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Atmospheric Measurement Techniques Discussions

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Interactive comment on "Calibration of an airborne HO_X instrument using the All Pressure Altitude based Calibrator for HO_X Experimentation (APACHE)" *by* Daniel Marno et al.

Anonymous Referee #2

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In this paper Marno et al. demonstrate the first results from the "APACHE" chamber designed to calibrate and characterise the Mainz airborne "HORUS" OH and HO2 instrument. The results show the APACHE chamber operating on the ground under controlled conditions to calibrate HORUS, but it is designed also to be operated on the HALO aircraft when OH and HO2 measurements will be made, in order to calibrate in flight.

The development of a device to calibrate for OH and HO2 measurements in flight is a very difficult challenge, not only does the sensitivity of the instrument vary with a change in the pressure and temperature sampled (which changes with altitude),and also the level of water vapour, but also the losses between the point of OH and HO2 generation in the calibrator and sampling by HORUS change also (there would be losses also for ambient OH and HO2 which are to be measured). For the former, the change in sensitivity owing to changes in parameters with altitude after the HORUS inlet can be experimentally determined via the calibration – but in this paper these are investigated through calculations also. For the latter, i.e. losses in OH from the point of generation (lamp) and the HORUS inlet need to be characterised experimentally – and understood. CFD calculations are used to simulate the flowfield within APACHE for comparison with experiment.

The description of a device to generate known concentrations of OH and HO2, and its characterisation and comparison with simulations, given the range of parameters, is complex. Likewise the sensitivity of the instrument measuring OH and HO2 and how this varies with sampling pressure is also complex – and so naturally this paper is complex and many parameters have to be explained and how they change with pressure explained. However, this is critical, as OH and HO2 are highly reactive and can be lost both in the gas-phase and at surface. The authors have made the paper fairly clear – as the characterisation is quite complex – but some further clarity is needed. The experiments appear to have been carefully performed, and many of my comments are aimed to help improving clarity for the reader.

It is not clear from the paper whether the APACHE/HORUS device has been used in flight already, as this reports experiments done in a controlled environment on the ground – and perhaps something about how it performs in flight would be useful to include, and comparison with the ground performance. The paper is an impressive piece of work – and the APACHE/HORUS is quite a feat of engineering and the thorough characterisation of APACHE and HORUS is critical to give confidence in the OH and HO2 measurements from HORUS on HALO. The paper is suitable for AMT, and the development of a calibration source for use inflight for OH measurements is very important, and a considerable achievement. There is a lot covered in this paper, but some further details/clarifications are needed in some places. See comments below.

More specific comments.

Abstract.

A key result is that the two actinometric approaches agree fairly well, and as well as the average it would be good also to give the level of agreement also. Say what the two approaches are. What pressure is relevant for the value stated, as you say “depending on pressure”, which is not clear?

Although the paper is about APACHE and its characterisation, I think readers will want to know what the sensitivity is of HORUS determined with APACHE. Could the expected C factors be stated for OH and HO2, and the derived limits of detection, and how these are predicted to vary with altitude, also be given in the abstract.

The overall accuracy of the calibration ought to be stated also in the abstract from the use of APACHE. This is given in some detail in the paper but there is nothing here. A few more numbers summarising actual performance needed in the abstract.

Also, “controlled environment” is a bit unclear, please make clear that this is on the ground, rather than results being presented of APACHE used under “a controlled environment” on the aircraft in flight.

Introduction.

46. The referencing is rather selective, please also include Juelich and Leeds LIF references (zeppelin and aircraft measurements also). For CIMS include some Eisele group references also (and subsequent including Mauldin/Cantrell which have also flown).

Figure 1. The APACHE shown here is for the controlled environment on the ground – make clear in the figure caption. Looking at Figure 2, the left hand side of APACHE would be a bit different when on the aircraft? (no inflow from mixing blocks?)

96, replace “being” with “is”

107. Is the 0.9 to 1.5 ms-1 in APACHE over the pressure range the same as the flow velocity at the same pressure when sampling on the aircraft. In line 132 the “choke” on the aircraft nacelle is used to lower the flow velocity to < 21 ms-1, but not clear if < 21 ms-1 means it will be similar to the 0.9-1.5 ms-1 as in the controlled experiments on the ground? < 21 ms-1 could cover a wide range.

124 – say also there is a critical orifice at the end of the IPI, this was not clear (and not labelled in Figure 2).

There is both a HORUS inlet, and a IPI critical orifice, and I think the presence of these two needs to be clearer. In figure 2 I suggest, that both the HORUS inlet and also the IPI critical orifice have a label. Also both “IPI orifice”, “HORUS inlet” and “IPI critical orifice” are used. In line 128, is “IPI orifice” the “HORUS inlet” which samples from APACHE, or the “IPI crictical orifice” which is between the IPI and the 2 fluorescence cells? I think the former as the choke point is then mentioned which slows the flow from the aircraft speed to a slower flow in APACHE?

132. “sample velocity of HORUS”, this means the flow within APACHE at which HORUS sampled perpendicularly? Is 44-53 ms-1 what is expected on the aircraft?

