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RESEARCH ARTICLE

Estimation of methane emissions from the U.S. ammonia fertilizer industry using a mobile sensing approach

Xiaochi Zhou[*,†], Fletcher H. Passow[*], Joseph Rudek[‡], Joseph C. von Fisher[§], Steven P. Hamburg[‡] and John D. Albertson[*]

To date, estimation of greenhouse gas (GHG) emissions from the natural gas (NG) value chain have focused on upstream (production) and midstream (gathering, transmission, and storage) operations. In this study, we estimate methane emissions from an important downstream consumer of NG, the ammonia fertilizer industry, which commonly uses NG as a feedstock and a fuel for the production of ammonia and other upgraded products. Using a Google Street View (GSV) car equipped with a high-precision methane analyzer, we adopted a mobile sensing approach to measure methane mixing ratios along public roads that are downwind of the ammonia fertilizer plants. Useful data were collected from six plants, which represent >25% of the total number of U.S NG-based ammonia fertilizer plants, and use >20% of the total NG consumption by this industry. Based on the measured data, a source characterization model was applied to estimate the methane emission rates from the upwind plants. Assuming that the estimates are representative of emissions during normal operations of a plant, we calculated the NG loss rate (i.e. the ratio between NG emission rate and NG throughput). If the sampled plants are representative of the U.S. ammonia fertilizer industry, the industrial-averaged NG loss rate (± standard deviation) is estimated to be 0.34% (±0.20%), and the total methane emissions (± standard deviation) from this industry are estimated to be 29 (±18) Gigagram per year (Gg CH_4/yr) in 2015–2016. This is significantly higher than the reported methane emissions of 0.2 Gg CH_4/yr from the U.S. EPA's Facility Level Information on Greenhouse Gas Tools (FLIGHT). This study begins to fill an important knowledge gap in quantifying methane emissions along the NG value chain, and demonstrates the capability of mobile sensing for characterizing airborne emissions.

Keywords: Methane emission; Ammonia fertilizer industry; Bayesian inference; Mobile sensing

Introduction

Natural gas (NG) is considered an attractive "bridge fuel" towards clean energy due to its potential lower greenhouse gas (GHG) emissions compared with other fossil fuels (e.g. coal) (Alvarez et al., 2012). However, NG is the largest industrial source of anthropogenic emission of methane, a potent GHG with a global warming potential (GWP) of 86 and 34 on a time horizon of 20 years and 100 years including carbon-climate feedback, respectively (Stocker, 2014). Leakage of NG can happen anywhere along the value chain from production to end-use, reducing its potential short- to medium-term GHG advantage over competing fossil fuels. A study proposed a tipping point of 3.2% (2.7% using revised GWPs (Stocker, 2014)) loss of NG along the NG value chain, beyond which NG-based electrical generation will cause more net warming for some period of time than would using coal for power generation in the U.S. (Alvarez et al., 2012). Many studies have focused on characterizing methane emissions from production (Allen et al., 2013), and gathering, processing, and distribution (Lamb et al., 2015; Marchese et al., 2015; Mitchell et al., 2015) along the NG value chain. Some studies sought to estimate methane emissions (e.g. pipeline leaks) in metropolitan areas, which is dominated by local distribution and residential/commercial use of NG (Lamb et al., 2016; McKain et al., 2015; von Fischer et al., 2017). In contrast, methane emissions from industrial consumers of NG are largely unexplored. One rare example is a recent effort to quantify methane emissions from NG-fired power plants and oil refineries (Lavoie et al., 2017).

The ammonia fertilizer industry is a major industrial consumer of NG, which is used as feedstock and energy source to produce ammonia and some upgraded products (e.g. urea) (Elvers et al., 1989). In 2015, the U.S. gross production capacity of ammonia (including ammonia as a

* School of Civil and Environmental Engineering, Cornell University, Ithaca, New York, US
† California Air Resources Board, Sacramento, CA, US
‡ Environmental Defense Fund, New York, US
§ Department of Biology, Colorado State University, Fort Collins, Colorado, US
Corresponding author: John D. Albertson (albertson@cornell.edu)

feedstock for upgraded products) was >12,000 Gigagram per year (Gg NH_3/yr) (United States Geological Survey, 2017). Due to stable low NG prices resulting from the U.S. shale gas boom, the U.S. ammonia production capacity is expected to increase in the next several years (United States Geological Survey, 2017). Consequently, a large amount of NG is and will continue to be consumed in the production of ammonia fertilizer. According to the Manufacturing Energy Consumption Survey (MECS) conducted by the Energy Information Administration (EIA) (Energy Information Administration, 2017), 383 trillion British thermal units (BTU) of NG (approximately 7,700 Gg of NG) was consumed by the U.S. ammonia fertilizer industry in 2014. Ammonia fertilizer plants are large GHG emissions sources mainly because CO_2 is a major byproduct of ammonia production. However, there is less understanding of methane emissions despite the large amount of NG consumption by this industry.

In this study, we evaluate the scale of methane emissions from the U.S. NG-based ammonia fertilizer industry. Since ammonia fertilizer plants are dispersed all over the U.S., a mobile sensing approach was adopted to be able to quickly cover a representative number of plants. Using data collected with a mobile sensor, a Bayesian approach was used to characterize methane emissions from the plants surveyed. These estimates were up-scaled to infer methane emissions from the U.S. ammonia fertilizer industry.

Material and Methods

Field experiments

Two sampling campaigns were conducted, the first from Jun. 16th to 19th, 2015, and the second from Sep. 12th to 29th, 2016. Three and eight ammonia fertilizer plants were sampled, respectively, during the two campaigns. Two of the three plants sampled in the first campaign were revisited in the second campaign. As shown in **Figure 1**, the plants surveyed were mostly located in the mid-western U.S. surrounded by relatively flat terrain, resulting in few obstacles impacting plume transport. The downwind measurements were conducted without notifying plant operators with intent of encountering routine plant operation. Water vapor from cooling towers and effluent flares were observed when sampling all the plants, suggesting that they were in production.

