

Monitoring of atmospheric Methane and Ethane using two Continuous-Wave Interband Cascade Lasers

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Abstract: A sensor system for simultaneous detection and monitoring of ppb concentration levels of methane and ethane using two interband cascaded lasers was developed. Long-term monitoring of these two atmospheric gases was performed in the Greater Houston area.

OCIS codes: (280.3420) laser sensors; (300.6340) spectroscopy, infrared; (140.5965) Semiconductor lasers, quantum cascade;

1. Introduction

Methane (CH₄) is a main contributor to the greenhouse effect and a safety hazard in the production of chemicals. Hence the monitoring of CH₄ concentration levels in urban or rural areas is critical.¹ Ethane (C₂H₆) is the second-largest component of natural gas after CH₄, which is used in the chemical industry. Tunable diode laser absorption spectroscopy (TDLAS) is an increasingly important method for trace gas detection.^{2,3} In TDLAS, either a near or mid-IR semiconductor laser source is used. Commercially obtainable quantum cascade lasers (QCLs) are limited to wavelengths above 4 μm,⁴ which cannot be used for targeting the strongest absorption lines of CH₄ and C₂H₆ at ~3.3 μm. An alternative is to employ, interband cascade lasers (ICLs) providing continuous-wave (CW) radiation in the 3.0~4.0 μm spectral region at room temperature.^{5,6} Compact ICLs also possess an intrinsic distributed feedback (DFB) structure, which permits CW tuning with spectral linewidths of less than 10 MHz. Therefore, the combination of TDLAS and ICL provides ultra-high sensitive detection on CH₄ and C₂H₆ in the 3.0~4.0 μm spectral region.

2. Sensor system and integration

Fig. 1 shows a diagram of the dual monitoring system, where the upper part is a CH₄ sensor system and the lower part is a C₂H₆ sensor system. Two Nanoplus GmbH, CW, DFB ICL mounted in a TO66 header emitting single-mode radiation at a center wavelength of 3291 nm (ICL #1) and 3337 nm (ICL #2) were employed as light sources. Detailed parameters of the two ICLs are shown in Table 1.

Table 1 Parameters of the two ICLs for CH₄ and C₂H₆ detection

ICL	Center wavelength	Operation temperature	Temperature tuning coefficient	Current tuning coefficient	Target absorption line	Target gas
#1	3291 nm	30.95°C	-0.240 cm ⁻¹ /°C	-0.232 cm ⁻¹ /mA	3038.5 cm ⁻¹	CH ₄
#2	3337 nm	10°C	-0.301 cm ⁻¹ /°C	-0.142 cm ⁻¹ /mA	2996.88 cm ⁻¹	C ₂ H ₆

The two optical sensor systems possess similar structure. The infrared laser beam and a visible alignment beam were combined by means of a dichroic mirror (DM, ISO optics, model BSP-DI-25-3). The co-propagating beams were coupled into the multipass cell (MPC) using a mode matching lens of 200-mm focal length. Two plane mirrors fold the necessary optical path into a reduced space as the focal point of the lens must be positioned at the MPC entry aperture. An effective optical path length of 54.6 m was obtained after 435 beam passes in the MPC. The exiting, collimated ICL beam from the MPC was subsequently focused onto a TEC, mercury-cadmium-telluride (MCT) detector (Vigo, PVI-4TE-4) using a 35-mm focal length parabolic mirror.

For the CH₄ sensor, a computer laptop equipped with a NI DAQ card (NI 6062E) was used to generate the 500 Hz sawtooth wave with an offset of 2.1 V and a peak-peak amplitude of 0.4 V, which was sent to the ramp input of the ICL current driver, resulting in an ICL current scan from 38.3 mA to 46.1 mA. This current range corresponds to a wavelength range from 3038.04 cm⁻¹ to 3039.03 cm⁻¹. The TEC driver was programmed to automatically control the ICL temperature at 30.95 °C using an onboard temperature controller. The sensing signal was directed to the DAQ card for data processing and recording with a sampling rate of 250 k/s. The acquired CH₄ spectrum was processed using a four-step algorithm. Finally, a Lorentzian line shape was fitted to the absorption peak using a

Levenberg-Marquardt least-squares fit procedure. The fitting parameters were used to determine the CH_4 concentration.

