An intercomparison of water vapor measurements in the TTL and lower tropical stratosphere during AVE-WIIF, CRAVE and TC4: The importance and implications of laboratory calibrations with water vapor mixing ratios from 0-10 ppmv

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Introduction

- Accurately forecasting UV dosage levels has become far more important as the rate of climate forcing has accelerated.
- The NRC Decadal Survey made clear that it is the irreversible nature of potential increases in water vapor in the stratosphere coupled with decreases in lower stratospheric temperatures resulting from CO_2 forcing and water vapor feedback will increase the catalytic destruction of ozone in the stratosphere.
- Unexpectedly high relative humidities observed in the cold tropopause region in clear air and clouds challenge current microphysical and dehydration models.

Consequently, accurate or "benchmark" quality water vapor measurements are needed to:

- Maintain an accurate observational database for stratospheric trend measurements.
- Provide measurements in the TTL to help distinguish proposed strat-trop exchange mechanisms.
- Provide the accuracy necessary for categorizing relative humidity measurements in contrails, in the tropopause region and in the upper troposphere near and within cirrus clouds.
- Provide accurate water vapor measurements in polar regions where heterogeneous ozone loss critically depends on ambient water vapor.
- Provide "benchmark" quality water vapor data for satellite validation.

Intercomparison of water vapor measurements in the UT/LS have highlighted systematic instrument differences:

- Water vapor measurements as summarized in Figure 1 of SPARC 2000 illustrated significant differences between water vapor measurements in the UT/LS.
- We focus on systematic differences observed between Harvard Lyman α, the balloon-borne NOAA CMDL Cryogenic Frost point Hygrometers (e.g. CMDL and CFH), and the Microwave Limb Sounder (MLS) on the Aura satellite. First intercomparison example shown here:

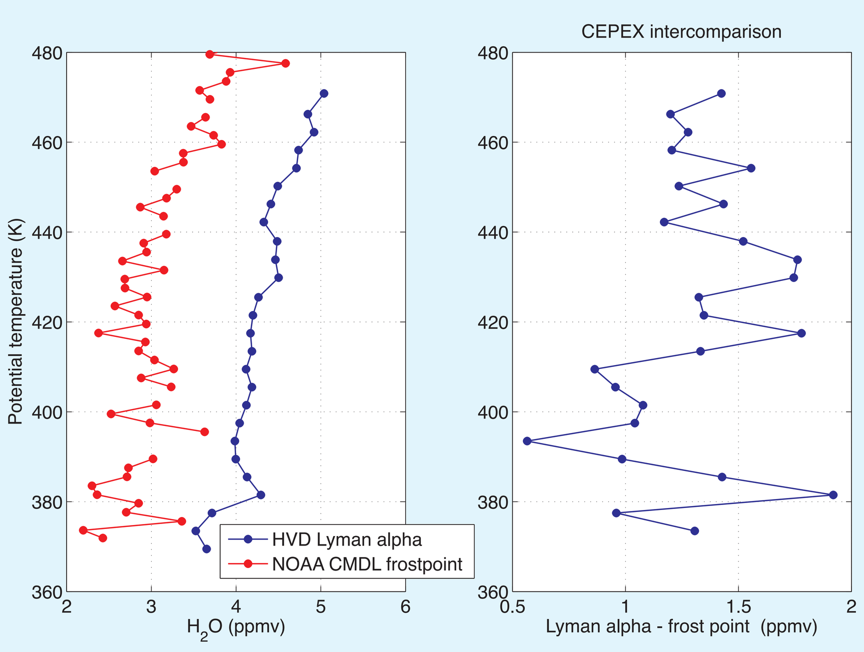

FIGURE 1. Intercomparison between Harvard Lyman α on the NASA ER2 and CMDL during CEPEX. Harvard points are binned and averaged at 2K intervals for data taken during aircraft dives at 2° S latitude on March 18, 21, 24, 29, 31 and April 4.

