

ACTIVE STAND-OFF DETECTION OF GAS LEAKS USING A SHORT RANGE HARD-TARGET BACKSCATTER DIFFERENTIAL OPTICAL ABSORPTION SYSTEM BASED ON A QUANTUM CASCADE LASER TRANSMITTER

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ABSTRACT

Fugitive gas emissions from agricultural or industrial plants and gas pipelines are an important environmental concern as they can contribute to the global increase of greenhouse gas concentration. Moreover, they are also a security and safety concern because of possible risk of fire/explosion or toxicity. This study presents gas concentration measurements using a quantum cascade laser open path system (QCLOPS). The system retrieves the path-averaged concentration of N₂O and CH₄ by collecting the backscattered light from a scattering target. The gas concentration measurements have a high temporal resolution (68 ms) and are achieved at sufficient range (up to 40 m, ~ 130 feet) with a detection limit of 2.6 ppm CH₄ and 0.4 ppm for N₂O. Given these characteristics, this system is promising for mobile/multidirectional remote detection and evaluation of gas leaks. The instrument is monostatic with a tunable QCL emitting at ~ 7.7 μm wavelength range. The backscattered radiation is collected by a Newtonian telescope and focused on an infrared light detector. Puffs of N₂O and CH₄ are released along the optical path to simulate a gas leak. The measured absorption spectrum is obtained using the thermal intra-pulse frequency chirped DFB QCL and is analyzed to obtain path averaged gas concentrations.

1. INTRODUCTION

In order to reduce greenhouse gas concentrations, there is a need for tools able to detect fugitive gas emissions from agricultural, industrial plants, or natural gas pipeline infrastructure. This is of concern for the global warming [1] as well as possible toxicity or fire and explosive risks. Optical methods to measure fugitive emission flux has been developed in order to detect greenhouse (or hazardous) gases [2-5], these methods are appropriate for fugitive emissions as they cover larger area than point sensors.

In this contribution, we present a potential system for fast gas concentration measurements based on a distributed feedback quantum cascade laser (DFB - QCL), its central wavelength of emission when used in a long pulsed (200+ nanoseconds) mode is rapidly chirped during the course of the pulse resulting in a well defined frequency sweep allowing this type of laser to be use for intra-pulse spectroscopy [6]. Using the backscattered light of a hard target, this system is promising for mobile/multidirectional remote detection and quantification of fugitive gas leak. Laboratory measurements have been performed on methane (CH₄) and nitrous oxide (N₂O), respectively the second and third most potent anthropogenic greenhouse gases [1] over a distance of 40 meters (~ 130 ft) and with a high temporal resolution (68 ms).

2. METHODOLOGY

The system is based on differential optical absorption spectroscopy (DOAS) [7]. A quantum cascade laser (QCL) emits a laser pulse also referred to as the reference signal $I_0(\nu)$. This pulse is pointed towards a topographic scattering target (in this demonstration, a well-characterized near-Lambertian scattering target). A coaxial optical Newtonian telescope is used to collect the backscattered light which is then focused on an optical detector resulting in a measured signal $I(\nu)$. During propagation, the laser pulse undergoes optical extinction due to the presence of the gas leak.

A remarkable feature of the QCLOPS system is that it does not use a spectrally resolved detector. In order to measure an absorption spectrum, we use the intra-pulse frequency chirp of a pulsed quantum cascade laser (1 cm^{-1} in ~ 200 nanoseconds in our case), which is remarkably stable and repeatable [6, 8]. Therefore, the time resolved intensity measurements $I(t)$ can be converted to a spectrally resolved intensity $I(\nu)$ using the experimentally-determined chirping rate of the QC laser.

The negative logarithm of the ratio between the reference signal $I_0(\nu)$ and the measured signal $I(\nu)$ is equal to the gas absorbance times a constant, from which the target gas concentration can be retrieved using the least-squares fitting technique [9-11]. The range to the target is measured using time of flight of the hard target backscatter (echo). The intensity of the signal $I(\nu)$ measured by the system may vary due to laser intensity fluctuations or changes in scattering efficiency of the target, as a consequence only the relative absorbance can be measured. However, using the least-squares fitting technique there is no need to measure the absolute absorbance and therefore no need to calibrate the signal intensity.

3. EXPERIMENTAL RESULTS

Measurements of N_2O and CH_4 have been performed illuminating a scattering target 40 m away (~ 130 feet) in the mid-infrared spectral range on rotational absorption bands located between 1295 and 1296 cm^{-1} ($\sim 7.7 \text{ }\mu\text{m}$). This spectral range has been chosen since N_2O and CH_4 can be addressed within the tunable range of the QC laser used. The repetition rate is 20 kHz and the average power reaches 0.20 mW .

Figure 1 shows an example of the recorded signals using our system for the N_2O leak through the laser optical path. The reference signal $I_0(t)$ is plotted as a function of time, represented by a green line (laser pulse in absence of a leak); the measurement signal $I(t)$, is displayed as a red line, showing the strong absorption effects on the laser pulse when a puff of N_2O is released from a tank.