Useful data were collected along downwind roads at six of the nine plants sampled, all during the second field campaign in 2016. These six plants represent >25% of the total of 23 NG-based ammonia fertilizer plants in the U.S. that were operating in 2015–2016. No useful data were obtained at the other three plants (two were visited during both campaigns) due to poor meteorological and road conditions, and hence were not included in the analysis. The data quality control protocol ruled out some data from the six plants surveyed (Supplemental materials Text S1). Key characteristics and sampling conditions related to the six plants that were successfully surveyed are summarized in **Table 1**.

The mobile sensing approach utilizes a Google Street View (GSV) car equipped with a fast-response and high-precision methane analyzer to repeatedly measure methane mixing ratios (c, in ppm) along public roads downwind of the ammonia fertilizer plants. Here, we only offer a brief description of some components of GSV car that are relevant to this field campaign, and more detail on the GSV car can be found elsewhere (von Fischer et al., 2017). A Picarro cavity ringdown analyzer (Model G2301

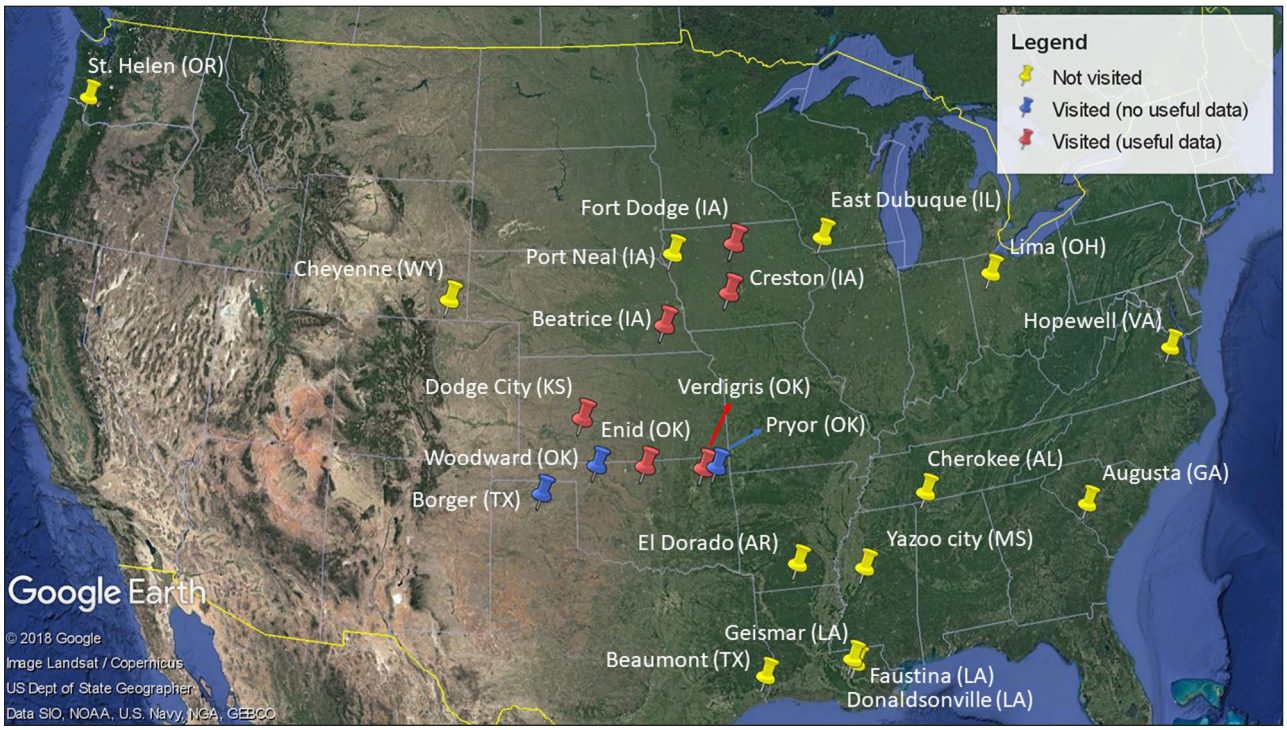

Figure 1: Map of NG-based ammonia fertilizer plants in the U.S. A Google Earth representation of NG-based ammonia fertilizer plants in the U.S that were operating in 2015–2016. DOI: https://doi.org/10.1525/elementa.358.f1

Table 1: A summary of information about the sampled ammonia fertilizer plants, including facility location (latitude and longitude), sampling date and time, number of valid mobile passes (N), plume travel distance (x_s), and local meteorological conditions during the sampling period, such as the mean stream-wise wind velocity ($\bar{u}$), and the mean wind direction (θ_m) clockwise from the north. DOI: https://doi.org/10.1525/elementa.358.t1

Facility location (Latitude, Longitude)	Date [MM/DD/YY]	Local time [HH:MM]	N [-]	x_s [m]	$\bar{u}$ [m/s]	θ_m [deg]
Verdigris, OK (36.2338, –95.7190)	09/12/16	10:42–14:48	52	1730	2.90	164
Enid, OK (36.3794, –97.7624)	09/14/16	10:24–14:33	45	350	2.80	8.7
	09/15/16	10:02–12:38	48	1230	3.84	184
Dodge City, KS (37.7777, –99.9299)	09/20/16	16:16–18:58	8	2170	5.97	183
	09/21/16	10:19–13:49	15	1880	5.41	196
	09/22/16	10:17–12:01	14	1800	5.67	195
Beatrice, NE (40.3202, –96.8401)	09/26/16	09:47–12:04	24	590	2.50	291
Creston, IA (41.1170, –94.3551)	09/26/16	17:41–18:41	16	650	2.56	305
	09/27/16	10:38–13:08	37	590	3.51	304
	09/27/16	14:38–15:38	12	730	3.73	291
Fort Dodge, IA (42.4992, –94.0171)	09/28/16	13:08–15:38	34	1400	3.43	355
	09/29/16	10:52–12:22	5	1440	2.77	352
	09/29/16	14:52–16:52	9	1410	3.12	347

