

Dear Prof. Surratt,

Thank you very much for your support and suggestions for our manuscript. Also, we thank the reviewer again for careful reading and thoughtful suggestions that help us improve our manuscript. According to the comments, we have added the related statements in the Results and Discussion section. In the following, please find our responses to the comments one by one and the corresponding revisions made to the manuscript (highlighted in yellow color). The original comments are shown in italics, and our point-by-point responses are listed below. We believe the new version is now suitable for publication in ACP.

The referee believes that the revised manuscript becomes much better than before and the current form will satisfy almost requirements from referees. However the following minor revisions will still be necessary for the publication:

(1) Referee #2 commented that it has been recently proposed that the toluene oxidation proceeds dominantly via the cresol pathway (Ji et al., 2017). The authors cited this suggested paper, but they did not discuss appropriately. They cited it after the sentence, “small alpha-dicarbonyls compounds such as glyoxal and methyl glyoxal are important intermediate products of toluene that undergo polymerization to produce low-volatility oligomers”, but this paper reports the formation of highly functionalized products such as polyhydroxytoluenes during the toluene oxidation. Please survey previous knowledge of UV/visible absorption spectra for polyhydroxytoluenes (e.g., methylcatechol and methylbenzenetriols), and please consider how polyhydroxytoluenes affect aerosol optical properties to respond the comment. If necessary, please add discussion to the manuscript. Referee #2 suggested that poly hidroxytoluene products are just an example of newly found products. According to knowledge of this referee, other newly found products are highly oxygenated molecules (HOMs) (Molteni et al., 2018, please see the previous reference list of referee #1). In addition to products proposed by Ji et al., it may be useful to discuss whether or not the products found by Molteni et al. affect aerosol optical properties by surveying absorption spectra of organic peroxides.

Response: Thanks for the suggestions. We have re-read the reference and re-sited the paper in the manuscript: “Ji et al. have reported that toluene oxidation proceeded dominantly via the cresol pathway and formed highly functionalized products such as polyhydroxytoluenes (Ji et al., 2017), which would undergo subsequent reactions with OH to form the precursors including α -carbonyl compounds, organic acids, and other highly oxygenated low-volatility products. Although the peak of 3-methylcatechol, one of the major polyhydroxytoluenes of toluene oxidation products, appeared around 275 nm in UV-Vis spectrum (Ferris et al., 1971), the subsequent products like nitrocatechol and low-volatility oligomers formed by small α -carbonyl compounds might absorb light near ultraviolet and visible. Parenthetically, the reaction rate of 3-methylcatechol with OH radicals was 7.44×10^{-11} cm³/molecule·s, and the estimation of atmospheric lifetime was 22 min approximately (Coeur-Tourneur et al., 2010). Small α -carbonyl compounds and organic acids might undergo particle phase reactions, e.g., acid-catalysed aldol condensation reactions under SO₂ conditions, plausibly contributing to the observed light absorption (Fu et al., 2009;Fu et al., 2008).” (Line 20-31, page 9).

We also added the discussion of HOMs: “As proved in the study of Molteni et al. (Molteni et al., 2018), highly oxygenated molecules (HOMs) were also important oxidation products of toluene. HOMs that contained a structure of phenyl ring could not absorb light above 300 nm, however, the subsequent HOMs of nitration and ring-opening might absorb UV-Vis light. HOMs were detected in our experiments, especially under high humidity conditions.” (Line 20-24, page 8).

Reference:

Ferris, J., Briner, R., and Boyce, C.: Lythraceae alkaloids. IX. Isolation and structure Elucidation of the alkaloids of *Lagerstroemia indica*, *Journal of the American Chemical Society*, 93, 2958-2962, 1971.

Coeur-Tourneur, C., Cassez, A., and Wenger, J. C.: Rate coefficients for the gas-phase reaction of hydroxyl radicals with 2-methoxyphenol (guaiacol) and related compounds, *The Journal of Physical Chemistry A*, 114, 11645-11650, 2010.

Molteni, U., Bianchi, F., Klein, F., El Haddad, I., Frege, C., Rossi, M. J., and Dommen, J.: Formation of highly oxygenated organic molecules from aromatic compounds, *Atmospheric Chemistry and Physics*, 18, 1909-1921, 2018.

(2) *Line 22, page 2. According to knowledge of this referee, oxides of nitrogen are a mixture of gases that are composed of nitrogen and oxygen and it contains gases other than NO and NO₂. The accurate terminology is “nitrogen oxides (NO_x)”.*

Response: Thanks for the suggestion. We have changed the words to “nitrogen oxides (NO_x)”, “... the effects of nitrogen oxides (NO_x) on the optical properties of ...” ([Line 22, page 2](#)).

(3) *Line 1, page 3. According to the format, you may need to remove the initial of first name from the citation, “M. Jaoui et al. (2008)”*

Response: Thanks for the reminding. We have converted the citation to “Jaoui et al. (2008) showed that ...” ([Line 1, page 3](#)).

(4) *Line 31, page 3. You might have to insert the following sentence written in your response, “unlike NO_x analysers equipped with molybdenum converters, HONO was not detected in T200UP NO_x analyzer combined with a patented high efficiency photolytic converter.” to satisfy a requirement from referee #1.*

Response: Thanks for the suggestion. We have added the sentence in the manuscript, “Parenthetically, unlike NO_x analysers equipped with molybdenum converters, HONO was not detected in T200UP NO_x Analyser with a patented high efficiency photolytic converter.” ([Line 16-18, page 4](#)).