So how do we resolve this systematic difference?

Foundation for measurement accuracy must be laboratory calibrations tied to SI traceable standards.

Harvard calibrations

Table 1. Calibration techniques for each Harvard water vapor instrument in our laboratory, each traceable to physical properties of water.

Instrument	Vapor pressure of liquid water (Bubbler)	Liquid water droplet injector	121.6 nm Abs.
Lyman α	X		X
ICOS*	X	X	
Hoxotope*	X	X	

* Both ICOS, (Integrated Cavity Absorption Spectrometer) and Hoxotope (Modified from the OH laser-induced fluorescence instrument) were developed at Harvard to measure water vapor isotopes and both calibrate in the laboratory under flight conditions.

We illustrate the Lyman alpha calibration setup in Figure 2.

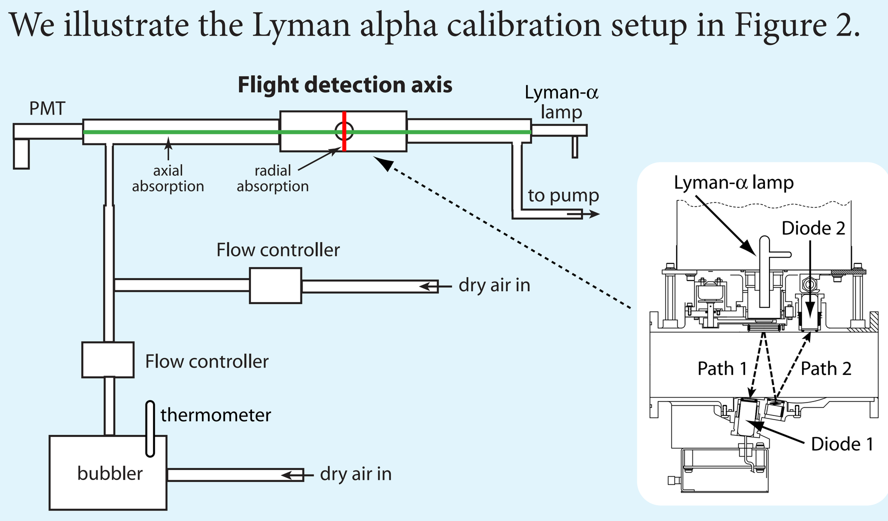

FIGURE 2. Schematic of the Lyman alpha calibration system (left); Lyman alpha detection axis (right); PMT (not shown) perpendicular to the air flow and lamp flux.

CONCLUSION: Instrument sensitivity in laboratory tied to physical and chemical properties of water.

However, systematic differences illustrated in Figures 1 and 8, and a report summarizing the CRAVE water vapor workshop suggested the need for low water calibrations and intercomparisons, examples of which we show in Figure 3 below.

