Response to Anonymous Referee #1

We want to thank the referee for the positive words, helpful comments and suggestions. We have revised the manuscript according to the comments

(1)

Reviewer: Page 30626. Line 1. It is not clear how the pressure dependent calibrations are performed. Were different pinhole diameters used to simulate the change in pressure in the cell owing to sampling at different altitudes? Or was a constant size pinhole used with a varying pumping speed. This procedure, even if it is described elsewhere, needs some attention there as the pressure dependence is critical. Is the assumption made that for sampling pressures (different altitudes) any losses of HOx on the sampling assembly is the same? A way around this assumption is to perform an in flight calibration, even being able to see a relative change in sensitivity with altitude is useful. In the GABRIEL paper an in flight calibration was discussed for this instrument (but not here).

Response: The calibration unit used provides different known concentrations of OH and HO_2 radicals in moistened synthetic air. The calibration air is drawn into the detection system through a nozzle. The pressure dependent calibration is performed using different pinhole diameters. Quenching and wall loss depend on pressure. While quenching increases with increasing pressure, wall losses (due to longer mean free diffusion way) become less. The pressure dependent instrument sensitivity considers both effects (see Figure 6 in the paper).
Martinez et al. (2010) report HO_x observations obtained in the tropical boundary layer and mid-troposphere. Good agreement was observed between ground-based and in-flight calibrations. However, HO_2 produced during in-flight calibration was always less than atmospheric HO_2 and comparable in magnitude to its natural variability during the calibration period. Therefore the in-flight calibration was used only to characterize the OH sensitivity of the instrument during the GABRIEL campaign.
The good agreement between the OH calibrations is also assumed for the HOOVER mission. According to the GABRIEL in-flight calibrations no HO_2 in-flight calibrations could be performed due to even lower H_2O mixing ratios in the mid-latitude upper troposphere over Europe.
We added to the calibration subchapter:
"The internal pressure was changed by using different pinhole diameters during calibration."

(2)

Page 30626, line 12. Is there a reason why a significant H2O dependency in the calibration was not seen in HOOVER compared with GABRIEL? It is a very similar instrument?

During the GABRIEL and HOOVER campaigns the same instrument was used in a similar setup. However, minor changes were applied.
In contrast to the findings of the GABRIEL campaign, calibrations of the HORUS instrument performed during the HOOVER campaigns indicate no additional water effect.
Also, a fit is applied to describe the pressure dependence of the instrument sensitivity. The calculated fit with an additional water dependence results in a slightly lower chi-square compared to the fit without an additional water dependence since an added degree of freedom is applied. However, the chi-square is not that much lower as it would be expected for a significantly better physical description.

(3)
Page 30626, line 12. It is interesting that the CFD calculations show no wall contact until after the OH cell when the jet broadens and there is wall loss prior to the HO2 detection cell. Extending the calibration to different temperatures is an important landmark, as this is not typically reported. Although the ambient temperatures varies quite a bit, the temperature within the instrument sampling tube within the wingpod varies much less. It is interesting that the OH sensitivity did not really change much with lowering the temperature (a small increase was seen) but it dropped quite a bit for HO2 (about a factor of 2, which is a lot considering that the reported concentrations will directly mimic this). It is interesting that it is thought that the cooling down of the calibration tube or the electronics. I think there will be uncertainties in translating this dependence found in the laboratory to those that may prevail on the aircraft. However, the fact that a temperature dependent calibration has been performed on the ground is a step forward, but further studies of the temperature dependence are needed, particularly in flight.

We agree. Little is published about temperature dependences of OH-LIF-FAGE instruments. Hence further studies and the publication of the results are necessary to provide a broader picture about characteristics and performance of OH-LIF-FAGE instruments in cold environments.
A more detailed response is given to reviewer #2.

(4)
Page 30632. For INTEX-A where the model underestimates HO2 by up to a factor of 3 where there is a higher load of pollutants. Is this due to an interference for this instrument, was HO2 being measured which is significantly more than HO2 (as in the model)?*

(Fuchs et al., 2011) reported a potential HO_2 interference originating from oxidized alkenes and aromatics when NO is added in order to convert HO_2 into OH. The RO_2 interference signal can significantly enhance the HO_2 fluorescence signal, in particular under high NO addition flows.
In contrast to observations within the planetary boundary layer, airborne side-by-side measurement of OH and HO_2 in the troposphere comparing a LIF-based instrument (ATHOS) and CIMS-technique shows, in general, reasonable agreement between the techniques (Ren et al., 2012).

(5)
Fig 5. In order to calculate the flux some assumption needed to be made about the average photolysis time in the flow-reaction calibrator. What was this time?

We assume a turbulent flow. The Reynolds number (Re) gives a measure to access if a flow is turbulent or not. To a constant gas flow of 7 L/min of nitrogen different flows of N_2O were added, usually between 1 L/min and 7 L/min.

The Reynolds number is calculated through:

$$Re = \frac{\rho \cdot v \cdot d}{\eta}$$

ρ is the characteristic density of the fluid [kg/m³]

v is the velocity [m/s]
d is the characteristic diameter of the inner volume [m]
η is the dynamic viscosity of the gas [kg/(ms)]

The characteristic density of the gaseous nitrogen at 298 K is 1.15 kg/m^3 and 1.97 kg/m^3 for N_2O resulting in a mean characteristic density of 1.56 kg/m^3. With a pressure increase of 16%, the calculated velocity in the photolysis tube is about 4.08 m/s at a volume flow of 14 L/min. 0.007 m is the “mean” diameter of the inner volume. The dynamic viscosity of the gaseous nitrogen at 298 K is $17.9 \bullet 10^{-6}$ kg/(ms) and $15.0 \bullet 10^{-6}$ kg/(ms) for N_2O resulting in a mean dynamic viscosity of $16.5 \bullet 10^{-6}$ kg/(ms).

$$Re = \frac{1.56 \cdot 4.08 \ \cdot 0.007\, kg\, m^3\, s}{16.5 \ \cdot 10^{-6}\, kg\, m^3\, s} = \frac{0{,}044}{16.5 \cdot 10^{-6}} = 2700$$

The calculated Reynolds number for the flow of 14 L per minute is 2700 at 298 K. Since we observe similar actinic fluxes in the range of flows used (see Figure 5), we expect turbulent flow being established in the whole range.

Resulting from the gas flow of 14 L/min the residence time (average photolysis time) t of a molecule within the photolysis volume with a length of 3.6 cm is

$$t = \frac{0.036\, m\, s}{4.08\, m} = 0.009\, s$$

(6)
Fig 6. Say how the pressure was varied (changing pinhole, or same pinhole with different pumping speed?)

See above.

References

Fuchs, H., Bohn, B., Hofzumahaus, A., Holland, F., Lu, K. D., Nehr, S., Rohrer, F., and Wahner, A.: Detection of HO(2) by laser-induced fluorescence: calibration and interferences from RO(2) radicals, Atmos Meas Tech, 4, 1209-1225, DOI 10.5194/amt-4-1209-2011, 2011.

Ren, X., Mao, J., Brune, W. H., Cantrell, C. A., Mauldin, R. L., Hornbrook, R. S., Kosciuch, E., Olson, J. R., Crawford, J. H., Chen, G., and Singh, H. B.: Airborne intercomparison of HOx measurements using laser-induced fluorescence and chemical ionization mass spectrometry during ARCTAS, Atmos Meas Tech, 5, 2025-2037, DOI 10.5194/amt-5-2025-2012, 2012.