.g., S6) as if they were self-interpretive. The manuscript is written for the audience being other scientists in their research group. There is considerable jargon and the writing is highly imprecise hardly worthy of a scholarly journal. (4) To the extent possible, the authors should give a greater physical interpretation of the metrics they present in Section 3. Some are obvious (e.g., Sec. 3.2.2) others far less so (Sec. 3.2.1; 3.4).

2) SAPRC-11 is geared to predict O_3 formation under low NO_x conditions especially for aromatic hydrocarbons as described by Carter and Heo (2013). Literally, the NO_x range we used in current work is within the used range of NO_x when the model is updated to SAPRC-11. In fact, the aromatic experiments used to develop the SAPRC-11 update are included in this work. We agree that it could not well predict SOA formation since the gas phase products are not well demonstrated as suggest by Carter and Heo (2013), especially for those associated with gas to particle partitioning. However, SAPRC-11 should be sufficiently good to predict gas phase radical concentration, which is closely associated with ozone formation. 3) Section 3.2.1 extends the traditional f_{44} vs f_{43} (C₂H₃O⁺) chemical composition analysis by including fragments (C₃H₅O⁺ m/z 57 and C₄H₇O⁺ m/z 71) from longer alkyl substitute other than methyl since longer alkyl substitutes are included in the isomers investigated. The goal of Section 3.2.1 is to provide insights into the SOA formation mechanism from different isomers as discussed in the later part of Section 3.2.1.

Some changes are already made in Section 3.2.1 (please see reply to referee #1, Comment 8). Also, we have added the following sentences on Page 11 at Line 3 to Section 3.2.1.

This work extends the traditional f_{44} vs f_{43} (C₂H₃O⁺) chemical composition analysis by including oxidized fragments (C₃H₅O⁺ m/z 57 and C₄H₇O⁺ m/z 71) of the longer (non-methyl) alkyl substitutes. Therefore, f_{44} vs f_{43} + f_{57} + f_{71} is plotted instead of f_{44} vs f_{43} .

We demonstrate the calculation of H/C and O/C in section 2.2 Page 7 Line 16-19. We also described how the Figure S6 graph is made in the title of Figure S6. We add following sentences in the revised manuscript to better interpret Fig. S6 and other graphs (e.g. Fig. S3).

Further, we have added the following sentence on Page 7 Line 19:

Evolution of SOA composition (Heald, et al., 2010; Jimenez, et al., 2009) refers to the bulk SOA chemical composition changes with time. f_{44} and $f_{43+57+71}$ evolution and H/C and O/C evolution refer to the change of f_{44} and $f_{43+57+71}$ with time and the change of H/C and O/C with time, respectively.

Additionally, to address jargon concerns, we have had a couple of non-SOA focused experts in the air field review the paper to help identify and remove jargon along with the suggestions provided by the review.

4) The following revisions are made in Section 3.2.1 and Section 4 to improve the physical interpretation of the metrics used.

Sec. 3.2.1: The physical interpretation is improved after the revision described in 3) comment and the referee #1's Comment 8. Sec. 4: The physical interpretation stated at the beginning of Section 4 as "Methyl dilution theory (Li, et al. 2015a) is extended to alkyl substitute dilution theory in order to investigate the influence of longer alkyl substitutes compared with methyl group substitutes." Additionally, the following sentence has been added to clarify the physical interpretation at Line 19 Page 17:

A robust prediction of SOA H/C and O/C trends for longer (C2+) alkyl substituted aromatics based on the methyl substituted aromatics will suggest a similarity in the role of methyl and longer alkyl to SOA formation; an underestimation or overestimation will indicate different oxidation pathways for aromatics with differing alkyl substitute length.

We also update the Fig 7 a & b according to referee #1's Comment 3 about standard deviation.

The implication from the difference between the measurement and prediction from the aromatics is updated correspondingly in the later part of Sect 4.

Some comments and suggestions:

2. *P5*, *L27*. *The goal of the research states the obvious. Perhaps more insight will motivate the reader to actually read the paper.*

We add the following sentence on Page 6 Line 2:

The effects of molecular structure impact on SOA yield, chemical composition (H/C, O/C, OS_c , f_{44} , f_{43} , f_{57} and f_{71}) and physical properties (density and VFR) are demonstrated. Alkyl substitute dilution conjecture is further developed from methyl dilution theory (Li, et al., 2016).

