Review of Atmos. Meas. Tech. Manuscript (#amt-2019-439) "Calibration of an airborne HOx instrument using the All Pressure Altitude based Calibrator for HO_X Experimentation (APACHE)" by D. Marno et al.

We are thankful to the reviewer for the helpful and constructive comments.

General Comments

In this work, a new airborne HOx calibration system was developed and evaluated to mimic the conditions (e.g., varying pressure, temperature, and humidity) during a typical flight. This kind of work is important to determine the response of HOx instruments for the accurate airborne measurements of OH and HO_2, which is the key to understand the atmospheric photochemistry. A computational fluid dynamics model (COMSOL) was used to simulate the fluid dynamics in the calibrator. Two actinometric methods based on the photolysis of ozone and N2O (used in ground-based calibrator) were used to determine the actinic flux of the mercury lamp that is used to generate OH and HO2. Overall I found this manuscript needs major revisions. The difference in actinic flux measurement using the two methods is quite large. I would suggest conducting the actinic flux measurement in APACHE using the photolysis of N2O to rule out any uncertainties in transferring the ground calibration to airborne calibration. Section 5 is particularly lean and not well organized. More details and discussion should be included in this section (see details below). I also found many errors in equations and units and tried to point most of them out. Please check out the entire manuscript. I would ask the authors to consider the following special comments in their revision.

In light of the comments provided, we have made changes and provided clarification to the paper. Regarding the actinic flux measurements, we originally considered the lamp being a point source, which is wrong as the diameter of the lamp tube is 19mm. When considering the lamp as an respectively extended source of light with the corresponding beam profile we achieve a convergence between the two flux measurements with the HORUS transfer standard flux of 6.9 (± 1.1) $x10^{14}$ photons cm^{-2} s^{-1}, and the Ozone experiment yielding 6.11 (± 0.8) $x10^{14}$ photons cm^{-2} s^{-1}. The agreement between the two experiments have improved from a zeta score of 0.88 to 0.59, with the overall flux value being 6.37 (± 1.3) $x10^{14}$ photons cm^{-2} s^{-1}. This value is calculated by taking the average of the two methods weighted by their uncertainties. Section 5 has been merged with section 4, as we feel that the whole of section 4 entails results and discussion. Discussed elements that were in section 5 have been organized and expanded upon, please see comments and revised document.

Special Comments

1. L.18: For ground-based HOx instruments … (remove systems)

   Deleted "Systems"

2. L.26: Define COMSOL at its first appearance.

3. We have described COMSOL as a computational fluid dynamics model. Its origin is FEMLAB, a former toolbox of Matlab, which name is derived from ‘Finite-Element-Method-Laboratory’.
4. L.47: “Other methods have also been … (Remove “However”)

   Removed “However“
5. L.48: the CIMS work by C. Cantrell and L. Mauldin should also be cited here.

   “Cited C. Cantrell and L. Mauldin” .
6. L.61-69: Start this with a new paragraph. At the end of this paragraph (or maybe start a third paragraph), you might want to mention what was done in this work (e.g. establishment and evaluation of the APACHE, etc.)

   Now is a separate paragraph. Containing what was done in this work.
7. L.75: Define APACHE at its first appearance in the main text even though you have defined it in the abstract.

   Defined APACHE in its first appearance in the main text.
8. L.92: Figure 2 (capital F). Please check this throughout the manuscript.

   All references to a figure or table in text or otherwise have been capitalized as Figure or Table.
9. Fig. 1: “Controlled humidity airflow of 300 sccm”: is the 300 sccm of humidified air is enough to vary the humidity in the total flow of 200-900 sL/min mentioned in L.105?

   Typo. It was 300 sL min^{-1}. Figure corrected.
10. Caption of Figure 1: Maybe change it to “Overview of the APACHE system and the premixing setup. A picture at the bottom shows the perforated stainless steel plates with wool mesh.”

