

Janecek et al. provide new laboratory measurements of SOA formed from the OH oxidation of cyclic volatile methyl siloxanes. The authors produce SOA via an OFR and test various aerosol properties, including morphology, composition, volatility, and hygroscopicity. The authors also explore various experimental parameters on SOA yield, and report new yields that are higher than previously measured.

The results from this study are useful for models aimed at understanding organosilicon aerosol properties. I'm particularly interested in the TEM measurements and how these could be useful for identifying organosilicon aerosols in the atmosphere. Overall, I find the methods to test aerosol properties to be good and the conclusions about volatility, hygroscopicity, and composition to be sound. I have some concerns about the aerosol yield conclusions (see below). I would appreciate if the authors addressed these concerns prior to publication.

**Major Comments:**

Page 11, lines 1 -15 and Page 11, lines 29-32.

I have some concerns with the interpretation of the yield experiments. How certain are the authors that the yield measured at the highest OH exposure is statistically significant? Looking at panels A and C, it seems like this point could be an outlier, but the authors attribute the uptick in yield as an increase in semi- or low-volatility oxidation products. The authors argue that the fate of gas-phase species is condensation to aerosols (line 27), but then note that the experiments were run in particle-free air. I interpret this to mean that the authors are relying on nucleation processes to drive particle formation. If so, this significantly complicates the interpretation of the yield because there is no way of understanding the nucleation rate and its relationship with the vapor condensational sink. Consequently, I believe that the authors are overestimating the aerosol sink and underestimating the aerosol yield. The authors even show this experimentally in the following section (3.2.2). When the authors add 50 nm ammonium sulfate particles, the aerosol mass increases by 44%. Would this not suggest that the maximum yield was not observed for any of the experiments conducted with particle-free air, and that significant condensational losses were to the walls of the chamber?

In yield experiments, it is crucial that sufficient aerosol be added in order to drive oxidation products away from the walls and to the aerosol phase (Zhang et al. 2014). Consequently, I believe that the seed aerosol experiment described in section 2.2.4 should actually be treated as the yield experiment. If this input of aerosol is significant enough drive most of the oxidation products to the aerosol phase, then I believe the authors can interpret the effect of OH exposure on aerosol yield; otherwise, I would treat this yield as a lower-limit estimation.

Section 3.3

I find this section to be quite interesting, though the discussion is very brief. I understand that the primary purpose of this section is to show that the aerosol collected from antiperspirant oxidation exhibits a qualitatively similar TEM spectra as aerosol produced from D5 oxidation. Does this also suggest that D5 was the predominant SOA precursor in the antiperspirant experiment and that other components of the antiperspirant did not contribute to SOA formation? I presume that there were a lot of other components in the antiperspirant that was tested, including fragrances (which were likely a mixture of highly reactive compounds like monoterpenes and terpenoids) and possibly other ingredients, such as glycols. Can the authors provide more details of the ingredient list so that the reader has a sense of what precursors may have been present in the antiperspirant stick? Was D5 listed at the top of this list, which presumably implies that it was a major ingredient?

Did the authors also test the composition of the aerosols formed via oxidation of conditioner emissions? I'm curious if the TEM measurements show the same relative elemental composition, or if other peaks (such as carbon) are more abundant. If this spectra is different, then this could point towards the influence of other ingredients on SOA formation.

Ultimately, I think the authors can do a bit more here to put the personal care product experiments into perspective. As written, a reader could interpret these results to suggest that aerosol formed from personal care products containing D5 is exclusively composed of D5 oxidation products! This is unlikely, of course, and not what the authors are intending to show, so some further discussion should be provided. Furthermore, could the TEM technique be useful in understanding organosilicon aerosol in the atmosphere? Presumably, most particles containing silicon in modes < 100 nm would have been derived from secondary processes. Could these TEM measurements be a useful tool in identifying aerosols resulting from organosilicon oxidation?

#### Page 15, lines 3-11.

In this section, the authors discuss the relevance of cVMS SOA formation and potential shortcomings of this work. The authors rightfully demonstrate that cVMS will contribute a small fraction to total SOA; however, I believe the authors are overextending when they try to frame these results into the measurements conducted by Bzdek et al. (2014). Specifically, I take issue with the extrapolation of the organosilicon SOA content. The nano-particle measurements conducted by Bzdek et al. provide a constraint to nucleation processes close to the source, but downwind of major cities like L.A., chemical and meteorological conditions could change and limit the oxidation of organosilicon precursors. Furthermore, an extrapolation to 1.5 ug/m<sup>3</sup> seems quite unrealistic. That amount of SOA is comparable to the background SOA in LA believed to come from regional biogenic sources (~2 ug/m<sup>3</sup>, Hayes et al. 2015).

