

## Responses to Referee #1's comments

**General comment:** In this manuscript, the authors report the first measurements of the rate constant for the reaction of eugenol, an atmospherically abundant methoxyphenol, with the hydroxyl radical in the gas phase. The results are placed in the context of other previously investigated methoxyphenols, including a helpful discussion of substituent effects. The authors also present a detailed characterization of the SOA yield and its response to SO<sub>2</sub> and NO<sub>2</sub>, including a surprising enhancement in SOA yield due to the presence of NO<sub>2</sub>, which may apply to other methoxyphenols. The experimental work is thorough and precise, with appropriate controls, and the manuscript is well organized. I recommend the manuscript for publication following minor revisions.

**Response to comment:** Many thanks for the reviewer's constructive comments and valuable suggestions, which would be much helpful to improve the scientific merits of this manuscript. The concerns raised by the reviewer have been carefully addressed in the revised manuscript.

**Comment 1:** 144: Though photolysis does not contribute to the decay of eugenol, could it contribute to the evolution of the oxidation products, either in the gas or particle phases? Though it is not a focus of this study (one can imagine forming SOA in one OFR, scrubbing any remaining ozone, and then irradiating the products in a second OFR), perhaps the possibility of photolysis of the oxidation products should be acknowledged at the end of this paragraph.

**Response to comment 1:** Thank you for your constructive suggestion. As you pointed out that it is unclear that how UV lights affect the evolution of oxidation products. Using a second OFR, this question can be answered. Unfortunately, we do not have a second OFR in our laboratory. This will be investigated in the future. According to your valuable suggestion, the discussion about the possibility of photolysis of the oxidation products in the OFR has been added in the revised manuscript.

**Revision in the manuscript:**

**Lines 154-155, Add:** "However, it cannot be ruled out that photolysis under UV irradiation might have influence on the evolution of the oxidation products."

**Comment 2:** 164: I enjoyed the comparison of the experimental and AOP WIN-predicted rate constants. I wonder if the comparison should be placed in the context of other methods of prediction. For example, the DFT-predicted rate constant for the reaction of guaiacol with the hydroxyl radical (DOI: 10.1002/poc.3713) is about 1.6 times greater than the experimental value (DOI: 10.1021/jp1071023). In this context, the present agreement, with a predicted value about 0.8 times the experimental value, seems quite good.

**Response to comment 2:** According to your constructive suggestion, the comparison of rate constant of guaiacol with OH radicals obtained by density functional theory (DFT) calculation and experiment study has been added in the revised manuscript.

**Revision in the manuscript:**

**Lines 178-183, Add:** “In addition, the difference between density functional theory (DFT) calculation and lab study has been also observed. For example, the DFT-predicted rate constant of 2-methoxyphenol with OH radicals ( $12.19 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ) is higher than that ( $7.53 \times 10^{-11} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$ ) obtained by lab study (Coeur-Tourneur et al., 2010a; Priya and Lakshmipathi, 2017)”

**Comment 3:** 252: The lack of kinetic limitation to condensation is very interesting. Could this observation be related broadly to the viscosity of the SOA derived from eugenol and guaiacol? How does the present relative humidity of about 44% compare to that in the previous OFR and smog chamber experiments discussed in the comparisons?

**Response to comment 3:** The lack of kinetic limitation on SOA condensation might be mainly related to the physico-chemical properties of the SOA derived from the OH-initiated reactions of eugenol and guaiacol, such as viscosity, low volatility, and high oxidation state, etc.

The relative humidity (RH) in this work was similar to that in the previous OFR study about SOA formation from guaiacol oxidation by OH radicals, while was higher than that in the previous smog chamber experiments conducted by Lauraguais et al. (2014) and Yee et al. (2013).

**Revision in the manuscript:**

**Line 272, Add:** “conducted at low RH (Fig. S6) (Lauraguais et al., 2014b; Yee et al., 2013)”

**Comment 4:** 365: Perhaps the detailed discussion of the effects of NO<sub>2</sub> on the SOA yield would benefit from a mechanistic schematic.

