Response to reviewer RC2 of manuscript https://doi.org/10.5194/egusphere-2024-4019

JLH Mark2 - An Improved Opto-Mechanical Approach to Open-Path *in situ* Water Vapor Measurement in the Upper Troposphere / Lower Stratosphere

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We would like to thank reviewer RC2, Dr. David Sayres, for his excellent and detailed comments on our manuscript. Below are our responses to each comment in order.

Minor Comments:

1. Line 28: addition should additional

Reply: We will change this word to "additional".

2. Line 37: Be consistent with UTLS or UT/LS

Reply: We will change this (and all uses of this word) to "UTLS" to be consistent with widespread usage.

3. Lines 83-90: Where are these temperature and pressure measurements compared to JLH. Could you comment on the suitability of these measurements for the water absorption. Do you expect there to be T and P differences between where these measurements are made and the Herriott cell?

Reply: These instruments are situated several meters distant from each other in both water line coordinate and fuselage station coordinate; however, Computational Fluid Dynamics (CFD) modeling indicates that these instruments sample beyond the boundary layer in the freestream.

At approximate air speeds of 250 m/s, instantaneous variations in atmospheric water vapor mixing ratio, static air temperature, and static air pressure at the integrated instrumentation locations are assumed to be trivial in comparison to the variations in these *parameters in the total atmospheric volume sampled during the data integration period of 1 Hz. Consequentially, the atmospheric volumetric average parameter value is expected to be identical beyond instrumental precision at each location.*

We have revised the text to clarify that these instruments sample the freestream.

[line 79] "and flights of opportunity (2015-2023). For these flights, JLH Mark2 was mounted on top of the aircraft fuselage where it samples the atmospheric freestream (Figure 1b)."

[line 86] "The probe was routed to an instrumentation system for data collection. Static pressure was also sampled outside the boundary layer. The ..."

4. Line 107: Can you comment on boundary layer depth of the fuselage.

Reply: CFD alpha and beta sweeps encompassing expected parameters for cruise, ascent, and descent at relevant Mach numbers and altitudes were performed. In no condition investigated did the boundary layer extend to the optical measurement.

We note that this conclusion is also provided on line 330 of the original manuscript. We revise line 107 to provide the reader with an earlier indication that the instrument optics lay in the free-stream. The new sentence on line 107 is:

"The optical open-path multipass absorption cell of JLH Mark2 is mounted external to the aircraft and situated beyond the expected boundary layer in order to avoid contamination or..."

5. Line 117: This additional pathlength seems potentially an important source of error. Do you believe this air to be flushed in the same way the air between the mirrors? Have the same temperature and pressure? I realize it's only 0.8% of the pathlength, but if this volume is "dead air" it will have much higher mixing ratio of water.

Reply: The additional optical path is flushed, so water vapor remains relatively close to environmental mixing ratios. This optical path is an open cavity between the hole in mirror M_0 and the free stream between the air foil and laserhead housing (Figure 1) and air flows through this as explained below. We will add several sentences at line 117:

"The mirror airfoils were designed to maintain the air density between the two mirrors equal to the free stream density (see Section 4.2). Outside the mirror airfoil, between the M_0 mirror airfoil and the laser housing, the geometry of the parts forms a venturi. At typical flight air speeds, the pressure between the airfoil and the laser housing will be lower than the ambient; there will be air flow from the hole in mirror M_0 through the airfoils due to this differential pressure."

6. Line 119: How is the pathlength calculated? Is this by ray trace or knowing the angle that the light bouncing between the mirrors?

Reply: The pathlength is calculated by ray trace. We will add an additional sentence to line 119:

"The total absorption path length is calculated based on a ray trace, knowing the Herriott configuration for fifty passes between mirrors of known radii of curvature (Altmann et al., 1981; Herriott et al., 1964). This value is thought to be precise to one part in 10,000 over the ambient temperature range of 100 K in flight because the length of the long axis of the multi-pass cell is defined by invar spacing rods, which have a coefficient of thermal expansion of 10^-6 K^-1."

7. Line 166: 8 Hz is a little ambiguous here. You mean that you have 8 ramps a second, but the actual rate of data acquisition is much higher. Probably best to have another sentence that specifies the acquisition rate and ramp rate and remove the 8 Hz at the end of the sentence.

Reply: To make this more clear, we will modify line 166 to:

"The TDL is driven with a sawtooth-shape current ramp at a ramp repetition rate of 8 Hz to scan across the targeted water absorption transition. Data are acquired at 4 kHz (512 points per ramp)."

