

Comments:

This manuscript aimed to study SOA formation from furan under photooxidation conditions with varied NO_x and RH. SOA mass, O₃ concentration, and SOA composition were carefully measured. The authors concluded that furan photooxidation is dominated by RO₂ + NO chemistry that leads to formation of carbonyl-rich products. SOA formation was found to enhance under higher relative humidity and higher VOC/NO_x concentration. The results are clearly presented, but there are a few important issues that need to be addressed before becoming publishable. My major comment is that the manuscript did not provide sufficient chemistry insights given the suite of instruments used.

Major:

1. The authors claim that the O₃ maximum concentration decreases with increasing RH likely due to that high RH favors the partitioning of the NO_x reservoir, RNO₂ and RONO₂ into the particle phase. Thus the release of NO_x is limited. However, it is unclear how much more pronounced is the RNO₂ and RONO₂ uptake at higher RH and whether this enhancement is sufficient to affect O₃ formation. These species need to be very soluble to show distinct partitioning behavior under different RH. What are their possible structures and could the Henry's law constants be estimated? Were any RNO₂ or RONO₂ species enhanced in the SOA samples under higher RH? From the ESI-MS results, the authors said that the SOA composition is similar between higher RH and low RH. Does this contradict what the authors concluded earlier? You have a max SOA yield of 5% and only a small fraction of that is RNO₂ or RONO₂. Thus are they important enough to affect gas-phase NO_x concentration? Evidence is needed. Alternatively, could wall loss of the RNO₂ and RONO₂ under high RH better explain the observation?

2. The ESI-MS results presented grouped ions and observed m/z 200-299 as the most abundant group. What does this mean? What are the major chemical formulas observed in this range? I think a lot more discussion of the ESI-MS results could be included here. For example:

(1) Could the authors group all nitrogen-containing species and discuss their presence?

(2) What are the ranges of O:C ratio and oxidation state?

(3) Any variation as the furan/NO_x and RH changed?

(4) For soluble species uptaking onto aqueous particles, they will likely oligomerize. Do the authors see oligomers in the ESI-MS?

(5) There is a substantial decrease of large molecules under lower furan/NO_x ratio and higher RH in the negative ion mode (but not in the positive ion mode). What does that suggest?

(6) What is the ion in the negative ion mode around m/z 380?

In the current form, there is very little discussion (page 11, line 16-24, page 12, line 2-7, and page 13, line 8-11).

3. The observation of enhanced SOA formation with increased NO is interesting, because most previous work has shown that increase of NO tends to promote RO₂+NO chemistry which leads to fragmentation. Why are SOA yields higher with higher NO in the case of furan photooxidation? The authors gave these possibilities in the text, but did not further conclude it based on the mechanism. Is this because furan is a cyclic compound and the fragmentation does not break the

C4 backbone? Or were OH concentrations very different between different furan/NOx experiments (as in the cases for the two referred studies)? Or both?

4. The discussion of the RH effect on SOA using the mass spectral data was very vague. The authors only discussed two ions, m/z 187 and m/z 255. First, it seems the m/z 187 ion is C₄H₄O₄Cl₂ from high-resolution fitting. But how is it formed? The authors proposed Cl adduct for the major ions m/z 187, 255, and 281. It is probably the case for m/z 255 and 281, since they contain nitrogen and have only one Cl on the ion. But m/z 187 is probably not. The C₄H₄O₄ compound does not exist in Scheme 1. The compound D is C₄H₄O₃. How does it form C₄H₄O₄ in the mechanism? Further, for the m/z 255 ion, the formulas provided in Figure 6 is incorrect. [C₄H₄O₄]³⁵Cl⁻ has an exact mass of 185.949; [C₄H₄O₃N]³⁵Cl⁻ has an exact mass of 255.938; [C₈H₈O₈N]³⁵Cl⁻ has an exact mass of 280.994. The authors need to make sure the chemical formula assignments are correct before discussing the mechanism.

Minor and technical comments:

1. Abstract: The abstract should deliver the most important scientific findings. It is unnecessary to describe how the measurements were performed in the abstract (e.g., how particle mass concentration and size distribution were determined). Additionally, “the SOA mass concentration and yield increase with increasing humidity, because higher aerosol liquid water content brings more aqueous phase reactions” does not make sense. More aqueous phase reactions do not mean higher product yield.

2. Second sentence in the introduction puts different categories together. Remove black carbon and brown carbon.

3. Page 2, line 1. Change volatile organic compound to VOC.

4. In the introduction, some statements about cited studies are incorrect. For example, page 2, line 3-4, the Chan et al. (2010) study demonstrated that isoprene SOA formed under high NO₂/NO is large, not under high-NO_x conditions; the Zhang et al. (2012) as well as the Zhang et al. 2011 studies show different compositions under dry and wet conditions.

5. Page 10, line 10. What is C₄H₄O₄Cl? Does the authors have a prediction of the possible structure? How does the NaCl interacts with the organic components in SOA to form this species? On page 11, line 24, some discussion was made, but it is still unclear how “Cl adduction of SOA product” happens.

6. Page 10, line 13. The carboxylic acids may not generate enough acidity to catalyze reactions as the authors mentioned, unless the authors could provide evidence.

7. Page 12, line 28. The authors said “it is highly probable that the particle surface area increases with the amount of ALW increasing”. Could the SMPS measurements provide quantitative evidence?

8. Page 13, line 4. Under high RH, the acidity, if any, is likely lower due to water dilution.