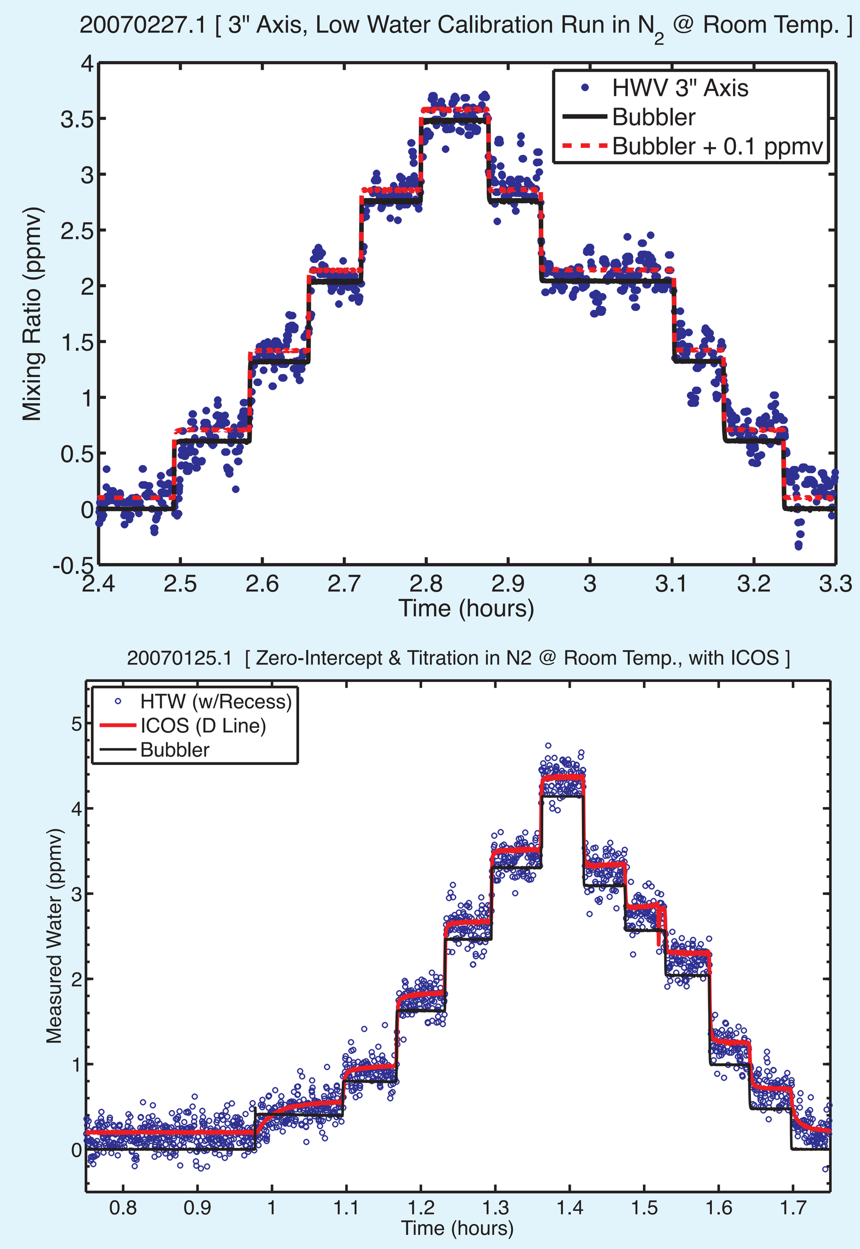

FIGURE 3. Low water vapor calibration runs for the ER2 (top panel) and WB57 total water (bottom panel) detection axes. For the ER2 axis, the plot of water vapor as determined by the water vapor addition system (bubbler) with 0.1 ppmv added illustrates the measured water vapor in the system prior to water being added and no measurable offset. A virtually identical background water vapor as measured by both the total water axis and the ICOS instrument is shown in the bottom panel.

CONCLUSION: Laboratory calibrations at low water vapor constrain any offset of the ER2 or WB57 Harvard Lyman α instruments to < 0.2 ppmv.

Because typical laboratory calibrations are carried out at room temperature, their insensitivity to temperature must be established. We show an example of the calibration's insensitivity to temperature in Figure 4.

