

Mobato et al., studied the aqueous-phase photochemistry of vanillin (VN) in cloud/fog relevant conditions leading to aqSOA. They conducted two types of photochemistry, photolysis of VN and nitrate-mediated VN (irradiation of VN + Ammonium Nitrate mixture) and analyzed VN decay, product mass spectra, O<sub>2</sub>/pH/ammonium effects, and UV spectra for light absorbance of products. They indeed conducted full analyses of aqSOA chemistry. However, novel findings with compelling experimental evidence in this manuscript are not well written. Instead, their arguments are mostly speculative:

1. The purpose of conducting two types of photochemistry is not clear. Didn't you compare the photolysis and OH-radical reactions of VN? Then, it is not clear how much of OH radical was generated in their condition. According to Fig. S3 and also the main text, photochemistry by OH radical is negligible. Then, why not set the concentration of OH radical relevant to cloud/fog condition adjusting nitrate concentration, which can be obtained by a kinetic model based on decay rate of VN and table 1? If the purpose is to study the nitration, then why was ammonium nitrate (AN) used? Ammonium clearly complicates the system. Since authors used sodium nitrate (SN), why did not authors conduct explicit comparison between AN and SN that could lead to chemical insights in the aqueous phase?
2. The explicit mechanisms that could support their arguments are lacking. Fig. 2 is oversimplified. Their arguments about explaining chemistry were heavily based on chemical mechanisms, but the arguments were not convincing because authors did not propose explicit mechanisms—radical-based full mechanisms containing steps in details.
3. Most of the discussions sound speculative (words like "may, probably" were used often). Experimental data and analyses do not seem to support their arguments. Although they attempt to show their results to be consistent with others previous studies, it is difficult to find the novelty of this manuscript.
4. Therefore, substantial revisions and restructure are recommended for publication.

Line 55: There needs to be a reason why NO<sub>3</sub><sup>-</sup> in the aqueous phase is the source of nitration in this work. In most of cases, nitration (organonitrate formation) occurs in the gas phase. Why your aqueous phase study is suitable instead of multiphase experiments.

Line 100-101: I don't understand this statement. Shouldn't you want nitrates to affect the kinetics of VN photolysis by generating sufficient OH radicals? The purpose of adding ammonium nitrate should be clearly stated.

Line 126-127: I disagree the equal ionization efficiency for different compounds. This cannot be true.

Line 133-138: The term, "normalized abundance of products" is scientifically meaningless, unless ionization efficiencies for each product were taken into account.

Line 149-156: A schematic containing radical-based full mechanisms is required. This should be Fig. 2, which is currently oversimplified.

Line 179-181: Building a kinetic mode based on Table 1 is recommended. You can verify it by simulating a kinetic model.

Line 192-194: I do not understand the purpose of conducting N<sub>2</sub> experiments in the first place. Clearly, O<sub>2</sub> is the oxidant that required for the oxidation. Therefore, you expect better oxidation with O<sub>2</sub>. There always exists O<sub>2</sub> in cloud/fog droplets. This is not a novel finding.

Line 225-227: Negative-mode analysis sounds more suitable for oxidized products. The reason for conducting positive-mode analysis that would bring a benefit and unique results in this work needs to be addressed.

Line 234-237: This statement is weak. You need to make a strong statement based on your compelling evidence.

Line 244-246: It is difficult to conclude that products are mainly conjugated pi system based on your ESI analysis (even MS-MS analysis).

Line 266: The range of pH 2.5 and pH 4 seems too narrow to study a pH effect.

Line 270-271: This is speculative, again. You need to make a strong statement based on your novel analysis.

Line 296-300 & 305-306: Speculative

Line 315: The term, "the 50 most abundant products" provides no scientific meaning. You cannot tell the abundance of products by ESI signal. Besides, why did you choose 50?

Line 330-332: Speculative. Propose an explicit mechanism to support your argument.

Line 350-352: How can you prove this based on your experiments?

Line 352-354: What are the anticipated product based on NH<sub>4</sub><sup>+</sup> chemistry? What are the corresponding products you have found? How does that related to BrC absorbance or conjugate bond?

Line 366: speculative