Response to reviewers for the paper "A comprehensive evaluation of enhanced temperature influence on gas and aerosol chemistry in the lamp-enclosed oxidation flow reactor (OFR) system" in Atmos. Meas. Tech. Discuss. Doi: 10.5194/amt-2023-230

By Tianle Pan et al.

We appreciate the three reviewers' comments and support for the publication of this manuscript after revisions. Following the reviewers' suggestions, we have carefully revised the manuscript. To facilitate the review process, we have copied the reviewer's comments in black text. Our responses are in regular blue font. We have responded to all the referee comments and made alterations to our paper (**in bold text**).

Anonymous Referee #3

General Comments

Tianle Pan et al. present a study on the influence of the heating effect of oxidation flow reactor (OFR) UV lamps on oxidant chemistry, flow conditions and secondary organic aerosol (SOA) formation. The authors find that while the increase in temperature does not greatly affect the oxidant chemistry, the effect of temperature gradient on residence time distribution shortens the mean residence time. Thus, the OH exposure at higher light intensities is affected by the heating effect. The temperature increase in the OFR influences the SOA yields and composition, so that the SOA yield is lower at higher temperatures and the O to C ratio of SOA is higher at higher temperatures.

The heat caused by the OFR lamps is an important aspect of OFR studies that has not been well characterized earlier. The measurements and analysis in this study are comprehensive and thoroughly done. However, the quality of reporting and language is not fully adequate. Another concern is the relevance of the SOM model for the SOA formation in the OFR. Thus, I recommend publishing this study in Atmospheric

Measurement Techniques, but only after the following remarks have been addressed. Especially the language needs to be improved.

Major Specific Comments

R3.1: The reporting should be concise, and all statements should be well-defined and unambiguous. Currently, the text contains unnecessary extra words (like 'around', 'might'), which are sometimes necessary but could be mostly removed. The authors often use word 'support', which in many places could be replaced by a more exact wording. Furthermore, for many sentences in Section 3 it is unclear what the authors actually mean. Some of these are included in the following specific comments, but I recommend the authors read through the paper and for each sentence ponder whether the sentence is necessary, and whether the statement is unambiguous.

A3.1: We appreciated the reviewer's suggestions and comments. We carefully examined the manuscript sentence by sentence and revised the whole manuscript thoroughly. As shown in the tracked version of the manuscript, most of the sentences (>70%) were carefully revised. Here are some examples:

(1) Line 399-401: "**This indicated that the primary T/RH sensor in the backplate with default OFR settings lead to underestimation of the temperature inside of OFR, which should be verified and corrected by the users based on the configuration of their instrument.**" We replaced "**suggests**" with "**indicated that**" and "**might lead**" with "**lead**"

(2) Line 243-245: "**These results indicated that the temperature increase inside of the PAM-OFR was mainly due to the heat from the lamps, which was further confirmed by Fig. 3b**". We replaced "**supported**" with "**indicated**" and "**confirmed**".

(3) Line 255: "**assuming a mixing water ratio of 1.88%" and Line 255: "assuming ambient OH concentration of 1.5x10^{6} molecules cm^{-3}**". We deleted "**around**" before "**1.88%**" and "**1.5 x10^{6}**".

(4) Line 271-273: We modified "**This inconsistency is mainly due to the lamps starting at 10 V with colder conditions (e.g., room temperatures or lower**

voltage settings), meanwhile, the OFR reactor has a thermal mass that needs time to accumulate or dissipate heat." to "**This discrepancy was primarily attributed to the fact that the lamps were initiated at 10 V under cooler conditions (e.g., room temperatures or lower voltage settings), while the OFR reactor had a thermal mass that required time to accumulate or dissipate heat.**"

(5) Line 629: "**cold air can also be effective ways**". We replaced "**might**" with "**can**".

R3.2(a): It is unclear why the authors used the ring flow manifold in RTD measurements when it was not used in the simulations. In my opinion, this is justified since the ring flow is typically used in actual measurements. However, the authors could discuss this justification when they describe the RTD measurement and simulation. This difference between measurement and simulation should also be mentioned in Fig. 8 caption.

