EPJ Web of Conferences **362**, 09005 (2026) https://doi.org/10.1051/epjconf/202636209005
ILRC31/CLRC22-2024

Open-path Dual-Comb Spectroscopy Observations of Greenhouse Gas Spatial and Temporal Variability over Km-scale Paths

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Abstract: A dual-comb spectrometer is deployed for kilometer scale open-path measurements of CO_2 and CH_4 at building-top heights in Manhattan, New York City. The system operated for more than 3 months in summer 2023 and more than 1 month in winter 2024. The measurements show the high spatial and temporal variability of greenhouse gas emissions in an urban setting.

1. Introduction

Estimates attribute 50% of all greenhouse gas emissions in the United States to urban areas, and this fraction is expected to grow as urban populations continue to increase. Greenhouse gas emission reduction in urban areas is crucial for meeting national and global climate goals, and many cities have made commitments to reduce their emissions in the coming years. For example, New York City is targeting 40% reductions in emissions by 2030 and 80% by 2050, relative to 2005 levels. City policies encourage emissions reductions to meet these goals, however effective monitoring of emissions is necessary to track progress. Furthermore, the ability to identify and quantify specific source contributions is needed. Contributions from different source-sectors is normally determined using emissions inventories, which uses activity data such as natural gas usage, traffic counts, etc. to scale emissions factors and estimate total emissions.[1] In order to accurately reflect total emissions and source contributions, inventories must be informed by accurate knowledge of emissions sources and their associated emissions factors. Recent research suggests that urban greenhouse gas emissions may be severely underestimated by current inventories, especially for CH_4[2], [3].

Urban areas include strong discrete point sources of emissions such as wastewater treatment plants and power generation facilities alongside distributed sources such as natural gas distribution and combustion.

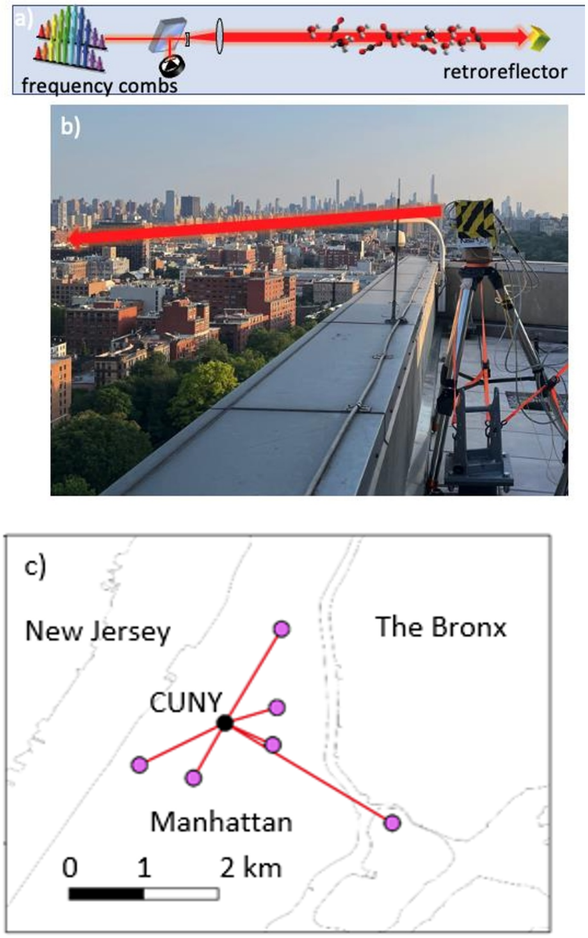

Figure 1. a) Diagram of DCS open path measurement system setup. b) Photograph of signal transmit/receive telescope on the rooftop of the CUNY ASRC building in New York City. c) Map of the measurement region in Harlem, New York City including the CUNY ASRC building, measurement beam paths, and retroreflector locations.

Discerning the relative contributions of discrete and dispersed sources remains a significant challenge for urban emissions monitoring. Most greenhouse gas observations are made using stationary single-point sampling sensors, and additional area-wide measurements are made by aircraft or satellite observations. However, satellite observations have limited temporal and spatial resolution, and research aircraft flights are costly and infrequent. To date, there are few long-term, kilometer sale observations of greenhouse gasses. Here we use dual-comb spectroscopy (DCS) measurements to provide new insight into greenhouse gas variability with long term, minute resolution, kilometer scale observations.

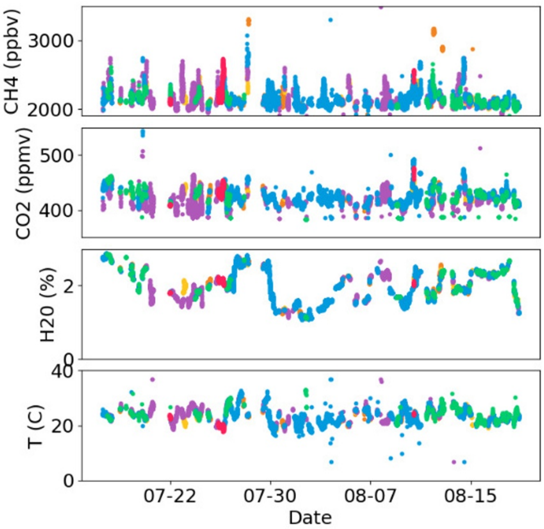

Figure 2) Time series of retrieved concentrations from a subset of the measurement period using 1 minute time resolution. Colors represent different beam paths.

