An Observational and Modeling Study of Natural Gas Leakage in Urban Houston

Final Report

Year 1 (April, 2015-April, 2016)

Two mid-infrared tunable diode laser-spectroscopy-based sensor systems were developed for detection of atmospheric methane (CH_4) and ethane (C_2H_6) concentrations. These systems employed continuous wave (CW), distributed feedback (DFB) interband cascade lasers (ICL) coupled into multi-pass gas cells (MPGC) with an optical path length of 57.6 m. Methane and C_2H_6 absorption lines at 3038.5 and 2996.9 cm^{-1} were targeted, respectively, achieving minimum detection levels (MDL) of ~1.5 and 5 ppbv in the respective CH_4 and C_2H_6 sensing units (60-sec integration time).

The developed CH_4 sensor system was deployed in the Rice University Laser Science Group laboratory in May 2015, and monitoring of CH_4 atmospheric levels was conducted for ~2 weeks. After the stability and detection capabilities of the sensor system were evaluated, the instrument was deployed in a compact vehicle for monitoring of CH_4 concentrations at a Clean Energy compressed natural gas (CNG) station located in Northeast Houston in December 2015. This test demonstrated the suitability of the sensor unit for mobile-mode deployment and for future mapping of potential CH_4 leaks associated with the natural gas distribution system (NGDS) in the Houston area.

Subsequently, the C_2H_6 system was integrated into the field tests and the two sensing units were deployed in a vehicle for monitoring of CH_4 and C_2H_6 levels in a North Houston Freedom Energy CNG station in March 2016. No evident enhancements in atmospheric CH_4 and C_2H_6 background concentrations were observed at this location and its perimetric area.

Concurrently with the development of the CH_4 and C_2H_6 instruments, indicators of the expected probability of CH_4 leakage in residential areas were established. Natural gas (NG) consumption levels and NG infrastructure age were defined as appropriate indicators of the expected occurrence of CH_4 leaks associated with NG usage in the Houston area. The NG heating unit density (HUD) and the median housing age were selected as proxies for NG consumption levels and infrastructure age, respectively. Four levels of expected CH_4 leak probability were defined based on these variables: High- zones with predominantly pre-1980 housing units and high HUD, Medium A-zones with predominantly pre-1980 housing units and low HUD, Medium B- zones with predominantly post-1990 housing units and high HUD, and Low- zones with predominantly post-1990 housing units and low HUD.

Year 2 (April 2016-April 2017)

Data from the 2014 American Community Survey were used to identify block groups in the Houston area falling in the CH_4 leakage probability categories defined previously. Two residential zones were selected in each category (High: H1 and H2, Medium A: MA1 and MA2, Medium B: MB1 and MB2 and low: L1 and L2), and monitoring of CH_4 and C_2H_6 concentrations was conducted in these locations during summer 2016.

To this end, a new CW DFB-ICL-based sensor system was developed for simultaneous detection of CH_4 and C_2H_6. This single system replaced the separate CH_4 and C_2H_6 sensing units initially developed, overcoming power consumption and footprint-related limitations associated with the field deployment of these instruments.

The CH_4-C_2H_6 instrument included a CW ICL centered at 3.337 µm and targeted absorption lines at 2999.06 and 2996.88 cm^{-1} for CH_4 and C_2H_6 detection, respectively. A MPGC with a 57.6 m optical path length was utilized in the sensor unit, achieving a MDL of 17.4 and 2.4 ppbv for CH_4 and C_2H_6 (averaging time of 4.6 sec), respectively.

A weatherized version of the dual-gas sensor system was deployed in a vehicle along with a weather station (Airmar 150WX), allowing acquisition of CH_4 and C_2H_6 concentrations as well as wind direction, wind speed, ambient temperature and GPS coordinates of the vehicle. Sampling was conducted during August and September, 2016 in the selected neighborhoods and while en route to/from these locations. Over 90 hours of data collection covering 14 days was completed during the field campaign.