A3.2(a): Thanks for reviewer's comments. Previously, we measured the RTD by sampling through ring flow. Then, we also realized that the simulation and measured results shall use the same flow set. When we submitted our manuscript to AMTD, the measured RTD results were already updated to these measured by sampling only from aerosol line at 5 L min^{-1}. However, we forgot to change the discussion. Thus, the modelled and measured RTD shown in Fig. 8 shall be consistent with each other. The sentence of "**This inconsistency is probably because i) we only considered the airflow sampled from the center outlet in the backplate, but not the ring flow manifold, which caused more recirculation**" was deleted.

R3.2(b): In addition, I think the description of RTD measurement (p. 13, l. 412-415) should be moved to Section 2.

A3.2(b): Yes, we moved the description for the RTD measurement to Sec. 2.

"In addition to the temperature measurement in OFR, we measured the residence time distribution (RTD) at different voltages to explore the effect of temperature on RTD. Specifically, we first turned on the lights to make the temperature stable. Then, a 2 s pulse of 50 ppm SO_2 was injected into a 5 L min^{-1}

carrier gas (N_2) with RH <10%. N_2 was selected as the carrier gas to prevent the reaction between SO_2 and the generated oxidant when UV lamps were turned on. We measured the RTD with lamp driving voltage set at 0 V, 5 V and 10 V. Note that we only used the outlet for aerosol line for sampling (5 L min^{-1}) during the RTD measurement for better comparison with simulation results in Sec. 2.3."

R3.2(c): What were the flow rates in aerosol line and exhaust line during RTD or temperature measurements? The authors provide the flow rates in different occasions, but it is not clear whether this is the total flow rate (aerosol line + exhaust line) and what is the ratio between the aerosol line and exhaust line.

A3.2(c): Most of the time, sampling through the exhaust line was used for the temperature experiments. However, the ratio between aerosol line and exhaust line shall play very minor impact on the absolute temperature enhancement measured in side of OFR. We clarify the flow sampling strategy in the maintext: "**Most of the temperature experiments were done with sampling exhaust line from the ring flow.**". For the RTD experiment, the flow sampled from the aerosol lines was displayed in Fig. 8. We clarify this in the main text "**Note that we only used the outlet for aerosol line for sampling (5 L $min^{-1)}$ during the RTD measurement for better comparison with simulation results in Sec. 2.3.**"

R3.3: The authors use term 'Enhanced temp.' in figures to describe the difference between OFR and ambient temperature. In my opinion, e.g. 'Temp. enhancement' or just ΔTwould be better.

A3.3 We replaced all the 'Enhanced temp.' with $\Delta T_{(OFR\text{-}amb.)}$ in the figures in the revised manuscript.

R3.4: When discussing the approaches to reduce the heating effect in Sect. 3.5, the authors actually only discuss approaches to reduce the effect of increased temperature. For example, when the fans are used to cool down the OFR external surface, the heat transfer inside the OFR is improved and this affects the RTD since the heat transfer occurs via convection. However, the authors did not characterize the effect of different

cooling methods on the RTD, which at least should be mentioned in this section or the section headline should be changed.

A3.4: We appreciated the reviewer's reminding. This is a really good point. Corresponding explanation about the potential influence on RTD was added in this section. **"Cooling down the OFR would also affect RTD since the heater transfer occurs via convection inside of OFR, which needs to be further investigated in the future."**

R3.5: The authors sometimes mix past and present tense. See e.g. Fig. 3 caption: four lamps were turned on, flow rate is 5 lpm.

A3.5: We examined all the tenses used in the text and made corresponding corrections. All the revisions can be seen in the tracked version of manuscript.

R3.6: Based on the current description of the SOM model, it seems that the authors first model the SOA formation in "normal" temperature and then study its evaporation in elevated temperature (e.g. p. 17, l. 494: "for the newly formed SOA in the OFR, the temperature impact was simulated based on SOM model"). In that case, what is the temperature where the SOA formation is modeled? If this is the case, the model results regarding e.g. the SOA yield are not very relevant, because in the OFR the SOA formation would take place in the elevated temperature.

The authors should change the heading of Section 3.4 to "Temperature influence on OA evaporation", and describe more clearly that they are modeling the evaporation of SOA that was formed in temperature X and then injected into the OFR at elevated temperature. This needs rewriting of Sect. 3.4 so that the authors discuss OA evaporation instead of SOA formation.