from Picarro Inc., Santa Clara, CA, USA) configured to sample at 1 Hz was used in the first sampling campaign, and a fast methane/ethane gas analyzer (Los Gatos Research, Inc., San Jose, CA, USA) sampling at 2 Hz was used in the second sampling campaign. We did not re-calibrate the Los Gatos Research (LGR) fast methane/ethane gas analyzer during deployment. After initial factory calibration, the instruments largely operate on a self-calibrating basis, accounting for variation in performance of the analytical cell using a wavelength that was not absorbed by any gases. Otherwise, the analysis is derived from first-principles (cell dimensions, pressure, temperature, and methane's permolar absorptivity) (Gupta, 2012). Together, these concentration measures yield methane values that are accurate to better than 60 ppb (Robert Provencal, LGR, personal communication, 04/16/2019), which is relatively small considering that our methane data vary from ~2.0 ppm (ambient level) to ~30 ppm (peak mole fraction). Though ethane concentration were also measured by the gas analyzer, it was not used for analysis since the concentrations were low and the noise was high, resulting a low signal to noise (SNR). The inlet of the methane analyzer was located at the front bumper of the GSV car (~30 cm above-ground) to avoid measuring exhaust of the car. Real-time location of the GSV car was determined by a roof-mounted GPS unit (Model A100 from Hemispheres GNSS, Scottsdale, AZ, USA) with an acquisition frequency of 1 Hz. A portable 3-D sonic anemometer (Model 81000 from R.M. Young Co., Traverse City, MI, USA) was installed near each visited fertilizer plants in a relatively flat and open location to measure local meteorological conditions (**Table 1**). The height of the anemometer was ~1.5 m above the ground.

During the sampling, the GSV car first circled around the plant to be surveyed to map the presence of elevated methane mixing ratios, and to determine whether the pattern of emissions was clear enough such that attribution of any emissions could be made based on wind direction (similar to triangulation). An example is shown in **Figure 2**, a well-developed methane plume was observed downwind of the ammonia fertilizer plant, while ambient background methane mixing ratios were found when sampling upwind of the plant. After identifying the facility-introduced plume, the GSV car would pass through the plume multiple times to make repeated samplings. Local emissions, such as small pipeline leaks, can be identified as small spikes in the data (**Figure 2**). In contrast, emissions from the ammonia plant (sampled approximately 500 to 2,000 m downwind as shown in **Table 1**) are characterized by a broad plume (**Figure 2**). In the data post-processing, elevated mixing ratios due to local emissions were excluded from the analysis, and we focused on the broad plumes. A low-pass moving average filter was established based on the low frequency drift of the internal sensor pressure, which is likely caused by partial clogging of the sensor inlet due to dust. The filter was later used to mask and adjust the raw mixing ratio data to remove the adverse effects caused by this issue. The background methane mixing ratio (c_b) was estimated as the 5th percentile of the ranked time series of raw methane mixing ratios (c_r) (Albertson et al., 2016; Brantley et al., 2014), and the above-ambient mixing ratios $c = c_r - c_b$. The estimated c_b is very close to the ambient methane mixing ratios measured upwind of the plant, suggesting that the determination of c_b is robust.

Emission characterization using a Bayesian approach

Using data collected during multiple sensor passes, a Bayesian approach was adopted to characterize methane emission from the ammonia fertilizer plants. This

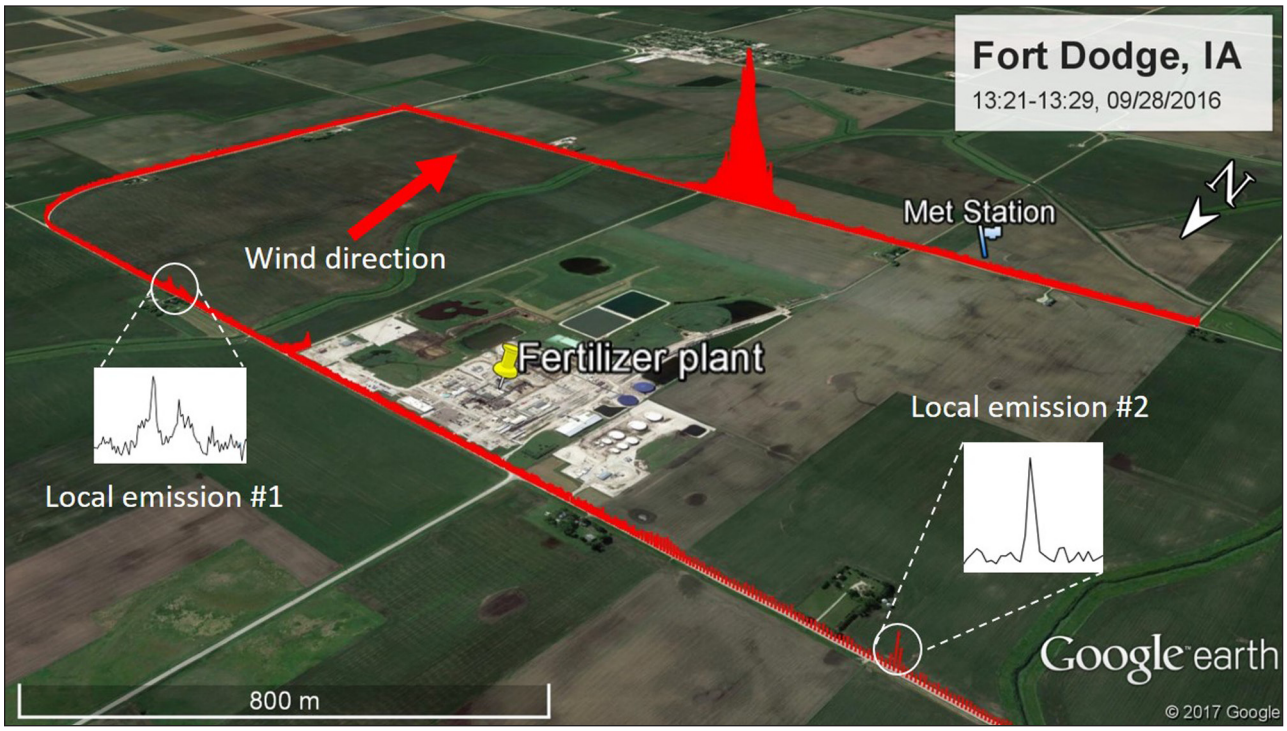

Figure 2: An example of downwind methane plume measurement. A Google Earth representation of an example of methane mixing ratios (red bars) measured by the GSV car along its pass around the ammonia fertilizer plant located in Fort Dodge, IA. Small methane spikes, which may be caused by unground pipeline leaks and not related to emissions from the fertilizer plant, are identified as local emissions. DOI: https://doi.org/10.1525/elementa.358.f2

