

This manuscript by Gong and Chen describes a series of laboratory experiments, aiming to elucidate the contribution of stabilized Criegee intermediates (SCIs) to the formation of SOA from limonene. The authors used a creative flow tube setup to investigate SCIs arising from ozonation of the endo- and exo-double bonds (DBs) separately. By employing an SCI scavenger in the system, the authors claimed that they have quantified the contribution of SCI chemistry towards the SOA yields as a function of RH. The major conclusion is that water plays a complex role in the reaction system, suppressing SOA formation under low RH, while facilitating SOA formation at high RH. Over the past few years, the importance of SCI chemistry in the atmosphere has become evident. With SCI being a reactive intermediate that is difficult to detect, quantitative evaluations for the importance of SCI is lacking. This manuscript aims to provide quantitative information that fills our gap in understanding. The topic is timely and is within the scope of ACP. The writing and data analyses were conducted with caution. However, I have concerns regarding a few approaches and assumptions that authors employ in the study. I recommend a major revision before publication on ACP.

### Major comments:

- 1) I'm afraid that the contribution of SCI to the SOA formation may be exaggerated in the current setup due to the presence of a high concentration of butanol (OH scavenger). The authors provided an estimate that 12.5% of SCI has reacted with butanol, using a lower-band estimate for the SCI reactivity with alcohol. It is not convincing that the effect of butanol is "not important (Line 304)". Can the author perform an experiment with an aprotic OH scavenger (e.g., hexane) to experimentally confirm their assumption?
- 2) The fraction of SCI reacting with water ( $SCI_w$ ) was estimated solely from the formation of  $H_2O_2$ .
  - How did the authors measure  $H_2O_2$ ? Was it the gas phase  $H_2O_2$  or particle phase?
  - Although I agree that  $H_2O_2$  is the major decomposition product of  $\alpha$ -hydroxyhydroperoxides (product of  $SCI + H_2O$ ), the decomposition of  $\alpha$ -hydroxyhydroperoxides is an equilibrium process and may not always proceed completely.
  - $\alpha$ -hydroxyhydroperoxides are not the only source of  $H_2O_2$ . It's known that  $H_2O_2$  is generated in SOA extracts, likely due to the decomposition of larger organic peroxides.
  - In particular, Zhao et al. 2018, J. Phys. Chem. A reported  $H_2O_2$  arising from the decomposition of hydroperoxyester, which is formed from  $SCI +$  organic acids. Although the mechanism is not completely clear, this implies that the product of  $SCI + AA$  may also give rise to  $H_2O_2$ .

Citation: Ran Zhao, Christopher M. Kenseth, Yuanlong Huang, Nathan F. Dalleska, Xiaobi M. Kuang, Jierou Chen, Suzanne E. Paulson, and John H. Seinfeld  
The Journal of Physical Chemistry A 2018 122 (23), 5190-5201 DOI: 10.1021/acs.jpca.8b02195

- 3) The flow tube experiments employ tens of ppb of limonene with an excess amount of O<sub>3</sub> for reactions. While these concentrations are rather typical for flow tube experiments, I think the author should discuss the feasibility of extrapolating their flow tube results to the real environment. As the authors point out, limonene mixing ratios are at the sub-ppb level for forest and urban environments. In my opinion, SCIs will predominantly react with water when organic concentrations are low. Thus, the SOA formation potential of SCIs they determine in the flow tube may or may not be applicable to the ambient conditions.

### **Minor and Technical Comments**

- Line 155 “that” to “than”
- Line 307 the sentence: “The reactions of SCIs with the compounds formed from SCIs scavengers would not compensate the effect of the consumption of SCIs on SOA formation.” is unclear. Please rephrase.
- I’m supportive of the authors’ idea to provide an atmospheric implication of their findings by simulating three scenarios: forest, urban, and indoor. Instead of investigating all the RH for each scenario, I wonder if authors can constrain the RH to ranges that are more relevant to each scenario? For instance, the most comfortable RH range for human occupancy in an indoor environment is between 30 to 60%. It is unlikely we see an indoor that are extremely dry or wet.
- Acid anhydrides can be hydrolyzed to form organic acids. By any chance, can the acid anhydrides arising from SCI + AA be hydrolyzed at higher RH, regenerating AA?
- Line 391-392 - “On the other hand” is repeated in the same sentence.
- Line 173 - k(other) - I wonder if wall loss of SCIs should be included in k(other)?