

This manuscript described photooxidation of cyclohexene with changing SO₂ concentration and concluded that both SO₂ competing OH and acid-catalyzed heterogeneous reactions are important to result in the observed SOA yield trend. Organosulfates are also observed in SOA composition. The main results are clearly stated, but the discussion could have been more comprehensive and in depth, regarding the complementary measurements. I think there are a few major issues that the authors need to address before the manuscript can be published.

Major comments:

1. As initial SO₂ concentration increases, it becomes more competitive of OH radicals against cyclohexene, whose initial concentration remain stable. Note cyclohexene could also react with O₃. From the Figure S1 in supplemental, it seems O₃ concentration was already high at 20 min. If OH is mostly reacted with SO₂, when cyclohexene remains, cyclohexene + O₃ could become the dominant pathway for cyclohexene loss. At page 9, line 8-9, the authors argued that the ozonolysis rate is 6 orders of magnitude lower than OH oxidation. But the much higher O₃ concentration than OH could offset this difference, especially in the case of the current study where OH was insufficient. Therefore, varying only initial SO₂ could cause very different cyclohexene chemistry (OH oxidation vs. ozonolysis). I think this is important to discuss. The authors at least need to provide convincing data to show ozonolysis is not important in this study. A statement like “In addition to the kinetic limitation of the cyclohexene reaction with O₃, the typical concentration of O₃ in our chamber was 200 ppb and hence the importance of cyclohexene reaction with O₃ was expected to be less significant than that of its reaction with OH under any relevant SO₂ conditions” is too vague. The authors did not include cyclohexene concentration in the supplementary figure, which is an important indicator of the VOC chemistry. Also, it will be evident if the authors could show cyclohexene decay curves at different initial SO₂ concentrations. It might be best if the authors could provide a figure (could include that in Figure 2) estimating how much cyclohexene reacts with OH vs. O₃ under the studied SO₂ concentrations.

2. Comparing Figure 3 and Figure 5, it is unclear to me what the amount of sulfate indicates in either case and they obviously represent different measurements. Is the amount of sulfate in Figure 3 only from IC measurement (inorganic SO₄²⁻)? And the amount of sulfate in Figure 5 from IC measurement normalized to SOA mass? I think it is the main results of this manuscript and need to be clearly stated in the figure caption. Also I wonder how does the ratio of the FTIR band 1100 cm⁻¹ to IC SO₄²⁻ as a function of initial SO₂ look like? This ratio might tell how efficiently organosulfates are formed under changing SO₂ concentrations.

3. As indicated by the title, I think more discussion regarding chemical composition is needed. From the results, only organosulfates are focused. From Figure S1, it looks NO_x gets lost to organic nitrates. From Figure 4, the IR data suggest –ONO₂ presents in SOA. I suggest the authors discuss more on organic nitrate in SOA. Only a paragraph at the very end seems insufficient. For example, does –ONO₂ IR data correlate better with SOA yield? What N-containing chemical formulae present in the ESI-HR-MS data? Any suggested mechanisms?

4. Based on Page 8, Line 35, it seems both NO- and NO₂-initiated experiments were conducted. But it is unclear according to Table S1. The authors used NO_x in most experiment description. I think it is better to state clearly whether they used NO or NO₂. The SOA yields might be similar, but chemistry and timescales of SOA formation might be different, as the authors already indicated.

Minor comments:

I have a big issue with the literature citing quality of this manuscript (and I do not know how to make suggestions because there are too many of those). Some examples: Page 1, Line 27. A few important review papers need to be cited in the first paragraph of introduction, such as the Hallquist et al. 2009 ACP, Kroll et al., 2008 AE. Page 8, Line 33. Many papers were published demonstrating acid-catalyzed heterogeneous reactions and enhanced SOA formation before and around 2010. The authors did not cite the most important studies.

Page 2, Line 1. Jaoui et al., 2012 citation was not in the reference list.

Page 4, Line 6. It should be specified, whether NO or NO₂ was injected.

Page 4, Line 23. What were the TD temperature and time?

Page 5, Line 9. FTIR analysis uses 300L of air sample, >75% of total chamber volume. Discuss potential artifact.

Page 8. Line 8. It is problematic to say “NO₃-initiated reaction was a poor source of SOA”. Presto et al., 2005a and some later studies did find out that NO₃ oxidation of alpha-pinene does not make a lot of SOA, but not necessarily for cyclohexene.