Atmos. Meas. Tech. Discuss., doi:10.5194/amt-2018-443-RC2, 2019 Author Response (AR) to Referee #1

We thank Referee #1 for the time spent on reviewing the manuscript and providing constructive comments. We will work on the revised manuscript accordingly. Answers to the comments are given below.

General Referee Summary: This paper reports the characterization of a new commercial laser-based HCHO sensor from Aeris Tech Inc. The 3σ detection limit of the sensor was 690 pptv with 15 min integration time. A comparison with LIF instruments was performed. The Aeris sensor provides a small and easier-to-operate HCHO sensor, which can be potentially adopted in networks. This work is interesting and meets the scope of AMT.

The new sensor takes advances in the design and data processing method, which should be included in the manuscript. Without these key informations, I do not see the compelling advances to publish in AMT in its present form.

Author Response: We appreciate the general and specific comments of Referee #1 and have added more information about the instrument design that is not proprietary or patented information. A newly-made schematic diagram now appears in the supplemental information to make the operating principle of the Aeris sensor easier to understand. We have also added a few more clarifying details about HAPP fit since the fast-fitting routine of ART fit is proprietary and closed-source (this was one of the primary reasons why HAPP fit was created for the Aeris sensor and is publicly available on GitHub).

Manuscript Changes: Modified part of Section 3: "Spectral parameters (such as the line position or the Doppler and Lorentz widths for each transition) are dynamically fixed or floated depending on a specified threshold, and spectral lines of the same molecular species are grouped together to better constrain the final fit. All spectral line information can be easily sourced from the HITRAN database. While HAPP fit itself is written in C++, the program is supported by a suite of MATLAB scripts to assist in setting up the necessary configuration files from the Aeris raw data and to process the output of HAPP fit into finalized HCHO mixing ratios."

Comment 1: A schematic diagram is useful to make the principle of the sensor clearer.

Author Response: We agree with the referee and have added a schematic of the sensor.

Manuscript Changes: The following figure was added to supplemental information:

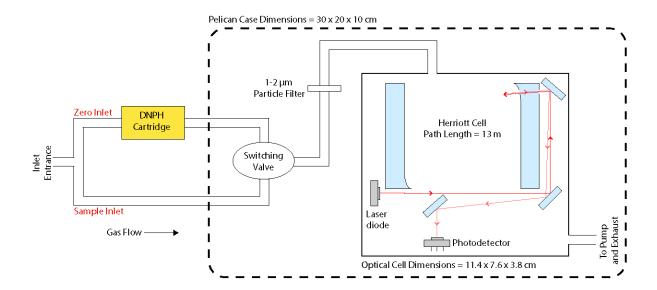


Figure S1. Schematic diagram of the absorption-based Aeris HCHO sensor. Air flows through the inlet entrance, and the switching valve either allows air to pass directly into the instrument via the sample inlet or is first scrubbed of HCHO via the zero inlet. Before entering the Herriott cell, all dust is removed from the air flow with a 1-2 μ m particle filter. The patented folded Herriott cell (US Patent #10,222,595) has a path length of 13 m and dimensions of 11.4 x 7.6 x 3.8 cm (Paul, 2019). The laser diode, photodetector, filters, and mirror coatings are proprietary information.

Comment 2: Details about the multi-pass cell (the type of the cell, diameter, coating of the mirrors, and some related references) are not clear.

Author Response: Additional details about the multi-pass cell have been added to make it clearer for the reader. The multi-pass cell is a folded Herriott cell with a 13 m path length, dimensions of 11.4 x 7.6 x 3.8 cm, and a volume of 60 cm³. The cell is also patented (US Patent #10,222,595) and details about the coating on the mirrors are proprietary. Moreover, the Herriott cell shown in Figure S1 aims to accurately capture the light path of the laser beam as it enters and exits the cell (this information was reproduced from Figures 3 and 4 of US Patent #10,222,595).