Figure 2. label the critical orifice in the IPI and also HORUS inlet for clarity (as discussed above).

144. As an IPI is used, it would be worth mentioning OH-WAVE (on to off resonance) and OH-CHEM, otherwise not clear of the purpose of the IPI. All the experiments performed here are OH-WAVE – presumably results of OH-CHEM in a controlled environment (to show all OH removed etc.) will be discussed in another paper. The IPI is present here but not used.

149. Again the referencing of papers is selective to a couple of groups only who use LIF.

153. Quantitative conversion is mentioned here. can a % be given, as it is not possible

to achieve 100% owing to OH+NO + M = HONO + M meaning that not all of the HO2 conversion to OH remains as OH. What is the % that is achieved here? What flow of NO is added?

180 “where” small w

202 – state the size of the critical orifice here. (diameter)

Fig 3 – make clear this is a schematic only – rather than any actual performance of the HORUS. Could point to fig 10 where this is shown. Also in the caption, the dotted blue line is for “OH transmission”, whereas in the figure it is “wall loss”.

219 – split – and 1 in the units

230. Juelich showed that the reaction of H* with O2 did not lead to OH, rather that 100% of H went to HO2, so worth referencing that.

Table 1. For (IV) CSTR, was the OH generated through UV irradiation of the VOC, or of another precursor? Certainly the decay rate of the VOC is used to determine the OH. Also reference Winiberg et al. 2015 (in the reference list) who used the decay of a hydrocarbon to calibrate for OH in a chamber with a LIF instrument (agreeing well with method I, water paper photolysis).

238, “where”, small w

268. The exhaust from the pumps are at a different pressure when in flight compared to when the exhausts are exposed 1 atm, and this is taken account of by matching to ambient pressures in flight – that is good. Was the same pumping system used for the APACHE testing on the ground as the pumps that will be used (or are used) in flight (which might be 400 Hz pumps from the aircraft power)? (different pumps or pumps used with different motors may have different capacities).

305 “from the measured...”

Figure 6. Can it made clear what is meant by “internal wall of APACHE”, perhaps by

cross-referencing to figure 1?

240. The number of sig figs in the error 179 +/-20 does not seem consistent with the sig figs quoted in the errors in brackets for the other units.

361. L, C, and R term are introduced, to make clearer, say which figure they are in – otherwise not clear what referring to.

371. How is 22.2 % loss known for OH and HO2 the inlet? (HORUS inlet). Also, one might expect the loss to be higher for the more reactive OH? Please expand a little.

Figure 8. What [H2O] the same for all the pressures? Perhaps add this value.

Tabel 2. Right hand column – OH (ppt) also?

395. The IPI critical orifice diameter is given here – but needs to be given earlier as well when this orifice is first introduced. What is the reason that the diameter of this orifice is changed from 1.4 mm to 0.8 mm for the controlled experiments on the ground?

439 and 441, another “where” to change

457 and elsewhere, for the units of flux of the light should this be “photons s-1”, or even also per unit area?

Section 5 is the results, and quite a few are shown, but compared with the rest of the paper this is fairly short, and the discussion ought to be extended a little to fully exploit the results – what behaviour is therefore expected from aircraft measurements based on the lab work?

495. The losses at the inlet were the same for OH and HO2? Some further discussion of this as might expect OH to lost more.

498 “where”

Page 20 – I found this page difficult to follow, there were a lot of losses discussed, quantified by the alpha values, for various stages of the airflow, e.g. the meanings of equations 16-18 and the discussion around this was confusing.

522. Remind reader of the two actinometric methods again (as not much detail was give on these two methods earlier).

Section 5.2 seems to be a series of tables 5-8, and a big figure, and there is virtually no text to go with this? Some further discussion is needed to bring this all together, given it is the main results from the paper. From the C factors presented , e.g. in Table 8, can the LOD of the instrument be presented, and this compared with expected levels of OH and HO2 in the atmosphere during the flights?

Figure 10. For the second row on quenching, link this to an equation used in the text – the label of the plot "Overall quenching" is unclear – and some link to the relevant part of the text is needed. Likewise for the other panels. for the first row, the y label is "Overall sensitivity" which I assume is the C(OH) factors etc., and an explicit link should be made. Likewise ALHPA (total) – refer to the equation where that is in the text.

554. The losses of HOx is discussed for the operation of APACHE during the controlled conditions ground testing. Can this be compared with the expected losses during flight when the flow velocity within APACHE may be a somewhat different (or a statement making clear the velocity within APACHE will be the same as here, or similar).

566 "is" missing after "system"

567 – experienced in flight is mentioned, but make clear again that the tests presented here are on the ground.

568. 17-18% overall uncertainty (1 sigma) – explain why this is "suitable" for a calibration approach. Mention is needed of what the measurements will be used for – to compare with OH and HO2 calculations from an atmospheric model – for which there is an uncertainty also – and a robust comparison can only be done if the measurements are accurate to a certain %, etc.

The paper focusses on pressure and water vapour, can any comments be made about the expected change in performance (e.g. losses on surfaces, or lamp) with changes in temperature during flights?

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