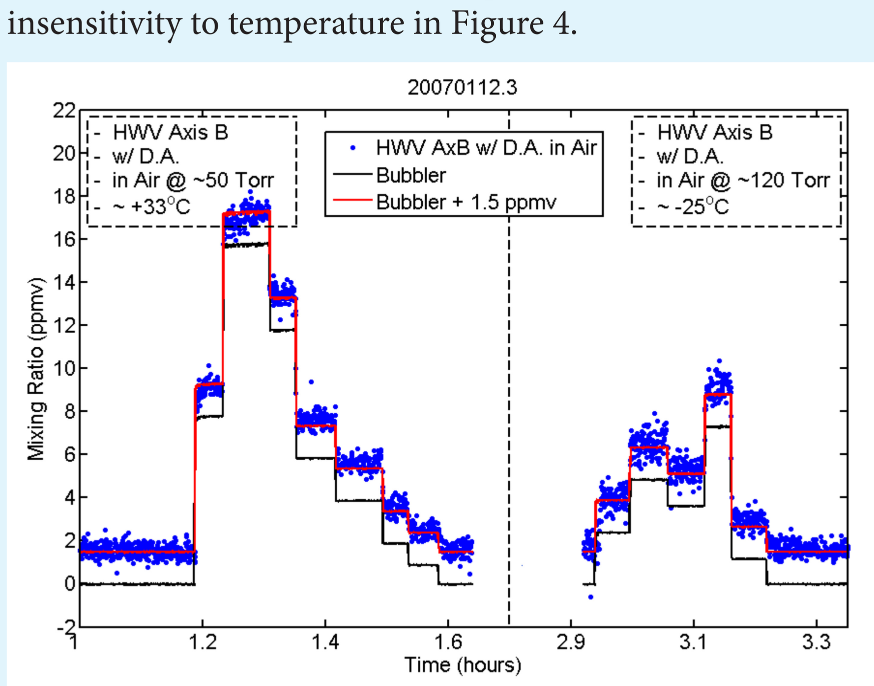

FIGURE 4. Repeat laboratory calibrations at temperatures 60°C apart illustrate temperature independence. All data are analyzed using the same calibration constants.

CONCLUSION: The temperature independence of laboratory calibrations carried out at atmospheric pressures Illustrates applicability of room-temperature calibrations to flight conditions.

Flight intercomparisons

AVE-WIIF July, 2005 Houston TX

Recent in-flight validation of Harvard Lyman α during the AVE-WIIF mission from Houston, TX in July, 2005 is shown in Figure 5. We show data from six instruments but emphasize here agreement among all the Harvard instruments: four different instruments used three completely independent detection methods and four different sampling strategies. That agreement was demonstrated over a range of atmospheric pressures, temperatures, water vapor concentrations, and for Harvard water vapor flow velocities.

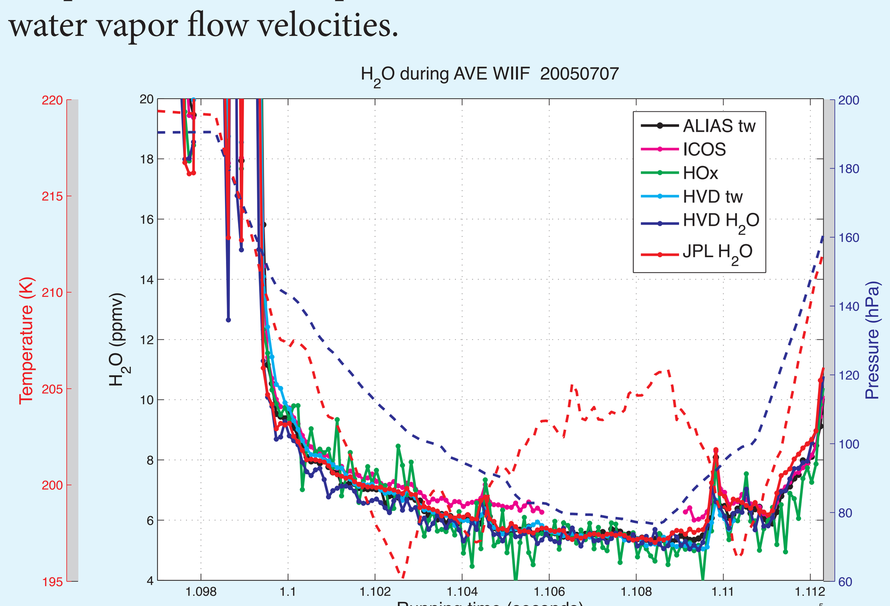

FIGURE 5. Water vapor data taken during the last stratospheric segment of the 20050707 flight. Ambient pressure (dashed blue line) and temperature (dashed red line) are plotted as well.

