RESPONSE TO REVIEWER 2: Manuscript ID: amt-2017-352

Mitroo et al. introduce a method to deconvolve the measured residence time distribution (RTD) from sampling tubes to get the real RTD inside the Washington University Potential Aerosol Mass (WU-PAM) reactor, which is also validated by computational fluid dynamic (CFD) simulation. The idea of this paper can help improve the understanding of RTD for the oxidative flow reactor (OFR) user community. This paper is well-written and fits the scope of AMT. I suggest for publication after considering the following aspects:

We thank the referee for their time in reading and reviewing this manuscript.

[In reference to: General Comments]

1. I agree with Review #2's comments about the expansion of Section "5 Potential implication". My concern is that how your method can be applied to simulation rather than just used to explain RTD. In other words, how does the incorporation of CSTR tank-inseries (TIS) model framework behave when compared with the PFR framework? For example, most of your inversion results indicate that the number of TIS, N, is a little bit larger than 1. Does that mean it is CSTR rather than PFR that can better represent OFR? So to simulate what happens in OFR, we should use CSTR model instead of PFR? Then the question is to what extent the difference will be introduced to the simulated results by shifting from PFR to CSTR. I think the authors should clarify these points in this section.

The referee is correct that given the number of TIS is a little bit larger than 1, the reactor is considered to behave more like a CSTR than as a PFR. We address comments for both referees in the revised manuscript (lines 406-434):

"Initial PAM modeling work assumed plug flow behavior in OFRs (Li et al., 2015). Li et al. stated that correcting for the non-ideal E-Curve in their OFR would account for ~10% error in their oxidant exposure results, which is less than the overall model uncertainty. However, recent work incorporates the effect of non-ideal RTDs on model outputs (Palm et al., 2017, 2018, Peng et al., 2015, 2016; Peng and Jimenez, 2017). Peng et al. (2015) show that for three OFR operational modes (that is, modes of different oxidant formation mechanisms denoted by 'OFR185', 'OFR254-70', and 'OFR254-7'), a comparison between model output for ideal plug flow vs. non-ideal RTDs (using the RTD experimentally obtained by Lambe et al., 2011a) for OH exposure (OH_{exp}) generally agree within a factor of 2 for low OH_{exp}; the model disagreement exacerbates at high OH_{exp} beyond a factor of ~4. Peng and Jimenez then extend OFR operational modes to include N-containing chemistry (in modes referred to therein as 'OFR185-iNO', 'OFR185-7-iNO', and 'OFR185-70-iNO') where at moderate-to-high OH_{exp}, the deviations exacerbate significantly, although the authors argue those conditions represent unrealistic chemical pathways. It is worthwhile noting that the chemistry modeled by Peng and Jimenez may find a workaround by utilizing N_2O as NO precursor (Lambe et al., 2017) rather than NO itself, potentially minimizing RTD-related errors. Palm et al. (2018) report data from OFR field deployment where the same comparison (ideal plug flow vs. the RTD experimentally obtained by Lambe et al., 2011a) suggests RTD-related errors overpredict (for CO) or

underpredict (for toluene and monoterpenes) photochemical age (that is, the ratio of OHexp to tropospheric average OH number concentrations) in the reactor, generally within a factor of 3 of model error. Considering this work employs the compartmental model RTD described by Lambe et al. (2011a), which for reasons mentioned in the previous section may not be the true PAM RTD, and given that non-ideality in RTDs affects certain OFRs more than others, implementing the method presented here to obtain a more representative reactor RTD can either help constrain error uncertainty in the models, or possibly extend the OH_{exp} range in which OFRs can be operated, a reportedly nontrivial task (Palm et al., 2018). Considering our results indicate that OFRs like the WU-PAM exhibit an RTD closely matching that of an ideal CSTR, which is more well-mixed than the Lambe et al. RTD, the sensitivity analysis conducted so far could represent a lower bound for error analysis because the Lambe et al. RTD is closer to a PFR-like RTD than a CSTR-like RTD."

2. TIS model can have different forms. The authors assume the same residence time for each CSTR-tank and find the tank number. One can also take the form with a fixed CSTR (or PFR or mixed CSTR/PFR) number but to find each residence time, which looks more reasonable given the CFD simulation. Can the authors discuss this a little bit more? For example: How does the number of TIS, N, depend on the flow rate, or in other words the average residence time? Since the flow rate changes the fluid field, the mixing style could be different at different flow rates (e.g. Fig.3a-c). But I cannot see any trend. Can the author give some explanation for that?.

