Comment on acp-2021-953
Anonymous Referee #2

Xu et al. presents laboratory studies where they investigated the heterogenous oxidation of an alpha-pinene organosulfate surrogate, sodium 2-hydroxy-2,6,6-trimethylbicyclco[3.1.1]heptan-3-yl sulfate, or apOS-249, using an oxidation flow reactor. The reaction and products were measured with an UHPLC-ESI-QToF-MS, HPLC-ESI-QTRAP-MS, and IC. The authors observed various products they provided chemical formula and compared against ambient and lab studies where these compounds had been observed before. Further, they determined the heterogenous OH oxidation of apOS-249 and found the lifetime to be comparable to the lifetime of aerosol (~10 days). They explored potential reaction mechanisms. Finally, they investigated the potential amount of inorganic sulfate (SO4) produced from the heterogenous reaction of apOS-249 with OH. The paper is generally well written with interesting results that would be of importance for the community that reads ACP. I recommend publication after the authors address some of the comments below.

1. In general, more information concerning the oxidation flow reactor (OFR) is needed to better understand and replicate the experiments. The following details needs to be included:
   1a) How was the aerosol and gases introduced into the system? How were they sampled from the OFR?
   1b) What material is the OFR made of?
   1c) Was this made in laboratory or purchased from a company?
   1d) What was the temperature for the experiments?
   1e) How was OH reactivity determined during the experiments?
   1f) What were the losses within the OFR? Was this included in the calculations?

2. Other information within the methods that would improve the paper:
   2a) Was a drier used after atomization?
   2b) How were O3 and gas-phase species removed prior to sampling? What was the particle loss through this method to remove gas-phase species?
   2c) How was the SMPS operated? What SMPS system was used?
2d) What assumption was used to assume an aerosol mass loading of 2000 ug m⁻³? Was this mass concentration too high for heterogeneous oxidation (e.g., OH-limited at the beginning due to too high aerosol mass loading compared to OH concentration)?

2e) Description of the aerosol collection/filters are needed -- size, type, and pore size of filter, were they cleaned before use, how backgrounds were collected, stability of products on filters, and any impactors that may have been used.

2f) A simple diagram or an actual photo of the experimental set-up would be beneficial.

3) As reviewer #1 mentioned, sample preparation and/or sampling of the products with ESI may lead to side reactions. Though many of the products may be hard to synthesize, have surrogates been used to investigate their stability on filter, during preparation, and during sampling? One thing that would help with this question is stating that apOS-249 shows high extraction efficiency (pg 8, ln4-5) sooner and maybe discuss other organosulfates.

4) Section 3.1. These results are really interesting and important for the community. However, as has been studied more within the community, phase state, ionic strength, and "shell" formation are most likely extremely important parameters in the OH heterogeneous reaction of apOS-249. As this is a single phase experiment, there is likely little phase state and shell formation concerns (which might be useful to explore in future experiments including with changing RH). I strongly recommend the authors briefly put this experiment into this context and how these results are most likely upper limits. Similar thoughts in regards to ionic strength, especially as this may impact the reaction mechanism and what intermediate and final products are stable.

5) Section 3.4. Though the authors show that sulfate formation is minimally important, there is a question if the sulfate they observe may be due to the hydrolysis of the primary, secondary, etc. products. The authors investigate and determine that apOS-249 is likely stable but it is currently not clear if the other products may be.