## Review of "Modeling Day and Nighttime SOA Formation via Multiphase Reactions of Biogenic Hydrocarbons"

## **Overall Comment:**

This manuscript uses both chamber experiments and modeling to explore photochemical and nocturnal production of SOA from three different biogenic VOC precursors. The approach and questions asked are scientifically relevant to the readership of ACP. However, I have several concerns about the manuscript's readability, overstatement of some conclusions, and missing methodological details that should be addressed before publication.

## General remarks:

Understanding the mechanistic processes that contribute to SOA yields is an important question for the atmospheric chemistry community. The authors use a nice general approach of using chamber experiments to validate their model and then using their model to explore mechanistic details of the chemistry.

However, the results/discussion section is very hard to follow. As Referee 1 noted in their remarks, "some parts of the manuscript seem to be more a review paper than an original work." I think this sentiment still applies to the results/discussion section. The original conclusions of this work are deeply buried and hard to follow in this section. I recommend revision of this section to make the conclusions more easily identifiable to the reader. Moreover, many of the mechanistic conclusions stated in the paper are backed up only by previous literature, rather than a detailed analysis of the authors' modeling work. Including some process-level modeling results to support the stated conclusions would greatly improve the science presented (some specific examples of this are listed below as "specific comments").

Similarly, the abstract is very long-winded and makes it hard to understand the science question that the authors are trying to answer. I recommend revising the abstract to focus more on the big picture impact of this work rather than a list of detailed conclusions.

Some methodological details are missing, including: Was the chamber filled with outdoor air or zero air prior to perturbation by VOC, seed, NOx, and O3 injections? If filled with outdoor air, how did the composition vary between experiments? How was the chamber cleaned between experiments?

Figure S2 seems like an important figure that would be worth considering moving to the main text since it relates to some of the main conclusions about the work involving the interaction of different oxidants.

Because the chamber used in this study is outside, the experiments presented here are not explicitly 'controlled experiments,' i.e., multiple variables change between each experiment. This variability is fine when using these experiments to validate their model since the variability is captured in the model inputs, and a framing that emphasizes that the model can capture the observed variability is useful. However, it does become problematic when drawing process-level

conclusions in Section 4.1 since variation in multiple variables could be contributing to the observed trends. As such, I think some of the conclusions in Section 4.1 are overstated.

Lastly, the paper contains numerous grammatical errors that inhibit its readability. I recommend thorough proofreading.

Specific comments:

Lines 15-19: These three sentences are hard to understand and should be rewritten for clarity.

Lines 39-41: This sentence contains redundant statements about the dominance of biogenic HC emissions.

Lines 51-52: Note that O<sub>3</sub> titration is possible in some environments.

Line 54: It's unclear what the quoted 80% refers to.

Section 2: Referencing Table 1 in this section would be helpful to the reader.

Line 101: What was the CCl<sub>4</sub> from?

Line 124: What process does the "stoichiometric coefficient" refer to? Is it for each lumped bin?

Line 141-142: What model does "the current regional model" refer to?

Line 148: It could be helpful to include the C\* saturation vapor pressure equivalents for the vapor pressures listed here.

Line 149: Can you give quantitative descriptions for the reactivity scale?

Section 3.4 and Figure 1: I'm confused by which parts of the model mechanism uses MCM gasphase chemistry and which parts use SAPRC chemistry. Please clarify.

Fig. 2(a): Why isn't there a line for  $OM_p$  here?

Fig. 2: An indication of when t=0 is (e.g., injection time at night or sunrise in the daytime) would be helpful in these plots. Additionally, I recommend noting in the caption that meteorological parameters (e.g., temperature, relative humidity) varied between these cases in addition to the chemical parameters already noted. It might also be more appropriate to label the middle and right columns as "VOC +  $O_3$  +  $NO_3$ " instead of just "VOC +  $O_3$ ".

Line 225: This conclusion about an increase in SOA formation because of aqueous reactions of ozonolysis products is unsupported. Can you include a figure that shows process-level detailed analysis of the model to support this?

Line 242-243: Reporting the atmospheric range of pHs would be more useful if the manuscript also included information about the pH of the seeds used in the experiments.

Line 248-250: The sentence beginning with "Evidently..." is redundant.

Lines 258-259: The conclusion about the impact of photolysis of isoprene and a-pinene oxidation products on SOA production is unsupported.

Lines 261-263: Why is the high reactivity relevant here?

Lines 283-285: The speculation presented here can be supported by the modeling work in this paper. Including supporting plots/calculations would be useful.

Line 316: Is the difference between nighttime and daytime SOA for the same amount of initial VOC? Does this statement account for variation in emissions / PBLH between day and night?

Line 320: Note that NO<sub>3</sub> can also be lost at night by reaction with fresh NO emissions.

Lines 371-373: How does the ratio of alpha-pinene to gasoline correspond to that in the atmosphere?

Lines 383-385: Can you provide a plot to support the conclusion of this sentence, or at least a quantification of the conclusion presented here?

Fig 3(f): Why is there no dotted black line in Figure 3(f)?

Fig 4: Because RH=45% is right around the efflorescence point for ammonium sulfate, do these high and low RH cases actually probe significant differences in aerosol liquid water content? Also, for clarity, I recommend noting in the caption that this figure presents modeling results.

Fig 7: What is driving the oscillations in the fraction of SOA from alpha pinene (solid red line)?