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Key Points:
• Methane emissions from natural gas and agriculture can be separated using the concept of excess column measurements of source tracers.
• Column integrals over boundary layer height are complementary to in situ methods and useful to quantify and source apportion methane on regional scales.
• Natural gas sources dominate over agricultural and other sources, but the latter are relatively more important when excess CH₄ is smaller than 5 ppb.

Supporting Information:
• Supporting Information SI

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Abstract
This proof-of-concept study demonstrates that methane (CH₄) emissions from natural gas (NG) and agriculture can be disentangled using the concept of excess column observations. A network of cost-effective sensors measured excess column-averaged dry-air mole fractions for CH₄ (ΔXCH₄), ethane (ΔXC₂H₆ as NG tracer), and ammonia (ΔXNH₃ from agriculture) in the Denver-Julesburg Basin during March 2015. ΔXCH₄ varied up to 17 ppb and was 3 times higher with winds from directions where NG is produced. The ΔXCH₄ variance is explained by variations in the C₂H₆/CH₄ tracer pair, attributing 63 ± 17% to NG, 25 ± 10% to agriculture, and 12 ± 12% to other sources. The ratios ΔXCH₄/ΔXNH₃ (16 ± 2%; indicates wet NG) and ΔXNH₃/ΔXCH₄ (43 ± 12%) were compatible with in situ measured ratios. Excess columns are independent of boundary layer height, characterize gases in the open atmosphere, are inherently calibrated, average over extended spatial scales, and provide a complementary perspective to quantify and attribute CH₄ emissions on regional scales.

Plain Language Summary
Methane is the second most important anthropogenic greenhouse gas. Knowledge about methane sources is increasingly relevant as energy production continues to shift toward natural gas and becomes complicated by collocated emissions from natural gas production and agriculture due to shared land use. There is a need for methods to better decouple emissions from multiple sources that contribute to local enhancements in methane, which are small compared to the regional methane background concentrations, and depend on atmospheric transport and planetary boundary layer height. In this study, we show that the concept of collocated excess column measurements of methane and chemical tracers shows great promise as a viable approach to disentangle methane emissions from multiple sources by means of cost-effective networks of ground-based sensors. Excess columns are independent of boundary layer height, which makes quantification and source attribution of methane more straightforward.

1. Introduction
Methane (CH₄) is a major greenhouse gas in the atmosphere (Intergovernmental Panel on Climate Change, 2014). Globally, CH₄ sources can generally be grouped as biogenic, thermogenic, and pyrogenic; biogenic sources include wetlands, ruminants, and waste such as landfills; thermogenic sources are from fossil fuel combustion and the surface via coal and oil and natural gas (NG) extraction; or pyrogenic sources include wildfires and combustion of biomass (Kirschke et al., 2013; Saunois et al., 2016).

In Colorado, the dominating CH₄ sources are the NG industries, coal mining, and agriculture (which includes concentrated animal feeding operations [CAFOs]), with coal mining only occurring in the western part and not in the Front Range area (Arnold et al., 2014; Petron et al., 2012). Other sources in the state include waste combustion, landfills, and wastewater treatment facilities. According to Arnold et al. (2014), NG accounts for 38.5% of CH₄ emissions statewide, coal mining for 28.9%, agriculture for 22.3%, waste and landfills combined for 8.4%, and remaining sources for <2%.

