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 #1

General Comments

The manuscript by Pan et al. investigates the impact of lamp-induced heating in an Aerodyne PAM-OFR, assessing the temperature distribution, flow dynamics, and chemical consequences resulting from UV lamp heating. The authors have used CFD simulation, KinSim kinetic model, and SOM model to investigate how the temperature affects the flow and average OH exposure and how the enhanced temperature impacts the chemistry of gas-phase reactions and SOA formation. They find that the temperature enhancement can be up to 15 ℃ and it has impacts on the gas-phase chemistry and the yield, size, and oxidation levels of SOA. Overall, this manuscript gives a relatively comprehensive evaluation of the increased temperature on the chemical processes in the PAM-OFR. However, some concerns need to be addressed before the manuscript can be considered for publication in AMT.

Major Specific Comments

R1.1: The authors find that the heating inside PAM-OFR is mainly from the heat transfer of the hot quartz sleeve (heated by the lamps) but not from the optical radiation.

This is true since UV radiation generates little heat. Based on this finding, I would expect that the authors recommend moving the lamps out of the reactor, which can overcome the heating issue caused by the lamps. This can be found in the design of other OFRs in previous studies (e.g., Huang et al., Atmos. Meas. Tech., 10, 839–867, 2017; Simonen et al., Atmos. Meas. Tech., 10, 1519–1537, 2017; Li et al., Atmos. Chem. Phys., 19, 9715–9731, 2019) and should be discussed in "Section 3.5 Approaches to reduce the heating effect".

A1.1: We agree with that moving the lamps out of the reactor will help reduce the temperature increase. However, we do not think this method can overcome the heating issue. For examples, additional cooling methods were also applied for OFRs with UV lamps mounted outside. e.g., Huang et al. (2017) used circulating water to cool down the system; Li et al. (2019) used 30 L min^{-1} N_2 through the quartz tubes; Four fans were used to dissipate the heat in Xu and Collins (2021); The temperature increase of the tube wall could be 8 **°C** inside the Go:PAM when the intensity of UV lamps was maximum and the fan was turned on (Watne et al., 2018).

Following the reviewer's suggestions, we declare that moving the UV lamps outside of OFR is a method to mitigate the heating issue.

"**Moving the UV lamps outside the tube and designing a cooling system on the outer surface of OFR with circulating water or cold air can also be effective ways to improve the temperature control inside of OFR (Watne et al., 2018; Xu and Collins, 2021; Huang et al., 2017; Liu et al., 2018; Chu et al., 2016; Zhao et al., 2021; Li et al., 2019), however, these will require a substantial redesign of the hardware of OFR tubes and are beyond the scope of this manuscript. And mounting the lamps outside of the OFR limits the use of OFR185 mode due to the low transmission efficiency of quartz glass for light at 185 nm (Simonen et al., 2017) and OFR254 mode is usually used**."

R1.2: The authors use the SOM model to investigate the influence of temperature on SOA formation, which highly relies on the performance of the model under different temperatures. It would be helpful to conduct SOA formation experiments with different temperatures to get accurate decreases in SOA yield under high temperatures. This

comparison can be done with or without efficient heat removal methods including a high volume of N2 purge air and external fans as the authors have shown in the manuscript.

A1.2: Thanks for reviewer's suggestions. We have incorporated these suggestions and did two experiments to prove that the SOA formation in the OFR was indeed decreased when the lights were on, as shown in Fig. S19 the revised manuscript. However, we cannot calculate the yield due to the PTR-MS which can be used to measure the VOCs mass concentration was broken in recent several months and still in repairing. Thus, we cannot compare the measured results with the SOM model simulation. More experiments will be done in the future.

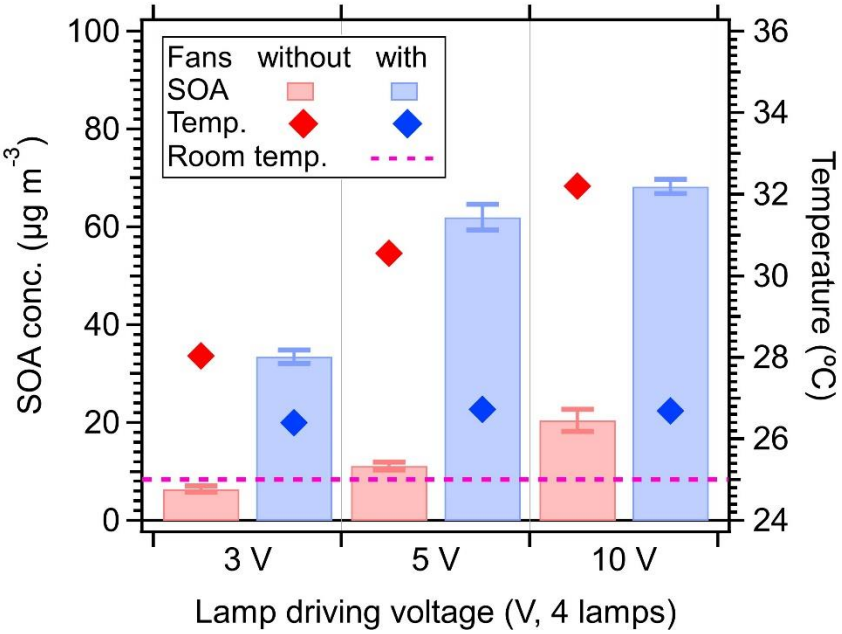

Figure S19. The SOA formation from benzene and OH radicals in the PAM-OFR as a function of light intensity. Two cases including PAM-OFR was blown with fans and without fans were both shown. The room temperature and temperature measured with the primary Temp&RH sensor set in the back panel were shown in the right axis. Note the OFR temperature reported here is the lower limit as discussed in section 3.1.5. The gas-phase benzene (99.80%, Sigma-Aldrich) was generated with syringe pumps. Benzene was used as gas-phase precursor in this experiment due to its lower k_{OH}, since benzene will not be totally consumed under the high OH exposure at high voltage settings in OFR. The flow rate in this experiment was 4.5 L min^{-1}, and the RH was ~30%.

