Reply to comments by Anonymous Referee #2

General Comments

1. **There is a growing body of literature suggesting that adding 185 nm irradiation in OFRs – along with 254 nm – is advantageous to use of only 254 nm radiation with externally added ozone, especially with respect to organic peroxy radical (RO2) chemistry, resilience to OH suppression and UV photolysis, and easier operation in the field (e.g. Peng et al., 2019; Peng and Jimenez, 2020; Rowe et al., 2020). It isn't clear to me from the text (L137-L141, L151-L155) if the PFA OFR can implement the OFR185 mode or not. If OFR185 operation is possible, it's worth clarifying that, and explaining why it wasn't evaluated here. If it is not possible, please clarify, and discuss the associated tradeoffs.**

   Answer: The PFA OFR is operated only in 254 mode. An explanation was provided in what is now Section 2.1.2:

   "Though the combination of materials results in sufficiently high reflectance for the 254 nm emission peak of a mercury lamp. Silva et al. (2010) showed that the reflectance of ePTFE at 175 nm is significantly lower, with the difference thought to be due to absorption by O2 trapped in pores. Reflectance at the 185 nm emission peak of a mercury lamp is expected to be slightly higher than that at 175 nm, but it is likely that a significant intensity gradient would still exist and so a 254 nm-only lamp is used and ozone generated externally and introduced with the sample flow."

   Nevertheless, we appreciate the advantages of operation with 185 and 254 nm and will continue to evaluate design modifications that might enable it with our OFR.

2. **Overall, the most novel aspect of the PFA OFR design appears to be the higher reflectivity achieved with the ePTFE gasket combined with the lower lamp power. This design modification enables the PFA OFR to achieve a higher OH exposure at a specific lamp power relative to other designs, as noted in L128-L130, which is noteworthy. The potential implications that are identified from the results seem to be better residence time distributions because of less recirculation and reduced temperature gradients. Aside from that, the implication on measurements of interest was less clear. The gas and penetration efficiencies are comparable to previous OFR designs with broader RTDs and less internal reflectivity, as are the α-pinene and m-xylene SOA yields. To me, this suggests that results of the sort described here are not sensitive to this design component, or that OFR applications that might be affected by higher internal reflectivity are not adequately discussed. I would strongly encourage adding a section that illustrates applications where this higher reflectivity demonstrably improve performance using metrics other than the OH exposure.**

   Answer: We thank the referee for the recommendation. We have substantially revised the text describing potential application in **Sections 2.1.2 and 2.1.3**, the importance of the

reflective ePTFE layers in **Section 3.1,** and the importance of the side flow **in Section 3.3.** Please refer to the revised paper.

The comparison of particle penetration efficiencies is incomplete, and in some places is misleading. Figure 7 shows that the size-dependent particle penetration efficiencies of PFA, PAM and TPOT OFRs, and as presented, suggests that the PFA performance is the best of the three. However, as noted in L215-L220, the PFA was conditioned for 12 hours prior to testing to suppress static discharge, whereas the other OFRs were not. Thus, results are a combination of OFR design and testing procedure, and how to isolate the relative importance of each factor is not clear. Either results for the PFA prior to conditioning also need to be shown for a direct comparison, or this difference needs to be more clearly identified in the figure/caption. Figure 7 also does not show published particle transmission efficiency data for several other OFR designs that were already referenced in this paper. Please see Figure 2 from Li et al., 2019, reproduced below for reference; to my knowledge, this is the most comprehensive comparison to date:

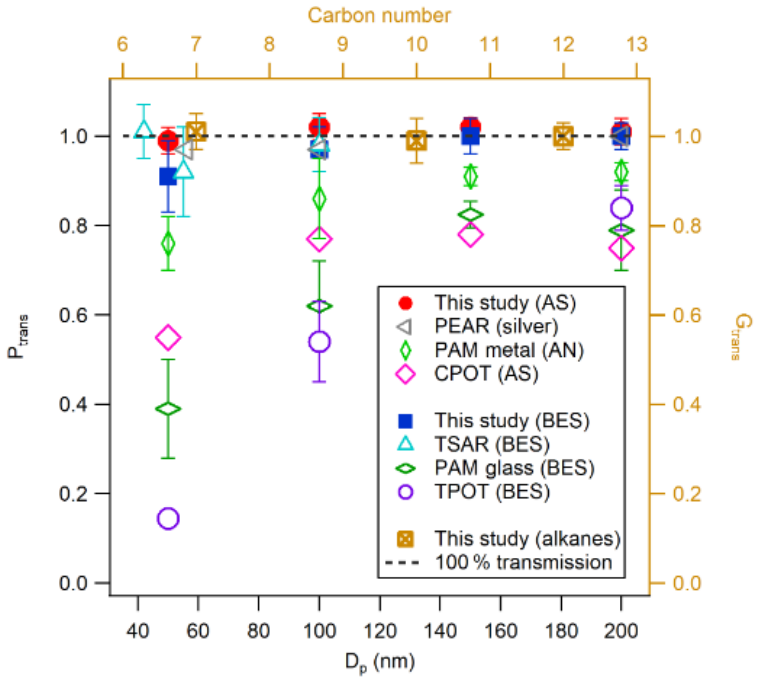

Figure 2. Particle (left and bottom axis) and gas (right and top axis) transmission efficiencies (P_{trans} and G_{trans}) for the ECCC-OFR. Particle transmission efficiencies of other OFRs are shown for comparison: PAM glass and TPOT (Lambe et al., 2011), PAM metal (Karjalainen et al., 2016), TSAR (Simonen et al., 2017), CPOT (Huang et al., 2017), and PEAR (Ihalainen et al., 2019).

Answer: We added the data for the CPOT, TSAR, and ECCC-OFR to the revised **Figure 6**. The results are not meant to be misleading. Our intent is to always minimize charge prior to

measurements. Though we have not tested this extensively yet, it is our belief that the charge will not return until and unless the OFR is disassembled or relocated.

3. **Similarly, the gas penetration efficiency may have been measured in different ways. For example, in measurements by Lambe et al. (2011) and Li et al. (2019), the OFR walls were first passivated by flowing the relevant gas(es) through the OFR. Based on the text (L199-L200), it doesn't appear that that was done here, in which case this may be a plausible explanation for the lower SO2 penetration efficiency in the PFA OFR.**

Answer: Thanks for pointing this out. We did first passivate the reactor by flowing the relevant gas through it for at least 15 minutes before the experiment and until the concentration measured at the outlet was constant. Even so, the SO_2 penetration efficiency was still relatively low compared with the others shown. We are unsure why the penetration efficiency is lower and plan to investigate this further.

Occasionally the reactor is referred to as "PFA". It might be less ambiguous to refer to it as the "PFA OFR" to distinguish it from perfluoroalkoxy alkanes.

Answer: Thank you for the recommendation, which we have followed. Please refer to the response to Anonymous Reviewer #1 general comment (3) for details.

Technical Comments

6. **L148-L150: The authors state that "Continuous operation for 6 hours resulted in a temperature rise of less than 2 ^{o}C" What is the temperature rise over 24 hours or longer, i.e. periods that would be relevant for continuous ambient OFR measurements?.**

Answer: We didn't observe significant temperature increase during the 3-day ambient measurements. We added this statement in the revised paper.

7. **L168: Please mention the OD of the copper bypass line, clarify the reason for using a 150 cm length of bypass inlet versus 200 ccm length of OFR inlet, and calculate the residence time in the bypass and OFR inlet lines to place in context of the OFR residence time.**

Answer: We mentioned the diameter (0.635 cm OD) in section 2.4. The residence time of the bypass is approximately 2 s. We added a sentence about it in the revised paper.