In Figure 6 we show a detailed examination of agreement between Harvard water vapor instruments.

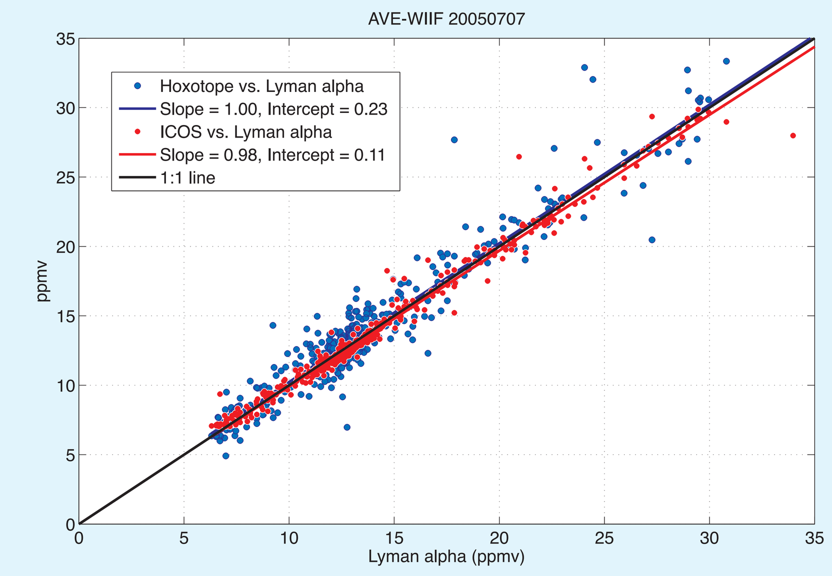

FIGURE 6. Least squares fits to the raw data for the last AVE-WIIF flight where Hoxotope and ICOS are plotted respectively against Lyman α.

CONCLUSIONS: laboratory and in-flight agreement between Harvard water instruments validate Lyman alpha:

- Laboratory calibrations apply in flight.
- Offset constrained to at most 0.1 ppmv in nitrogen corresponding to 0.2 ppmv in air.

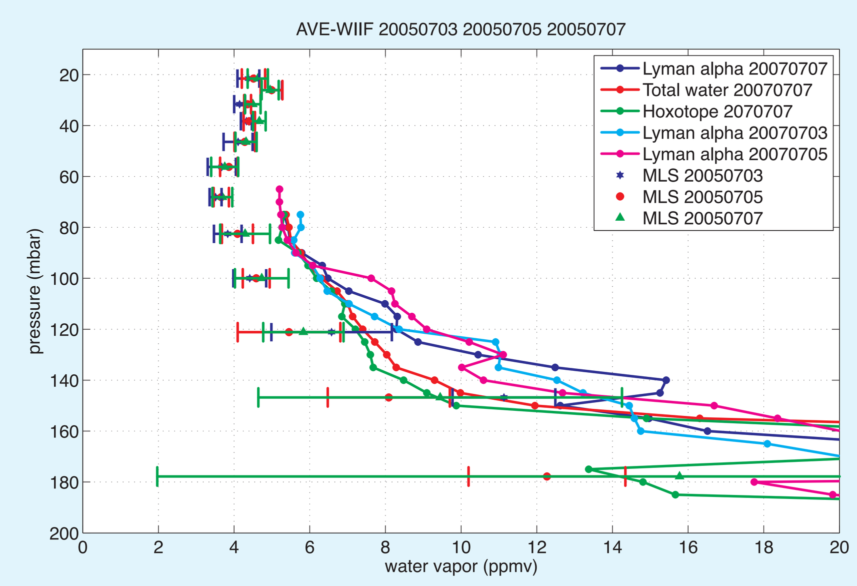

FIGURE 7. Intercomparison with MLS during Ave-WIIF.