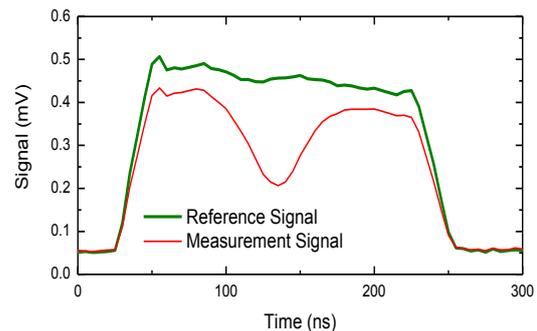


Fig. 1: Reference signal $I_0(t)$ (grey line) and measurement signal $I(t)$ (black line) displayed as functions of time. The measurement signal shows the absorption line due to the presence of the N_2O gas along the optical path.

Figure 2 presents the path-averaged concentration for methane and nitrous oxide, retrieved by the least-squares analysis, with a 68 ms temporal resolution. In both cases, a small amount of pure N_2O ($\sim \text{mL}$) or CH_4 ($\sim 10^2 \text{ mL}$) have been released near the laser beam.

The average N_2O ambient concentration is about 0.22 ppm with strong fluctuations ($\pm 0.2 \text{ ppm}$).

When the gas puff is released, the retrieved concentration peaks and reaches 11 ± 1 ppm. The methane concentration reaches 34 ± 3 ppm then oscillate for approximately 20 seconds. The concentration stabilized to a concentration slightly higher than ambient level (5 ± 1.5 ppm) and eventually returns to value ambient level (1.8 ± 1.3 ppm)

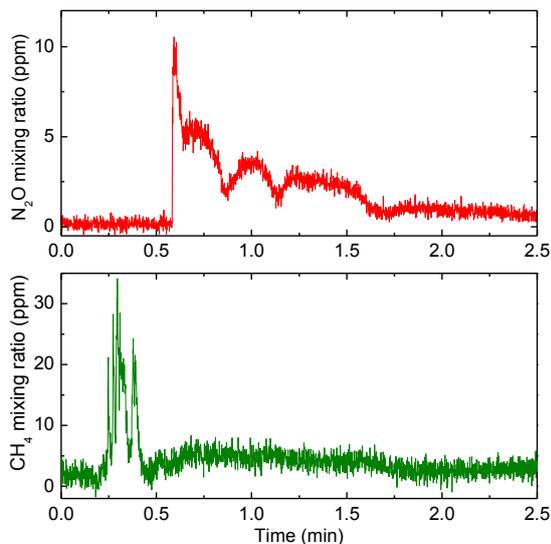


Fig. 2: Time evolution of the path-averaged mixing ratio of N_2O (top graph) and CH_4 (bottom graph).

Both measurements show that the system is able to detect small leaks of either methane or nitrous oxide with a high time response, as well as evaluating the integrate path mixing ratio with uncertainty better than 10 % while measuring the leak. The detection limit (at 2σ) of the instrument in this configuration (40 m range, 68 ms time resolution) is 0.4 ppm for N_2O and 2.6 ppm for CH_4 , which is very close to their respective ambient mixing ratio. The detection limit can easily be improved by reducing the time resolution or deploying a more powerful QCL.

4. SUMMARY AND CONCLUSION

In order to perform mobile or multidirectional remote detection of fugitive gas emission, we demonstrate in this contribution a backscatter

Quantum Cascade Laser Open Path System (QCLOPS) to measure trace gas with a high time resolution using topographic scattering. The trace gas concentration measurement is based on differential absorption spectroscopy, with the distance to the target determined by the hard target lidar return. The use of pulsed quantum cascade laser to carry out intra-pulse spectroscopy for lead detection in a backscatter configuration is, to our knowledge, new. Using the intra-pulse frequency chirp of the QCL, the absorbance along the optical path is measured, allowing to retrieve the target gas path-averaged concentration using a least-squares analysis. As experimental proof, we presented the detection of two gases, methane and nitrous oxide. Small puffs of gas have been released along the laser optical path and measured by the QCLOPS. With the backscattering target at 40 m (~ 130 feet) range, the QCLOPS has a detection limit of 2.6 ppm for CH_4 and 0.4 ppm for N_2O .

This experiment shows feasibility of this methodology. The use of a diffuse-reflectance target is an experimental proof that the gas sensor does not require setting a retro-reflector and can be directly pointed towards topographic targets such as pipes, gas tanks, buildings or walls. However, topographic targets present a wide range of backscattering efficiencies, leading to a variable optical signal and therefore various uncertainties and detection limits. Our current work is now focused on using the high time resolution and compactness of the instrument to achieve measurements to be taken from a moving platform. As an example, if set on a platform moving at 50 km/h, the instrument could perform one concentration measurement every meter. Thus, it makes possible the localization of gas leaks over large distances in a short time frame and with an excellent spatial resolution.

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