Open-path, Mid-infrared, Dual Comb Spectroscopy for Measurement of Ambient Ethane and Propane

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Open-path measurements of atmospheric gas species over km-scale path lengths are well suited to quantify emissions from sources like oil and gas, forest fires, and industry. Dual frequency comb spectroscopy (DCS) is a relatively new technique that combines high-resolution and broad spectral coverage with no instrument lineshape and near perfect frequency calibration. These features make DCS well suited for accurate measurements of multiple species simultaneously. Because the frequency comb lasers can be well collimated, such a system can be used for long open-path measurements with pathlengths ranging from hundreds of meters to several kilometers.

Previous demonstrations of open-path DCS have primarily been in the 1–2 μm spectral region; however, in order to reach the sensitivity necessary to detect many atmospheric trace constituents, including volatile organic compounds, operation in the mid-infrared (or UV/Vis) is required. Here, we show a mid-infrared, open-path, dual comb spectrometer operating in the 3–5 μm spectral region. We have used this spectrometer to measure methane, ethane, and propane (arising primarily from oil and gas activity) across a 1–km-long path in Boulder, CO for one week with an ethane sensitivity of ~0.1 ppb for a two-minute time resolution. In addition, we show quantitative measurements of intentionally released acetone and isopropanol with a 1-σ sensitivity of 5.7 ppm·m and 2.4 ppm·m, respectively. Finally, we discuss the outlook for detection of additional species such as N_2O, CO, and O_3 as well as a second-generation instrument that is more compact and has improved stability.

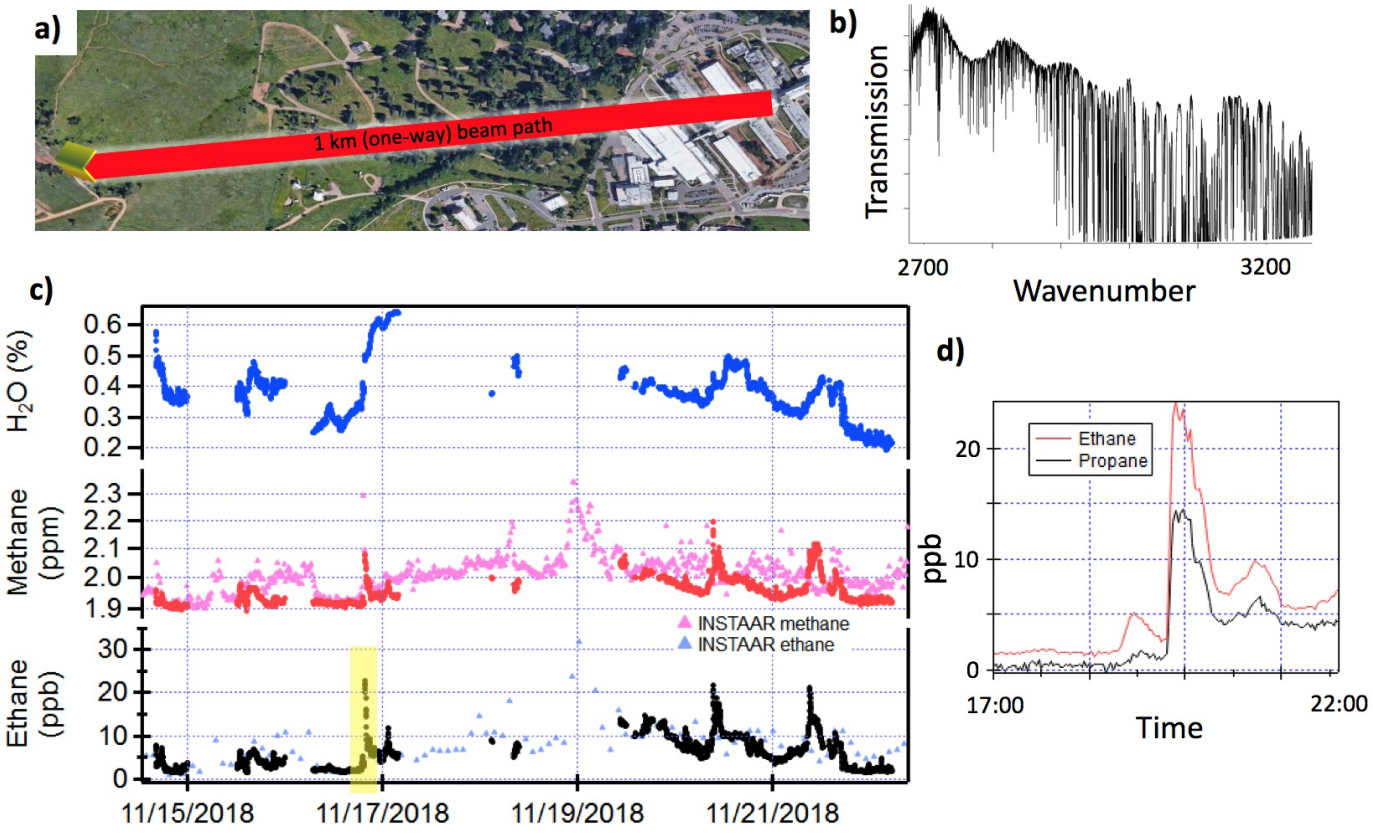

Figure 1. Open-path, mid-infrared, dual comb spectroscopy. (a) 1–km measurement path from NIST to Kohler Mesa. (b) Transmission spectrum showing strong water and methane absorption. (c) Retrieved path-averaged concentrations of H_2O, methane, and ethane over a one-week measurement series. The gaps are primarily due to poor weather. (d) Shorter time series showing retrieved propane.