Separation of methane emissions from biogenic sources and natural gas based on CH_4, C_2H_6 and NH_3 column observations in the Colorado Front Range

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I. Introduction

Atmospheric emissions of methane (CH_4) from anthropogenic and biogenic sources are important to air quality and climate.

In the Northern Colorado Front Range CH_4 is emitted from biogenic sources such as concentrated animal feeding operations (CAFO) and oil and natural gas (ONG) production and storage. Here, we demonstrate a novel approach to source apportion CH_4 based on column observations. A linear regression analysis explains excess CH_4 by relating it to ethane (C_2H_6) as a tracer for ONG and ammonia (NH_3) for CAFO emissions.

II. Site selection & Instrumentation

- Three COCCON (Collaborative Carbon Column Observing Network) type EM27/SUN Fourier Transform Spectrometers (FTS) observed CH_4, oxygen (O_2) and water vapor (H_2O) vertical column densities (VCDs) in Eaton and at two boundary sites in Boulder and Westminster, CO.
- CU mobile SOF[1] measured C_2H_6, NH_3 and H_2O VCDs in Eaton.

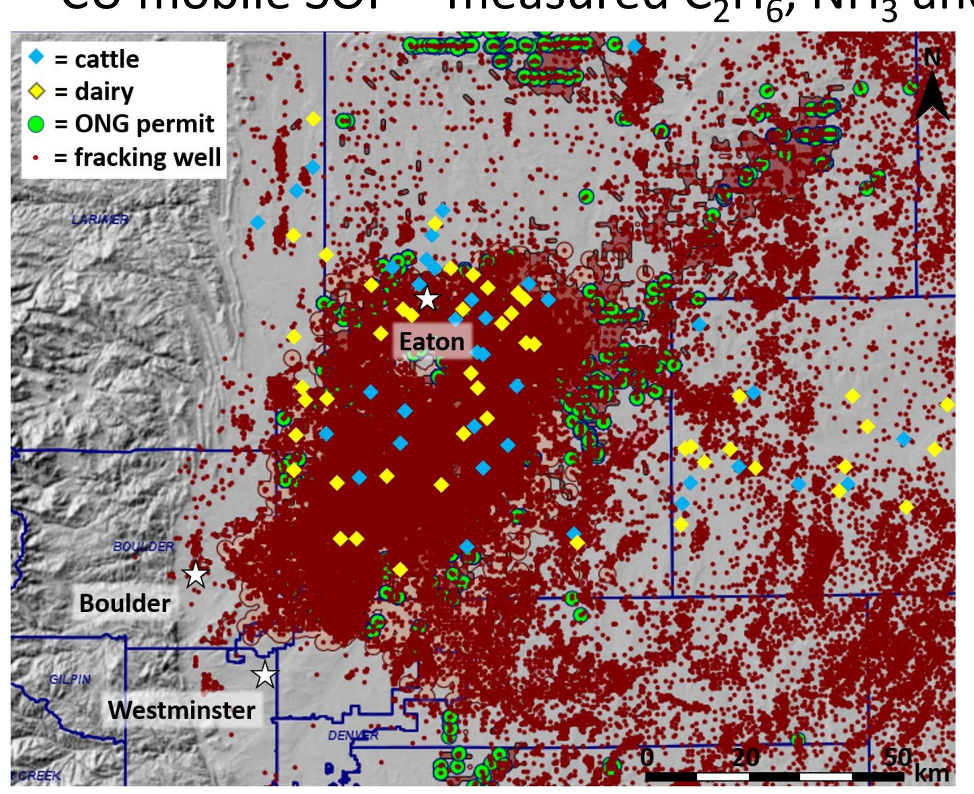

Figure 1: Map showing locations of ONG wells and CAFOs in the Northern Colorado Front Range.

Figure 2: The EM27 FTS are easy to transport. For calibration upon arrival to Colorado they were co-located at NCAR.