A3.6: I think there is a misunderstanding. The OA evaporation and SOA formation are two separate topics in our discussions. We simulate the SOA formation using SOM under different temperatures directly. No OA seed was considered. To clarify, we separate the original section 3.4 into two sections, which is "**3.4 Temperature influence on the evaporation of ambient OA**" and "**3.5 Temperature influence on**

the SOA formation".

Corresponding explanations were added in the section 3.4: "**Here, we estimated the potential losses of input ambient OA due to evaporation under enhanced temperature in OFR. This estimation is mainly based on literature results and modeling work.**"

Specific comments

R3.7: p. 1 l. 44: box model using radical chemistry → radical chemistry box model

A3.7: Corrected

R3.8: p. 2 l. 82: The high temperature inside the OFR does not cause the recirculating flows, it is the temperature gradient caused by lamp heating.

A3.8: Corrected

R3.9: p. 3 l. 88: SOA simulation and study → SOA simulations and studies

A3.9: Corrected

R3.10: p.3 l. 102: The acronym ARI has not been declared earlier.

A3.10: We added the definition in line 101: "**The PAM-OFR (Aerodyne Research, Inc., abbreviated as ARI) used in this study…**"

R3.11: p. 4, Fig. 1: What is the exhaust gas line? It seems that sensor (3) is measuring the ring flow outlet, but in the text it is unclear whether the exhaust gas means the ring flow or the N2 purge flow. Please clarify.

A3.11: The exhaust gas line means the ring flow. We added the explanation in line 336 and specify the exhaust gas line come out from ring flow in Fig.1:

Line 338: "The dissipation of energy through the exhaust air **(from the ring flow)** from the PAM-OFR"

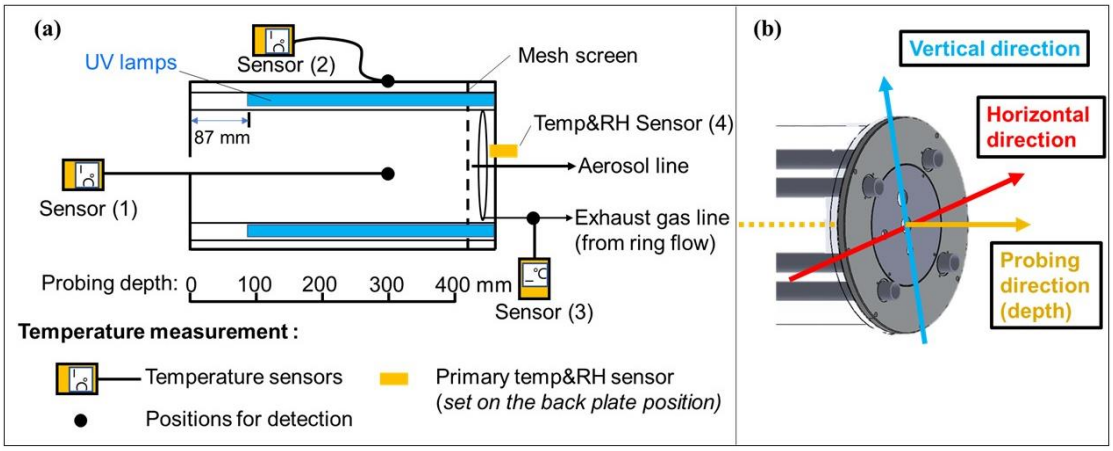


Figure 1: (a) Schematic plot for temperature measurement in the oxidation flow reactor of this study and (b) directions for temperature measurement. The center inlet, nut, and mesh screen near the front plate were removed when the temperature sensor was probed in the front direction. The information of different temperature sensors used can be found in Table S1.

R3.12: p. 7 Fig. 2 caption: "assuming ambient OH concentrations are around 1.5e6..." I suppose you have used an exact value of 1.5e6 in the calculations, so please remove the word 'around' (also earlier regarding the water mixing ratio).

A3.12: We removed the 'around' in the caption of Fig.2. And we modified the expression in line 238 for mixing ratio: "**(assuming the water mixing ratio is 1.88%, RH=60%, external OH reactivity=30 s^{-1})**."