3. P6, L7. UV-350 bulbs have considerable radiation in the UVB which accentuates the photolysis of carbonyl compounds to a considerable extent which accelerates PM formation by increasing the radical concentrations. Thus, the two-product parameters developed (Table 2) may not be applicable for predictions of ambient AHC PM (Tables2 and S3)

We agree that UV-350 bulbs do not provide the higher wavelength region which affects photolysis of certain carbonyl compounds. However, UV impacts on different carbonyl are different. The photolysis rate ratios with blacklights will be much lower in the chamber than in the atmosphere if carbonyls have action spectra similar to the α -dicarbonyls; however, blacklight photolysis rate ratios will be higher if carbonyls have action spectra more like that of acrolein (Carter, et al., 1995). The photolysis of carbonyl compounds are more likely to impact the radical concentration (e.g. OH) and may further impact the overall SOA formation by change the kinetic reactivity. Therefore, the light source impact on carbonyl photolysis turns out to be the influence of radical concentration on SOA formation. The difference in radical concentration between chamber and atmosphere is demonstrated in Li, et al., 2015.

Further work is needed to adjust the SOA yield concluded from current chamber studies to better predict the SOA formation under atmospheric conditions. The current work provides the fundamental data for further investigation. Therefore, we add to the paper the statement "Moreover, the five subcategories of aromatics and their two product modeling curve fitting parameters in this work at more realistic NOx loadings provide a more precise prediction of SOA formation form aromatic hydrocarbons under atmospheric conditions" in Section 5 for the atmospheric application as in referee #1 Comment 13. The current study is more focused on the isomer impact on SOA formation. The results and implications of the current study remain reasonable since all the precursors are studied under comparable conditions (see k_{OH} discussion in the reply to referee #1 Comment 4 last part).

4. P6, L18. Provide a chemical name for the standard OEKANAL.

OEKANAL is a Sigma-Aldrich Grade (purity) for 1, 2, 3-trimethylbenzene and is not a

chemical name. It is followed by "1, 2, 3-trimethylbenzene Sigma-Aldrich".

5. P6, L23. How is mixing achieved in this large chamber; fan, diffusion, other?

The mixing prior to commencing an experiment is achieved by fans. See Carter, et al., 2005 for more details "The two reactors are connected to each other through a series of custom solenoid valves and blowers. The system provides for rapid air exchange prior to the start of an experiment ensuring, that both reactors have identical concentrations of starting material. Each reactor can be premixed prior to the start of an experiment by Teflon coated fans located within the reactor." During the experiment the vibration on the chamber walls due to air circulation on the outside of the chamber provide sufficient mixing during the experiment.

6. P7, L10. Calling a bunch of peaks attributed to innumerable organic compounds completely fragmented by 70 eV electrons as a chemical composition stretches the concept of molecules beyond recognition. I would hardly call this metric a chemical composition in any traditional sense. Perhaps the word "effective" could be incorporated to indicate that this is simply a parameterized metric.

We agree that the chemical composition is derived from peaks from numerable organic compounds completely fragmented. There might be some difference in between the traditional definition and what is widely used nowadays to describe AMS chemical composition, which is a measure of the bulk chemical composition of the aerosol. We keep our manuscript consistent with recent publications using AMS results (eg., Crippa, et al., 2013; Lambe, et al., 2015).

7. P7, L23. The authors should reference the 2013 Atmospheric Environment article by Carter and Heo rather than the CARB report. The article went through peer-review and should be more reliable.

Done.

8. *P8, L11. The sentence is unneeded; include the information in the caption.*

Done.

9. P9, L3. To use the word "claimed" in a pejorative fashion is particularly bad form. If you believe the statement in Odum et al. is wrong, simply state it.

Fixed. Changed "claimed" to "stated".

10. P9, L14. Delete the sentence. The supplement does nothing to support the sentence other than to simply repeat itself and refer to a paper in preparation. Nothing is gain by including the sentence in the paper or in the supplement.

We keep this sentence to clarify the differences in the kinetics is insignificant (e.g., k_{OH}[OH],

 $[HO_2], ...)$ and therefore the molecular structure of the isomers is driving the difference in SOA formation. The referenced paper is now published.

11. P9, L23. Why is the assumption needed? The two-product model is just a fitting exercise anyway.

Similar products are expected to be formed from the aromatic isomers. Fixing the $K_{om,2}$ value provides for similar treatment of the high volatility products allowing us to focus on the low volatility products most important to SOA formation under atmospheric conditions.

12. P9. It might be worthwhile to examine partitioning using a volatility basis set (VBS) to see if any insight could be gained beyond the standard two-product fit which at this point is rather dated. This might provide a more useful metric for describing the partitioning of the AHC products.