approach is fully described in (Albertson et al., 2016); here only a brief introduction is included. Following Bayes's rule, the posterior probability density function (PDF) of the emission rate Q [kg/h] is (Albertson et al., 2016; Yee, 2008; Yee, 2012):

$$P(Q|c_y, I) = \frac{P(Q|I)P(c_y|Q, I)}{P(c_y|I)}, \quad (1)$$

where c_y (in ppm × m) is the cross-plume integrated methane mixing ratio. Practically, c_y can be estimated as $c_y = \sum c\Delta x$, where Δx [m] is the distance between the geo-referenced mixing ratio data. I is the ancillary information, including source information and the prevailing meteorological conditions. $P(Q|I)$ is the prior PDF, which represents the distribution of Q prior to the observation of c_y. $P(c_y|Q, I)$ is the likelihood function, which is the probability of observing c_y given Q and I. $P(c_y|I)$ is the evidence term that simply ensures that $P(Q|c_y, I)$ integrates to unity.

Before making any measurement, the prior ($P(Q|I)$) is unknown. Thus, a uniform distribution is adopted as a non-informative prior (Yee, 2007). After the first sensor pass (with a valid measurement of c_y), equation (1) is updated recursively such that $P(Q|I)$ is replaced by the posterior PDF ($P(Q|c_y, I)$) derived from the previous sensor pass (Albertson et al., 2016).

$$P(Q|I) = \begin{cases} 1/(Q_{max} - Q_{min}), & j = 1 \\ P(Q|c_y, I)_{j-1}, & j > 1 \end{cases}, \quad (2)$$

where j is a counter for successive sensor passes, Q_{max} and Q_{min} are the upper and lower bound of Q.

All the information provided by the measurement c_y about the unknown Q is contained in the likelihood function. Following (Yee, 2008; Yee, 2012) and (Albertson et al., 2016), the likelihood function is formulated as:

$$P(c_y|Q, I) = \frac{1}{\sqrt{2\pi}\sigma_e} \exp\left(-\frac{1}{2}\left(\frac{c_y - c_y^M(Q)}{\sigma_e}\right)^2\right), \quad (3)$$

where $c_y^M(Q)$ is the modeled c_y as a function of the candidate emission rate Q. σ_e [-] is the "error term" of the likelihood function, which is a measure of the uncertainty when comparing the modeled $c_y^M(Q)$ against the measurement c_y. The parameterization of σ_e is detailed in the Supplemental materials Text S2. When the vehicle's path is perpendicular to the wind direction, the effect of plume lateral dispersion vanishes and the modeled $c_y^M(Q)$ can be simplified as $c_y^M(Q) = \frac{Q}{\bar{U}} D_z$, where $\bar{U}$ is the plume advection speed, and D_z accounts for the plume vertical dispersion (Albertson et al., 2016). A common form of $D_z = \frac{A}{\bar{z}} \exp[-(\frac{Bz}{\bar{z}})^s]$, where z [m] is the height of the sensor inlet, $\bar{z}$ [m] is the mean plume height, and A [-], B [-], and s [-] are all empirical parameters of atmospheric stability and $\bar{z}$ (Gryning et al., 1987). In the case of a non-perpendicular sensor pass, $c_y^M(Q)$ can be calculated by numerically integrating along the vehicle's path (Albertson et al., 2016).

By updating the prior term $P(Q|I)$ with the posterior ($P(Q|c_y, I)$) derived from the previous sensor pass, $P(Q|c_y, I)$

is calculated recursively to incorporate data collected after each pass. After the final sensor pass (the N^{th} pass), the expected emission rate (Q_e, in kg/h) and the associated uncertainty (σ_Q, in kg/h) can be quantified as the expectation and standard deviation of the final posterior PDF $P(Q|c_y, I)$.

This approach considers the part of the facility that is related to NG-consumption (e.g. chemical reactors and boilers) as a point source, and the emissions are continuous and quasi-steady (i.e. Q is quasi-steady). The point-source assumption is based on the observation that the NG-related equipment is usually clustered within a radius of ~100 m from the center region (as visually inspected from Google Earth), while the source-to-sensor distances are 500 to 2,000 m (**Table 1**). We considered the point-source assumption as an added uncertainty in the source characterization model, and explicitly included it in the σ_e parametrization (Supplemental materials Text S2). The assumption of continuous and quasi-steady emissions comes from the premise that NG emissions originate from the chemical production and associated fuel combustion processes. These processes are often continuous and quasi-steady under normal operational conditions of typical chemical plants in order to maximize production efficiency (Green and Perry, 1973). Based on this assumption, the estimated Q_e and σ_Q (in the unit of kg/h) can be converted into the unit of Gg CH_4/yr, considering 340 days per year of effective production (i.e. excluding planned maintenance and normal outrages) as suggested by previous reports and surveys (Paul et al., 1977; The Fertilizer Institute, 2006; United States Geological Survey, 2017).

NG throughput of ammonia fertilizer plants

To relate methane emissions with operational characteristics of the ammonia fertilizer plants, we estimated NG throughput (T, in Gg/yr), which is the rate of NG consumption by the plant. For most ammonia fertilizer plants in the U.S., information about NG throughput is not publicly available. Therefore, we estimated T based on the known energy balance for the chemical reactions. Here, we assess the sources of NG-consumption for manufacturing fertilizer products at a typical ammonia fertilizer plant that uses NG as feedstock (**Table 2**).

The production of ammonia uses NG both as a feedstock and as an energy source. Ammonia is produced through the modified Haber-Bosch process, which refers to the reaction of nitrogen and hydrogen at a ratio of 1:3 under elevated temperature, pressure and the presence of catalyst (Wood and Cowie, 2004). The nitrogen is supplied from air, and the hydrogen is obtained in almost all plants in the U.S. from steam reforming of NG (United States Geological Survey, 2017). Meanwhile, NG is also used as an energy source to sustain this highly energy intensive process (Elvers et al., 1989).

The production of urea uses ammonia as a feedstock, and NG as an energy source. Urea is produced based on the Basaroff process, which includes the synthesis of carbamate from ammonia and CO_2, and the dehydration of carbamate to urea and water (Worrell et al., 2000). The synthesis of carbamate is an exothermic reaction and requires little energy. However, the dehydration step often uses NG as an energy source.