(5) *Line 31, page 3. You might have to use the singular for the subject in the sentence, “the concentrations of toluene were measured by a proton transfer reaction quadrupole mass spectrometry... ”.*

Response: Thanks for the suggestion. We have rewritten the sentence to “The concentration of toluene was measured by a proton transfer reaction quadrupole mass

spectrometry...” (Line 31, page 3).

(6) Line 28, page 4. The word, “200”, should be written in the roman font.

“,

Response: Thanks for the suggestion. We have changed the word “200” to the roman font, “...was achieved at m/z 200, with an absolute...” (Line 30, page 4).

(7) Line 38, page 7. A hyphen is missing between “200” and “400”.

Response: Thanks for the suggestion. We have added a hyphen between “200” and “400”, “products around m/z 200-400 and discussed ...” (Line 2, page 8).

(8) Section 3.2. According to a previous comment by referee #1, the subject, “the complex refractive indexes,” in line 2 of page 8 should be “the real part of the complex refractive indexes”. All three abbreviations, RI, appearing in the second paragraph of page 8 should be RI(n).

Response: Thanks for the suggestions. We have changed the “complex refractive indexes” to “the real part of the complex refractive indexes”, “ the real part of the complex refractive indexes increased from...” (Line 6, page 8) and all the “RI” to “RI(n)” in the second paragraph in Section 3.2 (Line 13-19, page 8). We have also corrected other abbreviations through the particle, e.g., “... overestimate the values of RI(n).” (Line 18, page 9).

(9) Line 12, page 9. The word, “oligomerization”, may be better than “polymerization” according to a previous comment by referee #1.

Response: Thanks for the suggestion. We have detected the word “polymerization” in the new version.

(10) Line 34, page 9. You might want to insert “of the WS condition” after the words, “toluene-derived SOA”.

Response: Thanks for the suggestion. We have added words “of the WS conditions” after “toluene-derived SOA”, “... RI(n) of toluene-derived SOA of the WS condition were lower than...” (Line 12, page 10).

(11) Figure S3. The vertical axis title, absorption, will be vague. Does it represent the absorbance based on common logarithm?

Response: Thanks for the suggestion. As the referee inferred, the word “absorption” in Figure S3 represents the absorbance based on common logarithm. To avoid misunderstanding, we have changed the vertical axis title of Figure S3 to “absorbance”.

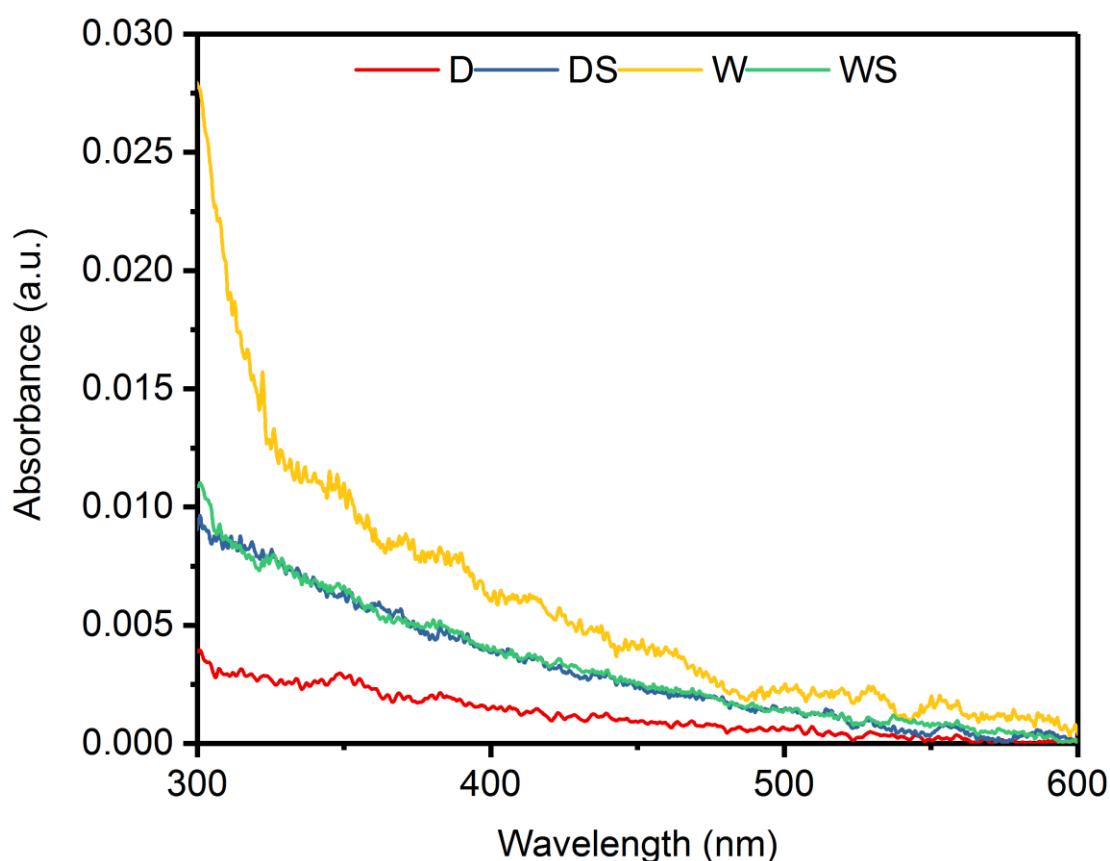


Figure S3. UV-Vis absorption spectrum of toluene SOA under the four different conditions.