Manuscript Changes: "A proprietary folded Herriott detection cell (Paul, 2019) inside the instrument has a 1300 cm path length, a volume of 60 cm^3 , and dimensions of $11.4 \times 7.6 \times 3.8 \text{ cm}$ ($4.5 \times 3 \times 1.5 \text{ inches}$) (Fig. 1 with a simple schematic in Fig. S1)."

Comment 3: Details about the fast-fitting routine (ART) and some related references are not clear.

Author Response: All details about the fast-fitting routine (ART) are proprietary to Aeris Technologies, which is one of the primary reasons why we developed HAPP fit as it would give the user complete control over fitting the raw 1 Hz spectral data rather than relying solely on the closed source ART fit. Developing our own non-linear least-squares fitting routine also allowed us to compare our fit to that of ART fit as we show in Eq. 2. The two fits agree to within a few percent of each other with very small offset. The fitting code for HAPP fit was further

made open source by making it available to the public via GitHub (https://github.com/nthallen/le-icosfit)

Manuscript Changes: No changes made due to the proprietary nature of ART fit.

Comment 4: Table 1, for absorption spectroscopy, what were the path lengths previously used? Then the readers can clearly see the sensitivity of the Aeris sensor with 13 m absorption pathlength and that with hundreds meters.

Author Response: We agree that including this information as a caption to Table 1 will help the reader in comparing the sensitivity of the Aeris sensor with a 13 m absorption path length to the research-grade instrumentation having path lengths that are 1-2 orders of magnitude higher than the Aeris.

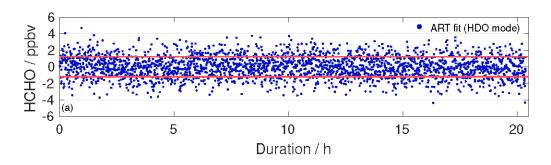
Manuscript Changes: "The path length of the astigmatic Herriott cell in the TDLAS and QCLS instruments are 100 and 200 m, respectively. The DOAS instrument has a light path of 960 m, and the BBCEAS instrument has an effective path length of 1430 m."

Comment 5: Fig. 3, for the Allan-Werle deviation, time series measurement results need to be shown. I found some disagreement between the Allan deviation and Fig. 4 and 5. The peak-to-peak variations were obviously larger than the value getting from Allan's plot.

Author Response: We will include time series measurement results for the Allan-Werle deviation curve.

Concerning the apparent disagreement when visually comparing the results of Fig. 3 to those of Fig. 4 and 5, we note that the peak-to-peak variation at each step is not equivalent to the 1σ standard deviation at each step (which is what the Allan plot shows). When we do calculate the 1σ standard deviation at each step in Fig. 4 and 5 (note that the integration time shown on the plots is 30 s), it's 1.0 - 1.3 ppbv HCHO for the HAPP HDO fits and 1.6 - 1.7 ppbv HCHO for the HAPP CH₄ fit. These results are in general agreement with what is shown in Fig. 3 and any discrepancies could be due to the fact that Fig. 3 was derived using zero air.

Manuscript Changes: The raw data used to derive the Allan-Werle deviation curves were added as Figure S2 to the supplemental information:



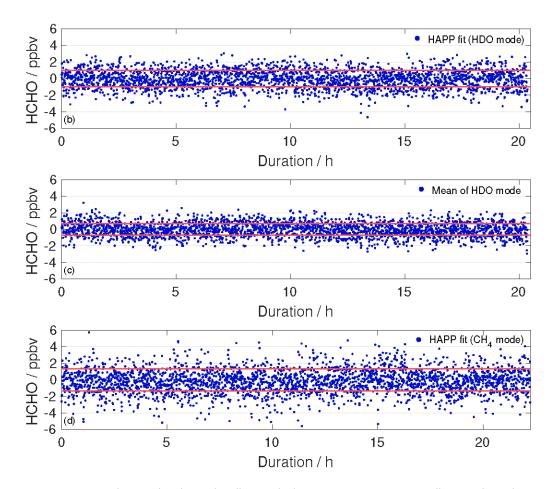


Figure S2. Raw time series data used to derive the Allan-Werle deviation curves in Figure 3. All points shown have an integration time of 30 s and were obtained by flowing ultra zero air through the Aeris sensor for a minimum of 20 h. Red lines indicate $\pm 1\sigma$ standard deviation from the mean of the data. Raw data for (a) ART fit (HDO mode), (b) HAPP fit (HDO mode), (c) mean of HDO fits, and (d) HAPP fit (CH₄ mode).

Comment 6: Please explain the abbreviations in the manuscript, "CAFE", "ISAF", "FILIF",...

Author Response: We agree with the referee and are defining the abbreviations for all the LIF instrumentation used in this study:

- (1) CAFE stands for the Compact Airborne Formaldehyde Experiment
- (2) ISAF stands for the In Situ Airborne Formaldehyde instrument
- (3) FILIF stands for the Fiber Laser-Induced Fluorescence HCHO instrument

Manuscript Changes:

 "During a HCHO multi-hour intercomparison performed at NASA Goddard in November 2017, the Aeris sensor was operated in HDO mode and compared against two NASA LIF instruments: NASA ISAF (In Situ Airborne Formaldehyde; Cazorla et al., 2015) and NASA CAFE (Compact Airborne Formaldehyde Experiment; operating principle described in St. Clair et al., 2017)." "The Aeris sensor was also operated in CH₄ mode in the laboratory and compared against the Harvard FILIF (Fiber Laser-Induced Fluorescence) HCHO instrument described previously (DiGangi et al., 2011; Hottle et al., 2009)"

Comment 7: Some explanations of the 2% disagreement of the Aeris sensor and NASA CAFE and ISAF are necessary. How were these two LIF instruments calibrated? Positive offset (180 to 210 pptv) was within the detection limit of Aeris sensor.

Author Response: The reviewer is correct that the positive offset is within the detection limit of the Aeris sensor (the same also holds true for the negative offset when comparing to Harvard FILIF). That being said, all fits (both with NASA and Harvard LIF instrumentation) show an intercept that is non-zero at the 95% confidence interval, which implies that we cannot definitively rule out a real, though minor, offset whose source is unknown.

The two NASA LIF instruments were calibrated previously with a HCHO gas cylinder from Air Liquide whose concentration was verified using Fourier transform infrared (FTIR) spectroscopy (following the procedure described in Cazorla et al., 2015). The Aeris sensor was also calibrated previously at Harvard using a different HCHO gas cylinder (but this cylinder's HCHO mixing ratio was also previously verified with the same FTIR instrument as the cylinder used for calibrating the LIF instruments).

It should be noted that during the comparison of the Aeris sensor with NASA CAFE and ISAF, a small humidified stream of zero air (158 sccm) was added to the Aeris sensor only, and even though a correction was applied to account for this added dilution, it still adds in extra uncertainty. This could in part help to explain why there was a slight 2% disagreement with the Aeris sensor and NASA's LIF instruments as opposed to the 0.5 - 1% error when comparing the Aeris to Harvard FILIF (both instruments sampled the same airflow that had a small flow of (<1 sccm) of ultra-pure CH₄).

Manuscript Changes:

"Prior to the intercomparison, all instruments were calibrated independently using
HCHO gas cylinder standards that had been verified by Fourier transform infrared (FTIR)
spectroscopy. In brief, the HCHO standard is verified by flowing it through an FTIR cell
for several hours to allow the signal to equilibrate, and the resulting HCHO mixing ratio
is scaled by a factor of 0.957 in order to tie the calibration to UV cross—sections by
Meller and Moortgat (2000) (Cazorla et al., 2015)"