Using ammonia and urea, fertilizer plants can produce other products (**Table 2**). For example, nitric acid (NA) and ammonium nitrate (AN) are produced by exothermic reactions, presumably requiring little energy from NG (Worrell and Blok, 1994). Urea ammonium nitrate (UAN), diesel exhaust fluid (DEF), urea liquor, and aqua ammonia are solutions of ammonia, urea, and NA in water. Therefore, their production only consumes a negligible amount of energy compared to the production of ammonia and urea. Thus, we assumed that the majority of NG consumption at ammonia fertilizer plants is as a feedstock for ammonia production and as an energy source for ammonia and urea production.

Given the above-mentioned NG use in ammonia fertilizer plants, we estimated T based on the energy balance:

$$T = \frac{C_N \times E_N + C_U \times E_U}{E_{NG}} \times R_c \tag{4}$$

where C_N [Gg NH_3/yr] and C_U [Gg Urea/yr] are the annual gross production capacities of ammonia and urea production capacity, respectively. C_N and C_U include ammonia and urea both sold directly in the market and produced

Table 2: A list of major chemical products, and their feedstock and energy source in a typical NG-based ammonia fertilizer plant in the U.S. (N/A = not applicable). DOI: https://doi.org/10.1525/elementa.358.t2

Chemical Produced	Feedstock	Energy source
ammonia	NG, N_2	NG
urea	ammonia, CO_2	NG
nitric Acid (NA)	ammonia, O_2, water	Exothermic
ammonium Nitrate (AN)	ammonia, NA	Exothermic
urea Ammonium Nitrate (UAN)	ammonia, NA, urea, water	N/A
diesel Exhaust Fluid (DEF)	urea, water	N/A
urea liquor	urea, water	N/A
aqua ammonia	ammonia, water	N/A

as a feedstock for upgraded products (**Table 2**). Note that both C_N and C_U are adjusted for planned maintenance and normal outages; hence they represent production under scheduled operating conditions. E_N and E_U are the energy demands for ammonia and urea production, respectively, in Terajoule per Gigagram of product produced (TJ/Gg). E_{NG} [TJ/Gg] is the energy produced by NG as a fuel, which is considered as 52.23 TJ/Gg (Boundy et al., 2011). Following U.S. convention, E_N, E_U, and E_{NG} are all expressed as higher heating value (HHV). R_c [%] is the capacity utilization rate, which is the percentage of a facility's production capacity that is actually used.

Given that most of the data were collected in the 2016 field campaign (**Table 1**), we obtained plant-specific C_N and C_U values from 2016 annual reports published by some operating companies (AdvanSix Inc., 2017; Agrium Inc., 2017; CF Industries Holdings Inc., 2017; LSB Industries Inc., 2017; OCI N. V., 2017; Potash Corporation of Saskatchewan Inc., 2017; The Mosaic Company, 2017). When 2016 annual reports were not publicly available (e.g. operated by privately held companies), C_N and C_U were obtained from the 2016 mineral yearbook published by the United States Geological Survey (USGS) (United States Geological Survey, 2017) and the International Fertilizer Association (IFA) (International Fertilizer Industry Association, 2015), respectively. The full lists of C_N and C_U are reported in the Supplemental materials Text S3.

E_N, E_U, and R_c depend on the manufacturing technology and operational performance, which can vary considerably among plants. E_N and R_c are reported by only a few plants. For the rest, E_N and R_c are considered as random variables, whose upper and lower limits are defined by the max and min of the reported E_N and R_c, respectively. This is partly validated by the industrial-averaged values of E_N and R_c reported in the literature (Supplemental materials Text S3). To the knowledge of the authors, plant-specific E_U values are not publicly available. Therefore, we used an industrial-averaged E_U of 2.8 TJ/Gg (Worrell et al., 2000) for all plants. Note that the industrial-averaged E_U of 2.8 TJ/Gg was reported for some U.S. plants in 1994 (Worrell et al., 2000), and it is expected to be lower today than it was in the early 1990's due to the technology advancement. However, we find that the estimation of T is not sensitive to the selection of E_U. This is because the industrial-averaged E_U is more than an order of magnitude smaller than the industrial-averaged E_N (i.e. 37.9 TJ/Gg (International Energy Agency, 2007)), and C_U is usually smaller than C_N. Therefore, the use of the industrial-averaged E_U of 2.8 TJ/Gg only introduces a small possible error in the NG throughput calculation (equation 4).

As discussed above, we considered the main sources of uncertainties in calculating T from the determination of E_N and R_c. Based on annual reports of operating companies, plant-specific E_N (from 34.7 to 40 TJ/Gg) and R_c (from 0.54 to 1.06) values were estimated for four and six plants, respectively (Supplemental materials Text S3). Meanwhile, an industrial-averaged E_N of 37.9 TJ/Gg and R_c of 0.8 were obtained from the International Energy Agency (IEA) and USGS reports (International Energy Agency, 2007; United States Geological Survey, 2017). Considering the limited number of plant-specific E_N and R_c values and the associated uncertainties, we decided to construct the "best", "worst", and "nominal" cases when estimating T. The best (worst) case for T, which implies the least (the most) amount of NG consumption by a plant, was calculated using the lower (upper) bound of the reported values for E_N and R_c in equation (4). Meanwhile, the nominal case for T was estimated using the industrial-averaged E_N and R_c. The full list of gross ammonia production capacity (C_N), urea production capacity (C_U), and the estimated NG throughput under nominal (T_N), worst (T_W), and best (T_B) case for NG-based ammonia fertilizer plants in the U.S. in 2015–2016 is provided in the Supplemental materials Text S3.

Upscale from the sampled ammonia fertilizer plants to the whole industry

To upscale the estimated methane emission rates from the sampled fertilizer plants to the whole industry, we first calculated the loss rate (LR, in %) of the sampled plants following (Zavala-Araiza et al., 2015). LR is defined as the ratio between the NG emission rate and the NG throughput. Considering that around 95% of NG is methane (Lavoie et al., 2017), LR can be estimated as:

$$LR = \frac{Q/0.95}{T} \times 100\% \tag{5}$$

Given the best, worst, and nominal case for T estimation, we can calculate the corresponding loss rate for the sampled plants by plugging in different T values in equation (5). Since T is in the denominator, the best case of LR (least amount of lose rate) will respond to the worst case of T (most amount of NG throughput). For each case, the uncertainty of LR only comes from the uncertainty of Q.

