Attention to detail is part of the scientific endeavour, and unfortunately this manuscript is missing important data to adequately assess the results. As it stands this manuscript is not fit for publication. Insufficient detail was provided, and other scientists cannot build upon the work, then the endeavour is fruitless. I encourage the authors to pay thorough attention to the details of their methods and results to clearly communicate to other scientists what was done and how to reproduce the data to subsequently build upon it. These steps include:

- Build on existing literature. The introduction has little information on what has been done in this field so far. And key words and examples are missing, leaving the reviewer wondering why this research was done in the first place.
- Communicate the mechanisms correctly. Lines 39-40 incorrectly describe how triplet state organic matter forms from the intersystem crossing of singlet state organic matter. (The reader is left concerned after reading this section.)
- Tell a story – why were PM and lab-generates filters made? What was the purpose of the comparison? Which hypotheses were being tested?
- Purify chemicals used, particularly the probes. (Line 107 explicitly states that all chemicals were used as is, and it is common practice in the community to distill furfuryl alcohol since it can easily dimerize and oxidize. Most papers in the field state that FFA is purified by distillation.)
- Describe all blanks in detail and show all the results. What were the field blanks, which controls were done? Line 158 is simply not good enough. Showing all the data of adequate background samples.
- Use the most up-to-date rate constants. For example, FFA decay was revised back in 2017 by (Appiani et al., 2017) to be $1.00 \times 10^8$ M$^{-1}$ s$^{-1}$.
- Caption all figures with thorough details. What do the colour wheels represent in Figure 1 and 2?
- The figure captions in the supporting information section contain so little data that figures are not understandable. (Striking examples include: Table S1 where all units are missing, copyright infringement for Figure S3, column with data not matching...
headings in Table S3 and in Table S4, poor resolution of Figure S10, no details in caption of Figure S12, no details of any of the 60 (!!) graphs in Figure S14.

- Rewrite the abstract to describe what was done: specific which aerosols were analysed and why, and what methods were used to study the processes.
- It is inaccurate to draw regressions through clusters of data as in Fig. 6
- Use acronyms consistent with the literature. (MSM and HP-SWM are not acronyms used by the community and are confusing, WSM in the community should be WSOC), use the word "probe" instead of capture agent.
- Rewrite the environmental implications. As it stands that section reads as a summary. However, the implications section should extrapolate the findings to their impact on the environment and future work.
- Proof read the manuscripts for typos (there were many, missing spaces and misspelt words)

There is a missing discussion on how this study builds upon past work. (and many missing references) For example, are the authors’ results consistent (or not) with what other have observed so far? I would encourage the authors to state clearly what their hypothesis was and why they specifically chose the aerosol samples listed to support their hypothesis. Furthermore, was the starting hypothesis validated? The authors should clearly state their scientific approach.

Specific comments:

- The concentrations used by the authors of 100 mgC/L is very high. How did the authors obtain such high concentrations from aerosol filters (about a factor 10-100 more than typically found on PM filters in polluted environments)
- How were the percentages in the first paragraph of the results calculated?
- The authors must compare their results with the literature (ex. Line 213-214, “moderate level” compared to what?)
- The EPR results are promising, but how do these measurements compare with quantitative methods?
- Show the blanks/controls in Fig. 5

References: