

Response to Anonymous Referee #1

The authors present here a description and investigation of a new oxidation flow reactor that is made entirely from Teflon. A variety of OFRs have been developed over the past decade from a range of materials, but few have been previously designed from Teflon despite its high inertness and low vapor wall losses. The specific geometry and design of each has important implications for interpreting the data they produce. It is consequently valuable and important to conduct (and submit for peer-review) the detailed sorts of testing described here, and this is a fitting journal for this work. This manuscript presents a detailed description of the design and examination of its residence times, wall losses, and results, and it should be published. My concern, however, is not so much with the science (though there are some specific comments below), but in the presentation. Overall, I think many of the Figures could be made more clear, and many of them (and some of their discussion) does not need to be included in the main body and could be moved to an SI.

General comment:

(1) In the descriptions of the operation of the PFA, the authors spend a lot of time discussed operating modes that are not necessarily pertinent to the final operational approach. For instance, in some figures they show the impacts of operating with the side purge flow off, or without the reflective ePTFE. These feel to me like advancements the authors figured out along the way that don't describe the actual recommended operation of the system (i.e., the authors do not seem to recommend ever operating with the side purge flow off). Consequently, they seem better suited to a brief mention in the main text and deeper discussion in a Supplementary Information for the more interested reader. The discussion of the static removal is similar, though I think may warrant a discussion in the main text as the static losses might be expected to be the main issue with a non-conductive OFR. Figures (and associated discussions) that I think are more detail than are maybe relevant to the main text are: Figure 3 - This figure is illustrative of the detailed description of the text, I don't think it really adds much Figure 5 - shows the importance of the reflective sheath, but since the reflective sheath is used throughout the manuscript and is "part" of the instrument, this mostly shows work done along the way, and is not of central importance to the rest of the work. One option would be to show the gradient by pairing Fig. 5a and Fig 6a as one figure, as Fig 5b (which is just the ratios shown in 5a) and 6b (which doesn't have much information) don't seem critical. Figure 9 - again, as with Figure 5, this mostly shows work done along the way, and is not really germane to the final instrument. The interested reader might want to dig into this, but since the average reader is probably more interested in the final configuration, this could go to the SI

This is a general issue, not just related to these specific figures. Overall, I think discussions of some of the methods are perhaps a bit too detailed, and then interpretation of the demonstration and scientific data at the end is a bit sparse.

Answer: We thank the reviewer for the constructive suggestions on the manuscript. Here we list the changes to the current version:

- (1) **Section 2.1 Reactor design** has been divided into three subsections.
- (2) Figures 3 and 9 have been moved to the supplement.
- (3) We have substantially revised the text describing the importance of the reflective ePTFE layer in **Section 3.1** and also state the importance of the side flow in **Section 3.3**

We prefer to keep Figure 5 in the main text of the modified manuscript because it provides justification for the use of ePTFE gasket and shows the significant improvement in the UV intensity and uniformity achieved when it was added.

Specific comments:

(2) Line 89-90: I am not sure, but I thought there was also a teflon film OFR in use by the Jimenez group at some point? Bill Brune also published one in 2007: <https://acp.copernicus.org/articles/7/5727/2007/acp-7-5727-2007.pdf> . Given that one of the big values/advances of the present work is the all-Teflon construction, some discussion of Teflon OFRs should be included.

Answer: We have added a brief description of that Teflon-film OFR (Kang et al., 2007) to the introduction.

(3) Line 107: It is confusing to call it the PFA. I get that it is a play on the fact that it is made of PFA, but it muddies the discussion somewhat.

Answer: We obviously chose the name because it's made of PFA and admit that this name is somewhat confused with the PFA material. We replaced "PFA reactor" with "PFA OFR" to make it easier to keep straight. "PFA" without OFR following is used only when referring to the material itself.

(4) Line 124: In Figure 1 the annulus and pinholes are not really clear, so it took a little bit of re-reading and examining to understand exactly how it is designed and works, and I'm still not totally sure I have it figured out.

Answer: Thanks for pointing this out. We have clarified the text as follows "The outer ~50 % of the flow that is most influenced by interactions with the reactor walls flows outside of the exit cone and into an annulus surrounding it. From there, it is pulled through 12 uniformly spaced ~0.15 cm ID pinholes drilled through the PFA pipe about 3 cm from the top. The flow extracted through those pinholes travels into a channel between the flow tube and the end cap and then through a port on the top cap where it is purged by a vacuum pump,"

(5) Line 280: Not really a 36% increase in transmission efficiency, more accurately a 36 percentage point increase, maybe state as "36% of particles that are no longer lost"

Answer: Thanks for pointing this out. We have rewritten the sentence "The similarity in the resulting 36% of the 50 nm particles that are no longer lost and the 41% of those 50 nm particles that are expected to be charged (Wiedensohler, 1988) suggests electrostatic loss was negligible after the static charge was minimized."

(6) Line 284: Not clear, is the data in Figure 7 directly from the referenced work?

Answer: Yes, the data are directly from the referenced work and we revised the figure and added the most recent published particle transmission efficiency data for several other OFR designs. Please refer to Figure 6 in the revised paper and to the response to Anonymous Reviewer #2 general comment (2) for details.

(7) Line 309: I'm not a statistician, so I'm not sure whether or not this is the right metric or approach – why adjust residence times by the average? Why not normalized to the nominal residence time (V/Q)? Isn't that the metric that is used for the laminar flow case? For example, in Figure 8b, SO₂ comes out sooner than expected from laminar flow or than CO₂, which the authors attribute to reversible uptake – but that should delay the SO₂ response, not speed it up. By delaying response, the average RT is shifted later, and the apparent speed of the early eluting SO₂ is faster. I think all of this would be easier to interpret (and perhaps more meaningful) if the nominal RT were used instead of the average.

Answer: We appreciate the noted advantage of using V/Q rather than an average that is calculated from the measurements. One complication for this OFR is that the residence time of the subsampled central flow should differ from that expected from V/Q because the velocity in the laminar flow profile is greatest in the center of the tube. In addition to the offset that would result, different sample to purge flow ratios were used for different tests, which would cause shifts in the distributions unrelated to effects such as wall interactions. Perhaps more importantly, we normalize by the average calculated from the data because that the approach that is used to present similar experimental data for the other OFRs (Huang et al., 2017, Lambe et al., 2011), which facilitates comparisons.

(8) Lines 342-361: How should the scientific community be dealing with or thinking about these yields? Looking at Figure 11, OFRs seem to be saying that the yield for a-pinene at a give Coa and OH_{exp} could easily vary by a factor of many (e.g. ~5-50% for Coa = ~100ug/m³ and OH in the range of 3-10x10¹¹). This is, of course, not a problem the authors are solely responsible for, but it makes me wonder about the utility of using yield comparisons to validate or understand the OFR. It is reassuring, I suppose, that the PFA is in the middle of all this noise (as opposed to outlying), but are we (the reader and/or the authors) really learning anything from this intercomparison? Is there some way to make sense of this spread other than the broad "uncertainty in OH_{exp}"? Maybe the points in Figure 11 could at least indicate different OFR modes, or something that might explain some of these differences?

Answer: We agree. There is, of course, variability in yields reported from smog chambers as well, though not as much as is evident for OFRs in the figure. We attribute some of the variation to differences in gas and particle wall losses and to the uncertainty in OH_{exp} noted by the reviewer. Because the comparison is with OFRs that have undergone much more extensive use and characterization than that described here, our primary objective was to show that results with this OFR are comparable to those from others. We hope to investigate run-to-run and OFR-to-OFR variability more in the future.

(9) Line 366: In Figure 12 it looks like the seed is multi-modal, but the test refers to a narrow mode at 200 nm - is this due to multiple charging in the DMA?

Answer: Yes, this is due to multiple charging. The multiple charge modes are pronounced because a volume (rather than number) concentration size distribution is presented. The 200 nm mode would be more amplified relative to the multiple charge modes if the surface area concentration or condensation sink distribution was instead presented.

(10) Line 374-376: Adding a panel to Figure 12 showing the trend in yield as a function of pinene:AS would aid this discussion

Answer: Thanks for the recommendation. We added **Figure S3** that shows the trend in yield as a function of precursor:seed ratio.

(11) Line 384: Both of these citations refer to specific conditions and locations in the early 2000's. How are the author's using these to estimate the stated OHR?

Answer: We only used this as a rough guide for OHR based on prior measurements in CA because we didn't measure it during the ambient measurement period. We agree that the OHR is considerably lower today than in the early 2000's. We have removed the statement in the manuscript.

Figure comments:

(12) Figure 2 is missing "(a)"

Answer: Corrected.

(13) Figure 3 labels are confusing because the arrows could refer to "that point onward" or to the region they are coming from. It would be clearer to indicate with colored regions or a binary on/off trace when UV was on or off.

Answer: Thanks for pointing this out. We moved it to supplement and added a binary on/off trace when UV was on/off. Please refer to **Figure S1** in the revised paper.

(14) Figure 6 could be made more clear - too much going on in the legend with lots of repetition. Also not clear if 6a is with or without the ePTFE. I gather it is without? Why are only two points from 6a reproduced on 6b? Why not just include the one point w/ ePTFE shown in 6b into 6a and make all the labeling more clear?

Answer: Thanks for pointing this out. We have combined **Figures 6 (a) and (b)** and show the enhancement in intensity/photon flux achieved by adding the ePTFE gasket. Please see **Figure 5** in the revised paper.

(15) Figure 9 - why not also include the modeled with the side flow on?

Answer: Our goal was only to show that the experimental results were consistent with the simulations. We did not include a similar comparison for operation with the side flow on simply because our CFD software did not have the package required to do so. However, we added a series of RTDs with different side:main flow ratios to the figure and moved it to the supplement. It is now **Figure S2**.

(16) Figure 10 legends are similarly busy and repetitive requires a lot of looking back and forth. Some estimate of OH exposure on these plots might help.

Answer: Thanks for pointing this out. We calculated OH exposure from the equation developed by Peng et al. (2015) and added it in the revised **Figure 8**.

(17) Figure 11 - make the legend markers darker so they can be seen more easily

Answer: We have adjusted the contrast of the legend markers. Please refer to **Figure 9** in the revised paper.