    Changed caption for Figure 1.
11. L.107: The word “respectively” is used but the air speed changes by a factor of less than 2 (0.9 to 1.5 m/s) while the pressure changes by a factor of 4 (from 250 to 1000 mbar). I understand the total mass flow rate was adjusted accordingly. Please clarify this and maybe remove respectively and say the pressure **from** 250 **to** 1000 mbar. Also because of ram effect during flight due to the installation of a choke point in the shroud (L.131) , the ambient air pressure can potentially more than 1000 mbar. Have the calibration system tested a little over 1000 mbar?

    During testing it was found that APACHE is capable of operating at pressures exceeding 1000 mbar. However, the main focus of this study was to investigate APACHE operation to calibrate the HORUS instrument for the HALO (High Altitude Long Range) aircraft at altitudes above the boundary layer. Only below 1.5km the pressure is due to the ram pressure larger than 1000 mbar
12. L.168, **where,** $W_{z1\ pwr}$ is …

Added "Where,"

13. Eq. (1) and (2): I would suggest using [OH] and [HO_2] for OH and HO_2 mixing ratios or concentrations. Please check this out for the entire manuscript. Also it seems to me that the last term ($C_{OH(2)}/C_{OH}*S_{OH}$) needs to take the laser power in the first and second axes into account (unless $Wz1_{power}$ and $Wz2_{power}$ are the same, which is unlikely) and assume there is little OH loss between the 2 axes. The OH signal in the second axis ($S_{OH(2)}$) should be:

   $S_{OH(2)} = [OH]*C_{OH(2)}*Wz2_{power} = S_{OH}/(C_{OH}*Wz1_{pwr})*C_{OH(2)}*Wz2_{power}$

   Please check and correct this.

   Checked and agree with the proposed changes.

14. L.179: I believe the term Wz_{pwr} the should be a denominator in Eq.(4) as the units for C_{OH} should be cts cm^3 molecule^{-1} s^{-1} mW^{-1}. Also here cm^3 molecule^{-1} is used, while in L.170 pptv^{-1} is used. Please be consistent and check this out for the entire manuscript.

   Eq. (4) has been adapted with the consideration of the following:

   The c0 coefficient actually has the units (cts pptv^{-1} s^{-2} cm^3 molecule^{-1} mW^{-1}) i.e in calibrations it is normalized by laser power, Boltzmann correction, quenching (s), internal density (molecules cm^{-3}). During flight and c0 is multiplied by Boltzmann correction, quenching (s), internal density (molecules cm^{-3}) resulting in the sensitivity C_{OH} having the units cts s^{-1} pptv^{-1} mW^{-1}. C_{OH} is then scaled by the actual power measured in flight resulting in the units, cts s^{-1} pptv^{-1}. Then the averaged 5 Hz measured signal (averaged to cts in a second) during flight (see Eq. 1) is subsequently divided by the laser power scaled C_{OH}, resulting in the units pptv for OH.

15. L.189: White cell (capital W)

   Capitalized W.

16. L.199: again the units in the denominator are not correct because C0 has units of cts cm^3 molecule^{-1} s^{-1} as mentioned in L.183, assuming S_{OH} has units of cts s^{-1}.

   See comment 13. Units for c0 was not correctly described.

17. L.206: see the above comment for the issue of units.

   See comment 13.

18. Caption of Figure 3: **dash-dotted** blue line and **dashed** red line.

   Corrected.

19. L.211: Table 1 (capital T)

   Capitalized the T.

20. L.213: change pure to purified.

   Pure in this statement means that only synthetic air is used to calibrate with. I.e. no other type of gas is used as a medium. We have changed "pure" to "only" to emphasize that only syn air is used as the medium for calibration.

21. L.241: the units for $F_{184.9\ nm}$ should be photons **cm^{-2}** s^{-1}.

    Units corrected to photons cm^{-2} s^{-1}.