The classic TIS model assumes constant mean residence time across the N tanks, which is $\bar{t}_i = \bar{t}/N$. In this work it is treated as a two-parameter model in which both N and $\bar{t}$ are scanned to find the optimum value pair that results in the best fit with experimental data. It is found that the calculated mean residence time $\bar{t}$ is similar to the space-time τ as expected. Theoretically we can use any well-defined reactor model in place of TIS, such as the axial dispersion model (ADM) (employed by Lambe et al., 2011a in tandem with compartmental modeling) which measures the non-ideality from PFR. Mixed CSTR/PFR is also possible, provided the mathematical derivation is properly carried out. Developing such a new model is out of the scope of the current manuscript, but can be recommended as future work in this field. Whether the reactor model selected is valid to represent the real reactor is subject to validation with experimental measurements, as performed in Figure 3. In this work we find the TIS model is satisfactory for the PAM reactor according to the close agreement between model prediction and experimental data, stating the caveat that the TIS model is *not* phenomenological. The reviewer raised the question about unclear trend between N and space-time in Figure 3(a-c), which is interesting to the authors too. Our guess is that under these conditions the reactor behaves so similarly to a single CSTR that the subtle differences are buried in the experimental uncertainties. Perhaps the trend would become more clear as the space-time is further raised, and we predict the trend to be N increasing with space-time. The reason is that the larger the space-time, the slower the flow, thus the weaker the turbulence and back-mixing, which means the further away the reactor is from a single CSTR. This reasoning is backed by Figure 3f, where N is more than doubled at a much higher space-time (although it is also a different configuration).

3. Equations in Appendix B should be carefully checked. For example, in Eq. (B3) it should be $E_{1|N-i}$ *instead of* $E_{1|N-1}$. *In addition, try to avoid "N", since the number of TIS is also "N", which may cause confusion. In Eq. (B8),* ***A*** *is a matrix, which should be listed as* $A_{i,j}$ *not just* $A_{N,i}$. *One more, as a vector,* ***B*** *should be listed as* B_i, *with* $i = 1, 2, \ldots, N - 1$ *and* $i = N$. *About time step* Δt, *see following comments for Figure 3.*

We thank the referee for pointing out the inconsistent notations in the equations. The misuse of indices can make the equations confusing and even wrong. We rewrote all of the equations in Appendix B with carefully checked syntax. We hope they bring much more clarity now. Please see revised manuscript lines 608-672.

[In reference to: Specific Comments]

1. Line 182: "create allow", delete either one.

Similar to Reviewer 1, we thank the referee for pointing this out, and have deleted "allow".

2. Line 218: "F-curve", define it here or mention it later

We restructured the sentences (L226-227 of the revised manuscript):

"After the simulation, the exit concentration is mixing-cup averaged to output a representative of a cumulative RTD (explained in the next section)."

3. It is unnecessary to list both dimensional and non-dimensional equations at the same time, e.g. EQ. 1-4 and 8-9, since the non-dimensional form has been introduced in detail in Appendix A

We respect the referee's point of view, however we choose to represent both dimensional and dimensionless equations in the main text for the audience.

4. Figure 2: Please use higher resolution figures and rearrange the figure locations (too compact, and x-labels are hidden)

We restructured Fig. 2 accordingly, incorporating requests from Reviewer 1 as well.

5. Figure 3: Why time resolution is different in Panel e? Does Δt *in Appendix B correspond to the time interval in Figures 3a-f?*

We had instrument problems that day, and could not take datapoints as frequently as for all other panels. We have noted it in the figure caption. This doesn't affect the output of our algorithm based on longer Δt.

"Lower frequency data for panel e) was due to instrument repair, and temporarily set on longer averages."

6. Figure 5: Please use an intuitive y-label instead of "F". Also please specify "N" value in the caption

We relabeled the y-axis with "Normalized Concentration" for easier interpretation The "N" for the N-CSTR acronym the referee is referring to in the legend is another acronym for the TIS model. We clarifiy this in the caption, and introduced this acronym along 'TIS' in Section 3.2 (lines 313-317):

"We chose to apply the tank-in-series (TIS) model (MacMullin and Weber Jr., 1935), also referred to as N-CSTR model, to the convolution integral since it is a one parameter model that, although not specific to flowtube, tubular, laminar, or plug-flow reactors, gives an idea of where the reactor lies on the spectrum of mixed flow vs. plugged flow based on the value of a parameter, N"

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