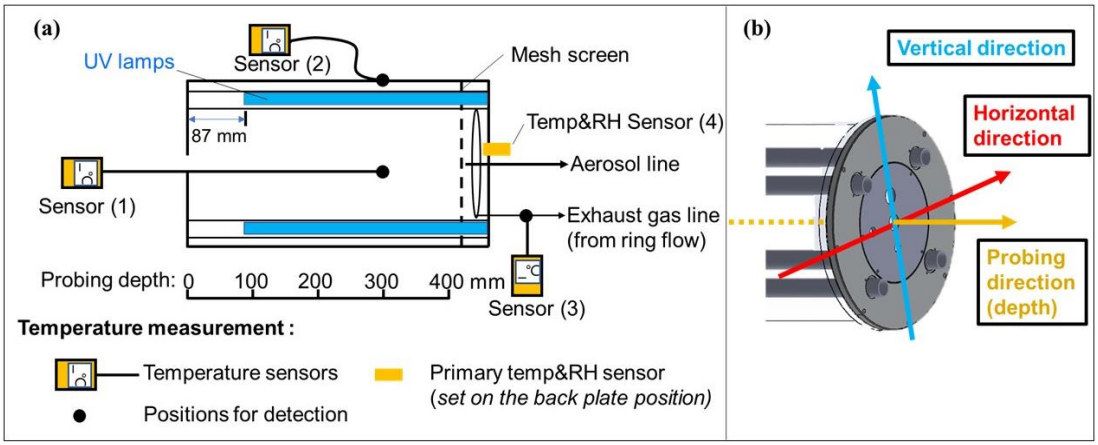
R3.13: p. 10 l. 296: "In our case, the..." → "In our case, when the..."

A3.13: Corrected

R3.14: p. 10 l. 298: What is "vertical axial direction"? I think axial would mean the direction of the central axis of rotation. It would be helpful to define the different directional terms (vertical, horizontal, probing depth) graphically e.g. by adding another panel in Fig. 1.

A3.14: Following reviewer's suggestions, we defined the direction of measurement in Fig. 1b and modified the text in line 126-128: "**Briefly, we measured the air temperature inside PAM-OFR at varied positions (vertical and horizontal**

directions, as well as different depths from inlet (Fig. 1b)) under different lamp configurations (e.g., number of lamps, types of lamps, intensity of lamps) and flow rates.”



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R3.15: p. 11 l. 319: “OFR chambers (OFR metal tubes)” → “OFR surface”

A3.15: We replaced the “OFR chambers (OFR metal tubes)” with “OFR surface”.

R3.16: p. 11 l. 331: surfaces → surface

A3.16: Corrected.

R3.17: p. 11 l. 333-339: The authors discuss in length why the temperature of the external surface is lower than that of the gas inside the reactor, while it is basic physics that since the OFR is not isolated and the ambient temperature is lower than the OFR internal temperature, there will be heat transfer from inside the OFR to the ambient, and the medium between these (the surface) will be at lower temperature than the OFR internals. This is correctly described in lines 338-339. The thermal mass of the OFR is not relevant, as it affects only the rate of temperature increase but not the final equilibrium temperature that is discussed here.

A3.17: We agree with the reviewer's comments on balanced condition. However, when the temperature equilibrium between OFR tube and air was not balanced, the thermal mass would be a reason as well. To reflect that we move the original cause "ii" (heating transfer) to be cause "i". Then explain the original cause "i" only work when the thermal system was not balanced.

" ii) When the temperature equilibrium between the air and OFR was not balanced, an additional reason will cause the lower temperature in OFR tube. The aluminum OFR chamber has a higher thermal mass than the air. Although the specific heat capacity of metal (0.908 J g^{-1} K^{-1} at 301.60 K) is similar to that of air (1.005 J g^{-1} K^{-1} at 300 K) (Giauque and Meads, 2002; Kieffer, 1956), the flow tube is considerably heavier than the air due to its significantly higher density (2700 Kg m^{-3} vs 1.29 Kg m^{-3}), resulting in a lower temperature for the OFR tube than the inner air."