Past studies in the Denver-Julesburg Basin (DJB) aiming to apportion CH₄ by agricultural and NG sources are based on in situ measurements and inventories and used carbon isotope ratios such as ^13C/^12C and...
hydrocarbons such as C₂H₆ (Smith et al., 2015). Here we present the first source apportionment of CH₄ from agricultural sources and NG using the concept of excess columns. Excess columns are vertical columns from which a regional vertical column background has been subtracted. Other studies have used the concept of excess columns to quantify CH₄ emissions (Viatte et al., 2017; Wennberg et al., 2012), but this requires coupling with a chemical transport model, which is beyond the scope of this study. Column measurements can also be used to determine emission estimates through a combination of enhancement ratios and emission inventories (Wunch et al., 2009) or long-term trends of enhancement ratios (Franco et al., 2016; Hausmann et al., 2016). However, we are not aware of a previous attempt to source apportion CH₄ emissions using collocated measurements of source tracers. Unique tracers from the NG industry and agriculture/CAFOs are ethane (C₂H₆) and ammonia (NH₃), respectively. Even though the agricultural processes responsible for CH₄ emissions are distinct from those emitting NH₃, the sources are collocated within CAFOs, and CH₄ and NH₃ enhancements have been shown to correlate downwind, on a regional scale (van Haarlem et al., 2008). Measurements of C₂H₆ and NH₃ are used in this study to separate the CH₄ emissions from those two sources.

Figure 1a shows the locations of oil wells in the DJB and CAFOs in Northern Colorado, indicating that the CH₄ emissions from agricultural sources and NG are co-located. Boulder, Westminster, and Eaton in Colorado indicated as stars represent the measurement sites. With winds predominantly from the south during daytime in spring, the Eaton site is located inside the Front Range CH₄ hotspot, and the Boulder and Westminster sites are considered background sites. We perform a linear regression analysis similar to Garcia et al. (2006) to separate the excess CH₄ columns in terms of excess NH₃ and C₂H₆ columns to determine the source. Using the excess C₂H₆ columns, and for agricultural sources corrected excess CH₄ columns, we are able to determine the NG source type.

2. Method
2.1. Measurement Setup
A mininetwork of four instruments was located at three different sites: one central measurement site in Eaton, CO, inside the DJB, and two background measurement sites outside the DJB in Boulder and Westminster, CO. The deployment period was from 14 to 23 March 2015, of which five clear days provided simultaneous measurements from all four instruments: 14, 15, 16, 22, and 23 March. Three COCCON (Collaborative Carbon Column Observing Network; Frey et al., 2019) type EM27/SUN from the Karlsruhe Institute of Technology, Germany, measured CH₄ and oxygen (O₂) at each of the three sites. In addition, the University of Colorado mobile Solar Occultation Flux (CU mobile SOF) instrument measured C₂H₆ and NH₃ in Eaton, CO. All instruments use direct sunlight to measure the trace gas absorption along the direct solar beam and infer the vertical column density (VCD) of an absorber gas above the site. Eaton is located 11 km north of Greeley, CO (Weld County). The temperature ranged from 8 to 28 °C (median and mean: 21 °C) at the Greeley-Weld County Airport and surface wind speed ranged from 1.5 to 13.9 m/s (median: 4.1, mean: 5.6 m/s), coming mostly from the South (median direction of 175°).

The background measurement sites Boulder and Westminster are located distant from the area in which most CAFO sites and NG wells are located. Surface winds ranged from 1.5 to 9.3 m/s (median: 2.6, mean: 3.1 m/s) in speed and had a median direction of 130° at the Boulder Municipal Airport and 2.1 to 7.7 m/s (median: 3.1, mean: 3.5 m/s) in speed and 150° in median direction at the Rocky Mountain Metropolitan Airport in Broomfield, CO, making these sites suitable to determine background measurements.

2.2. Retrievals
The EM27/SUN spectrometer is described in more detail in Gisi et al. (2012), Frey et al. (2015), and Hase et al. (2016). CH₄ is retrieved from EM27/SUN solar spectra (5,000–11,000 cm⁻¹) using the spectral region at 5,897–6,145 cm⁻¹ and O₂ at 7,765–8,005 cm⁻¹ (Frey et al., 2015); the PROFFIT code is used for the retrievals (Hase et al., 2004). The ratio of CH₄ over O₂ cancels systematic errors; therefore, the precision for XCH₄ (see equations (1)–(2) below for definition) is higher than for the VCD. The single sounding precision of XCH₄ for 10 coadded scans is ~0.33 ppbv based on Hedelius et al. (2016; did not coadd scans).