We agree that VBS is an attractive way to describe the SOA yield. However, VBS is fundamentally based on gas-particle partitioning theory which is the same as two-product model. The application of VBS only provide similar result in a different format. VBS presents the contribution of products with different volatility using bins and here two-product model use K_{om} . Therefore, we keep our analysis using the traditional two-product model.

13. Sec 3.2.1. This section suffers from a lack of an understandable interpretation of the various fragments from the AMS output and their combinations into the combined metrics (e.g. Eq 1). An annoying aspect of this section is the comparison with other work before any interpretation is provided (e.g., P11, L5, 16; P12 L6, 25,...). How do we even know that the conditions are applicable between these experiments and the ones being compared to?

The interpretation is improved according to the reply to Comment 3). We cited other's work to provide the AMS result found in other chamber work for selected isomer species. The initial hydrocarbon and NO_x conditions used in other's work are not completely the same as ours. Therefore, the AMS data is not exactly the same. However, we demonstrate the AMS data we use are reasonably in-line with earlier studies to contextualize the results and demonstrate that further discussion of the AMS data is reasonable.

14. P13, L7. LV-OOA and SV-OOA are presented both undefined and without context.

We change "LV-OOA and SV-OOA" to "low volatility oxygenated organic aerosol (LV-OOA) and semi-volatile oxygenated organic aerosol (SV-OOA)". The definition of LV-OOA and SV-OOA can be found in detailed in Ng, et al 2011.

15. P13, L8. The sentence as written belongs in the introduction. The intent of the sentence needs a rationale from the data. It is not self-evident.

We added a sentence to the introduction part to emphasize this part as a reply to Comment 2 (P5, L27). The sentence referred to here is to transition from evolution data to average data. We

will delete it here and rewrite it as a sentence below in blue. We mention in the manuscript that "The evolution trend agrees with Fig. S3 (Sect. 3.2.1)." This means that the evolution trend is not significant during the photooxidation similar to what is mentioned in Section 3.2.1 for f_{44} vs. $f_{43+57+71}$. Therefore, average value is sufficient to describe H/C and O/C. We add the following sentence in P13, L8 after "The evolution trend agrees with Fig. S3 (Sect. 3.2.1)." to provide the rationale.

..., which means no significant H/C and O/C evolution is observed in the current study. Therefore, average H/C and O/C with standard deviation provided is used to explore the impact of molecular structure on SOA chemical composition.

16. P14, L21. "a more accurate metric. ..." More accurate than what?

Fixed. Inserted "than H/C and O/C"

17. P16, L1. Eliminate first name for Borrás.

Fixed

18. P17, L15. Use of the term "theory" (in any scientific sense) strikes me as somewhat pretentious. I would consider it as more of a conjecture.

Good point. We use "Alkyl Dilution Conjecture" according to reviewer's suggestion; we keep "methyl dilution theory" as published in (Li, et al., 2016)

19. P18, L8-16. What does the term "extremely low" mean? Provide a value for comparison. I would characterize most of this part as speculation. Also, experimental limitations in Forstner et al., 1997 (their specific quartz filter configuration) suggested that the furan-type compounds were in the particle phase but were almost certainly in the gas phase. The metrics in the present paper refer only to particle phase OA. The reference should be avoided here.

We have now added the actual value for OS_c and delete the reference in the revised manuscript. This part has been slightly modified after considering standard deviation/uncertainty as mentioned in response to Referee #1's Comment 3.

20. Section 5. I seriously doubt that any of these parameterizations would appear in any wide-used air quality model. I would consider the work mainly for academic purposes.

The SOA yield parameters are widely used in current model (e.g. CMAQ see Carlton, et al., 2010; GEOS-Chem see Heald, et al., 2011, WRF-CHEM model Li, et al., 2011). Current work provides improved SOA yield parameters than previous work under high-NOx conditions (e.g. Odum. et al., 1997).

21. P19, L2. Replace "entend" with "extent".

Fixed

22. P19, L3. Nothing is "proved" here. The work simply provides "evidence for".

Done

23. P25, L8. Replace the ACPD manuscript with newly published ACP paper.

Done

24. Table 1, S2 and text. How many AHCs were studied: the text say twelve AHCs (P5,L7), Table 1 gives ten AHCs, and Tables 1 and S2 together give fourteen. Which is the right number? (For good measure, Figure 6 shows eleven precursors.)

Twelve is the right number. Table 1 and Table S2 combine to give the 12 unique AHCs used in this study.