CONCLUSION: Intercomparison of Harvard instruments with MLS illustrates systematic offset in UT/LS.

- In tropics MLS and CFH agree very well so MLS is surrogate for CFH during Ave-WIIF, and consistent with data in Figure 1.

CRAVE (Jan-Feb, 2006) and TC4 (August, 2007)

Differences consistent with these results have been most Recently observed during the CRAVE and TC4 campaigns which included intercomparison with MLS as shown in Figure 8.

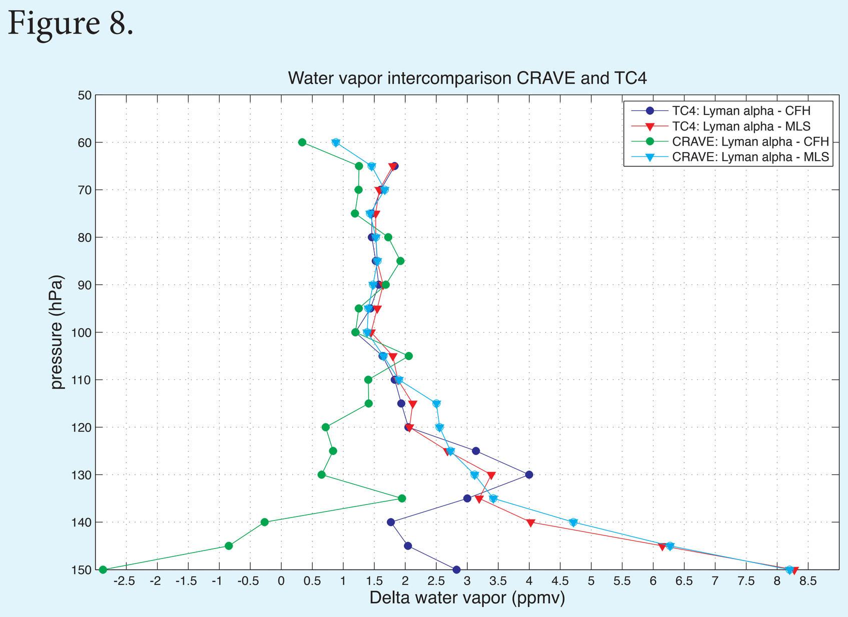

FIGURE 8. Intercomparisons between Harvard Lyman α and MLS and CFH during the CRAVE campaign and the TC4 mission, where the data are all binned and average at 5 hPa pressure intervals.

CONCLUSION: All intercomparison data between Harvard Lyman alpha and NOAA frost point instruments as well as MLS on the Aura satellite show a consistent difference of about 1.5 ppmv in the tropopause region and lower stratosphere.

So what can we learn from a carefully run laboratory intercomparison?

Recent AQUAVIT intercomparison

(see poster by Harald Saathoff et al. for further details)

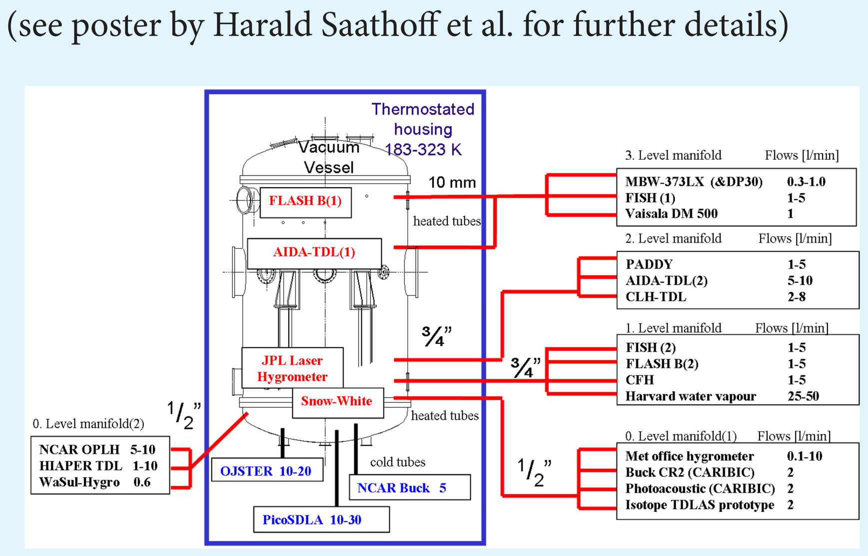

FIGURE 9. Instruments as configured during AquaVIT.