Assuming that these plants constitute a representative sample of the ammonia fertilizer industry, we constructed the best, worst, and nominal cases for loss rate representing the ammonia fertilizer industry, given the best (LR_B), worst (LR_W), and nominal (LR_N) loss rate for the six sampled plants. More specifically, the best case for loss rate of the whole industry will be the lowest LR_B among the sampled plants (i.e. best of the best): $LR'_B = \min(LR_B)$, and the worst case will be the highest of LR_W (i.e. worst of the worst): $LR'_W = \max(LR_W)$. The nominal case will be the averaged LR_N: $LR'_N = \overline{LR_N}$.

We believe the plants surveyed constitute a representative sample of the whole industry based on three statistics:

- The six sampled plants represent more than 25% of the total of 23 U.S. NG-based ammonia fertilizer plants that was operating in 2016.
- Total NG throughput (T) of the sampled plants was more than 20% of the total NG throughput of all NG-based ammonia fertilizer plants in the U.S. If we exclude the plant located in Donaldsonville, LA, which is estimated to be the largest NG consumer of all NG-based ammonia fertilizer plants in the U.S., NG throughput of the sampled plants was around 30% of the remaining industry.

- The sampled plants cover a representative portion of all ammonia fertilizer plants when ranked according to NG consumption, including both large (Verdigris, OK ranked the 2nd) and small (Creston, IA ranked 23th) plants.

With the assumption that 95% of NG is methane (Lavoie et al., 2017), the methane emissions from the ammonia fertilizer industry (Q^I) under normal operational conditions are:

$$Q^I = 0.95 \times LR^I \times \sum T, \quad (6)$$

where $\sum T$ is the sum of NG throughput of all NG-based ammonia fertilizer plants in the U.S. For Q^I, we will calculate three cases as well. More specifically, the best case ($Q^I_B = 0.95 \times LR_B \times \sum T_B$) and the worst case ($Q^I_W = 0.95 \times LR_W \times \sum T_W$) represent the least and the most amount of methane emissions from this industry, respectively. The nominal case ($Q^I_N = 0.95 \times LR_N \times \sum T_N$), however, is close to the "expected" methane emission scenario from this industry.

Results and Discussion

Methane emissions and loss rate from the sampled ammonia fertilizer plants

An example of $P(Q|c_y, I)$ estimate updating after each pass by the GSV car is provided in **Figure 3**, using data collected at Fort Dodge, IA from Sep. 28th to 29th in 2016. As shown in **Figure 3a**, it is evident that the shape of the $P(Q|c_y, I)$ "sharpens" after each additional measurement, from a relatively flat PDF after the first pass (black dashed line) to a much narrower and more defined peak after the final pass (black solid line). With the distribution of Q estimated from the Bayesian method (i.e. $P(c_y|Q, I)$), we calculated the expected emission rates (Q_e) and the associated uncertainties (σ_Q). Both Q_e and σ_Q can be derived from $P(Q|c_y, I)$ after each mobile pass, and are plotted in **Figure 3b**. It is clear that both variables are reduced significantly after the first several passes, when $P(Q|c_y, I)$ are strongly affected by the prior. After ~20 mobile passes, both Q_e and σ_Q becomes fairly stable, suggesting that the Bayesian inference might have converged.

To evaluate the short-term temporal variability of the methane emissions, we calculated $P(Q|c_y, I)$ using daily data collected downwind from each plant. Despite large uncertainties for the emission rate, we find that the estimated $Q_e \pm \sigma_Q$ for the same plant using different daily data shows a good overall agreement (**Figure 4**). This observation suggests that the daily variability of emissions maybe small, which partly support the applicability of the Bayesian approach and our conjecture of continuous and quasi-steady emissions from those plants.

Inspired by the good agreement from daily results, we upscale the estimated hourly emission rates to annual emissions considering 340 days per year of effective production (excluding planned maintenance and normal outrages) as discussed above. Here, we used all the data collected in each plant to derived $P(Q|c_y, I)$, and the resulting $Q_e \pm \sigma_Q$ for the sampled plants are shown in **Table 3**. Overall, our estimates are consistently higher (i.e. more than two orders of magnitude for most plants) than the facility-reported methane emission rates in the U.S. EPA's Facility Level Information on Greenhouse Gas Tools (FLIGHT) database (United States Environmental Protection Agency, 2017a), suggesting that methane emissions may be significantly underestimated by the EPA emission inventory. This finding highlights the need for the ammonia fertilizer production industry to better understand the source of these emissions.