8. Line 168: What is DC mean here, direct current, or did you mean DT?

Reply: We mean direct transmission (DT) here. "DC" refers to the direct current channel of the signal chain (as opposed to AC) (May, 1998). We will modify the text at line 168 to "water absorbance is quantified by direct transmission".

9. Line 211: Be good to mention how well this works. What was the flight stability of the laser temperature thermistor.

Reply: The temperature stability of the laser temperature thermistor (and the aluminum housing) is +/-0.01 deg C as already mentioned in line 120.

10. Line 249: should this be 'background noise'?

Reply: What we mean by "background" is that filtering passes the AC component of the signal and removes DC offsets caused by the slow ramp. Lines 248-249 will be rewritten,

"The time-varying signal is detected at frequency 2f = 256 kHz and filtered to remove DC offsets caused by the slow laser current ramp."

11. Line 287: I'm assuming the interpolation is linear, but inherently a laser's tuning rate with current does not have to be linear, so it might be worth noting whether using the methane lines you find it linear over this scan range.

Reply: The interpolation is a second-order polynomial that is very close to linear. We will clarify in the text that we fit the wavelength scale versus index to a second-order polynomial using seven identified methane lines and the known water line (pure methane with trace water in our calibration chamber). First, we rewrote this entire paragraph to better explain:

"Laser characterization is necessary for accurate water vapor measurements. We first determine the absolute laser wavelength to within ~0.18 nm (1 cm^{-1}) with a Burleigh wavemeter. The wavelength and laser tuning rate with wavenumber are refined by using seven identified spectral lines of CH_4 as wavelength standards in the same spectral region as the water line of interest (Brown et al., 2013; Rothman et al., 2013). The methane line positions are known to within 10^{-4} cm^{-1} (Brown et al., 2013), allowing the accurate determination of the water line positions by interpolation. The laser tuning rate here refers to wavenumber (cm^{-1}) versus index in the laser scan, which goes from 1 to 512. For this purpose, measurements are made with JLH Mark2 in a laboratory test chamber. These weak CH_4 lines are measurable by filling our calibration chamber with 27.6 hPa pure CH_4 (with trace water). We fit a second-order polynomial to wavenumber versus index. Ultimately this wavenumber scale is needed to fit the direct absorption using fundamental physical information."

"Other laser parameters of interest are spectral purity, linewidth, and laser modulation amplitude. Spectral purity means the fraction of laser emission centered on a single wavelength and not in side bands (e.g., at a shifted wavelength). The measurement of water mixing ratio is directly proportional to the spectral purity. The spectral purity of the laser is verified by measuring the laser transmission through a sample at very high water vapor concentrations, which absorbs all the light at the line center wavenumber of 7299.4311 cm^{-1}. The laser linewidth affects the amplitude of the pp2f signal and fitting of direct absorption, so it is important to characterize. The laser linewidth is determined in the laboratory by measuring the water absorption lineshape at low pressures where the absorption linewidth is solely due to the Doppler width. If the line is wider than the predicted Doppler width, it is assumed to be a convolution of the laser linewidth and Doppler width (Section 5)."

Second, we will modify the sentence at line 466:

"As discussed in Section 2.3.1, laser tuning rate (wavenumber versus index point) is determined by fitting seven CH_4 lines surrounding the targeted water line with a second-order polynomial."

12. Line 292: It would seem that this last sentence flows more naturally right after the discussion of determining the water line position (ending in the middle of line 287).

Reply: We agree with the reviewer and will move the text up to line 287 (see comment 11 above).

13. Lines 309 – 312: I find this procedure confusing or at least its description. Is the HITRAN database parameters being used to fit the direct absorption spectra? When you say you use the ratio, do you mean for the field data you scale the direct absorption measurements by this scale factor? So ultimately the standard that you are using to calibrate your instrument is the Thunder Scientific.

Reply: Yes, the standard that I am using to calibrate JLH is the Thunder Scientific. All the JLH measurements are scaled to the Thunder Scientific (TS3900), so we have revised the calibration text at lines 309-312:

“The instrument response is characterized through continuous flow of an accurately known air/water vapor mixture through the JLH sample cell when it is inside the laboratory calibration chamber (see below). JLH water vapor measurements are calculated as in the field data. The ratio of known source water data to JLH water data is used as the instrument response factor and is used to scale the field measurements.”