Key instrument of interest: Lyman α, CFH, JLH, FISH2, AIDA TDL, first four with extensive UT/LS data; last AIDA reference instrument.

Our approach:

1. Use analysis to distinguish between calibration errors (directly resolvable in lab), offsets or artifacts at low water, and sampling errors.

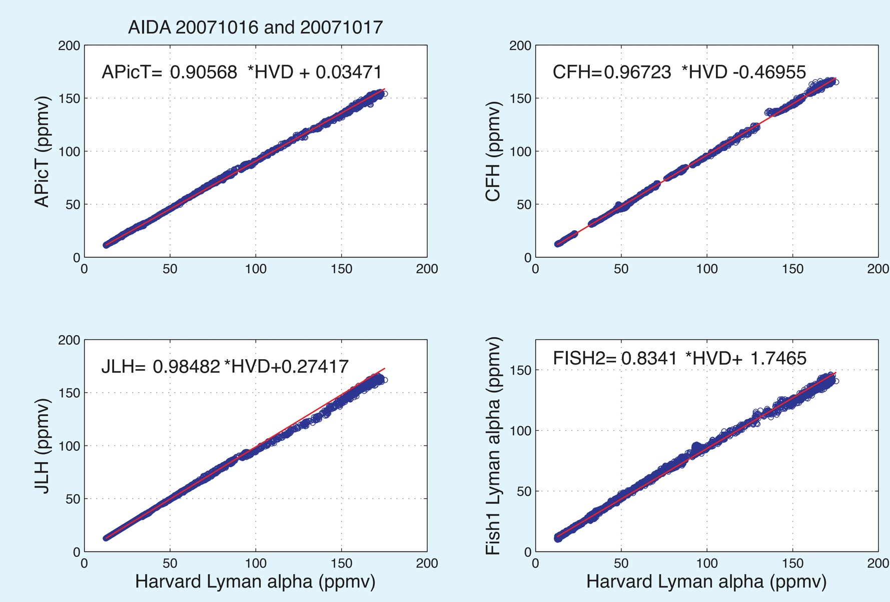

FIGURE 10. High water data on two days allows for calibration intercomparisons with Harvard Lyman α.

CONCLUSIONS:

- Agreement between Harvard, JLH and CFH is excellent; not quite as good between Harvard and AIDA TDL; still poorer between Harvard and FISH2.
- Large differences between HVD and FISH2 are resolvable in lab.

2. Examine low water results on 3 days while taking into account calibration differences. Magenta line represents modeled correction to Lyman α data because of insufficient flow.

