

We thank the reviewer for the examination of our work. Here are the point-by-point responses to their comments. In blue are our answers, and the text that has been added to the manuscript, is given in *blue and italic*.

## Reviewer 1:

### Comments and answer:

The hydroxyl (OH) radical is the pivotal oxidant in the atmosphere, its accurate measurement is essential for understanding atmospheric oxidation capacity, and thus air quality, and climate change. However, the accuracy of OH measurements using the laser-induced fluorescence (LIF) technique is unavoidably compromised by wavelength drift of the excitation laser. Therefore, a reference system capable of monitoring the laser output wavelength and actively locking it to the optimal OH excitation line is crucial for long-term, reliable measurements. In this manuscript, Chen et al. develop a compact real-time reference system for wavelength locking in LIF-FAGE measurements. Through comprehensive characterization of key parameters, an optimal operational window for stable and high-concentration OH generation and detection is identified. The system's high stability is convincingly demonstrated by continuous measurements over 12 hours, with exceptionally low drift (0.2% per hour) during the first 9 hours. The detailed description of this system will be valuable for other researchers aiming to implement wavelength-locking techniques to improve the stability of their measurement systems. Overall, I recommend its publication after considering the following minor comments:

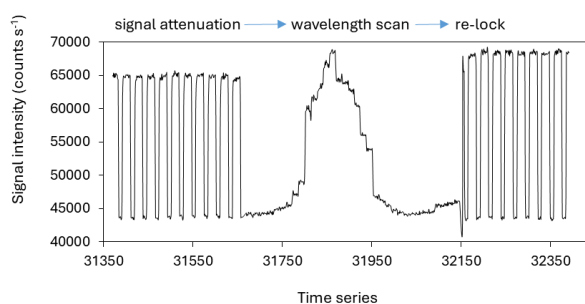
- In lines 74-75, it mentioned that the AIOFM group used a 282 nm laser as excitation laser, I know that they have updated the excitation laser to 308 nm, please see this paper (*Wang et al., "Development of a field system for measurement of tropospheric OH radical using laser-induced fluorescence technique"*.) for detail information, and add the corresponding updated description to the main text.

*Answer: We thank the reviewer for pointing this out. We have added this critical information to the manuscript, and corresponding descriptions has been adjusted:*

*The AIOFM group has provided initial characterization data for their earlier version reference cell, however, a 282 nm laser was employed (Xing et al., 2017). In their upgraded system, a 308 nm excitation laser was used, and good stability was reported: a fluorescence fluctuation of approximately 1.2% over 15 minutes at the resonance wavelength (Wang et al., 2019).*

- In lines 292-293, it says that, during the 12-hour test, the signal attenuation never exceeded the threshold to trigger the wavelength re-locking program. For a complete performance evaluation, it would be informative to know if and how the system performs when a drift event does trigger the full corrective sequence (wavelength scan and re-lock). Could the authors comment on this? Have they tested the system's response to induced, larger wavelength drifts?

*Answer: We thank the reviewer for this comment. The figure below illustrates a recorded event in which OH signal attenuation triggered the wavelength scan and subsequent re-lock process. It should be noted that this process is currently for display purposes only and was captured by an external standalone program, we can see that the x-axis represents time series rather than wavelength. For this reason, the data are not included in the main text. Nevertheless, this example helps demonstrate the capability of our reference system to effectively re-lock to the optimal OH excitation wavelength.*



*Figure s1. The full corrective sequence captured by an external standalone program.*

- In lines 296-298, the suggestion to normalize ambient OH data against the concurrent reference cell signal during wavelength drift should be treated with caution. The physical conditions (e.g., temperature, pressure) inside the reference cell and the ambient sampling cell are different. These differences can lead to slight variations in the OH excitation spectrum. Consequently, the rate of signal attenuation due to laser wavelength drift may not be identical in both cells. Using the reference signal for direct normalization

without carefully characterizing and correcting for this potential discrepancy could introduce additional error. Therefore, I recommend either removing this statement or significantly qualifying it with a discussion of the necessary corrections and associated uncertainties.

Answer: Thank you for this important comment. We agree that normalizing ambient OH signals using the reference cell signal requires careful consideration. In response, we have removed this statement from the manuscript to avoid potential misinterpretation.

- In Figure 4, there is a typographical error in the labeling of the OH excitation spectrum. The last characteristic peak within the red frame should be  $P_1(1)$ , not  $P_1(3)$ .

Answer: We have corrected the label in Figure 4.

- In Figure 4, an offset appears between the two OH excitation spectral lines, causing them to fail to correspond precisely. What might account for this phenomenon?

Answer: Thank you for pointing this out. Although the two OH excitation spectra show good agreement, a slight offset is observed. This may be attributed to different broadening effects due to slight differences in pressure and a more noticeable temperature difference between the reference cell and the OH detection cell. While both cells operate at approximately 0.4 kPa, minor pressure variations can occur depending on pump performance. More significantly, the reference cell uses a heated filament to generate OH radicals, resulting in a higher internal temperature than the detection cell, leading to different Doppler broadening effects. We therefore conclude that both temperature and pressure differences contribute to the slight spectral offset.

