

Atmos. Chem. Phys. Discuss., referee comment RC2
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Comment on acp-2021-965

Anonymous Referee #2

Referee comment on "Secondary organic aerosols from OH oxidation of cyclic volatile methyl siloxanes as an important Si source in the atmosphere" by Chong Han et al., Atmos. Chem. Phys. Discuss., <https://doi.org/10.5194/acp-2021-965-RC2>, 2021

Major Comment

The study was conducted to measure the formation of secondary organic aerosols (SOAs) during OH oxidation of cyclic volatile methylsiloxanes (cVMS) in an oxidation flow reactor (OFR). While the measured data of SOA formation was consistent with other published results obtained similar OFRs, the authors assumed that the results of OFRs would be used to determine the SOA formation in the real atmosphere. However, the assumption was proved incorrect by an earlier study by Charan et al. (2021). Their work was cited in Introduction of the current manuscript, but the main result of the paper was completely ignored (not mentioned/discussed). Charan et al. (2021) supported that the yield of SOA depended strongly on OH concentration and organic aerosol mass concentration (related to initial D5 concentration) among several experimental parameters (e.g., OH concentration, OH exposure, aerosol seed, NO_x, initial D5 concentration, reactor type). They also claimed that the formation of secondary aerosols is fairly small (<5% or more likely <1%) under environmentally relevant conditions (i.e., low OH and VMS concentrations). Thus, a large chamber might be a good option (rather than OFRs) to maintain environmental-relevant conditions and a low surface-to-volume ratio. Note that all the SOA yields from OFRs were high and in contrast with those from chamber reactors. However, the current manuscript (by Han et al.) did not discuss any of these important findings. They asserted SOA yield was dependent on OH exposure and applied the OFR results for the real atmosphere. Therefore, without providing a strong result against the conclusions of Charan et al. (2021), the current manuscript was built on an incorrect assumption. Accordingly, the estimated SOA production rate is not reliable.

The calculation of cVMS-SOA concentration (in Eq. 4) is not clear. The equation needs the amount of lost of cVMS, but the quantity was not measured or published. See more in Minor Comment below.

The manuscript states that the number and mass size distribution of aerosols was monitored using a scanning mobility particle sizer (SMPS), but no data was provided and

discussed except the statement that the data was used for calculation of effective aerosol densities. The results would be helpful to understand the characteristics of SOA formation under different conditions.

Minor Comment

L51-53: Is this (scarce knowledge) still valid? There have been many studies published since the reference you cited.

L135 & Table S2: Based on my extra calculations, the initial concentration range (i.e., 20-40 ppb & 20-25 ppb in Table S3) is equivalent to $\sim 10,000$ times greater than environmental concentrations or similar to the values of Janecheck et al. (2019). See Table S2. The authors should note that the concentrations are high enough to increase potential uncertainty in the extrapolation to the environmental concentrations. This manuscript does not provide individual initial concentrations for each of the cVMS species, but it would be helpful with the data.

L190 (& L29): This study claims that the production of SOA is dependent on OH exposure. However, this is partially true in the limit of OFR experiments. In contrast, Charan et al. (2021) discovered that OH radical concentrations are more important than OH exposure considering all experiment results with OFR and chamber reactors. If the latter is valid, it's better to use OH radical concentrations for the x-axis rather than photochemical age as the initial cVMS concentrations were in a relatively narrow range.

L255-256: This is under unseeded conditions. Please explain why there were more productions under seeded conditions at earlier PAs?

L321-322: The estimation would not be valid based on this study that used OFRs. See Charan et al. (2021) for the detailed discussion.

L323,330: There should be differences between C_{cVMS} and ΔC_{cVMS} . I assume SOA concentrations were calculated based on C_{cVMS} , not ΔC_{cVMS} . Eq.4 is valid only when $C_{cVMS} = C_{in-cVMS}$ and background $C_{cVMS-SOA}=0$. Thus, there should be more clarification. In addition, extrapolation should be highly uncertain due to 4 orders-of-magnitude differences of concentrations and overestimated SOA yields.

L332-333: Not clear what values were used in this study. The values were not directly measured.

L617-619: Check this reference: not used.