8. **L313: Were different side flow:center flow ratios studied to evaluate the influence of this flow ratio on the residence time distribution? Is a side:center flow ratio of 1:1 optimal, or could the RTD be further improved at a different value?**

Answer: We did some measurements to evaluate the influence of the flow ratio on the residence time, with the results added to **Figure S2** in the revised paper. It may be that increasing the ratio beyond 1:1, especially if accompanied by a reduction in the cross section of the exit cone, would result in narrowing of the RTD. However, that benefit would have to be balanced with the resulting decrease in sample flow rate or average residence time.

9. **L356: This statement is not correct – the TPOT and PAM OFRs were also operated in OFR254 mode in the study described here.**

Answer: Thanks for pointing this out. We have removed this statement in the revised paper.

10. **L412-L414, L423-425: Please apply the OFR254 OH exposure estimation equation developed in Section 3.7 of Peng et al. (2015) to calculate the OH exposure during these SOA yield measurements. As far as I can tell, the required inputs to this equation are available from the measurements that were described here.**

Answer: We added the OH exposure estimation in the SOA yield measurements. Please refer to **Section 3.4** in the revised paper.

11. **Figure 1 and Section 2.1: Please explain/justify the use of 35^{o} and 24^{o} inlet and outlet cone angles.**

Answer: We wanted to keep the angles close to the 30 degrees suggested by Huang et al. (2017), but they were also constrained by the diameters of the PFA tube and the exit flow port and by the capabilities of the machine shop we used.. We added a statement about this in **Section 2.1.1**.

12. **Figure 3 should either be moved to Supplement or deleted and described briefly in words.**

    Answer: Done.

13. **Figure 4 could be moved to Supplement.**

    Answer: We moved it to **Section 2.1.1**.

14. **Figure 6a: it would be better to present results in terms of the photon flux, which is an intrinsic property of the OFR that could be more easily compared with other OFR designs, rather than the fractional lamp power, which is only applicable to the specific lamp type used here. The photon flux could be estimated from the maximum lamp output normalized by the internal surface area of the OFR, or, preferably, constrained using ozone measurements measured at the exit of the OFR (as a function of humidity and lamp power) using a photochemical model such as the OFR- KinSim mechanism (Peng and Jinenez, 2019, 2020). Also, please change the y-axis to a logarithmic scale, or make the y-axis scale go to zero.**

    Answer: Thanks for the suggestion. We moved Figure 6 (a) to the supplement and combined Figure 6 (b) and Figure 6 (a). The recommended change was made in what is **Figure 5** in the revised paper.

15. **Figure 6b: I don't understand the utility of showing the single point obtained without ePTFE at 50% lamp power. With only 2-3 data points shown here, it might just be easier to integrate this data into Figure 6a.**

    Answer: Please see the response to the previous comment.

16. **Figure 7 is incomplete (see comment #3).**

    Answer: Done.

17. **Figure 8: Please clarify whether the literature data shown here were obtained with UV lamps on or off.**

    Answer: Done, please refer to **Figure 7** in the revised paper.

18. **Figure 9 could be moved to Supplement.**

Answer: Done.

19. **Figure 10 could be moved to Supplement.**

Answer: We revised this figure and pointed out the estimated OH during the SOA yield measurement. Please refer to **Figure 8** in the revised paper.

20. **In my opinion, Figure 11 should plot the SOA yield as a function of OH exposure rather than COA. The precursor concentration was not systematically varied, and COA is not really the independent variable here. The OH exposure can be estimated using the OFR254 estimation equation provided by Peng et al. (2015).**

Answer: We added a sentence in the figure caption to point out that the color represents the OH exposure, which was estimated from the OFR254 estimation equation provided by Peng et al. (2015). We didn't test the SOA yield over a wide range in OHexp, which is why we prefer to use C_{OA} as the x-axis. It also simplifies comparison with the other OFR studies presented in Lambe et al. (2011). Please see **Figure 9** in the revised paper.

21. **Figure 13 could be moved to Supplement.**

Answer: Though we moved some figures to the supplement, we prefer to keep this figure in the main text because it shows that the rapid particle concentration changes in the sampling period can be captured by the PFA OFR, which is one focus of our future studies.