The CU SOF instrument uses a custom-built digital solar tracker (Baidar et al., 2016), and is described in detail in Kille et al. (2017), where a detailed characterization of the NH₃ and C₂H₆ retrievals is also
presented. Briefly, NH₃ is retrieved from CU SOF solar spectra (700–5,000 cm⁻¹) using the spectral region at 950–980 cm⁻¹ and C₂H₆ at 2,970–3,000 cm⁻¹; the SFIT4 retrieval code is used for the retrievals (Hase et al., 2004). The precision of XNH₃ is 0.03 ppbv, and the precision of XC₂H₆ is 0.03 ppbv (using a campaign mean and median O₂ VCD of 3.90 × 10²⁴ molec/cm²).

Dry-air mole fractions (XGas) are derived from VCDs as

\[ X_{\text{Gas}} = 0.2095 \times \frac{\text{VCD(Gas)}}{\text{VCD(O}_2)} \] (1)

Furthermore, XCH₄ is divided by a calibration factor and scaled by a correction factor correcting for an air mass-dependent artifact that has been shown in Wunch et al. (2010) and Frey et al. (2015), see equation (2). This correction is dependent on solar zenith angle (SZA) θ and is <0.1% for XCH₄, insignificant for XO₂, and deemed irrelevant for XNH₃ and XC₂H₆

\[ X_{\text{CH}_4,\text{corr}}(\theta) = X_{\text{CH}_4}(\theta) \times \left\{ 1 + 3.796 \times 10^{-3} \left( \frac{\theta + 16.04}{90 + 16.04} \right)^2 \right\} \] (2)

The enhancement over background concentrations is determined by

\[ \Delta X_{\text{Gas}} = X_{\text{Gas}} - X_{\text{Gas,BKG}} \] (3)

Figure 1. (a) Measurement setup in context of cattle and dairy feedlots, natural gas permits, and fracking wells in the Colorado Front Range. The wind rose indicates observed winds near the Eaton site during the entire measurement period. The Greeley-Weld County Airport (KGXY), a wind measurement location near Eaton, the Rocky Mountain Metropolitan Airport (RMMA), and the Boulder Municipal Airport are indicated by red circles (background image from Google Earth 2018). (b) Dry-air mole fractions of NH₃ (blue), C₂H₆ (red), CH₄ (green), and their background values (solid lines) and uncertainties (shading) on 14 March 2015 (see text for details on instruments deployed at measurement and background sites). (c) Campaign ΔXNH₃ (blue), ΔXC₂H₆ (red), and ΔXCH₄ (green) as a function of wind direction (wind from north = 0°, wind from east = 90°, etc.). Light shading corresponds to 5th and 95th percentiles, darker shading to 25th and 75th percentiles; the dots represent the mean value. The gray bars in the ΔXNH₃ panel represent CAFO emissions, and the gray bars in the ΔXC₂H₆ panel represent natural gas emissions (see text for details).
This low bias is partially due to (lower) stratospheric CH$_4$ and includes $\sim$1% spectroscopic bias (Wunch et al., 2010). This uncertainty cancels out in calculating $\Delta$XCH$_4$ and does not affect the further analysis. The XNH$_3$ and XC$_2$H$_6$ backgrounds determined by this method correspond to $(0.76 \pm 0.32) \times 10^{16}$ molec/cm$^2$ NH$_3$ and $(1.77 \pm 0.28) \times 10^{16}$ molec/cm$^2$ C$_2$H$_6$ and are within error identical with the regional minimum values of $0.5 \times 10^{16}$ molec/cm$^2$ NH$_3$ and $1.5 \times 10^{16}$ molec/cm$^2$ C$_2$H$_6$ in Kille et al. (2017).

### 2.3. Data Analysis

The statistical data analysis software Stata (v14.1) was used to perform a linear regression analysis on the time series of $\Delta$XGas using the following equation:

$$\Delta$XCH$_4 = \beta_0 + \beta_1 \cdot \Delta$XC$_2$H$_6 + \beta_2 \cdot \Delta$XNH$_3$. \tag{4}$$

Regression parameters $\beta_1$ and $\beta_2$ have units of [ppb/ppb] and represent the $\Delta$CH$_4/\Delta$C$_2$H$_6$ and $\Delta$CH$_4/\Delta$NH$_3$ ratios at the source, respectively; $\beta_0$ is excess XCH$_4$ in units of [ppb] that exceeds the regional background XCH$_4$ and could not be attributed to one of the two sources.