25. Table 3. What's the point of the table if the p value are greater than 0.05. Certainly, the p-value for VFR and k (OH) is not zero.

It is included in note below the table that "Alpha (α) level used is 0.05. If the p-value of a test statistic is less than alpha, the null hypothesis is rejected". It the p value is greater than 0.05, the correlation found is not trusted within the α level. In another word, the larger the p-value the less confidence in the correlation provided. Certainly, the p-value for VFR and k (OH) is not absolute zero. However, it is <0.0005. Therefore, we change "0" into "0.000" for significant figure purposes.

26. Figure 1. The final points control the shape of the curve. The data below 20 ug m-3 would be of most interest for atmospheric applications.

The higher M_0 allows one to improve the parameters used to fit the overall aerosol formation trends, especially that for α_2 . The final points are therefore controlling the shape of the curve used to fit of the high volatility products, in this case α_2 . The identified curves reasonably represent the lower organic mass loadings (< 20 ug m⁻³) as seen in the quality of the fit where α_1 and $K_{om,1}$ (lower volatility products) dominate the shape of the curve. Curve fitting with and without α_2 and $K_{om,2}$ are presented below.



27. Figures 2, 3, 7. Am I missing something? Why are there no symbols for the values; is there that much uncertainty? The use of colors for the precursors in Table 2 is particularly annoying. Are not words sufficient without colors?

The exact locations of these values show up after adding error bar to each value (see Referee #1 Comment 6's reply). I think you mean Figure 2 instead of Table 2. The colors we use categorize all the 12 isomers into different subgroups (e.g. all xylenes are in green as what we labeled in the upper right). We think these colors help the audience to understand what kind of molecular structure impact it is (location vs. length). We would like to keep the colors to help us demonstrate the findings.

28. In Figure 4, what are the estimated uncertainties in the model-generate radical concentrations. Carter and Heo, 2013 suggests that these could be substantial.

We are unsure of what the reviewer is requesting. Figure 4 provides "Oxidation state (OS_c) of SOA formed from different aromatic hydrocarbon" which nothing about model generated radical concentrations. Table S4 lists model-generated radical concentrations. Generally speaking, the [OH] is fitted through precursor measurement from GC-FID and therefore [OH] has little uncertainty ($<\sim$ 5%). SAPRC-11 adjusted photoreactive product quantum yield parameters are used to minimize average biases in Rate (Δ (O₃-NO)) (Carter and Heo, 2013). The uncertainties of radical prediction is minimized since O₃ prediction relies on radical predictions. However, the uncertainties associated with SAPRC-11 is not a focus for current work. We provide the radical prediction provide by SAPRC-11 to rule out the impact of kinetic difference during the aromatic hydrocarbon photooxidation in order to emphasize the molecular structure impact.

29. Table S3. How about the C9-trimethyl compounds studied? Why not put the parameters obtained from the TMB compounds in the table? It would also be informative to include the data for toluene (which must have been studied at some point) for comparison with the other single position substituents, ethylbenzene and n-,i-propylbenzene.

Table S3 is used to support Fig. S1 to demonstrate the length effect among all C_8 and C_9 . We don't study triple alkyl substitute that contain longer chains and therefore there is no need to list C9-trimethyl compounds in Table S3. (The fitting parameters for trimethylbenzenes can be found in Table 2.) We don't include toluene since it is not in the range of C_8 and C_9 aromatics; instead, we refer to our earlier work (Li, et al., 2016) at Page 10 Line 5.

30. Table S4. The value of this table for predicting radical concentrations is very limited as noted above. It may be useful for urban NOx conditions but not where RO2 + RO2 is the dominant source of the aerosol. It doesn't surprise me that the p-values for virtually all comparisons in S5 are no different than the null hypothesis.

We agree that SOA formation is tied to peroxide radical reactions. It should be noted that peroxide radical reaction is associated with NO, precursor concentration and other radicals (e.g. OH) as is ozone formation. We maintain that SAPRC could predict radical concentrations sufficiently well for how they are used in this paper (see response to comments above comment 1-2) and comment 28). The insignificant correlations between yield and radicals are not due to the limitation of the model but the similarity in kinetics among all the isomer precursor we studied. We actually found pretty good radical (eg. HO2/RO2) correlation with yield in our earlier work (Li, et al., 2015). Therefore, we prefer to keep the radical discussion as part of the supplement supporting the manuscript.

31. SI Table 3 should be Table S6. Use lower case k in the table

Fixed

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