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Urban Methane Emissions – Advantages of Measuring Between Sunset and Sunrise for Isotopic Source Identification

Xinyi Lu (1), Rosanna Vazey (1), Frances Phillips (2), Zoe Loh (3), Rebecca Fisher (4), and Bryce Kelly (1)

(1) Centre for Ecosystem Science, School of Biological, Earth and Environmental Sciences, UNSW Sydney, 2052, NSW, Australia, (2) Centre for Atmospheric Chemistry, School of Chemistry, University of Wollongong, NSW 2522, Australia, (3) CSIRO Marine & Atmospheric Research, Aspendale, Victoria, Australia, (4) Royal Holloway, University of London, Egham Hill, Egham, Surrey TW20 0EX, United Kingdom

Recent developments in laser-based cavity ring-down and off-axis spectroscopy have enabled the rapid measurement of both spatial and temporal atmospheric methane. Surveys of the mole fraction of methane ([CH4]) in the ground-level atmosphere have been undertaken in a number of cities including Warsaw, Nagoya, Los Angeles, Boston, Washington (D.C.), Florence and London. These mobile surveys were recorded during daylight hours.

We present the advantages of undertaking methane source identification surveys between sunset and sunrise when the boundary layer is low. This maximises the range of [CH4] and isotopic composition (d13C_CH4) values recorded, and reduces the uncertainty in Keeling plot analyses. This is particularly important in temperate, subtropical, tropical and desert environments where there is a steep heat gradient from the ground surface to the upper atmosphere, which results in rapid upwards convection during daylight hours. To support our argument, we present continuous diel [CH4] and d13C_CH4 measurements recorded in a typical urban laneway located in the Eastern Suburbs of Sydney, Australia. These data were collected in May and June 2016 during a calm weather period.We continuously measured the [CH4] and d13C_CH4 using a Picarro G2201-i. The raw data were smoothed using a wavelet analysis method, sampled on a one-minute interval, and analysed using the moving Keeling plot method. Both the [CH4] and d13C_CH4 signals display strong diel fluctuations. The lowest ([CH4] = 1.79 ppm) and highest ([CH4] = 3.07 ppm) readings were recorded during daylight and night-time hours, respectively. The diel d13C_CH4 signature reflects the interplay between urban activities, changing wind speed, shifting wind direction, and the fluctuating atmospheric boundary layer. Background air, extracted from the afternoon signal, has mean [CH4] and d13C_CH4 of 1.81 $\pm$ 0.01 ppm and -48.7 $\pm$ 0.4 ‰ respectively. Between sunset and sunrise, as the boundary layer lowers and the air from blended sources higher in the atmosphere is compressed into a smaller volume, d13C_CH4 values become less negative, reaching as low as -44.4 ‰.This blended source air has a mean isotopic signature of -47.2 $\pm$ 1.7 ‰.

The dominant methane source in Sydney is the leaky gas distribution network, and to a lesser extent household hot water systems and stoves (indicated by an evening meal-time spike). Methane from these three sources had a clear thermogenic signature (mean -32.7 $\pm$ 2.0 ‰, which is consistent with the fact that conventional gas reservoirs supply most of Sydney's gas. These emissions have been poorly quantified in Australia's National Greenhouse Gas accounts, and they need to be better quantified for comprehensive energy sector life cycle greenhouse gas inventory assessments. Some parcels of air had methane that could be attributed to waste (mean -56.7 $\pm$ 0.02 ‰. No biological sources were detected using the moving Keeling analysis.

Stable conditions for when no drift corrections would be required for mobile surveys is restricted to a 4-hour window starting midday. However, the range of [CH4] and d13C_CH4 values recorded between sunset and sunrise provides the best opportunity for d13C_CH4 source identification of waste and town gas emissions.