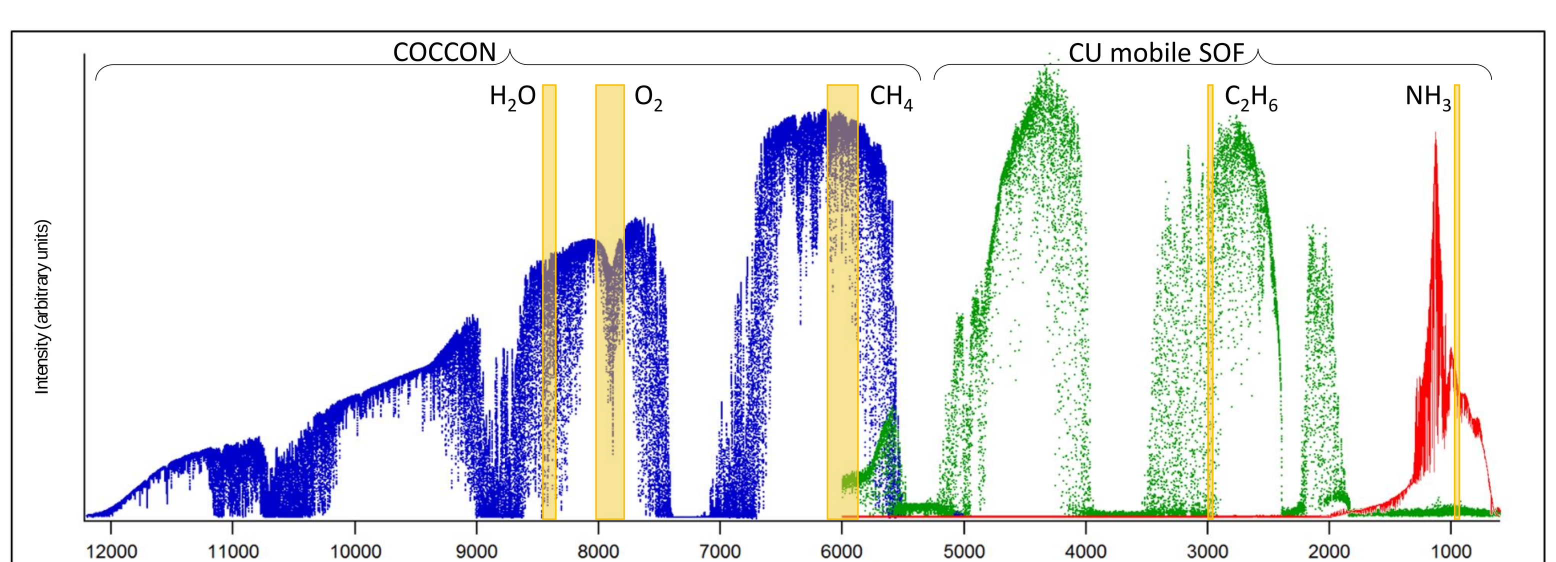

Figure 3: The solar spectrum measured with three detectors: InGaAs (blue), InSb (green), and MCT (red). The highlighted areas indicate the spectral windows used for the retrieval of the different gases.

III. Results

- Column averaged dry air mole fractions XCH_4, XC_2H_6, and XNH_3 (Figure 4) were determined using O_2 VCDs for air mass factor normalization[2]: $\boldsymbol{XGas = \frac{Gas\ VCD}{O_2\ VCD} * 0.2095}$.
- Background (BKG) concentrations of XCH_4 were determined using the 2nd percentile of hourly pooled data from all measurement locations, while BKG concentrations of XC_2H_6 and XNH_3 were determined using the 1st percentile of all days pooled together. The enhancement over BKG is given by: $\boldsymbol{\Delta XGas = XGas - XGas_{BKG}}$.
- Linear regression analysis[3] is performed using the following equation: $\boldsymbol{\Delta XCH_4 = \beta_0 + \beta_1 * \Delta XC_2H_6 + \beta_2 * \Delta XNH_3}$.

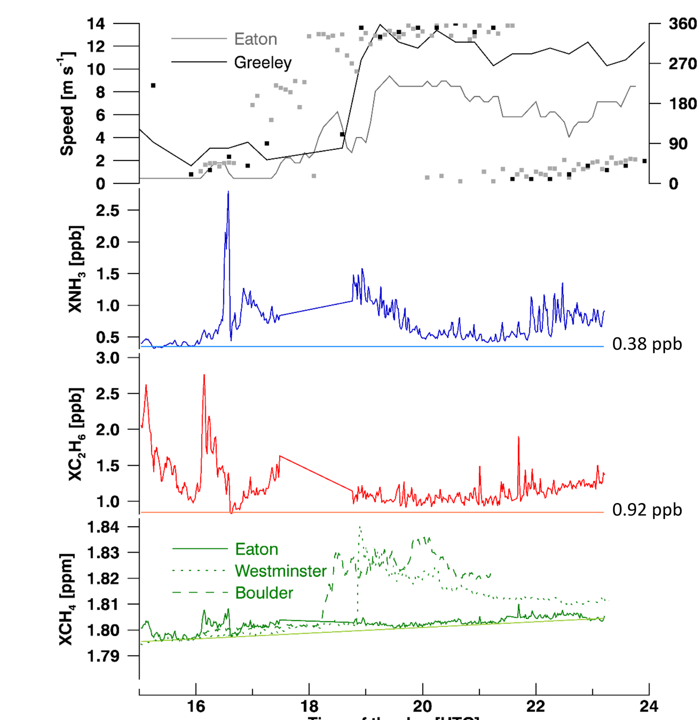

Figure 4: Timeseries of column averaged dry air mole fractions for 16 March 2015: XNH_3 (blue), XC_2H_6 (red), XCH_4 (green) and their background concentrations. Wind speed (solid line) and direction (squares) were recorded by meteorological stations near the Eaton site.