“By 2007, improved calibration methods were developed, which allowed for laboratory measurements of sample atmospheres in test chambers with water volume mixing ratios as low as 2 parts-per-million by volume (ppmV) (Troy, 2007). The source of the air-water mixture for determining the instrument response mentioned above is a commercial Thunder Scientific 3900 (TS3900) low-humidity generator (https://www.thunderscientific.com/), which can provide air with stable water mixing ratios from less than 1 ppmV to 12,000 ppmV. The carrier gas is ultrazero air from cylinders. The TS3900 uses a technique that generates air with a constant water mixing ratio to an accuracy of 0.1 K frostpoint (corresponding to better than 1% accuracy of mixing ratio). The technique is a primary standard recognized by NIST and used by commercial hygrometer manufacturers to check their instruments. It depends solely on accurate measurements of pressure and temperature to achieve a constant mixing ratio of water vapor in air flowing over a saturator. By scaling the JLH data to the TS3900, the standard we are using to calibrate the instrument is the TS3900.”

“For an open-path instrument such as JLH, it is necessary to place the laser/detector housing and sample cell inside a test chamber. The JLH test chamber used for laboratory calibration is a 304 Corrosion Resistant Steel (CRES) box vacuum chamber manufactured by the Kurt J. Lesker Company, and coated on the inside with a hydrophobic coating of Fluoropel® 1302IBA, manufactured by Cytonix Inc. of Beltsville, Maryland (Troy, 2007). For calibration of the JLH instrument in the lab, an air/water vapor mixture generated by the TS3900 flows continuously through the CRES chamber at constant pressure and temperature. The sampled air mixture is checked both upstream and downstream of JLH with a reference Vaisala DM500X precision surface-acoustic wave (SAW) hygrometer. Typically, half of the flow is directed through JLH and the rest through the Vaisala hygrometer, which has a quoted accuracy of 0.3 K frostpoint (corresponding to 2% accuracy of water mixing ratio).”

14. Line 312: I’d be careful of saying you are canceling the uncertainties in the HITRAN database. There may be many sources of error for fitting the direct absorption line. Line purity for one, which you mention. The choice of which line shape parameters you are using. At higher concentrations or for your studies using pure water self broadening can be important and also choice of lineshape (Voigt versus Galatry for example). How well you know your tuning rate and how linear it is. For the purposes of measuring water vapor in the atmosphere using methane lines to derive a tuning rate and position is perfectly adequate, but may miss some small nonlinearities in the laser.

Reply: We agree with the author that there are many sources of error for fitting the direct absorption line. We will remove the sentence at Line 312 ("This effectively cancels out the errors due to uncertainties in the line strength in the HITRAN database."). The tuning rate is known well because it is fit by a polynomial using seven methane lines. Uncertainties are addressed in our response to your final comment (below). Our methodology uses a Voigt lineshape for curve-fitting, but ultimately the measurements over the range of concentrations (3 to 1000 ppmv) are scaled to match the Thunder Scientific.

Furthermore, we will remove the paragraph at lines 527-534 because it overstates the accuracy of fitting the direct absorption line.

15. Line 361: Not sure why this section is called Laser stability. ‘Absorption line selection’ perhaps.

Reply: We agree that the topic of Section 3.2 is “absorption line selection” and not “laser stability.” This is a short paragraph that belongs better in section 2.3.1 Laser Characterization, so we will move the text to the start of Section 2.3.1 (line 282).

16. Line 362: Given that everywhere else you write ‘water’, I’d change H2O to water. Unless there is some more subtle point you are trying to make that I’ve missed.

Reply: We agree and will change “H2O” to “water”

17. Line 466: laser tuning rate here refers to cm-1 versus time or versus current? It should be defined somewhere. You also mention it earlier, so perhaps defining it there.

Reply: Laser tuning rate refers to wavenumber (cm-1) versus index in the scan (from 1 to 512). This is also addressed in our response to comment 11 above, where we rewrote Section 2.3.1.

Second, we will modify the sentence at line 466:

"As discussed in Section 2.3.1, laser tuning rate (wavenumber versus index point) is determined by fitting seven CH_4 lines surrounding the targeted water line with a second-order polynomial."

18. Lines 466 – 476: The discussion of line widths is a little confusing. On line 472 you say “the Gaussian is a convolution of Doppler-broadened linewidth and laser linewidth, then we calculate an effective instrument linewidth ...” Given the first clause of that sentence, what you are calculating is the laser linewidth. You then go on to say that the instrument line width is now a convolution of the true laser linewidth and electronic broadening which would seem to invalidate the presumption you make on line 472. I think it would be simpler to say that the difference between the Gaussian fit and the calculated Doppler broadening is dominated by the laser linewidth and any electronic broadening. You believe (I don’t think you say that you’ve measured the laser linewidth independently) the laser linewidth is smaller and therefore think that the electronics are broadening the line. Usually if the electronics are the issue they act as an RC filter of the data. That would not broaden the line symmetrically around the line center. You can also characterize the electronic time constant by chopping the laser light or cutting the current quickly and looking at the exponential decay on an oscilloscope.