Concentrations in the range of ~1.8 and 2.7 ppmv were observed for CH_4, while levels generally below 20 ppbv were detected for C_2H_6 during the specific sampling periods. Significant inter-neighborhood variation in the CH_4 concentrations was observed (based on one-way analysis of variance-ANOVA), with generally larger levels detected in the zones with higher expected probability of CH_4 leakage. This observation is likely related to higher and lower CH_4 leakage probability neighborhoods generally located in central and peripherical areas of Houston, respectively. This is evidenced by the correlation between the median housing age and the distance from downtown Houston for the selected residential zones (R= 0.92).

The inter and intra-neighborhood variation in CH_4 concentrations in the Houston area was further examined by cluster analysis using the Anselin Local Moran's statistics (ArcMap, ESRI) and by linear discriminant analysis (LDA). Results of LDA application to the CH_4 and C_2H_6 concentrations in the different sampling zones indicated that high and low CH_4 leakage probability zones can be mainly discriminated based on CH_4 concentrations, reinforcing the ANOVA results pointing out high inter-neighborhood variability in CH_4 levels. Cluster analysis conducted in each selected sampling zone showed high CH_4 concentration (HH) regions (ranging from 2.16 to 3.57 ppmv) dispersed across the H1, H2, MB2 and L2 zones and more grouped at H2, MA1 and L1.

The presence of HH clusters in the Houston area was also examined using the Anselin Local Moran's statistics. Six regions of high CH_4 concentration were detected in the sampled areas, and the influence of different CH_4 sources in these clusters during the sampling period was determined based on the C_2H_6/CH_4 ratio in these regions. This ratio indicated that three of the identified clusters were mainly associated with CH_4 of biogenic origin (e.g. bayous, landfills, etc), while three were related to thermogenic-like sources of CH_4 (e.g. NG).

A total of thirty-seven events of elevated CH_4 concentration (with concentrations below 3.60 ppmv) were observed during the field study. Elevated concentration events, defined as sustained increases in the CH_4 mixing ratios exceeding the local atmospheric background level plus three standard deviations with durations below 10 min were classified as peak events (20 instances). Elevated concentration events with longer durations were considered as large-area concentration enhancements (17 instances). The likely origin of the peak and large-area events was established based on the slope of the orthogonal regression between C_2H_6 and CH_4, which indicates the C_2H_6 volumetric content in the associated CH_4 emission source (Yacovitch et al, 2014). Null or low correlation between CH_4 and C_2H_6 concentrations during the peak episodes indicates a likely biogenic origin of CH_4 (microbial production of CH_4 is not accompanied by C_2H_6 formation), while significant correlation between these gas species indicates a likely thermogenic source of CH_4. For comparison purposes, a Keeling-like plot analysis was also conducted to gain insight on the origin of CH_4 in the observed elevated concentration events (Yacovitch et al., 2014).

Nine of the CH_4 peak events and ten of the large-area events were observed in the selected sampling zones. Eight of the CH_4 peak events detected in the residential zones were associated with thermogenic sources, while one of the large-area events in these areas had a likely thermogenic origin. The volumetric content of the thermogenic CH_4 sources varied between 2.7 and 5.9%, concurring with the composition of NG distributed in the Houston area (Eastern Research Group, 2012; EIA, 2016). Despite this fact, the association of the detected CH_4 peak events with leaks from the NGDS in Houston necessitates further consideration, as elevated concentrations were not generally observed when repeated sampling was performed at the same location, suggesting the influence of different NG-related sources on the observed CH_4 spikes.

The number of CH_4 peak events detected during the field campaign and the observed levels of increase in CH_4 concentration suggest that NG-leakage in the Houston area might be less frequent and of lower magnitude than previously reported for other urban centers in the United States. This observation concurs with newer NG infrastructure comprising only a small fraction of leakage-prone materials pipelines (e.g., bare steel and cast/wrought iron pipelines) in the NGDS in Houston (PHMSA, 2016).

References

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