Sensitivity studies that varied in the constraints to $\beta_i$ ($i = 0, 1, 2$) were performed: (I) no constraints, (II) only positive $\beta_0$, (III) $\beta_1$ and $\beta_2$ positive ($\beta_0$ unconstrained), (IV) only $\beta_2$ positive ($\beta_0, \beta_1$ unconstrained), and (V) $\beta_0$ held constant ($\beta_1, \beta_2$ unconstrained). For quality assurance nonphysical results, that is, $\beta_i < 0$, or $R^2 < 0.5$, were filtered out. Test (I) indicated that $\beta_0 = 0.64$ ppb and in test (V) the same value obtained the best $R^2$ (see Table S1 and Figure S1). The overall uncertainty on $\beta_0$ was determined as three times the standard deviation of $\beta_0$ from tests (I)–(IV). The uncertainties on $\beta_1$ and $\beta_2$ are the 95% confidence intervals from the linear regression analysis. In the final analysis $\beta_0$ is held constant and the quality-assured regression parameters taken from test (V) to allow for a better statistic compared to using test (I) (see Table S1) and resulted in $\beta_0 = 0.64 \pm 0.64$ ppb, $\beta_1 = 6.20 \pm 0.81$ ppb/ppb, and $\beta_2 = 2.34 \pm 0.65$ ppb/ppb (dashed lines in Figures 2 and S2). Figure S1 indicates that the choice of $\beta_0$ does not significantly change the percent-contribution from NG, agriculture, and other sources on CH$_4$ emissions. Possible error sources and an explanation for $\beta_0$ are discussed in section 3.2.

The $\beta_i$ parameters were used to calculate percent-contributions of NG, agricultural, and other sources as follows:

$$\%$NG = $\frac{\beta_1 \cdot \Delta$XC$_2$H$_6}{\beta_0 + \beta_1 \cdot \Delta$XC$_2$H$_6 + \beta_2 \cdot \Delta$XNH$_3$}, \tag{5}$$

$$\%$Agr = $\frac{\beta_2 \cdot \Delta$XNH$_3}{\beta_0 + \beta_1 \cdot \Delta$XC$_2$H$_6 + \beta_2 \cdot \Delta$XNH$_3$}; \tag{6}$$

$$\%$Other = $\frac{\beta_0}{\beta_0 + \beta_1 \cdot \Delta$XC$_2$H$_6 + \beta_2 \cdot \Delta$XNH$_3$}. \tag{7}$$

NG and CAFO emission information is represented as gray bars relative to the measurement site in Figure 1c. It is calculated as an effective distance that expresses in which geographic direction the source of interest is located weighted by the probability of wind coming from that direction during the data analysis period. Information about the feedlot locations and maximum head counts for Colorado in 2014 were provided by D. Bon at the Colorado Department of Public Health and Environment (CDPHE), and NG facility emissions were extracted from Environmental Protection Agency’s FLIGHT tool (http://ghgdata.epa.gov/ghgp). The effective distance is based on the inverse distance weight (IDW):

$$\text{IDW}(\alpha) = \frac{\sum_i \frac{Z_i(\alpha)}{x_i}}{\sum_i \frac{1}{x_i}}; \tag{8}$$

where $Z_i$ is the strength of source $i$ and $x_i$ is the distance between source $i$ and the measurement site. A proxy for expected emissions transported to the measurement site from a subset of azimuth directions is obtained as
Proxy Emission $\alpha_i + \Delta \alpha = IDW(\alpha_i + \Delta \alpha)f(\alpha_i + \Delta \alpha).$  \hfill (9)

where $(\alpha_i + \Delta \alpha)$ denotes the subset of azimuth directions beginning at direction $\alpha_i$ and $IDW(\alpha_i + \Delta \alpha)$ and $f(\alpha_i + \Delta \alpha)$ are the IDW of sources and frequency of wind directions located within the subset of azimuth directions.