Reply: The reviewer is correct that electronics would act as an RC filter of the data to produce an asymmetric line shape. We do not observe an asymmetric direct transmission line shape, but rather a symmetric Gaussian line shape (e.g., Figure 6). To produce a Gaussian line shape, we hypothesize that random fluctuations in either laser temperature or laser current are responsible. We have not proven this, however, so we will rewrite this paragraph similar to what this reviewer has recommended:

Lines 466-476:

"As discussed in Section 2.3.1, laser tuning rate (wavenumber versus index point) is determined by fitting seven CH_4 lines surrounding the targeted water line with a second-order polynomial. The instrument line shape is then characterized by comparison of a low-pressure absorption feature with the expected profile of a Doppler line shape. Figure 6 shows an experimental line shape obtained with the Herriott cell in a laboratory test chamber at a temperature of 297.54 K with pure water vapor at a pressure of $9x10^{-2}$ hPa. The observed absorptance, $(I-I_o)/I$, is only ~0.134, so the sample is optically thin and the line shape is due to only Doppler broadening. This Gaussian is 20% broader than the calculated Doppler-broadened lineshape for this measured temperature. We hypothesize that the difference between the Gaussian fit and the calculated Doppler broadening is dominated by fluctuations of the laser wavelength (associated with either temperature or dithering noise), but have not fully diagnosed the issue."

19. Line 518: Could you explain what you mean by the limited time response of the signal chain?

Reply: We have revised our explanation for the asymmetric negative lobes, which are also seen by other TDLAS. This effect is due to a shift between amplitude response and frequency response. In line 518 in the original manuscript, we speculated that the time response of the signal chain is the cause of asymmetric negative lobes (Figure 7). We will change the sentence at line 518 to:

"It has commonly been observed in the tunable diode laser (TDL) community that 2f spectra have asymmetric negative lobes, and other researchers have found reasons other than electronic filtering that can cause this artifact (e.g., Goldenstein et al., 2014)."

New reference:

Goldenstein, C. S., Strand, C. L., Schultz, I. A., Sun, K., Jeffries, J. B., and Hanson, R. K., Fitting of calibration-free scanned-wavelength-modulation spectroscopy spectra for determination of gas properties and absorption lineshapes, Applied Optics, 53(3), 2014, http://dx.doi.org/10.1364/AO.53.000356.

20. Figure 10: The y axis label says 'and Pressure'. I think that is a typo.

Reply: Good catch. We will change the y axis label to "Water (ppmv)"

21. Section 6. Performance: In all this discussion, you never state your accuracy. You talk about the calibration procedure in in section 2.3.2, but I expected at some point a statement or graph showing water vapor in your lab sample flow as determined by the Thunder Scientific versus water vapor mixing ratio calculated by JLH as you would do in flight. A plot of the points showing the linearity over a couple order of magnitude of water. A

difference plot and statement of accuracy in percent or ppmv. I think this is critical for an instrument paper. Comparison with other instruments is not sufficient.

Reply: here is the addition of accuracy discussion to the revised paper:

New text at line 323:

"For an open-path instrument such as JLH, it is necessary to place the laser/detector housing and sample cell inside a test chamber. The JLH test chamber used for laboratory calibration is a 304 Corrosion Resistant Steel (CRES) box vacuum chamber manufactured by the Kurt J. Lesker Company, and coated on the inside with a hydrophobic coating of Fluoropel® 1302IBA, manufactured by Cytonix Inc. of Beltsville, Maryland (Troy, 2007). For calibration of the JLH instrument in the lab, an air/water vapor mixture generated by the TS3900 flows continuously through the CRES chamber at constant pressure and temperature."

We will revise also Section 2.3.2 (at line 326) to discuss accuracy and to show recent calibration results:

"Below, we describe a recent example of JLH laboratory calibration. Fig. 4a and 4b show time series of JLH measurements within the lab calibration chamber as 200 ppmv and 50 ppmv, respectively, are flowed through the chamber.