22. L.288-289: were the air flow speed profiles measured at different pressures, e.g., such as pressures lower than 920 mb to simulate conditions at high altitudes during flight?

    With pressures below 920 mbar, the reading from the differential pressure sensor was close to or below its resolution. Hence the need to utilize other methods to parameterize the flow conditions within APACHE.

23. L.297: Spell out COMSOL.

    Defined COMSOL again as a computational fluid dynamics model (CFD).

24. L.309-315: the disagreement could also be due to the uncertainty in the COMSOL model simulation.

    Added a comment stating this.

25. Figure 6: the air flow speed within APACHE is really unified, even close to the wall. This is good.

26. L.316: do you mean discrete instead of discreet?

    Yes, this is a misspelling.

27. Caption of Figure 7: "The black arrows depict the flow direction." It is hard for me to see those arrows. Maybe include a big arrow on each plot to show the flow direction instead?

    We have increased the arrow size to make them clearer.

28. L.361: Please add "In Table 2" at the beginning of this sentence.

    Added Table 2

29. L.362: streamline (remove s or use streamlines in other places)

    By this, we mean literally on the leftmost streamline for L or rightmost streamline for R, C is in the middle of the streamlines. We have checked that such plural or singular usage is consistent.

30. L.366: **F**igure 8 and **T**able 2

    Capitalized

31. L.368: **On** the APACHE walls.

    Changed "at" to "on".

32. L.377: "between **the lamp** and a quartz wall" to be clear.

    Agreed and applied the change.

33. L.392-392: Martinez et al., 2010 is referred here, but I think at least a brief description of the ground-based calibration system should be given, especially the method to determine the actinic flux of the Hg lamp using the photolysis of N2O to provide the context for Table 3. Otherwise readers may have no idea why NO monitor/N2O cross section are suddenly mentioned in Table 3.

We have included a short description and equation with reference to Martinez et al., 2010, showing where the NO monitor and NO standard terms in table 3 are coming from.

34. Later I found the difference of the two methods is quite large (~20%). I wonder if it is possible to conduct the actinic flux measurement in APACHE using the photolysis of N2O directly so that any uncertainties in transferring the ground calibration to airborne calibration will not affect this difference.

    The difference in the original flux values may appear large however, given their uncertainties the zeta score was 0.88, suggesting agreement within the combined uncertainty of both measurements. However, in light of the comments and suggestions, the calibrations and terms therein have be checked, reevaluated and adjusted when considering the lamp as an extended source of light with a corresponding beam profile. By doing this, the two methods converge, 6.9 (± 1.1) $x10^{14}$ photons cm^{-2} s^{-1} for method A, and 6.11 (± 0.8) $x10^{14}$ photons cm^{-2} s^{-1} for method B. The agreement between the two experiments has a zeta score of 0.59, meaning they agree to within 59 % of one sigma of their combined uncertainties, suggesting agreement.

35. L.397: "…when the smaller 0.8 mm critical **orifice** was used."

    Added "orifice".

36. L.418: Do these OH and HO_2 occur inside APACHE during the transport of air flow from the UV radiation zone and HORUS inlet? Please specify.

    Yes, we calculate this based on the recommended reaction rates, number densities occurring in APACHE, and the calculated transit times that occur in APACHE between the lamp and the HORUS inlet.

37. L.426: Duplicate definition as this has been defined in L.235.

    The equation is used again here as the discussion is building up from it. We believe it is easier for the reader to follow the discussion if they do not have to flip back several pages to check what is being referred to in this section of the discussion.

38. L.457: units for F_β should be photons cm^{-2} s^{-1}.

    Corrected the units

39. L.458: Table 3 should be referred here.

    Table 3 in now referred here.

40. L.459-460: Martinez et al., 2010 should be referred here.

    Martinez et al., 2010 in now referred here.