**Response to comment 4:** Thank you very much for your valuable suggestion. In this work, the oxidation products could be not identified due to the lack of analytical instruments. Thus, it is very difficult to discuss the effect of NO<sub>2</sub> on SOA yield from the point of mechanistic schematic. According to the master chemical mechanism (MCM) of aromatic compounds, NO<sub>2</sub> has influence on not only O<sub>x</sub>/HO<sub>x</sub> chemistry but also the formation of nitrophenols and organonitrates. Therefore, we generally mentioned that NO<sub>2</sub> participated in the OH reaction of eugenol, consequently producing N-containing products. Based on NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratios measured by the HR-ToF-AMS, it is suggested that most of N-containing products are organic nitrates. Thus, the relative low volatility of these products could be favorable of SOA formation (Duport é et al., 2016; Liu et al., 2016).

In addition, the fraction of organic nitrates has been calculated to be in the range of 25.64% to 82.05% in the revised manuscript, using the NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratios obtained at different OH exposure, according to the method described by Fry et al. (2013).

**Revision in the manuscript:**

**Lines 418-421, Add:** “According to the method described by Fry et al. (2013) (shown in Supplement), the fraction of organic nitrate was calculated to be in the range of 25.64% to 82.05%, using the NO<sup>+</sup>/NO<sub>2</sub><sup>+</sup> ratios (3.98–6.09) obtained at different OH exposure.”

**Comment 5:** 427: I agree that it is very likely that the SOA derived from eugenol is light-absorbing. Other products, in addition to those containing nitrogen, could conceivably contribute to the proposed absorptivity. For example, products of oligomerization like biphenyls have been observed in the aqueous oxidation of phenolic species (DOI:10.5194/acp-14-13801-2014), and this pathway is likely relevant in the highly-concentrated aerosol phase.

**Response to comment 5:** According to your valuable suggestion, the formation of oligomers via OH-initiated reaction of methoxyphenols (Yu et al., 2014) has been added in the revised manuscript, and the discussion about their light absorption has also been added.

**Revision in the manuscript:**

**Lines 460-463, Add:** “In addition, the formation of oligomers in particle phase via OH-initiated reaction of methoxyphenols, which has been observed in aqueous oxidation of phenolic species (Yu et al., 2014), might also enhance light absorption in UV-visible region.”

**Comment 6:** Technical corrections:

77: In the phrase "a type of methoxyphenols", methoxyphenol should be singular. Perhaps this phrase is redundant and could be omitted.

115-116: Two instances of "approximate" should be "approximately".

126: "Vaccum" should be "vacuum".

199: In the caption to Figure S4, perhaps explain that the arrows indicate the maximum values (i.e, those listed in Table 2).

217-218: This phrase should be reworded to better reflect that the fragmentation occurs in the particles and that the products subsequently volatilize out of the particles.

238: This phrase is slightly confusing, since some measure of composition is determined using the AMS.

368: Should be "cyclic".

383: Should be "radicals".

414-415: This phrase should be reworded to give, for example, "more attention should be paid to SOA formation..."

442: Should be "formation".

**Response to comment 6:** Thank you very much, these technical errors have been corrected in the revised manuscript.

**Revisions in the manuscript:**

**Line 77, Delete:** “a type of methoxyphenols”

**Lines 116 and 118, Change:** “approximate” To “approximately”

**Line 129, Change:** “vaccum” **To** “vacuum”

**Supplement, Line 89, Add:** “The arrows indicate the maximum values (i.e., those listed in Table 2).”

**Lines 237-239, Change:** “which would generate a large amount of fragmented molecules that could not condense on aerosol particles” **To** “which would generate a large amount of fragmented molecules that subsequently volatilize out of aerosol particles”

**Line 258, Change:** “composition” **To** “product composition”

**Line 390, Change:** “yclic” **To** “cyclic”

**Line 410, Change:** “radicasls” **To** “radicals”

**Lines 442-443, Change:** “it should pay more attention on the SOA formation from the OH oxidation of biomass burning emissions and its subsequent effect on haze evolution”  
**To** “more attention should be paid to the SOA formation from the OH oxidation of biomass burning emissions and its subsequent effect on haze evolution”

**Line 472, Change:** “foramtion” **To** “formation”

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