2. Methods

The presented measurements were made using an open-path DCS in the 1.58-1.75 μm spectral range.[4] A transmit/receive telescope was installed on the rooftop of the City University of New York (CUNY) Advanced Science Research (ASRC) building in the Harlem borough of northern Manhattan, New York City. The DCS system includes two frequency combs generated by mode-locked erbium-fiber lasers. The combs have nominal repetition rates of 200 MHz, and the difference in repetition rate between the two combs is ~400 Hz. The two combs are spectrally broadened to the 1.58-1.75 μm spectral range and simultaneously launched from the fiber into the transmit receive telescope (Figure 1b). The light passes through a beam splitter and is then collimated on an off-axis parabolic mirror (10-cm aperture), as shown in Figure 1a, before being transmitted over a 0.72-2.87 km path. A high-precision programmed gimbal aligns the beam with one of the six retroreflectors placed throughout the measurement domain (Figure 1c), which returns the light back to the same transmit/receive telescope. After passing back through the telescope, the light is reflected by the beam splitter onto an InGaAs photodetector. The rf signal measured by the photodetector is the result of heterodyne detection of the two comb signals, allowing the high spectral resolution absorption information to be transferred to rf frequencies. The rf signal is digitized and passed to an FPGA that performs real-time phase correction and coherent averaging[5] to provide high-resolution spectra with 0.0067 cm^{-1} (200 MHz) point spacing at minute time resolution.

Spectral fitting is performed using cepstral domain free-induction decay analysis[6] to retrieve path-averaged H_2O, CO_2, and CH_4 concentrations as well as the path averaged temperature (Figure 2). HDO concentrations can also be retrieved but are not shown.

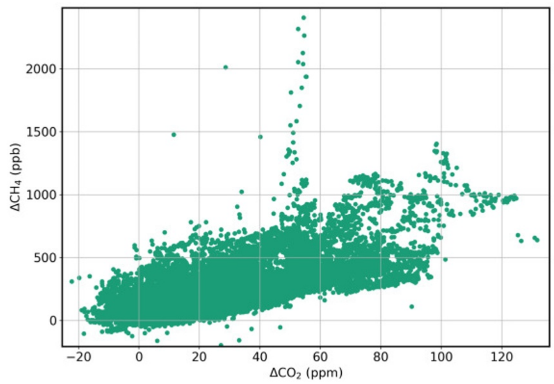

Figure 3) Scatter plot of CH_4 concentration and CO_2 concentration, showing a ratio of ~5 ppb CH_4/ppm CO_2 along with some datapoints with differing ratios. Recent anthropogenic emissions inventories predict a ratio of ~3 ppb CH_4/ppm CO_2 (not including biospheric contributions). [7], [8]

3. Results

Observations show large temporal and spatial variability of greenhouse gas concentrations, with diurnal and weekly patterns as well as non-regular plumes. CH_4 often reached concentrations of 3 ppm or higher, many of these plumes lasting for over 1-hour periods. While most measurements are consistent between beam paths, some time periods show significant differences in gas concentrations when switching beam paths. Similarly, DCS measurements often agree with point sampling measurements on the CUNY ASRC building, but sometimes show differences that reveal information about spatial extent of plumes. Point sampling measurements from CUNY ASRC are also available for other trace gasses, such as C_2H_6 and N_2O, which show enhancements corresponding to large plumes detected by the DCS.

High variability of CO_2/CH_4 ratios are observed, indicating a variety of combustion and not combustion related sources contributing to the atmospheric greenhouse gas composition. The background level of CO_2/CH_4 ratios constrains the relative emissions of the two gasses, and is consistent with levels previously observed in Los Angeles, California. Comparison between observations ratios predicted by emissions inventories suggest that the inventories underestimated CH_4 sources. We analyze these observations using meteorological data and inverse dispersion modeling to identify large greenhouse gas emitters in the area, such as nearby wastewater treatment plants, landfills, and power plants.

4. Conclusion

Urban areas contribute a large proportion of global greenhouse gas emissions, which must be decreased to meet climate goals. Accurate monitoring of greenhouse gas emissions is difficult in urban areas because of the wide variety of discrete and widespread emission sources that contribute to total emissions. A dual comb spectrometer was deployed in New York City, taking CH_4 and CO_2 measurements for over 3 months in the summer of 2023 and over 1 month in winter 2024. These measurements provide new insight as the first long-term, high temporal resolution, kilometer scale measurements of greenhouse gasses in New York City. Results show that CH_4 emissions are underestimated by inventories, and that greenhouse gas concentrations are highly spatially and temporally variable in an urban environment.

5. References

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