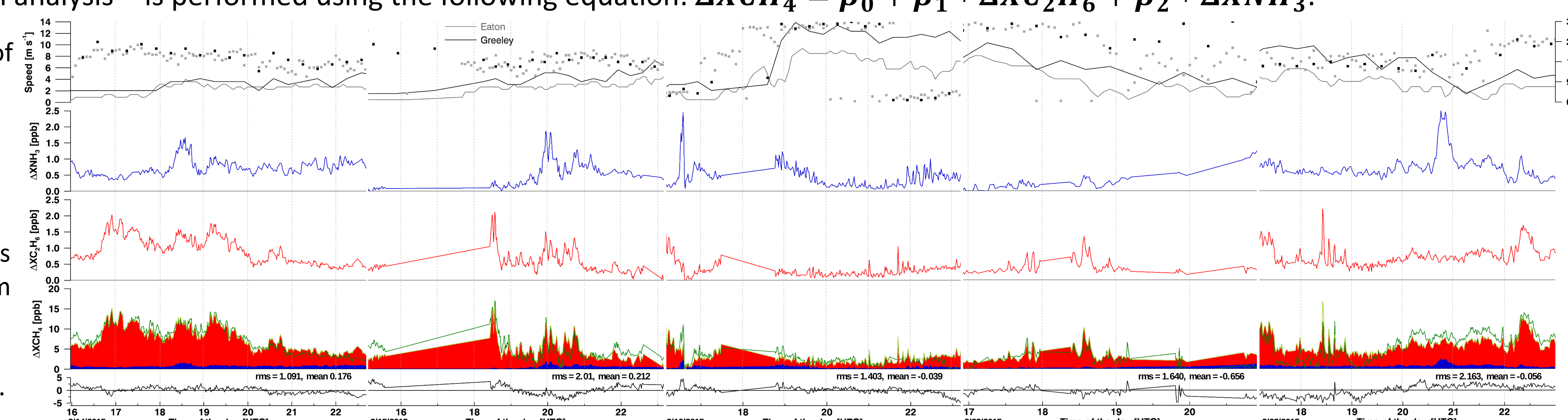

Figure 5: Timeseries of ΔXNH_3 (blue), ΔXC_2H_6 (red), ΔXCH_4 (green) and ΔXCH_4 (light green) calculated based on the regression parameters in Table 1. The bottom panel shows the residual of measured and calculated ΔXCH_4.

Table 1: Linear regression parameters. ΔXCH_4 is primarily explained by ΔXC_2H_6 though there are differences in the contribution from ΔXNH_3 depending on the wind direction.

Wind dir [°]	β_0	β_1	β_2	n (% of data)
All	**0.00**	**7.28**	**0.98**	1393 (100)
320-80	-0.02	4.88	2.46	293 (21)
80-200	0.34	7.44	-0.24	427 (31)
200-320	0.47	7.07	1.45	340 (24)

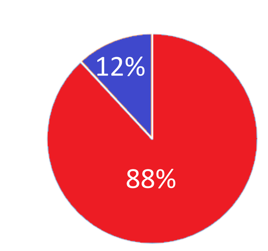

Figure 6: Pie chart distribution indicating the source apportionment of ΔXCH_4 based on the parameters in Table 1.

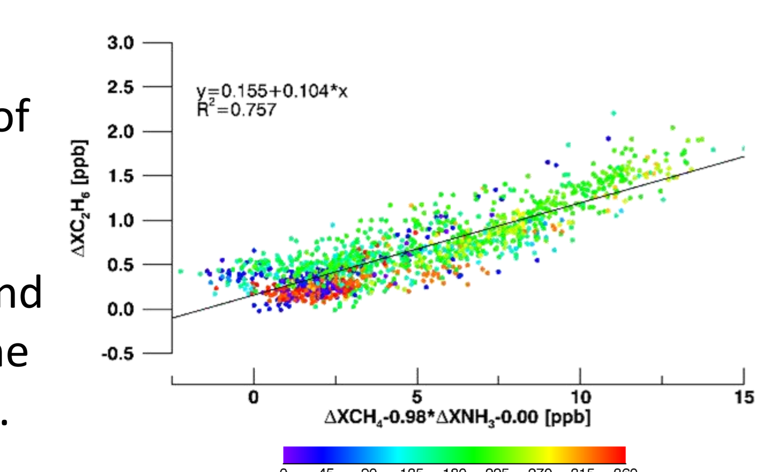

Figure 7: Correlation of ΔXC_2H_6 with ΔXCH_4 color coded by wind direction. The slope is 10%.

IV. Summary

Column observations are capable of characterizing the CH_4 dome as the difference between instruments located inside the CH_4 dome and at boundary sites. Four FTS have successfully been deployed in the Colorado Front Range to separate CH_4 emissions by source type.

- The ΔXCH_4 time series can be explained by tracers that represent ONG and CAFO emissions.
- It was found that in Eaton 88% of measured CH_4 can be attributed to ONG sources, and 12% to biogenic sources (here: CAFO).
- The ratio of ΔXC_2H_6 to ΔXCH_4 is 10%, which indicates that the CH_4 source is wet gas or pipeline grade natural gas.[4]

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