- In Section 3.2, the author did not specify the exact installation location of the THP sensor. Additionally, the filament's position and temperature were not considered in the author's experimental conditions. Would the filament's temperature rise during heating interfere with the humidity measurement results shown in Figure 8?

Answer: The THP sensor is installed on the inlet gas line upstream of the reference cell, providing real time measurements of the incoming gas temperature, relative humidity, and system pressure. Regarding the filament, its position and temperature are indeed critical for OH generation. The filament's mounting position is fixed mechanically. Experimentally, we align the laser beam to pass as close as possible to the filament surface in order to maximize fluorescence from generated OH radicals, however, the exact distance was not measured and is therefore not specified. While the THP sensor is located far from the filament and does not directly measure filament temperature, we could infer thermal behavior from background signal intensity. **Under stable operating conditions at fixed power, the filament temperature reaches thermal equilibrium rather than continuously rising. This is proved by the stability of the offline background signal in long-term tests, as shown in Figure 9, which indicates that thermal radiation, a major component of the background, remains steady.**

In the related humidity experiments, we observed a slight decrease in offline background signal (~1%) at RH above 60%, as shown in Figure s2, possibly because higher humidity slightly enhances heat removal by the pumped gas stream, and thus causes slight filament cooling. This cooling could also influence the OH production, although Figure 8 presents net OH signals increment (signal at online wavelength minus signal at offline wavelength), it is hard to demonstrate that subtle humidity-induced thermal interference is entirely ruled out.

However, in the RH range below 60%, humidity-induced variation in background signal, and by extension, in filament temperature, remains negligible. The observed change in the offline background signal across this range was only  $0.154 \text{ kcounts s}^{-1}$ , representing a relative variation of 0.23%. Over the same RH interval, the online signal increased by  $13.279 \text{ kcounts s}^{-1}$ , corresponding to a 7.0% rise in raw signal intensity. Even under the conservative assumption that the entire background decrease results from filament cooling, and that such cooling would proportionally affect the online signal, the corrected online signal increase would be  $13.309 \text{ kcounts s}^{-1}$ . The resulting increment remains 7.1%, which is statistically indistinguishable from the uncorrected value within typical measurement uncertainties.

In conclusion, within relative low RH regime, the influence of humidity on filament temperature, and consequently on the detected OH signal, is insignificant compared to the direct effect of water vapor concentration on OH production. However, at higher RH levels, multiple effects could slightly influence the OH signal. Water vapor not only contributes to high OH production but also enhances OH fluorescence quenching and may induce filament cooling. These likely contributes to the larger signal variability observed at elevated humidities. We will address this point in the revised manuscript to better support our recommendation that RH is better maintained within the 30–40% range for optimal system performance:

... Although the nonlinear response suggests that the optimal OH signal occurs around 74% RH, such a high humidity

level would cause slight filament cooling and thus complicate signal interpretation and increase system instability. In addition, high RH levels would also accelerate the aging of electronic components and increase long-term operational risk. Therefore, in practice, RH is maintained within the 30%–40% range, which balances signal intensity, sustained system stability and performance.

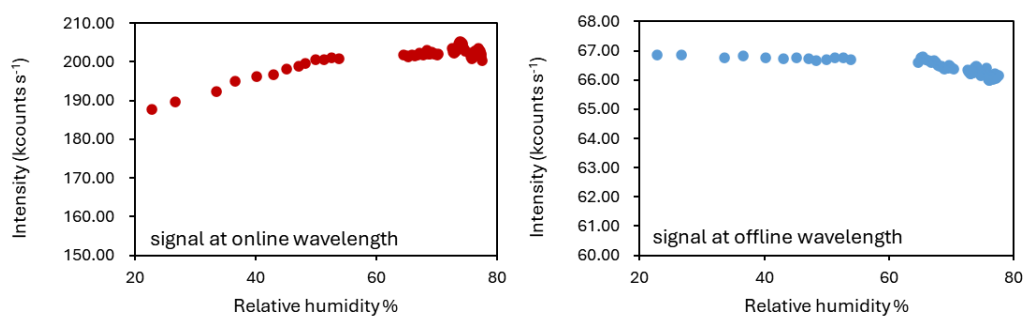


Figure s2. Measured raw signal intensity from the reference cell at online wavelength (left) and offline wavelength (right) as a function of relative humidity%.

## Reference

Wang, F., et al., Development of a field system for measurement of tropospheric OH radical using laser-induced fluorescence technique. *Optics Express*, 2019. 27(8): p. A419-A435.

Xing, X., et al., Study of a laser wavelength correction method applied to the measurement of OH Radical with laser-induced fluorescence. *Guang Pu Xue Yu Guang Pu Fen Xi*, 2017. 37(3): p. 692-6.