I recommend that the authors refrain from extrapolating to suggest that there could be unexplained sources of organosilicon precursors. I think it is sufficient to cite Tang et al. (2015) and McDonald et al. (2018) to show that organosilicon SOA precursors are emerging as an

important source of VOCs in urban areas, and that incorporating this work into models will help to constrain the VCP impact on urban SOA formation.

### **Other Comments**

Page 2, Lines 8 - 10. This sentence seems a bit out of place here, and doesn't necessarily requires its own paragraph. I suggest moving it elsewhere in the introduction ( perhaps after the last sentence at line 32?)

Page 3, Lines 1-20. This material would be better presented as a discussion rather than as an introduction. I would recommend moving this to section 3.1

Page 3, Line 23-24. This sentence reads as if concentration, size, and morphology were also measured by EDS. Please rephrase.

Page 4, Line 6. Please add "by photolysis of water" after "in situ"

Page 4, Line 11. It would be clearer to say "temperature-controlled"

Page 4, Line 14. It would be clearer to the reader if you mention that cyclomethicone and cyclopentasiloxane often refer to the same molecule.

Page 5, Line 5. Please note that the "D5 water bath temperature" translates to variations in precursor concentrations.

Page 5, Lines 11 - 12. From my understanding, it's not crucial that the concentrations be the same; rather, it's important that the OH reactivity be low enough that losses due to OH titration can be ignored. What concentration range of D5 and SO<sub>2</sub> was being injected into the OFR?

Page 5, Line 21. The term "disappearance" suggests that the SO<sub>2</sub> was lost by some unknown process. I would recommend replacing with "...measurement of reacted SO<sub>2</sub> in the OFR".

Page 5, Line 25. Please add "constant" after "rate".

Page 6, line 2. How well do the OH exposures calculated from Eqns 1 and 2 agree?

Page 6, Line 26. The semi-colons should be replaced with commas.

Page 7, Line 12. I would refer to "the DMT" or the "DMT Instrument" as a "CCN counter"

Page 7, Line 12-13. Consider removing the phrase "The controlled variable in the DMT instrument" and replace with "The thermal gradient was varied from..." for brevity.

Page 7, line 20. By "source", do you mean from the DMA monodisperse outlet?

Page 7, line 27 - Page 8, Line 10. The details of data QA are not needed here. I suggest removing, or placing in the supplemental information.

Section 2.5. I'm somewhat confused by the thermal degradation experiments. The authors don't explain in detail why these experiments were performed, or why these might be relevant in the atmosphere. Are there cases in which D5 might be exposed to high temperature that it would decompose and form lower volatility components? Or, is this a check to evaluate potential biases in the volatility experiments? If it is the latter, I would suggest moving this discussion to Section 2.4 and refer to these experiments as controls.

Page 10, line 21. What do the authors mean by "reasonable ways?"

Page 10, line 23. It seems to me that the experiment reached steady state long before 20 hrs! No need to change - I'm just impressed by the length of the experiment.

Page 10, line 24 - 25. This sentence is a bit awkward. I might consider rephrasing as "This was followed by a sampling period, during which four gas samples were taken to determine D5 SOA yields."

Page 10, line 31. I'm confused why the authors implicate the D5 injection rate as the primary cause for variability. Is it a mixing issue?

Page 13, line 19. Please add "that" between "shows" and "no". Also, it is incorrect to write "ka observed smaller particles" - please choose a different verb.

Page 13, lines 20-22. Can the authors please clarify the last statement about the higher oxidation state of smaller particles? Is that because the larger particles contain more semi-volatile components (e.g. dimers, or second-generation monomers?).

Section 3.5, Figure S13. The 10 nm mode is quite noisy. If it is difficult to quantify, why include this mode in the figure? I would find this figure more useful if I could see the zoomed-in traces for 20-110 nm experiments.

Section 3.6. Would thermal degradation be a significant process influencing the lifetime of D5? If so, please provide some justification. If not, I feel like these experiments are better characterized as volatility control experiments - i.e., to ensure that changes in particle concentrations during volatility experiments are not the result of residual gaseous D5 that could degrade and lead to particle formation.

## References

Zhang et al. (2014). Secondary organic aerosol formation in chambers. Proceedings of the National Academy of Sciences, 111 (16) 5802-5807

Hayes et al. (2015). Modeling the formation and aging of secondary organic aerosols in Los Angeles during CalNex 2010, *Atmos. Chem. Phys.*, 15, 5773-5801