3. Results and Discussion

A map of the Colorado Front Range is shown in Figure 1a, which also shows the wind rose for the measurement times. Figure 1b shows an example of the column-averaged dry-air mole fractions (XGas; see equations (1) and (2)) from 14 March as well as background concentrations with their respective uncertainty. The excess columns $\Delta X_{\text{Gas}}$ calculated using equation (3) are shown as function of wind direction in Figure 1c. Also shown are the expected emissions of feedlots and NG facilities based on IWD calculations (see section 2.3). $\Delta X_{\text{NH}_3}$ is enhanced evenly from east (90°) to south (180°) to west (270°); $\Delta X_{\text{C}_2\text{H}_6}$ is enhanced increasingly from east to south to west. This wind direction dependence is consistent with that of the expected emissions to a first approximation. $\Delta X_{\text{CH}_4}$ generally resembles the wind direction.
dependence of $\Delta C_2H_6$ more closely than that of $\Delta NH_3$—though both sources cannot be easily disentangled from a wind-direction-dependent analysis alone.

The approach for source apportionment is illustrated in Figure 2 for an example day (16 March 2015), when winds gradually shifted toward a northerly flow (wind direction of 180°). The regression parameters and calculated contributions of $\Delta NH_3$ and $\Delta C_2H_6$ to $\Delta CH_4$ are also shown; the full time series is included as Figure S2. Generally, when $R^2 > 0.8$ then $\beta$ was relatively constant. $R^2$ tends to decrease when $\Delta CH_4$ is low. The quality filter using $R^2 > 0.5$ was chosen to avoid eliminating too many data points.

Based on the regression parameters and observed tracer excess columns, $\Delta CH_4$ was calculated and compared to the observed $\Delta CH_4$. The mean and root-mean-square (rms) of the residual differences between observed and calculated $\Delta CH_4$ is shown in Figure 2 and in Figure S2 for each day. The overall mean = 0.10 ppb and rms = 0.87 during quality-assured periods, and during filtered out periods the mean = 0.07 ppb and rms = 1.34.

The filled polygons in Figure S2 indicate the contribution of $\Delta C_2H_6$ and $\Delta NH_3$ to the calculated $\Delta CH_4$. On 16 March $\Delta CH_4$ is smaller compared to 14 March, and the contribution of $\Delta NH_3$ is larger on 16 March compared to 14 March. The measured $\Delta CH_4$ median = 4.2 ppb, mean = 5.2 ppb, and max = 17.1 ppb. Figure S3 shows the change in percent-contribution as a function of $\Delta CH_4$; as $\Delta CH_4$ concentration increases so does the percent-contribution of NG emissions, whereas the percent-contribution from agricultural and other sources is more relevant during times of smaller $\Delta CH_4$. This matches the behavior in Figure 1c and establishes that both sources contribute to $\Delta CH_4$. It also emphasizes that during times of smaller observed $\Delta CH_4$ agricultural and other sources dominated the percent-contribution compared to times of higher $\Delta CH_4$ (>5 ppb) when NG dominated.

### 3.1. Comparison With the Literature

A few other studies that have apportioned (Peischl et al., 2018; Petron et al., 2014; Townsend-Small et al., 2016) or quantified (Eilerman et al., 2016; Fried et al., 2015; Gilman, 2017; Tzompa-Sosa et al., 2017; Yuan et al., 2017) CH$_4$ in the DJB study area exist, but none have attempted to source apportion CH$_4$ based on excess columns. These results can be used to assess how the concept of excess columns compares with results based on in situ observations. Based on the quality-assured linear regression parameters and the measurements of excess columns, 62.8 ± 17.1% of $\Delta CH_4$ is from NG sources, 25.4 ± 9.6% is from agricultural sources, and 11.8 ± 11.8% could not be attributed to a source. The attribution to NG agrees well with values reported in studies by Petron et al. (2014) for Weld County in May 2012 and Peischl et al. (2018) as shown in Table 1, where the dominating source of CH$_4$ is determined to be the NG sector. The data by Townsend-Small et al. (2016) remain somewhat ambiguous whether the majority of CH$_4$ is from NG or agriculture.

