Responses to the Reviewer 3

Quantitative measurements of total OH reactivity (k_{OH}) provide important insights into atmospheric photochemistry. This paper reports the development of a portable LP-FRS instrument for real-time and in-situ measurement of k_{OH} . The size of the instrument is reduced to $130 \times 40 \times 35$ cm. Its advantages in cost, operation, and transportation make it of great value in field observation and laboratory research. The optical and mechanical structure, the key MPC subsystem, parameter optimization, laboratory performance analysis, and field operation demonstration of the instrument are introduced in detail. I recommend acceptance after considering the following minor comments:

We thank the reviewer for the thoughtful and thorough reviews. Point-by-point responses to the comments are attached below. We have made corresponding modifications, and these changes are marked in the revised manuscript.

1. Attention should be paid to textual details, such as the missing "x" in Fig.6(a), line 159 (missing r_c unit), and the inconsistencies of time resolution value between line 299 and caption of Fig.7(a).

Done. We checked the manuscript and revised textual errors.

2. Table 1 does not seem to show the performance, but rather the compactness of pumpprobe multi pass cells. It would be better to make a modification.

For a pump-probe optical system, the effective absorption path length and overlapping factor are the best indicators of the system performance. Meanwhile, the overlapping factor reflects the system compactness which benefit in reducing instrument size. Therefore, we made the comparison presented in Table 1. To further clarify the point, we made modifications in the manuscript.

Table 1. <u>Overlapping factor</u> comparison of MPCs used for laser photolysis inpump-probe techniques.

In fact, from an optical structure perspective, the overlap factor characterizes both the utilization efficiency of the optical path length and the compactness of the pump-probe MPC, making it an ideal parameter for performance characterization. To demonstrate the efforts in reducing instrument size, a comparison of effective overlapping path lengths and overlapping factors with literature reported <u>pump-probe</u> MPCs is shown in Table 1.

3. What is model of the UV lamp used to generate O_3 ?

We added the model to the manuscript.

To produce sufficient OH in the MPC, a small flow rate (~ 0.25 L/min) of zero air which passed through a UV lamp <u>(UVP PenRay, Analytikjena)</u> and a bubbling bottle to generate O_3 and water vapour, is added to the main sampling flow (~ 6.0 L/min), resulting in a total flow rate of 6.25 L/min.

4. Adding some explanations to the beginning of line 67 of the Introduction section will be more helpful in explaining the problems of the current methods and instruments.

Done. At the beginning of this paragraph, we introduced several intercomparison experiments and then made a brief summary to describe the common limitations and advantages of current techniques and instruments.

Several intercomparisons of the techniques and instruments mentioned above have been conducted. Zannoni et al. (2015) reported a field intercomparison of two CRM instruments in the Mediterranan basin. Hansen et al. (2015) carried out an intercomparison between the CRM and LP-LIF techniques in an urban environment. The series of intercomparison experiments conducted in the SAPHIR simulation chamber at Forschungszentrum Jülich involved all types of existing methods and nine instruments from around the world (Fuchs et al., 2017). The result shown that the indirect or semi-direct methods exhibited more scattered in measurements and are most likely limited by the corrections for known effects, such as high NO concentrations for CRM and high reactivity conditions for FT-CIMS. In comparison, the direct methods (LIF) that combine laser-flash photolysis offer advantages in detection precision and accuracy.

5. The role of r_c is clear because it directly affects the overlap. However, the effect of g value is not obvious and needs to be briefly explained.

We added the description of g value.

where g ($g = g_1 = g_2 = \cos \theta = 1 - d / R$) is the parameter that describes the optical resonance stability of optical cavity or MPC. When the q value is less than zero, the base length of the multi-pass cell exceeds the curvature radius of the mirror, causing the resonance to become unstable. An incident parallel beam or a beam whose waist is not properly aligned with the cell's centre will quickly diverge and cannot be collected. However, a beam with its waist well matched to the centre can effectively prevent divergence. θ is half of the angle between two adjacent reflection light points on mirror surface. d is the base length, $R_1 = R_2 = R$ is the curvature radii of the mirrors. r is the radius of the spot distribution circle.

6. Why is the error bar of OH+NO in Fig.6 larger than the other two experiments?

This is due to two reasons. One is that the measurement averaging time is different from the other two experiments. The other may be that the reaction rate of NO+OH is very fast. The flow fluctuations can cause larger measurement fluctuations.

7. Line 261. The expression of "the slope errors of the fittings were less than 0.1" is not clear.

We directly put the slope errors into the reaction rate constant values in the revised manuscript to clarify the expression.

As shown in Fig.6, the obtained reaction rate constants for OH + CH₄, OH + CO, and OH + NO at 298 K were found to be $k_{\text{OH+CH}_4} = 6.4_{+0.1}^{-0.1} \times 10^{-15} \text{ cm}^3 \text{ molecule}^-$

¹ s⁻¹, $k_{\text{OH+CO}} = 1.6_{+0.1}^{-0.1} \times 10^{-13} \text{ cm}^3 \text{ molecule}^{-1} \text{ s}^{-1}$, and $k_{\text{OH+NO}} = 3.0_{+0.1}^{-0.1} \times 10^{-12} \text{ cm}^3$

molecule⁻¹ s⁻¹, respectively.

8. Line 312. The residual reactants in water are also one of the sources of uncertainty affecting the zero determination of the instrument.

We added the uncertainty to the manuscript.

The uncertainty of the instrument zero primarily originates from various influencing factors, such as changes in O_3 concentrations and residual reactants in zero air and water.