



Long-range open-path greenhouse gas monitoring using mid-infrared laser dispersion spectroscopy

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Accurate and sensitive methods of monitoring greenhouse gas (GHG) emission over large areas has become a pressing need to deliver improved estimates of both human-made and natural GHG budgets. These needs relate to a variety of sectors including environmental monitoring, energy, oil and gas industry, waste management, biogenic emission characterization, and leak detection. To address the needs, long-distance open-path laser spectroscopy methods offer significant advantages in terms of temporal resolution, sensitivity, compactness and cost effectiveness. Path-integrated mixing ratio measurements stemming from long open-path laser spectrometers can provide emission mapping when combined with meteorological data and/or through tomographic approaches.

Laser absorption spectroscopy is the predominant method of detecting gasses over long integrated path lengths. The development of dispersion spectrometers measuring tiny refractive index changes, rather than optical power transmission, may offer a set of specific advantages¹. These include greater immunity to laser power fluctuations, greater dynamic range due to the linearity of dispersion, and ideally a zero baseline signal easing quantitative retrievals of path integrated mixing ratios. Chirped laser dispersion spectrometers (CLaDS) developed for the monitoring of atmospheric methane and carbon dioxide will be presented. Using quantum cascade laser as the source, a minimalistic and compact system operating at 7.8 μm has been developed and demonstrated for the monitoring of atmospheric methane over a 90 meter open path². Through full instrument modelling and error propagation analysis, precision of 3 ppm.m.Hz^{-0.5} has been established (one sigma precision for atmospheric methane normalized over a 1 m path and 1 s measurement duration). The system was fully functional in the rain, sleet, and moderate fog.

The physical model and system concept of CLaDS can be adapted to any greenhouse gas species. Currently we are developing an in-lab instrument that can measure carbon dioxide using a quantum cascade laser operating in the 4 μm range. In this case, the dynamic range benefit of CLaDS is used to provide high precision even when peak absorbance in the CO₂ spectrum gets greater than 2. Development for this deployable CO₂ measurement system is still at an early stage. So far laboratory gas cell experiments have demonstrated a 9.3 ppm.m.Hz^{-0.5} for CO₂ monitoring. This corresponds to about 0.02% relative precision in measuring CO₂ atmospheric background over a 100 m open-path in one second.

¹ G. Wysocki and D. Weidmann, "Molecular dispersion spectroscopy for chemical sensing using chirped mid-infrared quantum cascade laser," *Opt. Express* 18(25), 26123-26140 (2010).

² N.S. Daghestani, R. Brownsword, D. Weidmann, "Analysis and demonstration of atmospheric methane monitoring by mid-infrared open-path chirped dispersion spectroscopy" *Opt. Express* 22(25), A1731-A1743 (2014).