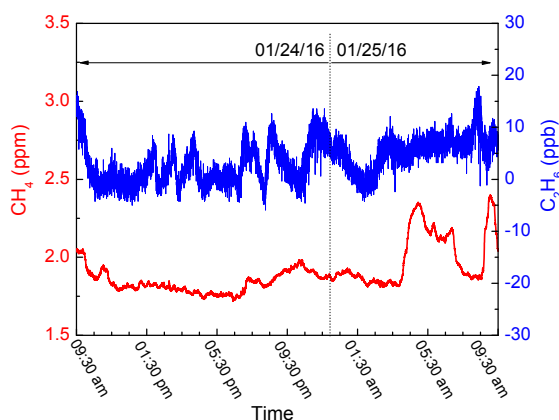
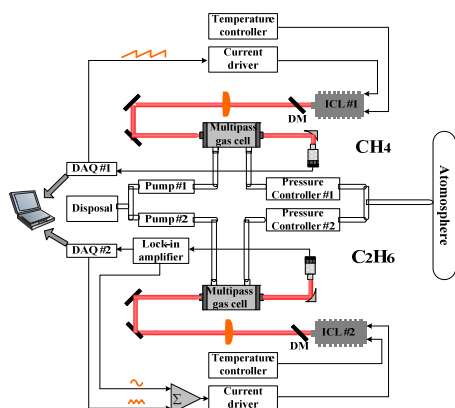


Fig. 1 Diagram of the CH_4 and C_2H_6 detection sensor system Fig. 2 Monitoring of atmospheric CH_4 and C_2H_6 for 24 hours's period

For the C_2H_6 sensor, the same laptop equipped with another NI DAQ card (NI USB6356) was used to generate the 0.3 Hz triangular wave with a peak-peak amplitude of 0.2 V; a lock-in amplifier (Stanford, SR830) was used to generate a 5k Hz modulation signal with a peak-to-peak amplitude of 0.026 V; the two signals were superimposed on each other and supplied to the commercial ICL current driver (Thorlabs, model LDC202C) with an bias current of 47 mA for targeting the absorption line of 2996.88 cm^{-1} . The TEC driver was programmed to control the ICL temperature to 10°C using a commercial temperature controller (Wavelength Electronics, model LFI-3751). The sampling rate of the C_2H_6 sensing signal was set to be 3k Hz, corresponding to 10^4 data points for one period ($10/3 \text{ s}$) of the triangular wave signal. The $2f$ signal was obtained from the lock-in amplifier, and was then transmitted to the DAQ card for sync sampling via the use of a triangular wave signal. The amplitude of the $2f$ signal represents the C_2H_6 concentration.

3. Monitoring of atmospheric CH_4 and C_2H_6

The sensor system was located at the Space Science and Technology building on the Rice University campus. Two pumps were used to sample air from outdoors. Due to the pressure gradient, the inside pressure of the MPC is 700 Torr for CH_4 , and is 100 Torr for C_2H_6 . Atmospheric CH_4 and C_2H_6 was measured for an entire day of continuous monitoring (9:30 am, 01/24/2016~9:30 am, 01/25/2016, CDT) as shown in Fig. 2, where the data recording period of the sensor system is $\sim 8\text{s}$. The average CH_4 concentration was 1.86 ppm, with maximum and minimum concentrations of 2.4 and 1.72 ppm, respectively. The average C_2H_6 concentration was 3.7 ppb, with maximum and minimum concentrations of 18 and -6 ppb, respectively. The diurnal profile of the CH_4 concentration shows an increase during the early morning between 4:30 am~8:30 am. The early morning peak and the diurnal trends observed in CH_4 mixing ratios during the sampling period are in agreement with previous research work reported.¹ Similar varying trends of CH_4 and C_2H_6 can also be observed, which will be verified by further experiments to study the concentration levels and changing trend of the two gases.

4. Conclusions

We demonstrated the integration and performance evaluation of a sensor system based on TDLAS technique using an ICL emitting at a center wavelength of 3291 nm for CH_4 detection and a second ICL emitting at a center wavelength of 3337 nm for C_2H_6 detection. Long-term monitoring on the two atmospheric gases in the Greater Houston area was performed.

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