41. Section 5: Results and Discussion: this section is very lean. Some results in Section 4 could go into this section (e.g., the results for the two methods to determine the Hg lamp actinic flux). There is also no mention how the individual measurements of overall sensitivity (1^{st} row of Figure 10) are used to calculate OH and HO2 mixing ratios in the real airborne measurements. For example, the HO_2 sensitivity in the 2^{nd} axis varied by a factor of 2 (20 vs. 10 cts/s/pptv/mW) at the internal density of 1.5E17 cm^{-3}. What sensitivity to use for the real measurements with internal densities between these two

calibration points? Also any plan/future work to conduct more calibrations to get a better statistics and possibly to draw a smooth calibration fitted line as a function of internal pressure as shown in Figure 3?

Section 5 is now merged with section 4, with some aspects expanded upon. We have included a description and equation showing how the c0, c1,c2 (otherwise labelled as a grouped term cN in the figure) are calculated.

In row A Figure 10 we have decided to provide a smoothed calibration curve much like the one shown in figure 3. Once c0, c1,c2 are known, and quenching, internal density and transmission efficiency are quantified with consideration of the measured internal temperatures and pressures, one can use equation 4, to adequately resolve the sensitivity within 2 sigma of the uncertainties. These points have been added into section 4.3.1. Additionally we have included a paragraph clarifying how these terms are then used to quantify the sensitivity for airborne measurements. Figure 11 has been included to show how sensitivity, HO_X transmission and detection limits look like when quantified using measured temperatures and pressures values in HORUS under flight conditions .

42. L.489: Table 6 is mentioned before the appearance of Table 5.

    Table 5 is now mentioned before mentioning Table 6.

43. L.495: "…resulting in **the transmission** for both OH and HO_2 to be…"

    Added "the transmission".

44. L.498: ".. the time it takes **for** air to flow…"

    Added "for".

45. L.522-526: this paragraph is out of the context of this section. I would suggest moving this paragraph and some actinometric results in Section 4 to a new subsection of 5.2.

    Paragraph moved to calibration uncertainty section, 4.3.2. Where a fuller discussion regarding uncertainty is present.

46. L.524-526: Again units for F_β should be photons cm^{-2} s^{-1} or cm^{-2} s^{-1}.

    Changed to units for F_β to photons cm^{-2} s^{-1} or cm^{-2} s^{-1}.

47. Again I would suggest conducting the actinic flux measurement in APACHE using the photolysis of N_2O directly.

    Addressed in previous sections, and opening statement

48. Section 5.2. Absolute Calibration Uncertainty: this section is very lean and more discussion can be included

    This section has been incorporated into the Evaluation of instrumental sensitivity section. In hindsight, we believe that it is clearer for the reader to follow the discussion and to realize where the uncertainties are sourced from and to what scale they impact the final sensitivity values.

49. L.531: **T**ables 5 to 8.

    Capitalized T

50. Table 5: units for F_β should be photons cm^{-2} s^{-1} or cm^{-2} s^{-1}. Also a temperature range of 282-302 K is given but no mention in the text how it was varied within APACHE.

    Temperature ranges now discussed in section 2.2

51. Table 7: this should go Section 5.1 where transmissions are discussed.

    See comment 47.

52. Table 6 and the 3rd row in Figure 10: details about how the term cN* internal density is calculated/measured should be given.

    Included equation and discussion regarding how cN is calculated se Eq 14.

53. L.559, and 562: the actinic flux of the mercury lamp should be photons cm^{-2} s^{-1}.

    Corrected the units

54. Figure 10: the 1st row: the units should be cts $\mathbf{s^{-1}}$ $pptv^{-1}$ mW^{-1}.

    Corrected the units

55. Figure 10: "Row C is (C) is internal density and cN". Do you mean "Row C is the product of internal density and cN"? I don't understand how cN is calculated.

    Included equation and discussion regarding how cN is calculated see Eq. 14,15 and 16.