The attribution to agriculture determined in this study falls between that from Petron et al. (2014) and Shale Oil and Natural Gas Nexus (SONGNEX; March/April 2015) CH$_4$ in the DJB study area exist, but none have attempted to source apportion CH$_4$ based on excess columns. This matches the behavior in Figure 1c and establishes that both sources contribute to $\Delta CH_4$. It also emphasizes that during times of smaller observed $\Delta CH_4$ agricultural and other sources dominated the percent-contribution compared to times of higher $\Delta CH_4$ (>5 ppb) when NG dominated.

The two advantages of using remote sensing column measurements are that they are independent of boundary layer height and do not use inlets, which present a challenge regarding NH$_3$ partitioning to surfaces. Ground-based solar absorption measurements offer good vertical sensitivity across the whole atmosphere. This is further illustrated in Figure S4, where the column sensitivities for NH$_3$, C$_2$H$_6$, and CH$_4$ are compared with vertical profiles measured in situ sensors during the Deriving Information on Surface Conditions from Column and Vertically Resolved Observations Relevant to Air Quality (DISCOVER-AQ; July/August 2014) and Shale Oil and Natural Gas Nexus (SONGNEX; March/April 2015) field campaigns. The median column sensitivity below 4.8 (2.4) km is 1.07 ± 0.04 (1.03 ± 0.04) for NH$_3$, 1.04 ± 0.03 (1.01 ± 0.03) for C$_2$H$_6$, and 1.05 ± 0.03 (1.06 ± 0.03) for CH$_4$ (errors indicate SZA variability for data used in this study; 42° < SZA < 70°). Most of the excess column resides in the lowest kilometer, and >99% resides below 4.8 km above sea level (or 550 mbar) for all gases. At higher altitudes, CH$_4$ during SONGNEX (same month as our study) agrees closely with the median CH$_4$ measured at MLO. Changes in free tropospheric mixing ratios contribute relatively less to the excess columns, since they are weighted by lower density. Figure S4 demonstrates that the vertical distributions are similar for the three gases and that the column sensitivities...
of individual instruments are virtually identical, and near unity for all gases at the altitudes that contribute to the excess columns.

We therefore believe that the consistency of excess columns as a complementary tool to in situ observations to source apportion CH₄ emissions on the regional scale can be evaluated by comparing the regression parameters $\beta_1$ and $\beta_2$ with the results from in situ observations in the literature. This is possible because the regression parameters $\beta_1$ and $\beta_2$ are directly indicative of the $\Delta C_{2H6}/\Delta C_{CH4}$ and $\Delta NH3/\Delta C_{CH4}$ ratios and have been determined as $16.1 \pm 2.1\%$ and $43 \pm 12\%$ in this study. Previous studies that have measured $\Delta C_{2H6}/\Delta C_{CH4}$ and $\Delta NH3/\Delta C_{CH4}$ using in situ sensors in the DJB are summarized and compared to our results in Table 2. There is excellent agreement with the aircraft study by Fried et al. (2015), and also Tzompa-Sosa et al. (2017). C₂H₆/CH₄ ratios reveal information about the source type; biogenic sources have a ratio of <0.2%, dry gas of 1 to 6%, wet gas >6%, pipeline grade NG <15%, and processed NG liquids >30% (Yacovitch et al., 2014). According to Smith et al. (2015) and Yacovitch et al. (2014) the $\Delta C_{2H6}/\Delta C_{CH4} = 16.1 \pm 2.1\%$ indicates that the NG source of CH₄ is wet gas. The NH₃/CH₄ ratio quantified based on excess columns in this work has robust error bars and tends to be compatible with the higher published values from previous in situ measurements. Altogether, the approach of quantifying and source apportioning CH₄ on the regional scale holds potential to decrease the error bars both of the ratios and the source attribution. A limiting factor in this work is the short study period.