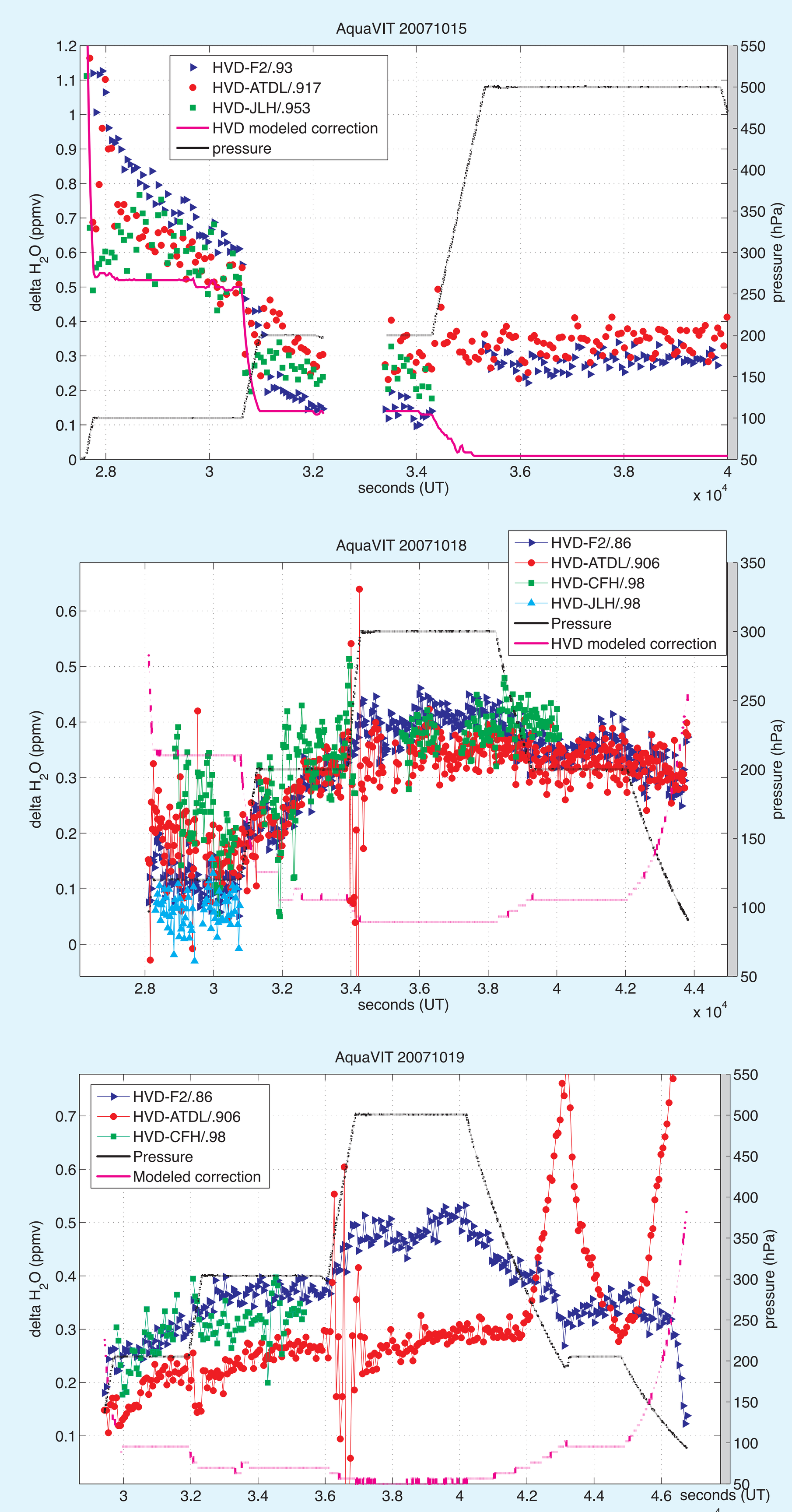

FIGURE 11. Low water data from the runs on the 15th, 18th, and 19th. The modeled correction is derived from diagnostic data taken as a function of flow and pressure.

CONCLUSIONS only from data where correction is negligible:

- Harvard Lyman α measures about 0.30±0.05 ppmv higher than the AIDA TDL
- Using the limited intercomparison data with CFH on the 18th and 19th, Lyman α is about 0.35±0.05 ppmv higher than CFH.
- The difference with JLH data is about 0.05-0.10 less than with AIDA TDL.
- The difference from Fish2 is slightly higher, about 0.40±0.10 ppmv.

MAJOR CONCLUSIONS from AquaVIT:

- Observed differences at low water are small, and do not approach those observed in-flight.
- Direct Laboratory intercomparisons under flight –equivalent conditions are needed for these instruments.