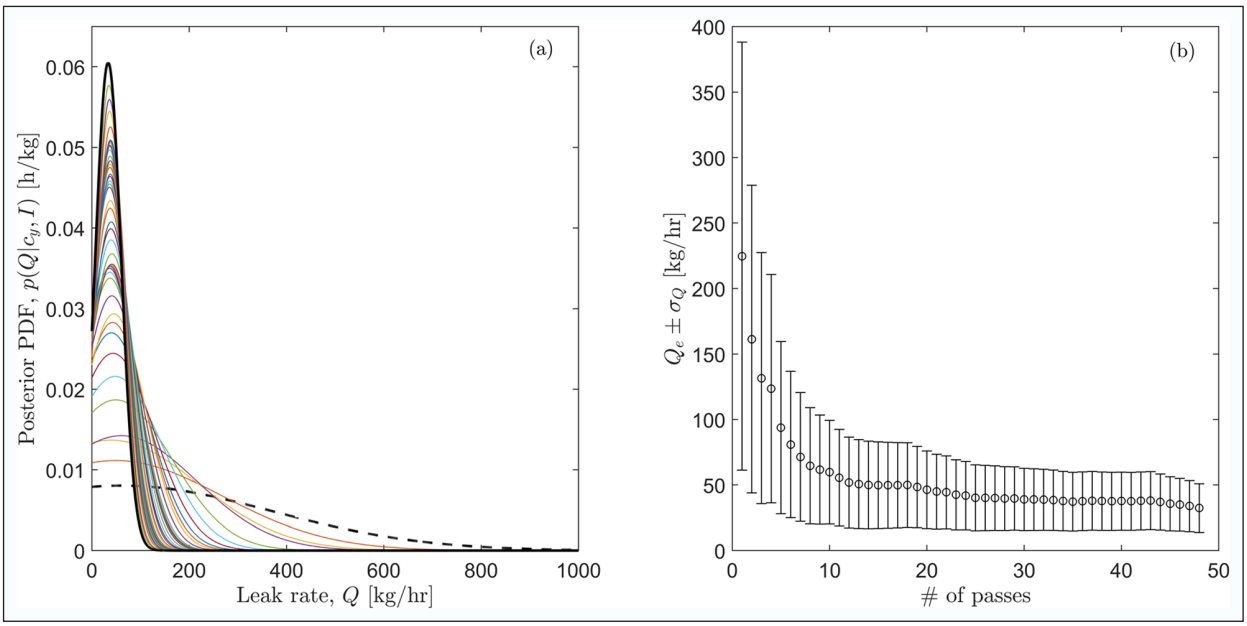

Figure 3: Emission estimates updated after each pass. (a) The posterior PDF, $P(Q|c_y, I)$, updated after each mobile pass for the plant in Fort Dodge, IA. $P(Q|c_y, I)$ after the first and last pass are highlighted in dash and bold black, respectively. **(b)** The estimated methane emission rate ± uncertainty ($Q_e \pm \sigma_Q$) derived from $P(Q|c_y, I)$ after each mobile pass. DOI: https://doi.org/10.1525/elementa.358.f3

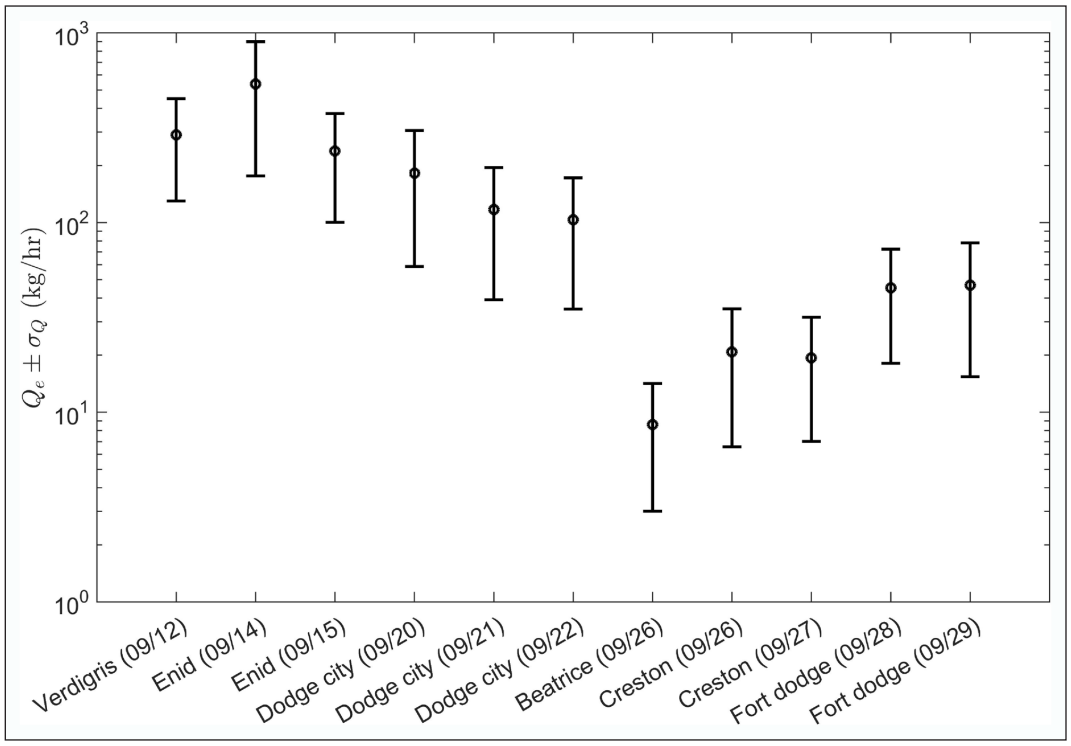

Figure 4: Emission estimates based on daily measurements. Expected methane emission rates and the associated uncertainties ($Q_e \pm \sigma_Q$), in logarithmic scale, derived from the Bayesian approach using daily data collected at each plant. DOI: https://doi.org/10.1525/elementa.358.f4

Table 3: The estimated methane emission (± standard deviation), $Q_e \pm \sigma_Q$, facility-reported methane emissions (Q_f) from the U.S. EPA's Facility Level Information on Greenhouse Gases Tools (FLIGHT) database, and the estimated NG loss rate (± standard deviation) under the nominal ($LR_N \pm \sigma_{LR}^N$), the worst ($LR_W \pm \sigma_{LR}^W$), and the best cases ($LR_B \pm \sigma_{LR}^B$) for the sampled fertilizer plants. DOI: https://doi.org/10.1525/elementa.358.t3

Facility	$Q_e \pm \sigma_Q$ [kg/hr]	$Q_e \pm \sigma_Q$ [Gg/yr]	Q_f [Gg/yr]	$LR_N \pm \sigma_{LR}^N$ [%]	$LR_W \pm \sigma_{LR}^W$ [%]	$LR_B \pm \sigma_{LR}^B$ [%]
Enid, OK	213 ±118	1.74 ± 0.97	0.01	0.33 ± 0.18	0.53 ± 0.29	0.23 ± 0.13
Verdigris, OK	290 ± 160	2.37 ± 1.31	0.02	0.37 ± 0.21	0.60 ± 0.33	0.27 ± 0.15
Dodge City, KS	75 ± 45	0.61 ± 0.36	0.004	0.39 ± 0.23	0.63 ± 0.37	0.28 ± 0.17
Beatrice, NE	9 ± 6	0.07 ± 0.05	0.004	0.05 ± 0.03	0.08 ± 0.05	0.03 ± 0.02
Creston, IA	16 ± 10	0.13 ± 0.08	0.00004	0.75 ± 0.47	1.22 ± 0.75	0.54 ± 0.33
Fort Dodge, IA	32 ± 19	0.26 ± 0.15	0.005	0.13 ± 0.08	0.21 ± 0.12	0.09 ± 0.05

Despite large differences in Q_e among these plants, the estimated loss rates (*LR*) are relatively more consistent, except for the facility in Beatrice, NE (**Table 3**). This observation is congruous with our assumption that NG emissions from these plants are associated with plant operation, and thus proportional to NG throughput. The plant located at Creston, IA shows the largest *LR* among all the sampled plants. However, it is accompanied by the largest uncertainty (σ_{LR}), making it more comparable to the *LR* of other plants.