3.2. Error Sources

Errors in the excess column method originate in the background correction and uncertainty on the regression parameters. The uncertainties on the background correction (included in Figure 1b) are 0.17 ppb on $\Delta NH3$, 0.15 ppb on $\Delta C_{2H6}$, and 0.62 ppb on $\Delta C_{CH4}$, determined as the standard deviation between the data analysis days. The uncertainties on the background correction and the 95% confidence intervals on the regression parameters were propagated through equations (5)–(7) for the overall error of the source apportionment. In this study, the number of data analysis points was limited to 1,294, and the number of points determining the instantaneous $\beta_i$ was set at 100. To decrease the 95% confidence intervals on the regression parameters using more than 100 points will increase the certainty of the source apportionment. A longer study with more measurement points would provide the data to determine how the wind direction affects the source apportionment.

The percent-contribution quantified due to the residual, $\beta_0$, of 11.8 ± 11.8% compared well with the attribution value in Petron et al. (2014) of 11% from landfills and wastewater plants. However, there is no unique

### Table 1

<table>
<thead>
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<th>Source attribution</th>
<th>Study period</th>
<th>Reference</th>
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<tbody>
<tr>
<td>NG</td>
<td>62.8 ± 17.1%</td>
<td>This work</td>
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<tr>
<td>Agriculture</td>
<td>25.4 ± 9.6%</td>
<td>Petron et al. (2014)</td>
</tr>
<tr>
<td>Other</td>
<td>11.8 ± 11.8%</td>
<td>Townsend-Small et al. (2016)</td>
</tr>
<tr>
<td>Study period</td>
<td>March 2015</td>
<td>Peischl et al. (2018)</td>
</tr>
<tr>
<td>Reference</td>
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### Table 2

<table>
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<th>$\Delta C_{2H6}/\Delta C_{CH4}$</th>
<th>Ratio (%)</th>
<th>Reference</th>
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<td>16.1 ± 2.1</td>
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<td>18.7 ± 3.2</td>
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<td>17</td>
<td>Tzompa-Sosa et al. (2017)</td>
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<td>10.2 ± 0.2</td>
<td>Gilman (2017)</td>
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<table>
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<th>$\Delta NH3/\Delta C_{CH4}$</th>
<th>Ratio (%)</th>
<th>Reference</th>
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<td>42.7 ± 11.9</td>
<td>This work</td>
<td></td>
</tr>
<tr>
<td>17 (9–30)</td>
<td>Eilerman et al. (2016)</td>
<td></td>
</tr>
<tr>
<td>~30–50</td>
<td>Yuan et al. (2017)</td>
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tracer from landfills and wastewater plants that could be measured and utilized in this study. Another possible explanation for the residual is tracer losses during transport; CH4 and C2H6 have lifetimes on the order of 8–10 years (Hodnebrog et al., 2018; Lelieveld et al., 1998; Sherwen et al., 2016) and 2 months (Hodnebrog et al., 2018; Hough, 1991), whereas NH3 has a much shorter atmospheric lifetime on the order of hours to a few days (Dentener & Crutzen, 1994; Gupta et al., 2003). Transport distances vary depending on the drying deposition velocity and height at which NH3 is located in the vertical column. Assuming that NH3 from feedlots further downwind of the measurement site has been deposited during transport, this would affect the regression parameters β0 and β1 and therefore overall source apportionment.

Altogether, the residual contribution β0 = 0.64 ppb implies that the magnitude of this error impacting the result is small because the measured ΔXCH4 varies up to 17.1 ppb.

**Data**

Data displayed in graphs are available from the following data archive: https://vokamerroup.colorado.edu/publications. DISCOVER-AQ data are available at: doi:10.5067/Aircraft/DISCOVER-AQ/Aerosol-TraceGas. SONGnex data are available at https://esrl.noaa.gov/csd/groups/csd7/measurements/2015songnex/P3/DataDownload/. MLO data are available at http://www.esrl.noaa.gov/gmd/obop/mlo/.

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