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Atmospheric Measurement Techniques Discussions

Interactive comment on "Intercomparison of Open-Path Trace Gas Measurements with Two Dual Frequency Comb Spectrometers" *by* Eleanor M. Waxman et al.

Anonymous Referee #2

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This paper compares the performance of two open-path dual comb spectroscopy (DCS) instruments over two weeks and analyzes the measurement differences, also between the DCSs and a Cavity ring down spectrometer measuring in-situ. The paper describes the function principle of the dual comb spectrometers, the optical setup, and the data processing. The precision of the individual DCS is determined using Allan analysis.

This kind of inter-comparison has not been done so far, and the DCS instruments deployed here are novel. The descriptions and analyses are clear structured. The reviewer recommends the publication of this paper, if the following items are addressed.

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Discussion paper

General comments:

1. As the authors noted, the dual comb spectrometer has negligible line width (120 kHz) and high spectral resolution, the absorbance resolution is below 10^{-3} ($5 \cdot 10^{-4}$), which is limited by the instrument noise. However, the laser spectroscopic instrument typically has an absorbance resolution of 10^{-5}, limited by the shot noise of the photodiode (also depends on the incoming light intensity). Could the authors be more specific about the origin of the noise? What kind of instrumental noises are these, photodetector noise, noise from the dual comb laser?

2. What benefits in the authors' opinion could be gained from the open-path DCS techniques compared to open path FTIR? There seems to be no benefit in terms of the achievable absorbance resolution, and the measurement precision is comparable. One argument brought by the authors is the "rapid scan rate" and "faster than turbulence-induced intensity variations", and "instrument-specific calibrations" are not needed. In the reviewer's opinion, it would be beneficial if the authors could discuss and summarize the benefits/drawback compared to open path FTIR in one paragraph.

3. The principle of dual comb technology should be briefly stated using mathematical formulas, so that it is easier for readers to follow. For example, formulas can be written below "two combs with nominal repetition rates of f_r and offset by Δf_r are phase locked together, transmitted through a sample, and their heterodyne signal measured on a photodetector. The resulting rf frequency comb can be mapped back to the optical domain to generate an overall spectrum".

4. The authors state that "the instrument line shape is the sum of two Delta impulses as shown in Fig. 1 (b)". The instrument line shape is not visible in Fig. 1 (b). It would be beneficial if the ILS can be schematically shown with the spectral spacing between the two impulses indicated.

5. It is difficult to assess the measurement precision of atmospheric open-path instruments because the measurement conditions (P/T/vmr) are usually not stable and constant. The authors use the measurements in 6 hours well-mixed time period to calculate Allan deviations. Can the authors draw any conclusions regarding to the 1000s turning point in the Allan plots? Is it instrument drift or given by variations of the atmospheric conditions?

6. The authors have done Allan analysis to assess the precision of individual DCS. The reviewer thinks that it would be interesting to conduct the Allan analysis for the measured concentration differences (Fig. 6). On one hand, it will give an indication of the precision of the differential measurements, on the other hand, the atmospheric influences will be cancelled out, which could be advantageous for conducting Allan analysis. Differential column measurements have been recently successfully used for determining local/city emissions [1, 2]. In [1] the measured differences between two side-by-side solar-viewing FTIRs are analyzed using Allan analysis, to determine the precision of the differential system that consists of two spectrometers (0.01% for XCO2 and XCH4 over 10-minute integration time) and the precision of an individual instrument (assuming the instruments are the same and the measurement noises are statistically uncorrelated). [1] presents a new way to determine the precision of atmospheric measurements, and could be included in the references.

7. Statistical distribution of the differences (caption of Fig. 7): if the Allan deviation of the differences follows a square root law (inversely proportional to the square root of the integration time), the distribution widths should be the third when increasing the integration time from 32s to 5 min (factor 10).

Specific comments:

1. Line 16: better to write "$5 \cdot 10^{-4}$ in absorbance"

2. Line 17: better to write “path-integrated concentrations for carbon dioxide (CO2)”

3. Line 19: averaging time interval information is missing: at 32 s integration time

4. Line 125: “with nominal repetitions rates f_r and the difference in the repetition rates Δf_r “

5. Line 132: absolute frequency accuracy is written in wavenumber, it would be beneficial if it is also converted to frequency, i.e. 1 MHz.

6. Line 220: which parallel surfaces cause these etalons?

7. Fig. 4: big discrepancies around 6290 cm^{-1} in both (c) the differences between absorption spectra and (d) fitting residual, please specify the reason for it.

8. Line 644, caption of Fig. 8: ...highlighted in Fig. 3 -> it should be Fig. 5

References:

[1] Chen, J., Viatte, C., Hedelius, J. K., Jones, T., Franklin, J. E., Parker, H., Gottlieb, E. W., Wennberg, P. O., Dubey, M. K., and Wofsy, S. C.: Differential column measurements using compact solar-tracking spectrometers, Atmos. Chem. Phys., 16, 8479-8498, doi:10.5194/acp-16-8479-2016, 2016.

[2] Hase, F., Frey, M., Blumenstock, T., Groß, J., Kiel, M., Kohlhepp, R., Mengistu Tsidu, G., Schäfer, K., Sha, M. K., and Orphal, J.: Application of portable FTIR spectrometers for detecting greenhouse gas emissions of the major city Berlin, Atmos. Meas. Tech., 8, 3059-3068, doi:10.5194/amt-8-3059-2015, 2015.

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