The explanation was also added in the revised mian text:

"To confirm the model results, we did a simple laboratory experiment and found the formed SOA masses was indeed substantially decreased in OFR due to the heating effect (Fig. S19), which is consistent with the simulated model results."

R1.3: Similarly, SOA formation experiments with different voltage setting strategies need to be added in Section 3.5 to show the effectiveness.

A1.3: The SOA formation experiment between benzene and OH radicals was done to prove the effectiveness of the cooling method. The detailed results can be found in A1.2.

R1.4: The high temperature also leads to lower RH. How would this influence the SOA formation?

A1.4: Thanks for reviewer's question. Higher temperature indeed led to lower RH due to the increased dew points. The literatures have suggested that although some studies have found that the variations of RH can influence the SOA formation, the influences were complex and in conflict. To reflect the question by the reviewer, we added the statement in the main text:

Line 598-606: "**In addition to the direct influences, the increase of temperature within OFR lead to the decreases of the relative humidity (RH), which can also impact SOA formation. However, the literatures show that the impact of RH on SOA formation remains inconclusive. For example, Tillmann et al. (2010) found the SOA yield was higher at humid conditions (RH: 40-70%) compared to dry conditions (RH: 0-10%) as the RH influenced the formation of products in α-pinene ozonolysis experiments. In contrast, Zhang et al. (2019) found the SOA yield of m-xylene-OH oxidation decreased as RH increased in a chamber study, as the high RH led to the less formation of oligomers and inhibited the reaction of RO_2 autoxidation. Thus, elucidating the influence of humidity on various SOA formations is still a challenge and falls outside the purview of our research topic here. In addition, given the short residence time within OFR (seconds to minutes), the impact of liquid phase reactions to SOA formation in OFR should be minimal.**"

R1.5: It is confusing when comparing Figure 3 and Figure 6b. (1) The horizontal distance is >400 mm in Fig. 3 but <200 mm in Fig. 6b. (2) The temperature shows a monotonic increasing trend from the inlet to the outlet in Fig. 3 but a minimum in the middle in Fig. 6b. Can the authors further explain the differences?

A1.5: To clarity, we added an illustration of vertical direction, horizontal direction, and probing direction (depth) in Fig. 1(b), as shown below. The horizontal and vertical directions formed a plane perpendicular to the probing direction (depth);

For the question (1), the x-axis in Fig. 3 was the probing depth from the inlet to outlet in the probing direction (460 mm in total, as shown in Fig. 1a). The x-axis of Fig. 6b shows the horizontal distance in horizontal direction (Fig. 6b). For the question (2), the x-axis of the two graphs did not indicate the same position. In Fig.3, all the temperatures were measured at the center line (the horizontal distance was at 0 mm) from the inlet to outlet in the probing direction. These positions were the same as the markers with a horizontal distance of 0 mm in Figure 6b, where a lower temperature at 100 mm were also shown.

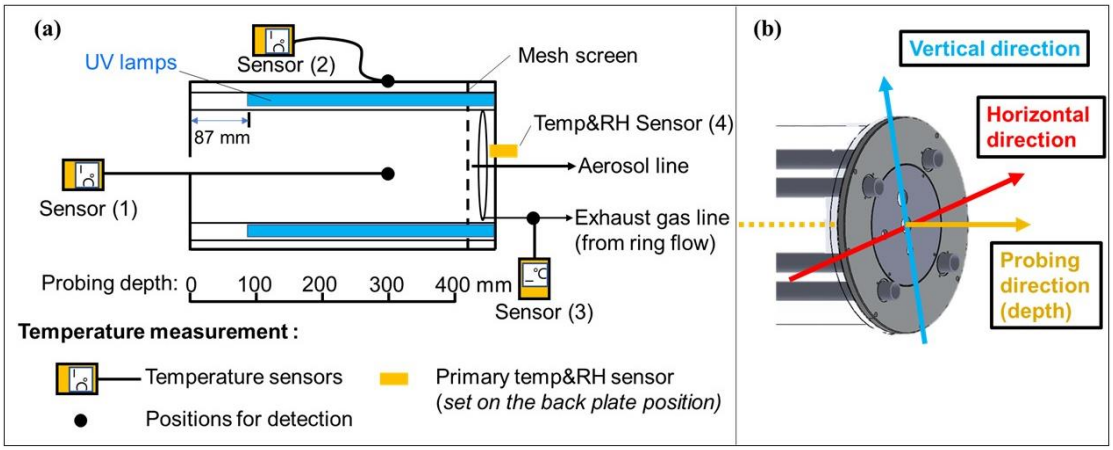

Figure 1: (a) Schematic plot for temperature measurement in the oxidation flow reactor of this study and (b) directions for temperature measurement. The center inlet, nut, and mesh screen near the front plate were removed when the temperature sensor was probed in the front direction. The information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

R1.6: Although PAM-OFR is the most commonly used OFR, there are many other types of OFRs. For other OFRs that put lamps outside of the reactor (like the ones listed

above), the heating issue is not as serious as PAM-OFR. Using the terminology “OFR” in the Conclusion may lead to misunderstanding. Therefore, I would suggest the authors use the terminology “PAM-OFR” rather than “OFR” throughout the manuscript.

A1.6: We replaced the “OFR” with “PAM-OFR” in the manuscript when the PAM-OFR was specifically referred to. This revision certainly makes our statements more rigorous.