Methane emission from the ammonia fertilizer industry

Based on equation (4), the nominal, worst, and best NG consumption of all NG-based ammonia fertilizer plants in the U.S. is estimated to be 8,750, 11,389, and 6,144 Gg/yr (approximately 434, 564, and 305 trillion Btu) in 2015–2016. As a reference, the MECS reported that the ammonia fertilizer industry consumed 383 trillion Btu in 2014 (Energy Information Administration, 2017), which is within the worst-best cases and fairly close to the nominal estimate, supporting the applicability of equation (4) and the underlying assumptions. The expansion of several facilities in the 2015–2016 period (e.g. facilities in El Dorado, AR (LSB Industries Inc., 2017), Port Neal, IA (CF Industries Holdings Inc., 2017; The Mosaic Company, 2017), and Donaldsonville, LA (CF Industries Holdings Inc., 2017) may be responsible for the higher estimate of nominal NG consumption relative to the 2014 MECS data.

Using equation (5), we estimate nominal, the worst, and the best case of the methane loss rate and the associated uncertainty of the ammonia fertilizer industry ($LR^I \pm \sigma_{LR}^I$)

to be 0.34% ± 0.20%, 1.22% ± 0.75%, and 0.03% ± 0.02%, respectively. The nominal case is comparable to the loss rate estimated from other NG-related industrial sectors, for example, 0.42% for U.S. oil and gas production sites (Allen et al., 2013), 0.47% for U.S. NG gathering and processing operations (Marchese et al., 2015), and ranging from 0.1% to 0.42% for NG fired power plants (Lavoie et al., 2017).

Using equation (6), we estimate the nominal, the worst, and the best case of methane emissions ($Q_e^l \pm \sigma_Q^l$) from the U.S. ammonia fertilizer industry to be approximately 29 ± 18, 139 ± 85, and 2 ± 1 Gg CH_4/yr, respectively. Even the best case emissions (2 ± 1 Gg CH_4/yr) is significantly higher than facility reported methane emissions from the U.S. EPA's FLIGHT database (i.e. 0.2 Gg CH_4/yr) (United States Environmental Protection Agency, 2017a). Comparing against U.S. EPA's 2016 GHG inventory (United States Environmental Protection Agency, 2017b), we find that the estimated methane emissions (nominal case) from the ammonia fertilizer industry are more than three times higher than the reported methane emissions from industrial processes and product use (i.e. around 8 Gg CH_4/yr, mainly from petrochemical production). As one of a major end-user of natural gas systems, it represents about 0.2% of methane emissions from the U.S. oil and gas supply chain (i.e. 13 ± 2 Tg/yr) under the nominal case estimation (Alvarez et al., 2018). Relative to other major sectors of methane emissions in the U.S. (United States Environmental Protection Agency, 2017b), such as enteric fermentation (6.8 Tg CH_4/yr) and landfills (4.3 Tg CH_4/yr), methane emissions from the ammonia fertilizer industry are quite low. Given the small number of ammonia fertilizer plants, however, the ammonia fertilizer industry has great potential for mitigation. As a reference, nominal methane emissions from this industry is almost equivalent to that from the entire city of Indianapolis, IN, USA (29 Gg/yr), which includes emissions from >6,000 km of natural gas pipeline, >30 metering and regulating (M&R) stations, a landfill, and a wastewater treatment plant (Lamb et al., 2016).

This finding begins to fill an important knowledge gap, industrial emissions of methane from NG end-uses, in the U.S. GHG emissions inventory. These results also suggest that this industrial process is likely a significantly larger emitter of methane than currently reported in the EPA inventory.

Limitations and future directions

This study is the first attempt to evaluate the scale of methane emissions from the U.S. ammonia fertilizer industry. One of the major limitations of this study is the relatively small number of sampled plants, which constrains our understanding of the variability of NG loss rate among different plants and causes the large range between the upper and the lower bound of emissions estimation. Enlarging the sample size will be critical for reducing the estimation uncertainty. Some plants have relatively poor road access, which may mean some emissions were missed in the selected facilities and a more complete sampling of the industry may require the use of airborne measurements. Validation of the adopted Bayesian inference method by co-sampling using other methods (e.g. airborne measurements and IR camera) will be very useful for future applications of mobile monitoring. The estimation of key plant information, such as the NG throughput and operational schedule, introduced errors and access to empirical data could resolve this uncertainty. It is worth noting that the limited number of days of observations means we were unlikely to capture any temporal dynamics in emissions, which we might expect to have a large impact on overall emissions rates if the results from other sectors apply (Zavala-Araiza et al., 2017). More extensive observations are warranted and knowledge of comprehensive routine measurements by plant operators would be valuable. A relatively modest investment in systematic measurements seems likely to result in a positive return on investment given that the loss of methane to the atmosphere represents the loss of a valuable resource.

Data Accessibility Statements

The above-ambient methane mixing ratios and GPS from the GSV, the wind data from the meteorological station, and the KML files of example plume measurements for each plant are available online (https://doi.org/10.15146/R3WT2N).

Supplemental files

The supplemental files for this article can be found as follows:

- **Text S1.** Data quality control. DOI: https://doi.org/10.1525/elementa.358.s1
- **Text S2.** The determination of the error term in the likelihood function. DOI: https://doi.org/10.1525/elementa.358.s1
- **Text S3.** Natural gas throughput for U.S. NG-based ammonia fertilizer plants. DOI: https://doi.org/10.1525/elementa.358.s1

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Competing interests

The authors have no competing interests to declare.

Author contributions

- Contributed to conception and design: XZ, JR, JVF, SPH, JDA
- Contributed to acquisition of data: XZ, JVF
- Contributed to analysis and interpretation of data: XZ, FHP, JR, SPH, JDA

- Drafted and/or revised the article: XZ, FHP, JR, SPH, JDA
- Approved the submitted version for publication: XZ, FHP, JR, JVF, SPH, JDA

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