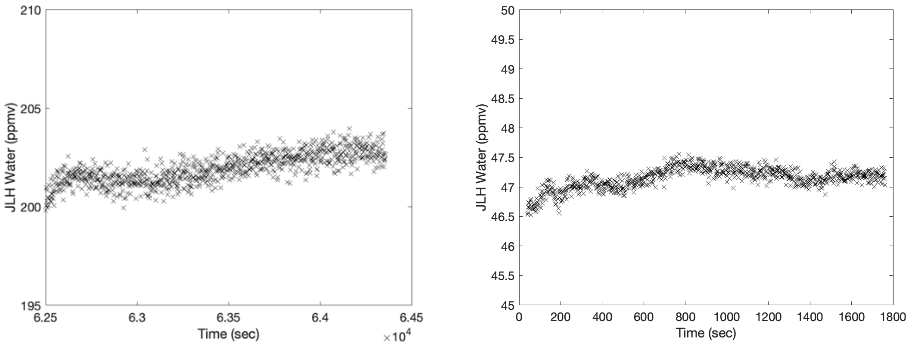

Figure 4. *Time series of JLH water vapor measurement inside the laboratory calibration chamber with continuous flow of air/water vapor mixture from the TS3900 humidity generator. (a) Left plot is 200 ppmv input at 57.2 hPa total pressure and 295.4 K temperature, (b) Right plot is 50 ppmv input at 120.3 hPa total pressure and 296.4 K temperature.*

Fig. 4c demonstrates the linearity of JLH response versus reference gas standard from the TS3900 from 3 ppmv to 200 ppmv. This is also shown in Table 3 along with the difference in percent and ppmv. Generally, results are within 9% but there is a pressure dependence as shown in Fig. 4d. This pressure dependence has been documented in our previous laboratory calibrations. At higher and lower pressures, the response changes systematically.

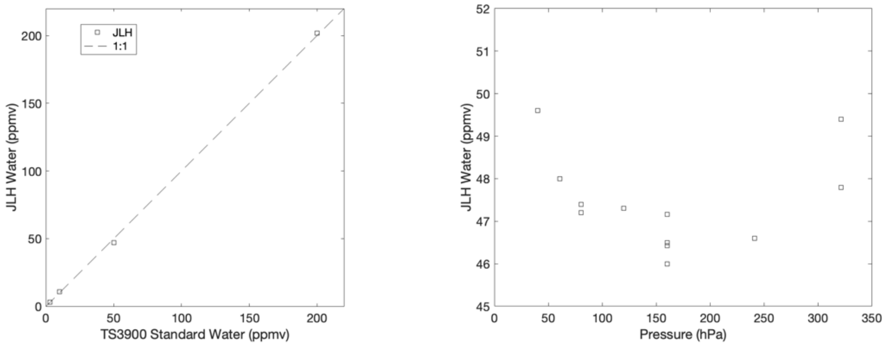

Figure 4. *(c) Left plot demonstrates that the relation of JLH water measurement to TS3900 water source is close to linear. A 1:1 line (dashed line) is also shown. (d) Right plot is JLH water measurement inside lab calibration chamber at different total pressures while the TS3900 provides a constant source of 50 ppmv water vapor at room temperature.*

Table 3. Summary of recent JLH laboratory calibrations

TS3900 Reference	**JLH Measurement**	**Drift**	**Fit**	**Offset**	**Offset (%)**
(ppmv)	(ppmv)	(ppmv)	(ppmv)	(ppmv)	(%)
200.00	201.98	0.7	199.51	2.47	1.2%
50.00	47.16	0.34	49.88	-2.72	-5.4%
50.00	46.43	0.14	49.88	-3.45	-6.9%
50.00	46.00	0.17	49.88	-3.88	-7.8%
10.00	10.85	0.06	9.98	0.87	8.8%
3.00	3.02	0.11	2.99	0.03	0.9%

The best estimate of JLH total error for NASA SEAC^{4}RS and more recent FTB flights is 12%, adding in quadrature the following terms. The largest error source is 9% in water measurement due to mismatch of the actual lineshape and the predicted lineshape as manifested as pressure-dependence (Fig. 4d). We estimate 5% uncertainty in water *measurement from uncertainty in the temperature dependence of the actual lineshape and 6% from scaling to laboratory calibrations (Fig. 4c and Table 3). There is 1% error in water measurement due to uncertainty in the in-flight temperature and pressure measurements. Finally, only 0.1% uncertainty in water measurement is due to uncertainty in the pathlength, and a fraction of a percent from other error sources such as wavelength, laser spectral purity, light scattering, et cetera.*