## Dual-Comb Spectroscopy of Carbon Dioxide and Methane Across a 14.5 km Long Outdoor Path

F. R. Giorgetta, <sup>1,2,\*</sup> E. Baumann, <sup>1,2</sup> B. R. Washburn, <sup>1</sup> N. Malarich, <sup>1</sup> J.-D. Deschênes, <sup>3</sup> I. Coddington, <sup>1</sup> N. R. Newbury, <sup>1</sup> and K. C. Cossel <sup>1</sup>

<sup>1</sup>National Institute of Standards and Technology, Communications Technology Division, Boulder, CO 80305, USA

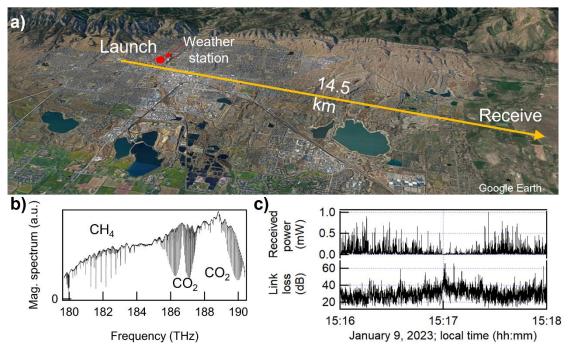
<sup>2</sup>University of Colorado, Boulder, Department of Physics, Boulder, CO 80309, USA

<sup>3</sup>Octosig Consulting, Québec, Québec GIV 0A6, Canada

Author email address: fabrizio.giorgetta@nist.gov

**Abstract:** Greenhouse-gas dual-comb spectroscopy is extended to a city-scale 14.5-km path length using remote receiver and data acquisition. This configuration enables lower link losses and longer path lengths compared to folded paths with a remote retroreflector. © 2023 The Author(s) Work of the US Government and not subject to copyright.

Open-path dual-comb spectroscopy (DCS) has been demonstrated for monitoring city emissions using path lengths up to 3.35 km [1]. However such paths are not truly city-scale and path lengths on the order of 10-100 km are desirable to quantify emissions from a medium to large size city. To date, open-path DCS mostly uses a configuration with DCS system, transmitter, receiver, and data acquisition co-located and the light probing a path between a transceiver telescope and a remote retroreflector. While convenient, such folded paths have an optical path length L which is twice the physical path length. Even worse, one invariably overfills the retroreflector leading to a  $L^4$  scaling for return power and thus SNR, making it very difficult to scale this folded geometry to longer distances.

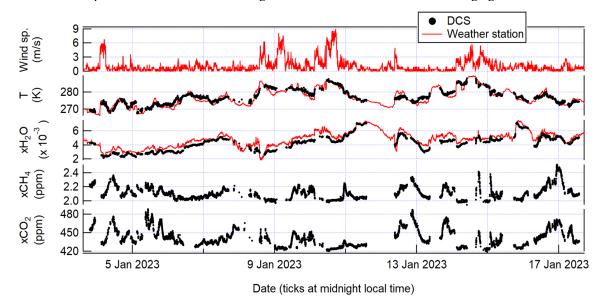


**Figure 1:** overview. **a)** The 14.5-km DCS measurement between NIST Boulder launch site and Table Mountain receive site. **b)** Raw DCS spectrum acquired across the 14.5-km link (two-minute averaging time). **C)** Turbulence impact for a typical 2-minute measurement window.

Here, we demonstrate an open-path geometry where the DCS transmitter and receiver are separated on the two ends of the path, which means splitting the launch site from the data acquisition at the opposite end of the path. This "unfolded" geometry has equal optical and physical path lengths L and results in a more favorable  $L^{-2}$  power scaling, allowing us to achieve high signal-to-noise ratio (SNR) over significantly longer paths. In this demonstration, we separate the transmitter and receiver by 14.5 km as shown in Fig. 1. However, this comes at the cost of some

increased system complexity as coherent averaging of DCS interferograms requires that the acquisition electronics be synchronized with the frequency comb repetition rate and dual-comb interferogram length. We introduce a common time base between the combs at the launch site and the data acquisition at the remote receive site by referencing both to local GPS disciplined oscillators. This along with real-time computational timing and phase correction of the acquired interferograms at an 8 Hz rate allows for successful coherent co-adding of interferograms for arbitrary averaging times without penalty in signal strength. Figure 1c) shows typical link loss behavior of the 14.5 km link during a 2-minute averaging window with an average received power of 60 uW for 20 mW launched light.

Using the remote receiver and data acquisition configuration, we measure atmospheric trace gasses across the 14.5-km path (see Fig. 2) over two weeks. The system is based on a pair of Er:fiber frequency combs measuring in the 1560 nm -1700 nm spectral region with a 200 MHz point spacing. The light from both combs is launched from a custom telescope with a 6-cm launched field diameter located on an azimuth-elevation gimbal. The telescope gimbal pointing was manually realigned every couple of hours to compensate for slow telescope pointing drifts. At the remote end, the light was received with a 15-cm-diameter telescope, which stayed aligned without adjustment during the measurement period. The measured interferograms were coadded for 2-minute averaging time.



**Figure 2:** Extracted path-averaged CO<sub>2</sub>, CH<sub>4</sub>, and H<sub>2</sub>O concentrations and temperature across the 14.5 km link during the 2-week measurement period. Also shown is data from a weather station located along the path.

Figure 2 shows the extracted atmospheric  $CO_2$ ,  $CH_4$  and  $H_2O$  concentrations over the twelve-day measurement period. The transmission spectrum shown in Fig. 1b) is baseline corrected and fitted to a model based on HITRAN 2020 [2] and Voigt line shapes, with path-averaged trace gas concentrations as well as temperature and pressure (not shown here) floating.  $CO_2$  and  $CH_4$  show significant correlated variation due to atmospheric variations coupled with local/regional emissions. The sensitivity is estimated to be 0.25 ppm and 0.003 ppm for  $CO_2$  and  $CH_4$ , respectively at two-minutes of averaging.

These results indicate that separating receiver and transmitter allows one to scale up dual-comb spectroscopy path lengths to encompass city-scale green-house gas emissions measurement. Additionally, the turbulence for this city-scale path length which stayed close to ground was quite strong ( $C_n^2 \approx 4 \times 10^{-14} \text{ m}^{-2/3}$ ) [3], reducing the average transmitted power. Vertical paths, or slant paths of a few degrees, would experience orders of magnitude less turbulence and could be pushed to even longer distances, potentially enabling ground to stratospheric balloon measurements of greenhouse gasses.

- 1. Waxman et al., Atmos. Chem. Phys., 19, 4177–4192 (2019)
- 2. I. E. Gordon, et al., J. Quant. Spectrosc. Radiat. Transf. 277, 107949 (2022)
- 3. Bodine et.al., APL Photonics 5,076113 (2020)